

Master Thesis in Mechanical Engineering

**Study of the impact of inorganic fillers on
the failure pattern mode in bonded joints**

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Abstract

The use of adhesively bonded joints has undergone a significant growth in the automotive, aeronautical, and aerospace industries, among many others. The reason for this growth is associated to some of the unique properties associated to this joining methodology. Modern adhesives allow for high specific strength and stiffness, characteristics which are ideal for structural applications in the transportation sector. Furthermore, their use provides uniform stress distributions and allows to easily join dissimilar materials. The combination of these characteristics allows to not only reduce the weight of the bonded structures but also allows to meet stringent safety requirements and reduce energy consumption. With this in mind, and specifically focusing on the automotive industry, ensuring that the joint's failure is cohesive rather than adhesive when critically loaded gains relevance, since in this failure mode is considered to be safer and it enables to precisely estimate the joint's strength.

This work is devoted to the analysis and prediction of the behavior of a bonded joint reinforced with inorganic fillers, regarding not only its failure mode but also the joint's strength. Moreover, the influence of introducing such particles into the adhesive in terms of its mechanical properties will be studied.

The strength and fracture toughness properties were determined through bulk tensile and double cantilever beam (DCB) tests were performed at quasi-static speed. These tests were carried out for different amounts, in volume, of hollow glass beads.

For the study of the reinforcements' influence on the failure mode and strength of the bonded joint, single lap joint (SLJ) tests were performed, at a quasi-static speed, for different amounts, in volume, of hollow glass beads. The load vs displacement curves were extracted, and the corresponding fracture surfaces were thoroughly analysed.

Since the bonded joints behavior and failure modes change with the different amounts of hollow glass beads, the SLJs were modelled numerically, using the eXtended finite element method (XFEM) in Abaqus[®]. The load vs displacement curves were extracted, and the evolution of the crack's path with the different amounts of hollow glass beads were determined.

The tensile strength did not vary significantly with the increasing amounts of glass particles. However, the strain at failure and the energy release rate in mode I continuously decreased, while the Young's modulus increased.

The bonded joints' performance was found to decrease with the addition of glass beads into the adhesive layer, while the failure mode changed from adhesive to cohesive failure.

The results of the numerical simulations were validated for the SLJ tests and were found to be in good agreement with the experimental results.

Resumo

O uso de juntas adesivas teve um crescimento exponencial nas indústrias automóvel, aeronáutica, aeroespacial, entre muitas outras. Tal deve-se a algumas propriedades únicas que lhes são inerentes, e que se revelam fundamentais nessas mesmas áreas. As juntas adesivas apresentam resistência e rigidez específicas elevadas, características ideais para esse tipo de aplicações. Para além disso, as juntas adesivas geram distribuições uniformes de tensões e permitem a ligação de materiais distintos, ao contrário do que acontece com o uso de outros métodos de ligação mais tradicionais. Todas estas características permitem reduzir o peso das estruturas a ser ligadas e, ao mesmo tempo, ir de encontro aos requisitos de segurança e reduzir os consumos de combustível, cruciais no caso de aplicações na indústria aeronáutica, por exemplo. Tendo em conta tudo o que foi referido acima, e concentrando particular atenção na indústria automóvel, torna-se relevante garantir que a rotura de uma junta adesiva seja coesiva e não adesiva, uma vez que este tipo de falha é considerada mais segura, sendo um modo de rotura que permite estimar com maior precisão a resistência da mesma.

Este trabalho tem como objetivo possibilitar a previsão, com precisão, do comportamento de uma junta adesiva reforçada com reforços inorgânicos, no que toca ao seu modo de rotura e à resistência da junta. Para além disso, será estudada a influência da introdução de tais partículas na camada adesiva, nomeadamente nos efeitos que tal provoca nas suas propriedades mecânicas.

As propriedades de resistência e tenacidade à fratura foram determinadas através de ensaios de tração e de "double cantilever beam" (DCB), que foram realizados em regime quase-estático, para diferentes quantidades, em volume, de esferas de vidro ocas.

Para o estudo da influência das partículas de vidro no modo de rotura e na resistência da junta adesiva, foram realizados ensaios de juntas de sobreposição simples, num regime quase estático, para diferentes quantidades, em volume, de esferas de vidro ocas. As curvas de força vs deslocamento foram retiradas e as superfícies de fratura foram analisadas minuciosamente.

Uma vez que o comportamento das juntas adesivas e os modos de falha que lhe estão associados mudam com as diferentes quantidades de partículas de vidro, as juntas de sobreposição simples foram modeladas numericamente, através do uso do eXtended finite element method (XFEM), com recurso ao software Abaqus[®]. As curvas de força vs. deslocamento foram retiradas e a variação do modo de rotura com as diferentes quantidades de esferas de vidro foi avaliada.

A resistência à tração não variou significativamente com o aumento da quantidade de partículas de vidro. No entanto, a deformação à rotura e a energia de fratura no modo I diminuíram progressivamente, enquanto que o módulo de Young aumentou.

A resistência das juntas adesivas diminuiu com a introdução de partículas de vidro na camada adesiva, e o modo de rotura revelou uma tendência para mudar de falha adesiva para falha coesiva com o aumento da quantidade de partículas de vidro.

Os resultados das simulações numéricas foram validados para os testes de juntas de sobreposição simples e revelaram uma boa concordância com os resultados experimentais.

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1 Introduction

1.1 Background and motivation

Adhesives are emerging as an alternative mechanical fastening option in detriment to the use of bolts, rivets and welding. This is happening as a consequence of a set of valuable characteristics that are inherent to them, such as their low weight and high stiffness, the capability of joining different materials and with small thicknesses, uniform stress distributions, the possibility of automating the joining process as well as the adaptability to different designs and contours, among others [1, 2].

For these reasons, adhesives are widely used in the aeronautical, automotive, and aerospace industries [1], all of which demand excellent mechanical performance, due to representing a key tool in the search for reducing emissions and energy consumption values. Besides, these industries have very strict safety requirements, which forces adhesively bonded joints to match these requirements.

The increasing search for ways of enhancing the mechanical behavior of adhesives has led to the use of a wide range of reinforcement materials, as an attempt to improve their performances. In addition, for the industries mentioned previously, making sure that the joint's failure is cohesive rather than adhesive when critically loaded is also a key design parameter.

For the reasons stated above, the present thesis focuses on the effects of the introduction of inorganic fillers into the adhesive layer, in terms of the adhesives' properties, joint's strength and its failure mechanism. A numerical model has also been developed, serving as a tool to simulate and predict the behavior of single lap joints' with and without inorganic fillers, regarding their strength and failure modes.

1.2 Objectives

The present work has three main goals, being the first to fully understand how are the adhesives' properties are influenced by the introduction of inorganic fillers into the adhesive layer, in terms of strength and fracture energy.

The second goal is to comprehend the effects caused by adding inorganic fillers to adhesives, in terms of the bonded joint's strength and the respective failure mode.

The third goal is to implement a numerical model which is capable of simulating the failure mechanism of bonded joints with and without inorganic fillers, and to predict the corresponding joints' strength.

1.3 Research methodology

To accomplish the desired objectives of the present work, the following steps were carried out:

- A detailed literature review, with the aim of analysing the state of the art related to the effects of using reinforcements in adhesives;
- Experimental tests, specifically bulk tensile tests, double cantilever beam (DCB) tests, and single lap joint (SLJ) tests for different amounts of inorganic fillers;
- Numerical simulation of the failure mechanism of single lap joints, making use of some of the materials' properties that were collected in the experimental study.

1.4 Thesis outline

The present thesis is branched into seven chapters. This introductory chapter briefly addresses the main focuses and goals of this work, its background and motivation, as well as the experimental and numerical approaches that have been adopted.

Chapter 2 presents a general review on adhesive joints, their classification types, loading conditions, failure modes, and joint designs. The strength prediction approaches for adhesive joints are also thoroughly described. Being this thesis' main subject the effects of reinforcements in adhesives and adhesive joints, these are also reviewed regarding their different scale sizes, mechanical behavior, and the damage mechanisms that their presence may lead to. Moreover, a particular focus is given to glass microspheres, which are the inorganic fillers being studied in this work. At last, a literature review on the finite element technique adopted during this work and on similar numerical simulation approaches is made.

Chapter 3 is devoted to the experimental procedures performed to obtain the mechanical properties of the adhesives and to analyse the bonded joints' strength and failure mechanisms, for different amounts of inorganic fillers.

Chapter 4 briefly describes the numerical study of the single lap joint models and the different configurations under analysis.

Chapter 5 exhibits the appended papers along with the corresponding abstracts.

Chapter 6 summarizes the main conclusions regarding the subjects investigated in the present work and presents suggestions of additional research works to be performed as a continuation of the topics studied throughout this thesis.

2 Literature review

2.1 Survey of adhesive joints

An adhesive consists of a material that can be applied to surfaces in order to join them together and resist separation. Structural adhesives are a subset of adhesives that are able to assure the strength and the stiffness of the structure itself, being able to resist significant loads [1].

The materials to be joined are called substrates, and after being bonded, they are generally referred to as adherends. The attraction between substances, that is a result of the intermolecular forces existing between them, can be defined as adhesion [1].

The interphase can be defined as the region between the adhesive and the adherend, while the interface is the plane of contact between the surface and the two materials [1].

When compared to other mechanical fastening options, the use of adhesives has some advantages associated to it [1,2]:

- Provides uniform stress distributions, which results in a higher stiffness and load transmission, leading to reductions in cost and weight, Figure 1.
- Enables the bonding of dissimilar materials;
- Has the ability to bond thin sheets of material;
- Allows to efficiently bond plates;
- Simplifies the creation of automatic processes for adhesive's application;
- Provides flexibility in joint design;
- Allows smooth surface finishes, since there is no longer the need to use holes for rivets or bolts.

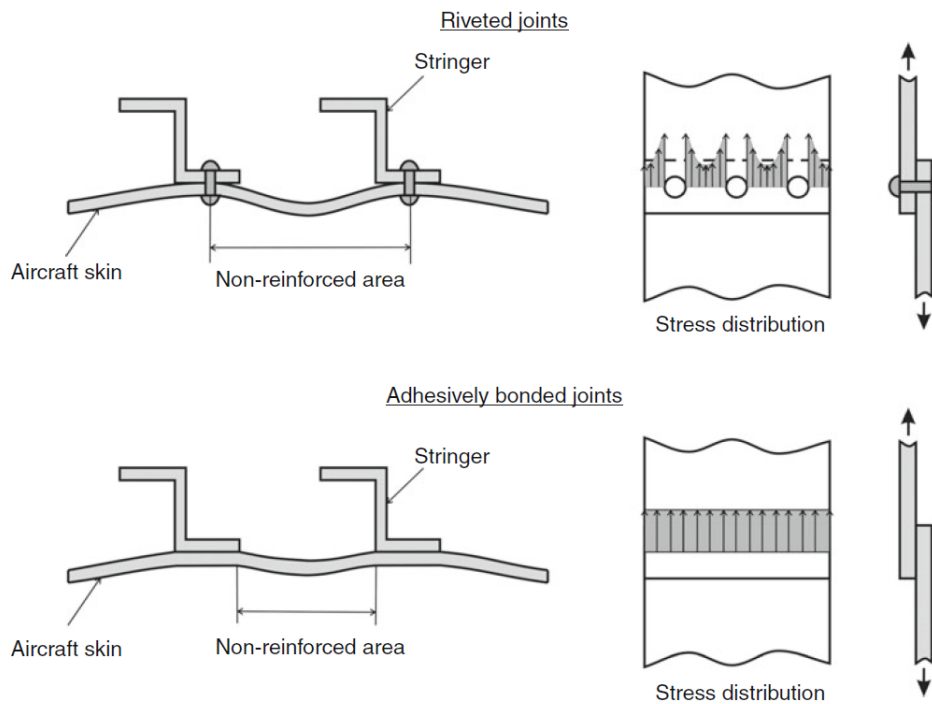


Figure 1: Comparison between riveted and adhesively bonded joints in terms of stiffness (left) and stress distribution (right) [1].

Nevertheless, the use of adhesive bonding has also some significant disadvantages, the most important of which are described below [1,2]:

- Requires the avoidance of peel and cleavage stresses, since these concentrate the load in a small area, leading to a poor joint strength;
- Has limited resistance to extreme temperature and humidity conditions, as a result of the adhesive's polymeric nature;
- Requires the use of tools to keep the substrates in position for a set period of time, since bonding is not an instantaneous phenomenon;
- The hardening of some adhesives requires high temperatures, which might represent an economic disadvantage;
- Demands careful surface preparation;
- It is often very difficult to perform quality control.

2.1.1 Classification of adhesives

Since it is important to distinguish the different adhesives and what characterizes them, there are a few main categories that are used to classify them, which are listed below [1]:

- Polymer base;
- Functionality in the polymer backbone;
- Method of application;
- Physical form;
- Chemical family;
- Functional type.

Regarding the polymer base, adhesives can have a natural or synthetic one. Natural adhesives comprise vegetable and animal-based adhesives, as well as natural gums. On the other hand, synthetic adhesives are all the adhesives which are not natural, such as epoxies and polyurethanes [1, 3].

In terms of the functionality in the polymer backbone, synthetic adhesives can be classified as thermosetting, thermoplastic, elastomeric or a combination of the previous ones (hybrid). Thermosetting adhesives are materials that cannot be heated and melted after the initial cure, which is a consequence of chemical reactions happening whether at room temperature or at high temperatures. These adhesives exist as one or two-part systems: the first one cures at high temperatures and has a limited shelf life; the second one can usually be cured at room temperatures and has longer shelf lives. Elastomeric adhesives are polymers that have a higher toughness, elongation, flexibility, and peel strength. Thermoplastic adhesives are one-component systems which are cured whether after cooling from a melted state or by evaporation of a solvent or water vehicle. These adhesives can be melted without affecting their properties in a significant way. At last, hybrid adhesives are a combination of the different adhesives that were described above [1, 3].

Adhesives can also be classified based on their method of application, which can be one of the following: coating, spraying, or brushing. Choosing one of the previous methods of application should take into account the adhesive's viscosity [1, 3].

In terms of physical form, adhesives can either be in the liquid, powder, film or paste state. The way as they are applied is usually related to their physical state. Below follows a brief description of each physical state and its characteristics [1,3]:

- Liquid adhesives are adhesives that are usually applied by the use of rolls, spraying or brushing;
- Paste adhesives are characterized by their high viscosity and little tendency to sag or grip, which allows their application on vertical surfaces;
- Film adhesives allow to have a uniform bond line thickness;
- Powder adhesives must be applied in a liquid form, which requires its heating or dissolution into a solvent.

The chemical families of adhesives can be divided into epoxies, polyurethanes, and many others. Regarding epoxy adhesives, these are probably the most industrially relevant family of structural adhesives, due to their versatility. They can be strong and brittle, or even flexible and tough, depending on their formulation. Polyurethane adhesives are frequently used in applications that call for an elevated toughened and resistance to tear and abrasion and can also be useful at low temperatures where high impact and flexibility is required [1].

Adhesives can also be classified according to their function [1]:

- Structural adhesives, which need to provide high strength and performance, since they are supposed to resist to high loading values;
- Non-structural adhesives, whose function is to hold the materials together, without needing to bear significant loads.

2.1.2 Failure modes, loading conditions and joint design

In terms of failure modes, there are mainly four types of them, being those (Figure 2) [1]:

- Cohesive failure in the adhesive, that represents the cases where the crack propagates through the adhesive and the surfaces of the adherends are still coated with adhesive. This type of failure happens when there is a strong bond between the adhesive and the adherend and the substrates are also stronger than the internal strength of the adhesive;

- Cohesive failure in the adhesive, which happens when the adhesive bond and the adhesive itself are stronger than the adherend;
- Interfacial failure, where the crack follows a path along the interface between the adhesive and the adherends. This type of failure can occur due to an incorrect surface preparation and it is usually not accepted in an industrial setting (especially in the automotive sector);
- Alternating or mixed failure, a failure mode that comprises the cases where the cracks propagate in a alternating way, varying between the adhesive and the adherend interfaces.

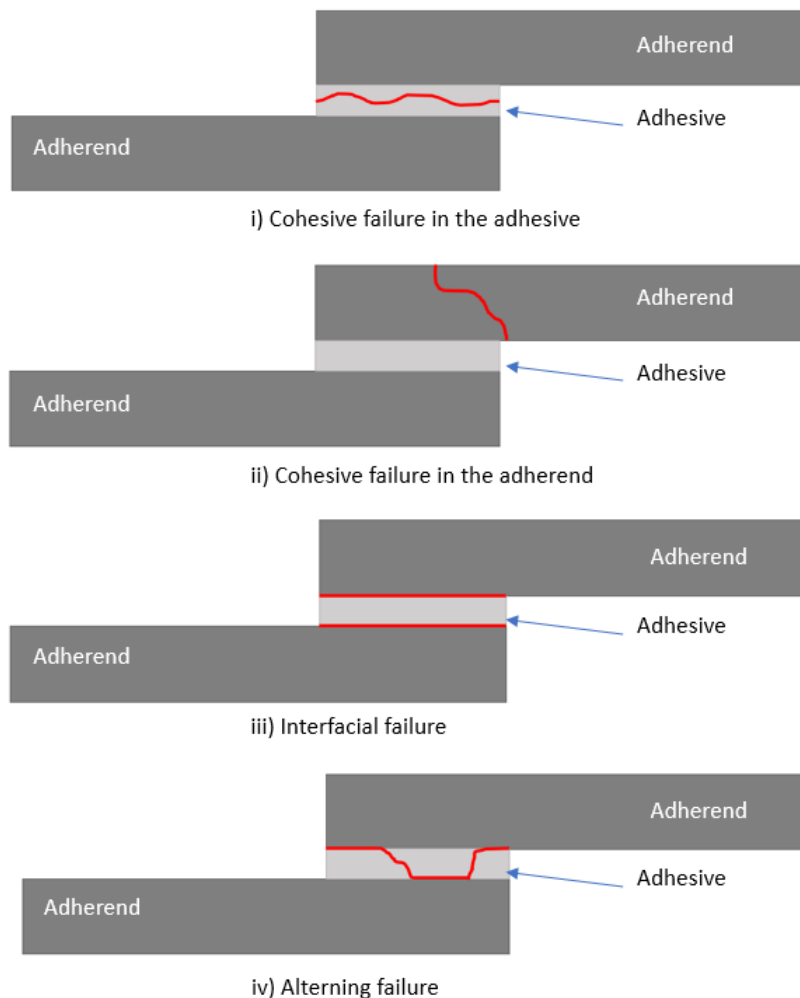


Figure 2: Schematic representation of the different types of failure.

Moreover, adhesives may be subjected to different loading conditions. Normal stresses appear when loads act perpendicularly to the joint's plane. On the other hand, shear

stresses are the ones that result from the action of loadings in the direction of the joint's plane. Finally, peel and cleavage stresses are the ones that most critically affect adhesive joints. Cleavage stresses are a consequence of loadings being applied in the ends of the bonded joint, perpendicular to it, and that tend to separate the adherends between themselves. Peel stresses are similar to the cleavage ones, but only exist when at least one adherend is flexible enough.

Finally, among the many possible configurations for bonded joints, the ones that stand out are the single lap joint, which is the simplest and the one that is studied the most, the double-lap joint, the scarf joint and the stepped-lap joint, being these represented in Figure 3 [4].



Figure 3: Most common joint configurations.

2.2 Strength prediction approaches for adhesive joints

The increasing use of adhesive joints in structural applications requires tools to better decide on the joint design and how to improve its performance. With this in mind, there are a few basic approaches that enable the characterization of adhesive joints performance and fracture.

2.2.1 Continuum mechanics

In the continuum mechanics approach, both the adhesive and the adherends are modeled using continuous finite elements, which presupposes a perfect connection between them. Doing so does not take into account the interface properties [5].

This strength prediction approach relies on the identification of deformations, stresses, and the maximum force that can be applied to the joint in the normal, shear, peel, and

cleavage loading conditions [5]. Maximum stress and maximum deformation are two different fracture criteria that are used in this matter. Yet, both fail to apply because of the singularities that are inherent to the joint tip geometry (Figure 4). Moreover, and by using the finite element analysis, the stress values increase with the mesh refinement, rather than converging, which prevents reaching the aimed solution [5].

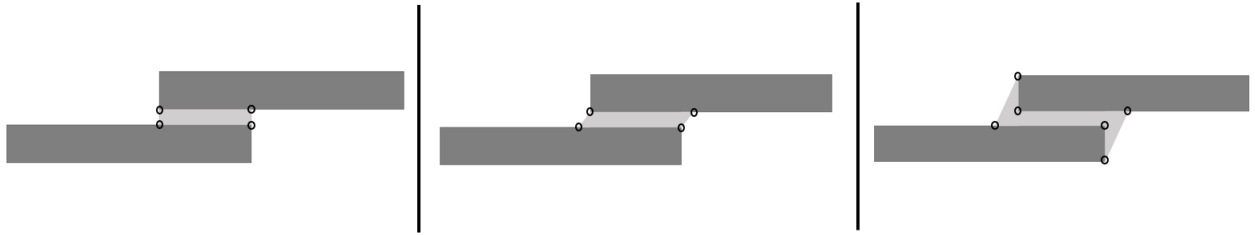


Figure 4: Singularities of the joint tip geometry.

To overcome this problem, many solutions have arisen. However, they all apply to continuous structures, and face the same difficulties when dealing with defects, or more than one material [5].

2.2.2 Fracture mechanics

Fracture mechanics, unlike continuum mechanics, considers that the structure is not continuous. On the contrary, this approach allows the presence of manufacturing defects and any other damage that may appear during its work life [5].

Not only cracks but also other imperfections behave as points of stress concentration and initiation points for the propagation of fracture and the failure of the material. Fracture mechanics analyses the size of each defect and determines if its size exceeds the critical failure size, which leads to structural failure. There are two main criteria for the study of fracture mechanics, which are the stress intensity factor criterion and the energetic criterion [5].

The stress intensity factor criterion is based on a stress intensity factor K , a scale parameter that reflects the stress state in the region near the crack tip. The fracture can generally be caused by three different loading modes. Mode I can be defined as the opening mode, while modes II and III are the modes related to shear (Figure 5). Mode II occurs when the crack surfaces move perpendicularly to the crack tip, and in mode III the crack surfaces move parallelly to the crack tip [5]. In the majority of the real situations, the load configurations lead to a combination of the fracture modes that were described.

For mode I, the stress intensity factor can be defined as follows:

$$K = Y \sigma_r \sqrt{\pi a} \quad (1)$$

where Y is a non dimensional factor, dependent on the geometry and load distribution, σ_r is the remote tension applied and a is related to the crack length. The crack will occur when the stress intensity factor, K , is equal to the material fracture toughness K_c .

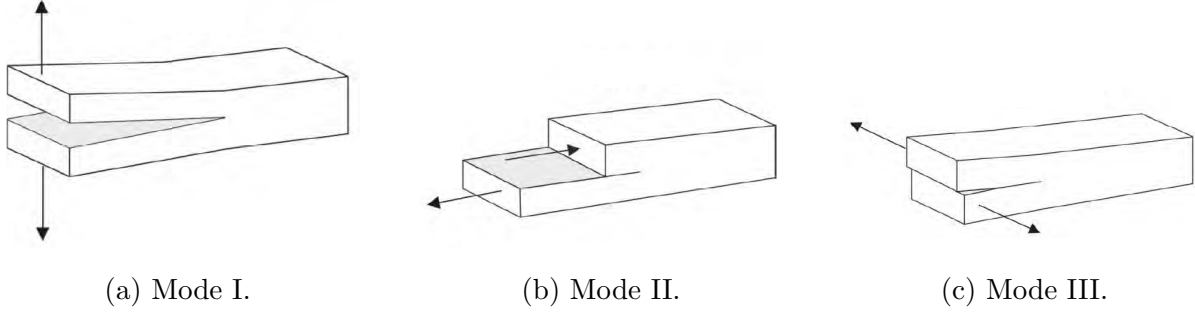


Figure 5: Fracture modes for an adhesive joint [5].

For each fracture mode, a stress intensity factor is defined, K_I , K_{II} and K_{III} for mode I, II and III respectively.

On the other hand, the energetic criterion assumes that the propagation of the crack will happen when the available energy at the crack tip reaches the energy that is needed for the crack to propagate [5].

The energetic and the stress intensity factor criteria can be related as follows, for the plane stress and the plane strain cases, respectively [6]:

$$G = \frac{K^2}{E} \quad (2)$$

where E is the Young's modulus. For the plane strain [6]:

$$G = \frac{K^2}{E} (1 - \nu^2) \quad (3)$$

where ν is the Poisson's ratio.

The energetic criterion is more commonly used since G is physically related to the crack energy absorption process. In addition, it is also very difficult to directly obtain the stress intensity factor values [5].

The mode ratio of the mixed-mode fracture can be defined as follows [5]:

$$\varphi = \tan^{-1} \sqrt{\frac{G_{II}}{G_I}} \quad (4)$$

Moreover, the energetic criteria for any type of mode combination can be defined by [5]:

$$\left(\frac{G_I}{G_{Ic}}\right)^\alpha + \left(\frac{G_{II}}{G_{IIc}}\right)^\beta = 1 \quad (5)$$

where G_{Ic} is the critical energy release rate in mode I, G_{IIc} is the critical energy release rate in mode II, and α and β are constants.

In order to obtain the strain energy release rate, there are multiple data reduction schemes that enable to collect these properties. One of the most widely used methods is the compliance-beam based method (CBBM), which was mainly developed by Moura et al. to determine the energy release rate in mode I.

CBBM defines the crack length based on the specimen's compliance, without requiring to directly measure it. This also accounts for the fracture process zone (FPZ) effects in the calculation of the fracture energy, which is defined by the existence of various micro-cracks and plasticized area ahead of the major crack which absorb some amounts of energy [7]. Figure 6 schematically represents the fracture process zone.

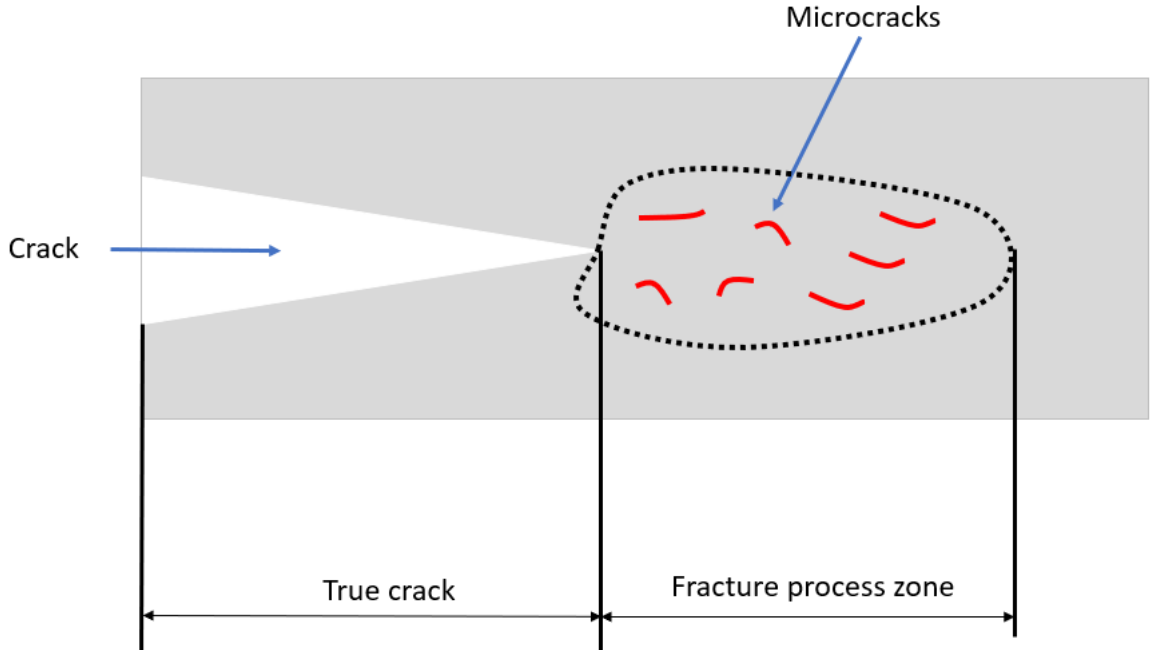


Figure 6: Schematic representation of the fracture process zone (FPZ).

The equivalent crack length, a_{eq} , is calculated based on the specimen's compliance [7]. Figure 7 is a schematic representation of the fracture process zone and the crack equivalent concept, being a the initial crack length, Δ the root rotation correction for the initial crack length and FPZ the correction to take into account the fracture process zone [8].

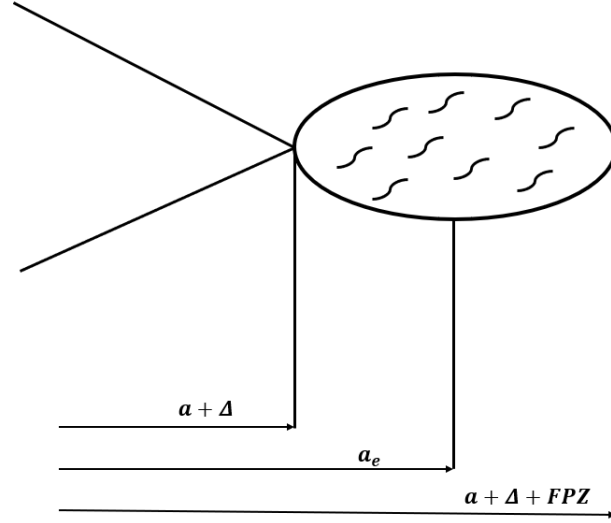


Figure 7: Schematic representation of the fracture process zone and the crack equivalent concept.

The equation below is the one used to calculate the fracture energy in mode I with CBBM [7]:

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

where P corresponds to the load, B is the specimen width, h represents the thickness of the substrates, G_{13} is the shear modulus, a_{eq} is the equivalent crack's length, and E_f refers to the corrected flexural modulus, due to the fact that CBBM does not consider the stress concentrations neither the substrates' rotation near the crack tip [7].

The equation below is used to calculate the energy release rate, in mode II [9]:

$$G_{II} = \frac{9P^2a_{eq}^2}{16B^2Eh^3} \quad (7)$$

where P is the load, a_{eq} is the length of the crack equivalent, B the specimen width, E_f is the corrected flexural modulus of the substrates and h represents its thickness.

Generally, the fact that fracture mechanics criteria are used already with the presence of defects, which are modeled as pre-cracks, makes this approach only suitable for the characterization of damage propagation, instead of damage initiation [5].

2.2.3 Damage mechanics

In damage mechanics, it is the combination of continuum mechanics and fracture mechanics that allows to model damage initiation and crack propagation, respectively.

In this approach, reaching a certain stress criterion at an integration point leads to a relaxation process. This process is based on a fracture energy criterion, resulting in the material's degradation [5].

For the study of adhesive joints, cohesive zone models (CZM) are widely used. These models are associated with interface elements, which have no thickness and assure the connection between two solid elements. The use of CZM does not require the introduction of any pre-cracks, and the crack propagation initializes independently of user intervention [5].

Putting into practice CZM requires the use of traction-separation laws to characterize the mechanical behaviour of the adhesive. For this matter, there are two main types of traction-separation laws. The triangular law is the most used one, since it is very simple to use and is generally suitable for stiff and brittle adhesives. However, the trapezoidal law can also be used, being usually more adequate for ductile adhesives. Triangular and trapezoidal traction separation laws are schematically represented in Figure 8.

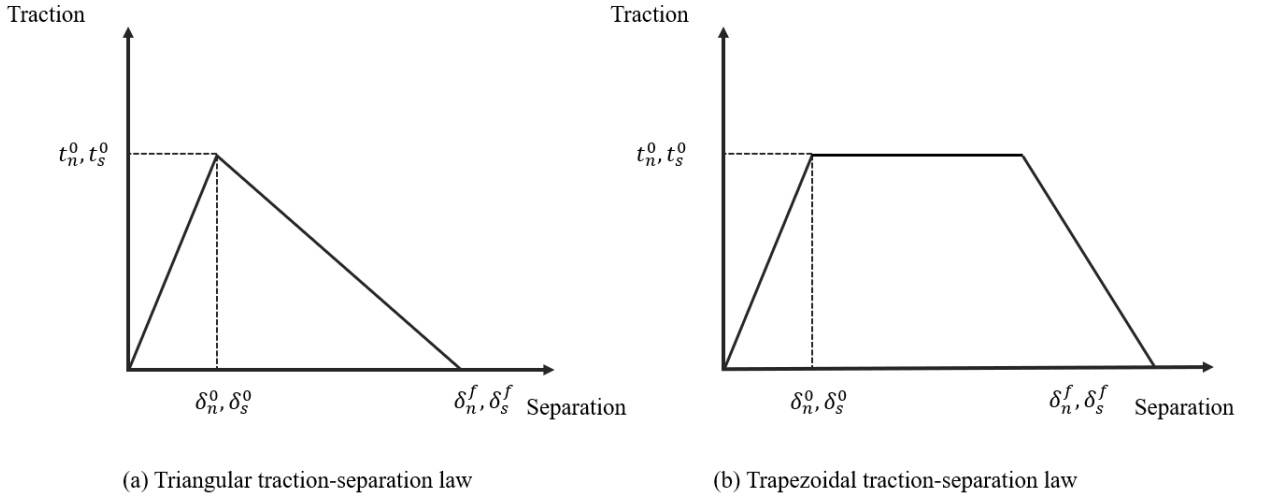


Figure 8: Schematic representation of traction separation laws.

2.3 Effect of fillers on adhesives

Since adhesive joints have a growing presence in high technology applications, many ways to improve their strength and failure behavior have been studied. Some of these methods involve varying the substrate's surface roughness, finding an optimum thickness of the adhesive layer, combining the adhesive joints with other conventional joining methodologies (hybrid joints), among others [10].

However, some of these methods are not only complex but also expensive. With this

in mind, adding fillers to the adhesive layer may appear as a more affordable and simpler solution to improve the adhesive's performance. Doing so forms what is in fact a composite material, since it becomes a combination of two or more materials [10].

Besides, and focusing on epoxy adhesives, these are usually amorphous and highly cross-linked materials, which enables them to have good structural properties. However, and since they are also a slightly brittle material, the addition of a second phase of particles, such as rubber particles, can lead to an increase in terms of toughness, without affecting other properties [11].

2.3.1 Different scales of fillers

Generally, fillers can be subdivided into three types [10]:

- Nanoscale fillers – Nanomaterials are materials that have at least one dimension below 100 nanometers. Their use as reinforcements has been proven to enhance not only the mechanical behaviour but also the thermal behavior of adhesives. This happens because of some properties of this kind of material, such as the large surface area, the high surface energy, and the reduced number of structured defects. Carbon-based nanomaterials and nano-silica are examples of groups of nanoparticles used in adhesives;
- Microscale fillers – Micro materials dimensions vary between 1 and 1000 micrometers, and their surface area to volume ratio is smaller than for nanomaterials. Thermally expandable particles and cork particles are a few types of micro materials that can be used as reinforcing fillers;
- Macroscale fillers – Macro fibers, unlike other types of reinforcing fillers, improve the adhesive's performance in terms of its curing process and electrical conductivity and provide a uniform adhesive thickness. The fibers' diameter may vary between 0.2 and 1.2 millimeters.

2.3.2 Mechanical behavior

The introduction of reinforcing fillers in adhesives has been proven to greatly influence the mechanical behavior of the structures being analyzed, more precisely the stiffness, fracture toughness, and strength [10].

Regarding the strength associated with adhesives and adhesive joints, it shows to vary with three main parameters: particle/matrix interfacial adhesion, particle size, and

content. In terms of interfacial adhesion, it can generally be said that a strong interfacial adhesion improves the tensile strength of the structure, as a result of the ability to transfer stress from the matrix to the particles. Moreover, the research work that has been done about the effect of particle size concludes that reducing it enhances the tensile strength of the composite. At last, the effect of particle content seems to be uncertain, since it depends on the two previous parameters [10].

The adhesive and adhesive joint's stiffness is shown to vary with the particle size and particle loading factor. The first one seems to provoke different variations to the stiffness, depending on the volume fraction. Generally, and if the particles have a higher modulus than the neat adhesive, increasing the volume fraction leads also to an increase in the modulus of the adhesive being reinforced [10].

Similarly, fracture toughness depends on particle size, particle-matrix interfacial adhesion, and particle loading. Regarding the particle size, its increase enhances the fracture toughness of the adhesive, due to the deflection that is caused by the bigger particles being introduced. However, this is only valid for microscale reinforcing fillers. Furthermore, the fracture toughness seems to improve with the increase of particle loading. Nevertheless, for much larger volume fractions, the agglomeration of particles leads to stress concentrations that worsen the adhesive's mechanical behavior. Finally, interfacial adhesion has also an important impact on the fracture toughness of the reinforced adhesive since well-bonded particles can perform the role of crack stoppers [10].

Barbosa et al. [12] has conducted a study focusing on the effect of cork particles on the strength of a structural adhesive. For this experiment, different amounts of cork (between 0.5 % and 5% of cork particles, in weight) were introduced into the adhesive and then tensile and single lap joints tests were performed to evaluate the variation of the adhesive's mechanical properties. This experimental study showed that, for 1% of cork particles, the adhesive became more ductile, and also presented a higher joint strength. However, increasing the amount of particles to 2 % already leads to a worse mechanical behaviour, since the particles start acting as defects.

Kinloch et al. [13] has presented an article which focuses on how the addition of nanosilica particles to a rubber toughened adhesive affects its toughness. Single-lap joint and tapered double-cantilever beam specimens were manufactured and tested to assess the behaviour of the adhesive. These particles, with an average diameter of 20 nm, led to improvements not only in terms of toughness, but also in terms of single-overlap shear strength for quantities between 1 % and 8 % in weight. This is a consequence of the interactions of the stress field around the particles and next to the crack tip, which helps

to enhance the plastic deformation of the adhesive's matrix.

Forte et al. [14] has studied the effects of adhesive reinforcement on the mode I fracture toughness of bonded joints. The study that was carried out compared the different energy release rates in mode I for different volumes of fiber-glass fibers in the epoxy resin. The results obtained showed that introducing fibers in the adhesive generally led to a significant decrease in terms of the energy release rate, when compared to the neat resin. Moreover, the Scanning Electron Microscopy (SEM) analyses allowed to observe fiber breakage for every combination of adhesive and fiber, as well as a matrix interface failure in most of the specimens.

Meng et al. [15] presented an article that compared the influence of nanosized and micrometer-sized rubber particles on the fracture toughness and strength of polymeric adhesives, for different adhesive thicknesses. For this study, the same volume fractions of 2.5 % and 5.0 % were used for both cases, and DCB, compact tension (CT) and lap shear tests were performed to evaluate the mechanical properties. The experimental results showed that nanoparticles lead to considerable improvements in terms of the fracture toughness and shear strength. This is related to the fact that, for a certain value of volume fraction, nanosized particles have shorter particle-to-particle distances and represent a higher surface area, which facilitates the energy absorption. Besides, microparticles resulted in a slight increase of the fracture toughness, depending on the adhesive thickness and the ductility of its matrix.

Kishi et al. [16] worked on assessing the differences regarding the adhesive properties of an epoxy resin toughened using polyamide-12 particles and core-shell rubber particles. This experimental study involved the manufacturing and testing of CT specimens, as well as the manufacture of specimens to evaluate the peel and the shear adhesive strength. The results obtained showed that the T-peel adhesive strength improved by increasing the amount of polyamide-12 particles. On the other hand, and by using the core-shell rubber particles as reinforcements, the adhesive's strength improved by increasing the number of particles, until reaching a plateau. Moreover, the polyamide-12 particles revealed a considerable ability to plastically deform until fracture in the peel process. This suggested the existence of a bridging mechanism that increases the adhesive's toughness, which will only occur when the reinforcement particles have a good adhesion to the matrix, the ability to absorb considerable amounts of energy, and a lower elasticity's modulus than the one relative to the adhesive's matrix.

2.3.3 Damage mechanism

Damage mechanisms can be highly influenced by the addition of fillers, which can alter the way the crack propagates along the adhesive. Below, the different effects that can be caused by the presence of second phase fillers are briefly described [10,17]:

- Bridging mechanism (Figure 9) – This mechanism can work as a way of improving the mechanical properties of the adhesive, being its effect related to the size and the geometry of the reinforcing fillers being added. Additionally, they break or pull out from one surface to another depending on the angle formed with the fracture surface and the interfacial bond strength with the adhesive [10];

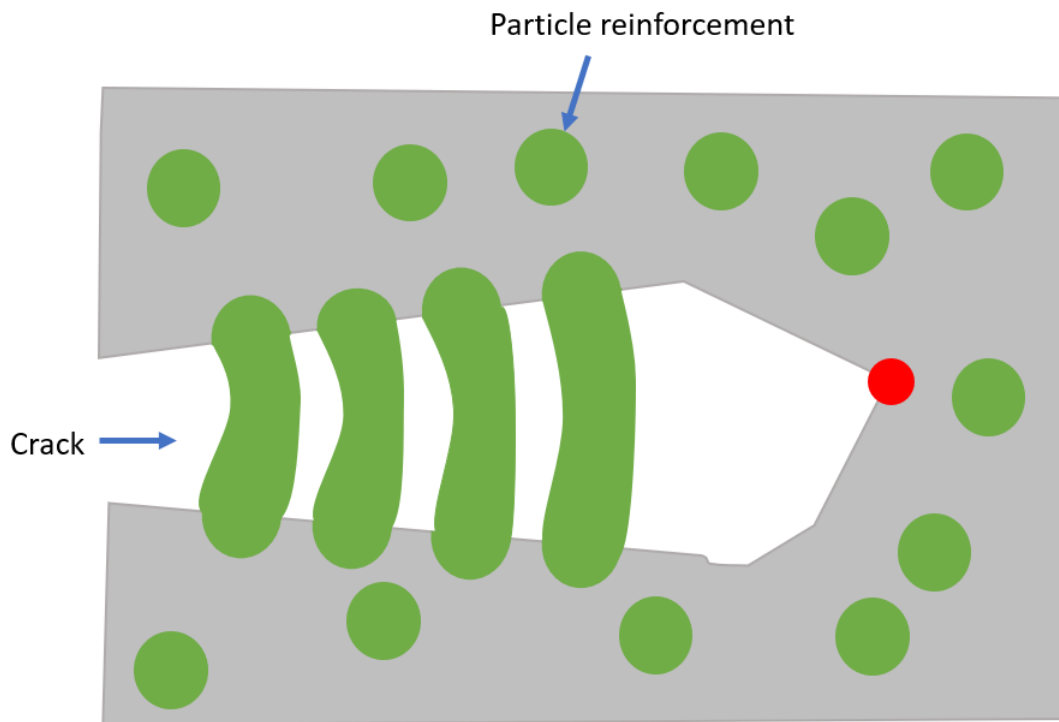


Figure 9: Schematic of bridging mechanism.

- Crack deviation mechanism (Figure 10) – The crack deviation mechanism is a result of the presence of particles in the adhesive's matrix since the crack reaches the spheres and changes its usual path. This phenomenon can both increase the fracture toughness and delay the failure of the reinforced adhesive [10];

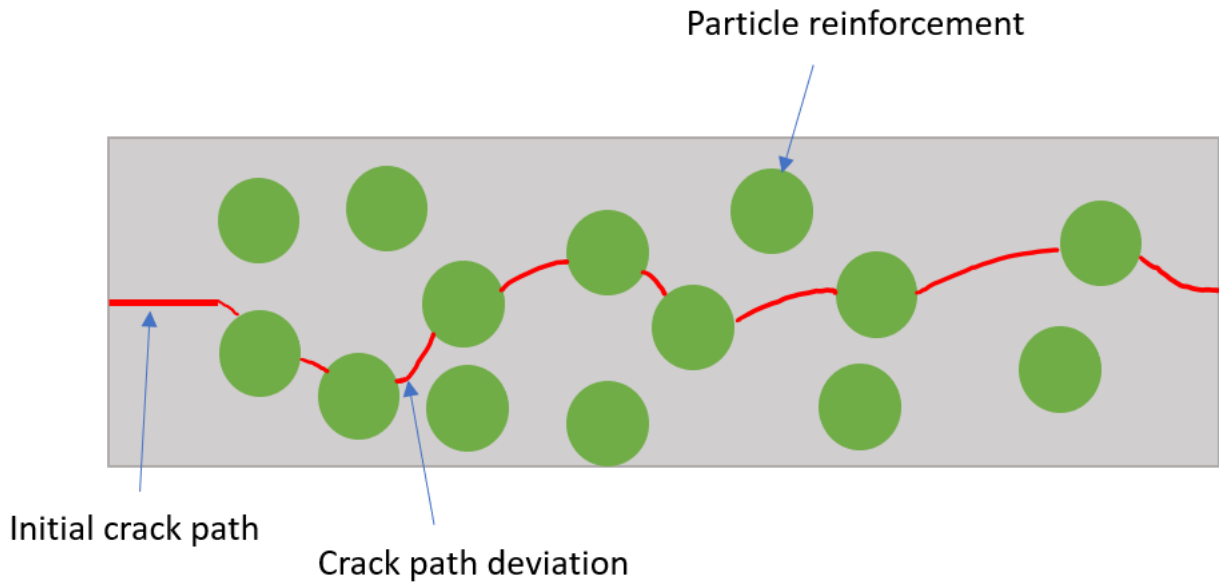


Figure 10: Schematic of crack growth deviation.

- Crack pinning and bowing mechanism (Figure 11) – This mechanism, which is thought to be more relevant for brittle materials than in ductile ones, is related to changes in the velocity and shape of the crack propagation near the second-phase particles, which act as obstacles [10]. This causes dissipation of energy along with the adhesive, since bowing out the crack front between the particles leads to improvements in terms of the fracture energy [18];

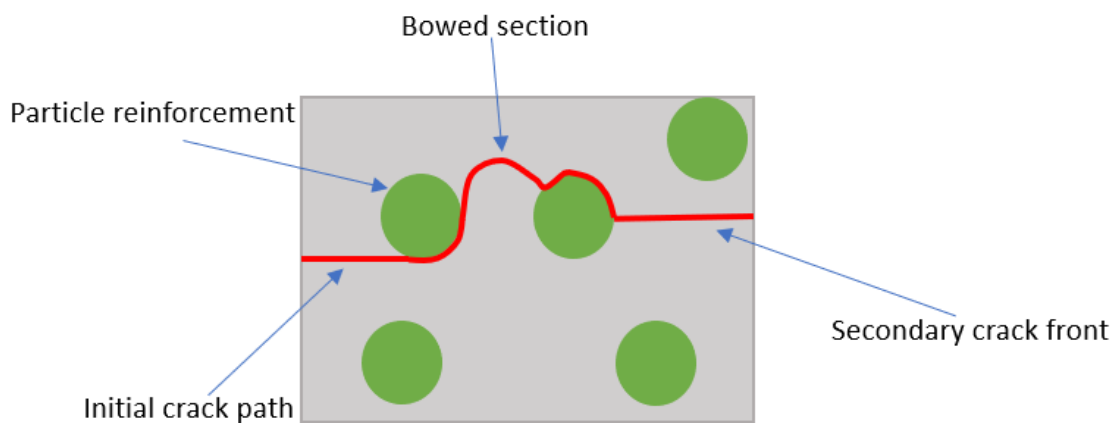


Figure 11: Schematic of crack pinning and bowing mechanism.

- Microcracking mechanism – The development of various cracks ahead of the main

crack tip, known as micro cracks, decreases both the modulus and the stress concentration near it, resulting in an improvement of the fracture toughness. This mechanism leads to the creation of new surfaces and particles debonding, and it varies with the size of the particles being used [10];

- Shear banding mechanism (Figure 12) – This mechanism results from local stress concentrations created by the reinforcing fillers and can be triggered by particle debonding and void nucleation. This decreases the stresses next to the particles and facilitates the propagation of the shear bands [10];

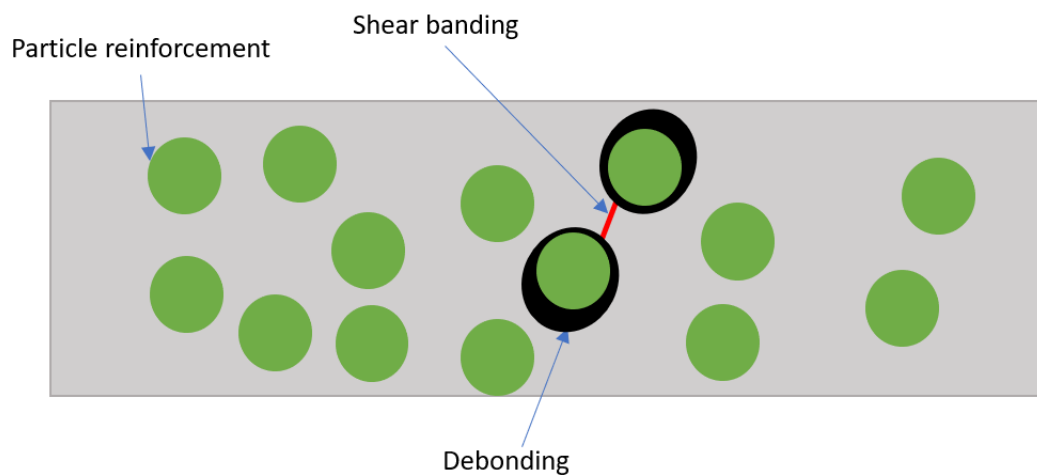


Figure 12: Schematic of shear banding mechanism provoked by particle debonding.

- Particle debonding and subsequent plastic void growth – Particle debonding occurs due to local stress concentrations at the particle-matrix interfaces. Then, local plastic void growth may appear as a consequence of the previous conditions. This leads to an increase in terms of the dissipated energy, due to the existing plastic voids [10];
- Stress fields around the particles – The presence of particles in the matrix, such as spherical rubber particles, introduces local stress concentrations around the equator of the particle. For the case of the rubber particles, and since they have the capability of bearing loads while functioning as stress concentrators, this makes them effective at toughening the adhesive [11, 19].

2.3.4 Glass microspheres

Glass microspheres consist of thin-walled hollow spheres made of soda-lime-borosilicate glass with a white powder appearance. They can vary in terms of size and density, and are known to be lightweight and to present a high strength-to-density ratio and a considerable isostatic crush strength [20,21]. Below, an example of the glass microspheres is displayed.

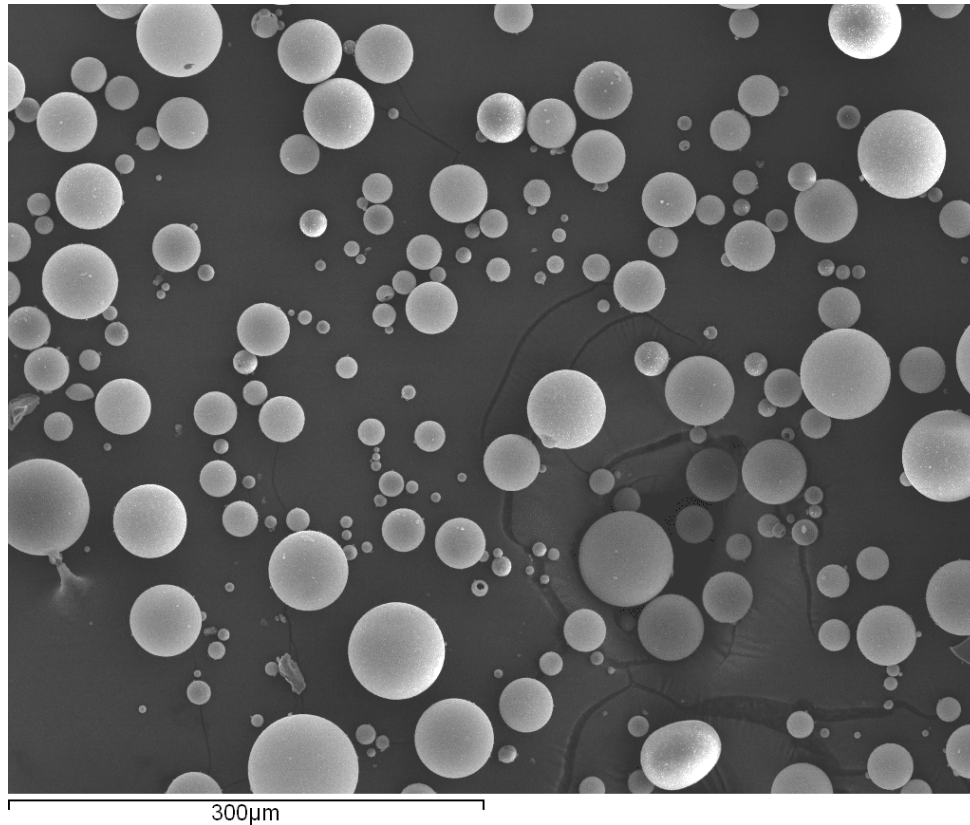


Figure 13: SEM micrograph of glass microspheres.

The association of its thin geometry with a considerable structural rigidity, along with a smooth and nonporous surface, allows hollow glass microspheres to stand out from other types of reinforcements or fillers. Besides, they are chemically inert and also water-resistant [20].

The use of hollow glass beads, when compared to other types of fillers with the same weight, allows to fill large volumes of reinforcement. For this reason, and by considering the cost per unit volume, it is safe to say that glass microspheres are a cost-friendly solution [20].

For an instance, Khalili et al. [22] studied the influence of adhesive reinforcement with glass fibers and micro-glass powder on single-lap joints with composite laminates as adherends. Focusing on the impact of using micro-glass powder, it has been concluded

that this improves the strength and stiffness of the joint. Moreover, thin-layer cohesive failure and cohesive failure were the predominant failure modes that resulted from this type of reinforcement.

According to Hunter et al. [23], who investigated how the presence of glass microspheres in the adhesive affects the mechanical adhesion of single-lap joints with fiber-reinforced polymers, its use has different effects which are dependent on how fast the adhesive is cured. For the case of the fast-curing epoxy adhesive, the joint's strength is shown to increase, for the 3% and the 10% weight concentration of glass microspheres, when compared to the neat adhesive's joint. On the other hand, for the slow-curing adhesive, the strength generally decreased for both concentrations. Regarding the failure modes, thin-layer cohesive and fiber tear failure were the most common ones for the slow-curing adhesive, while for the fast-curing adhesive, interfacial failure was also in addition to the previous modes .

2.4 Numerical simulation

2.4.1 XFEM

XFEM is a finite element formulation that was recently created as an extension to the traditional finite elements' method, having been firstly introduced by Belyschko and Black in 1999. Unlike cohesive zone modelling (CZM), it is not dependent on a predefined crack-path to properly model damage growth. Besides, it allows the crack to be modeled independently of the mesh, not requiring any kind of remeshing, and tracks its motion [24].

This method, which can be defined as a minimal remeshing finite element method, is based on the addition of discontinuous enrichment functions to the conventional finite element displacement function to account for the presence of a crack [25, 26].

In XFEM, when there is damage propagation, phantom nodes are created, which subdivide the elements that are intersected by the crack and enable the existence of discontinuities between the new elements. This allows the crack to freely propagate along an arbitrary path [24].

Figure 14 presents a representative scheme of the propagation of a crack in XFEM, and shows how this technique adds enrichment functions to account for its presence and the discontinuities that it creates. Initially, the element represented in a) had four nodes, $n_1 - n_4$, and after being crossed by the crack, four phantom nodes, $\tilde{n}_1 - \tilde{n}_4$, were added to allow the existence of the discontinuities between the new sub-elements. Therefore, the initial element was replaced by two new sub-elements, in b), being Ω_A composed of the

nodes n_1 , n_2 , $\tilde{n}_3$ and $\tilde{n}_4$, and Ω_B composed of the nodes nodes n_3 , n_4 , $\tilde{n}_1$ and $\tilde{n}_2$ [24].

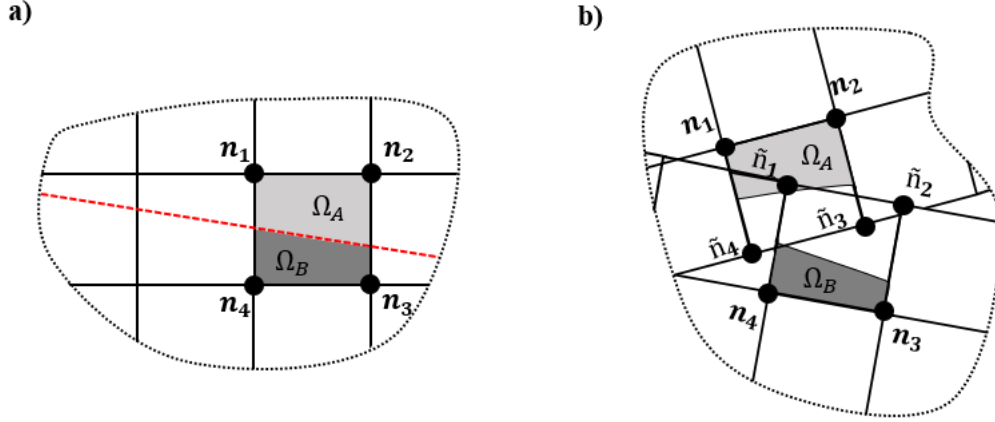


Figure 14: Representative scheme of the partition phenomenon associated to the propagation of a crack in XFEM.

Below, the equation for the displacement vector in the context of XFEM is presented, where vectors as displayed in bold lower case letters [24]:

$$\mathbf{u} = \sum_{I=1}^N N_I(x) \left[\mathbf{u}_I + H(x)\mathbf{a}_I + \sum_{\alpha=1}^4 F_{\alpha}(x)\mathbf{b}_I^{\alpha} \right] \quad (8)$$

where $N_I(x)$ corresponds to the nodal shape function and u_I represents the nodal displacement vector of the continuous part of the formulation [24].

Furthermore, $H(x)\mathbf{a}_I$, which is the generalized Heaviside enrichment function, is applied to all the nodes whose shape function are cut by the crack, as well as $F_{\alpha}(x)\mathbf{b}_I^{\alpha}$. The term $H(x)$ can be defined as the jump function or discontinuous shape function, which allows the modelling of the discontinuities across the crack over the points along the crack surface [25, 27].

$$H(x) = \begin{cases} 1 & \text{if } (x - x^*) \cdot n \geq 0 \\ -1 & \text{otherwise} \end{cases} \quad (9)$$

where x is a sample Gauss integration point, x^* is the point of the crack which is closest to x , and n represents the unit vector normal to the crack at x^* [27].

This term is multiplied by $\mathbf{a}_I$, which represents the nodal enriched degree of freedom vector [24].

Regarding the third term inside the brackets, $F_{\alpha}(x)$ is related to the asymptotic crack-tip functions, while $\mathbf{b}_I^{\alpha}$ also represents the nodal enriched degree of freedom vector [24].

XFEM can be used following two different approaches regarding the damage modelling: the cohesive segments approach; and the Linear Elastic Fracture Mechanics approach (LEFM) [24].

For the study of the adhesives' behavior, the cohesive segment approach is the one that is used, since it is governed by traction-separation-laws and the damage properties are defined as part of the bulk material definition. This approach can be followed using three stress-based and three strain-based damage initiation criteria, being the crack initiation dependent on the stress/strain value at the center of the enriched elements [24]:

- Maximum principal stress (MAXPS) and maximum principal strain (MAXPE), whose criteria are the following, respectively:

$$f = \frac{\langle \sigma_n \rangle}{\sigma_{max}^0} \quad (10)$$

where σ_n is the maximum principal stress at an integration point and σ_{max}^0 represents the material strength in terms of tension. Not only for this criterion, but also for the upcoming ones, the Macaulay Brackets are used in order to specify that a purely compressive stress does not induce damage [28].

$$f = \frac{\langle \varepsilon_n \rangle}{\varepsilon_{max}^0} \quad (11)$$

where ε_n is the maximum principal strain at an integration point and ε_{max}^0 represents the material strength in terms of deformation [28].

For these criteria, the crack initiates when $f = 1$ and its plane is perpendicular to the direction of the maximum principal stress [24].

- Maximum nominal stress (MAXS) and maximum nominal strain (MAXE), whose criteria are the following, respectively:

$$f = MAX\left\{\frac{\langle \sigma_n \rangle}{N_{max}}, \frac{\sigma_t}{T_{max}}, \frac{\sigma_s}{S_{max}}\right\} \quad \langle \sigma_n \rangle = \begin{cases} \sigma_n & \text{if } \sigma_n > 0 \\ 0 & \text{if } \sigma_n < 0 \end{cases} \quad (12)$$

where σ_n and N_{max} are the maximum nominal stress and the critical stress value in a normal-only mode at an integration point, σ_t and T_{max} are the maximum nominal stress and the critical stress value in a shear-only mode along the first shear direction,

and σ_s and S_{max} are the maximum nominal stress and the critical stress value in a shear-only mode along the second shear direction [24].

$$f = MAX\left\{\frac{\langle \varepsilon_n \rangle}{\varepsilon_n^{max}}, \frac{\varepsilon_t}{\varepsilon_t^{max}}, \frac{\varepsilon_s}{\varepsilon_s^{max}}\right\} \quad \langle \varepsilon_n \rangle = \begin{cases} \varepsilon_n & \text{if } \varepsilon_n > 0 \\ 0 & \text{if } \varepsilon_n < 0 \end{cases} \quad (13)$$

where ε_n and ε_n^{max} are the maximum nominal strain and the critical strain value in a normal-only mode at an integration point, ε_t and ε_t^{max} are the maximum nominal strain and the critical strain value in a shear-only mode along the first shear direction, and ε_s and ε_s^{max} are the maximum nominal strain and the critical strain value in a shear-only mode along the second shear direction [24].

For these cases, the damage initiates when $1.0 \leq f \leq 1.0 + f_{tol}$, being f_{tol} a tolerance value specified by the user. The user may also define the crack plane normal direction [24].

- Quadratic nominal stress (QUADS) and quadratic nominal strain (QUADE), whose criteria are the following, respectively:

$$\left(\frac{\langle \sigma_n \rangle}{N_{max}}\right)^2 + \left(\frac{\sigma_t}{T_{max}}\right)^2 + \left(\frac{\sigma_s}{S_{max}}\right)^2 = 1 \quad (14)$$

$$\left(\frac{\langle \varepsilon_n \rangle}{\varepsilon_n^{max}}\right)^2 + \left(\frac{\varepsilon_s}{\varepsilon_s^{max}}\right)^2 + \left(\frac{\varepsilon_t}{\varepsilon_t^{max}}\right)^2 = 1 \quad (15)$$

For these criteria, the user also defines the crack plane normal direction, as for MAXS and MAXE [24].

2.4.2 Numerical simulation approaches

Despite the lack of numerical studies regarding the influence of glass microspheres on the failure pattern of adhesively bonded joints, there has already been some research on the use of XFEM to model bonded joints.

Da Silva et al. [29] used the finite element-based XFEM technique to predict the failure mode of single lap joints reinforced with cork microparticles. This study proved that XFEM can be used as a precise method to predict the location of the crack initiation, the crack path and the mechanical performance of the joints. Regarding the failure modes that were obtained experimentally, which were then validated numerically, it has been

observed that, by increasing the size and the amount of particles, the crack evolves from an adhesive failure to a cohesive one, Figure 15.

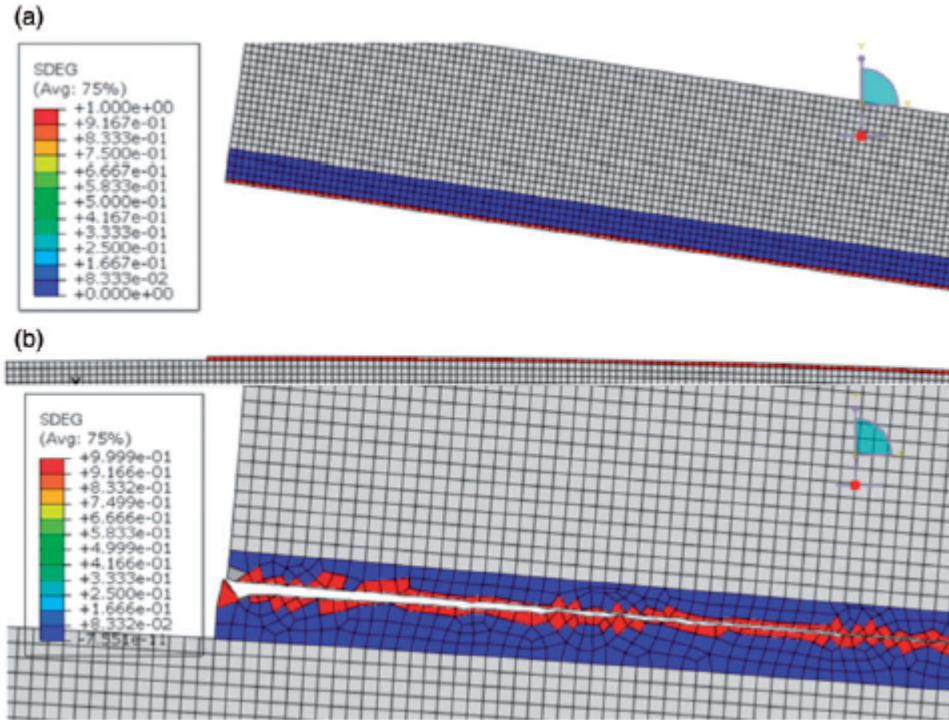


Figure 15: Single lap joints failure modes for two adhesive configurations: (a) neat and (b) 5% of 250–500 mm cork particles [29].

Campilho et al. [30] studied the applicability of XFEM to reproduce the mechanical behaviour of DCB specimens and their crack propagation, and to validate the experimental results regarding not only the load-displacement curves, but also the critical strain energy in pure mode I. This work concluded that XFEM allows to accurately simulate the $P - \delta$ curves, as well as the G_{Ic} values. Therefore, this method can be used as a way to predict the strength and the crack propagation of bonded structures.

3 Experimental procedures

3.1 Materials

During this work, two different adhesives were used, which will be referred to as Adhesive A and Adhesive B. Both are crash-resistant, one component adhesives, designed for the automotive industry. Their use aims to increase the body stiffness, the operation durability, and the crash performance. The general properties of the adhesive are listed in Table 1.

Table 1: General properties of Adhesive A and Adhesive B.

Adhesive	Type of adhesive	Supplied as	Cure cycle
Adhesive A	Epoxy resin	One-part	180 °C / 30 min
Adhesive B	Epoxy resin	One-part	180 °C / 30 min

The mechanical and physical provided by the suppliers of Adhesive A and Adhesive B are displayed in Table 2.

Table 2: Mechanical and physical properties of the adhesives used, provided by the supplier.

Property	Adhesive A	Adhesive B
Density/ $\text{g}\cdot\text{mL}^{-1}$	1.26	1.18
Viscosity/ $\text{Pa}\cdot\text{s}$	160	350
Tensile strength/ MPa	32	35
Elongation at break/ %	≈ 6	≈ 10
Young's modulus/ MPa	2100	1500

The substrates that were used for the manufacturing the DCB specimens and one of the two single lap joint geometries were machined from DIN 40CrMnMo7 steel, which is generally referred to as a high strength steel. The main goal of using high strength steel

was to avoid plastic deformation of the substrates. The main mechanical properties of these substrates can be seen in Table 3.

Table 3: Mechanical properties of the substrates used.

Young's modulus/ MPa	210000
Poisson's ratio	0.3

The other single lap joint specimens substrates were machined from a low strength, mild steel.

The hollow glass beads that were used were provided by 3MTM, with the commercial reference 3MTM Glass Bubbles K37 [20]. These consist of thin-walled hollow spheres made of soda-lime-borosilicate glass with a white powder-like appearance. They can vary in terms of size and density, present a high strength-to-density ratio and have a considerable isostatic crush strength [20].

In Table 4, their main properties are presented.

Table 4: Main properties of the hollow glass beads, provided by the supplier.

Property	3M TM Glass Bubbles K37
Density/ $\text{g}\cdot\text{cc}^{-1}$	0.37
Color	<i>White, powdery</i>
Composition	<i>Soda – lime – borosilicate glass</i>
Median particle diameter/ μm	45
Strength/ MPa	20.6

3.2 Testing methods

The present work's aim, in terms of the experimental study, is to understand how do the adhesives' properties change with varying amounts of hollow glass beads, and to assess how does the introduction of glass particles into the adhesive layer affects the failure mode and pattern and, ultimately, the bonded joint's strength.

To that end, strength and fracture tests were performed, as well as single lap shear tests, at a quasi-static speed, for different amounts of glass beads.

For the strength characterization, bulk tensile tests were realized as a way of determining the tensile properties of the adhesive, at $1 \text{ mm} \cdot \text{min}^{-1}$.

Regarding the fracture characterization, double cantilever beam (DCB) tests were performed in order to determine the fracture toughness in mode I, at $0.2 \text{ mm} \cdot \text{min}^{-1}$.

In terms of the single lap shear strength, single lap joint tests were completed to determine the bonded joints' strength, and to analyse the corresponding fracture surfaces.

For each condition at least three specimens were tested.

3.2.1 Strength tests

Bulk specimen tensile test

Bulk specimens are obtained by machining cured adhesive plates, which are manufactured by following the French standard NF T 76-142 [31], with a thickness of 2 mm.

These were machined to a dogbone shape, according to the British standard BS 2782 [32], which is represented in Figure 16.

The tests were conducted at a quasi-static speed of $1 \text{ mm} \cdot \text{min}^{-1}$, and the displacement was measured utilizing an extensometer. For each test, the load-displacement ($P-\delta$) curve was registered until failure occurred, and the tensile strength and the Young's Modulus were calculated.

Three specimens were tested for each condition.

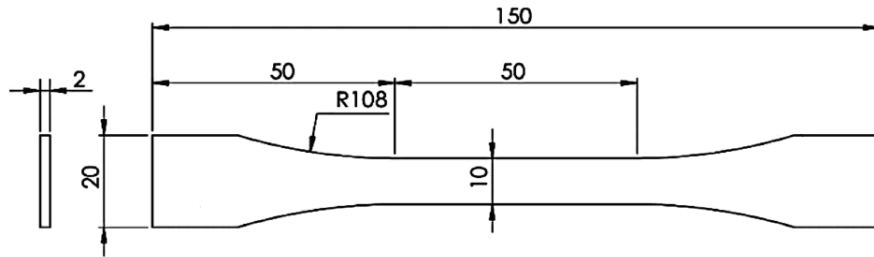


Figure 16: Representation of bulk specimen's geometry, dimensions in mm.

3.2.2 Mode I fracture tests

Double cantilever beam (DCB)

DCB specimens, which consist of two bonded beams with the same thickness and length, were tested to determine the critical energy release rate in mode I. The specimens'

geometry is shown in Figure 17.

The term a_0 refers to the pre-crack length, the length of an initial region without adhesive, h is the thickness of the adherends, and t corresponds to the adhesive's thickness.

The testing procedure consists of applying a tension load to the specimen, which is also represented in Figure 17.

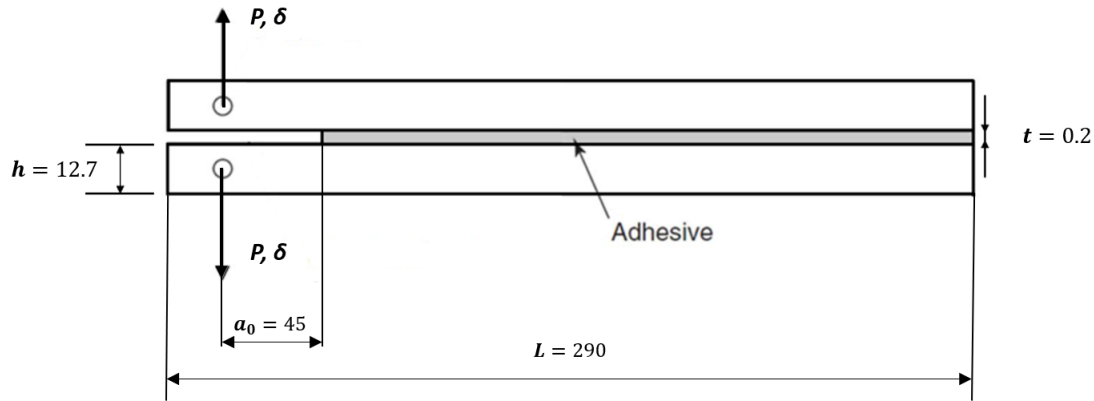


Figure 17: Schematic representation of DCB test.

For each test, the load-displacement ($P - \delta$) curve was registered until failure, and the critical energy release rate in mode I, G_{Ic} , was then calculated.

3.2.3 Single lap shear tests

Single lap joint with high strength steel

SLJ specimens with an adherend made of high strength steel (DIN C65 heat treated) were manufactured, according to the geometry represented in Figure 18.

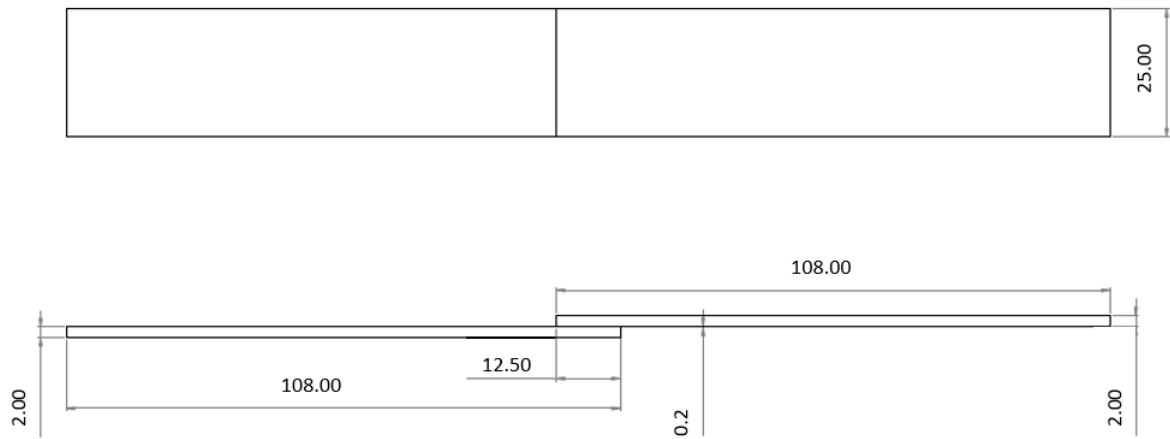


Figure 18: Representation of single lap joint specimens' geometry, dimensions in mm.

For each test, the load-displacement ($P - \delta$) curve was registered until failure, and the fracture surfaces were analysed.

Single lap joint with mild steel

SLJ specimens with mild steel were manufactured, according to the geometry represented in Figure 19.

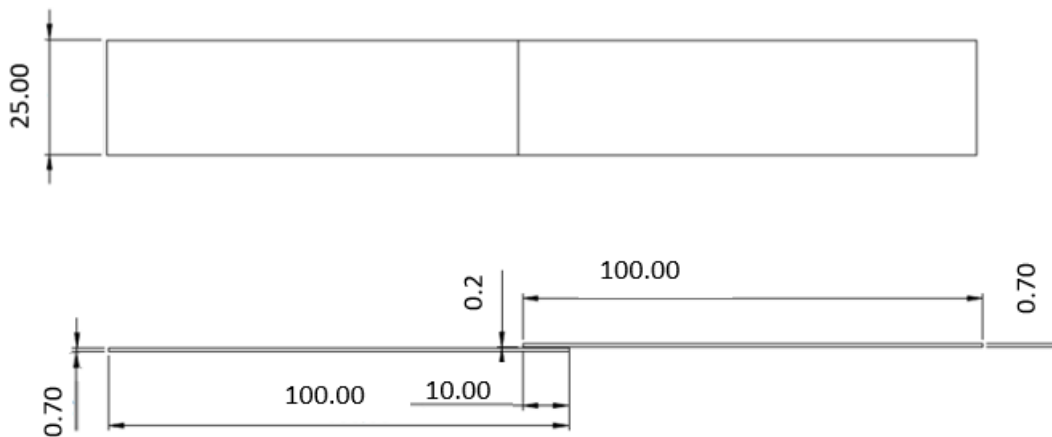


Figure 19: Representation of single lap joint specimens' geometry, dimensions in mm.

For each test, the load-displacement ($P - \delta$) curve was registered until failure, and the fracture surfaces were analysed.

4 Numerical details

In this chapter, a succinct description of the numerical procedures followed for the appended paper C is presented. A more meticulous explanation will be displayed in the corresponding paper.

4.1 Finite element method

Single lap joint specimens with mild steel adherends were tested experimentally, for different amounts of hollow glass beads, and for a quasi-static speed of $1 \text{ mm} \cdot \text{min}^{-1}$.

Three numerical models were created, with the single lap joint's geometry represented in Figure 19, and for the cases of neat Adhesive A, and Adhesive A with 5 and 10% volume of glass beads, since experimentally, these SLJs configurations already allow to behold not only a transition of the failure pattern but also the effects of introducing glass beads in the bonded joint's mechanical performance. These models were created for a quasi-static and implicit analyses, and the $P - \delta$ curves and the crack's propagation path were thoroughly analysed. Afterwards, the numerical results were compared to the experimental ones.

The numerical models were created in ABAQUS[®] (Dassault Systems, Providence, USA), adopting elements which support the XFEM technique. A triangular traction-separation law was adopted to model the adhesive behavior, and assuming a linear softening. This type of law is created with t_n^0 and t_s^0 , which represent the cohesive strengths in tension and shear, respectively, δ_n^0 and δ_s^0 , which are the corresponding displacements, and δ_n^f and δ_s^f , which are associated to the tensile and shear displacements at failure. The triangular traction-separation law considers an initial linear elastic behaviour, until reaching δ_n^0 and δ_s^0 . Then, a linear evolution until damage follows, which is associated to δ_n^f and δ_s^f [28].

During this study, a triangular traction-separation law was adopted to model the adhesive behavior, similar to the one which is portrayed in Figure 4, and assuming a linear softening. This type of law is created with t_n^0 and t_s^0 , which represent the cohesive strengths in tension and shear, respectively, δ_n^0 and δ_s^0 , which are the corresponding displacements, and δ_n^f and δ_s^f , which refer to the tensile and shear displacements at failure. The triangular traction-separation law considers an initial linear elastic behaviour, until reaching δ_n^0 and δ_s^0 . Then, a linear evolution until damage follows, which is associated to δ_n^f and δ_s^f [28].

The boundary conditions which were defined during these simulations were the ones that follow in Figure 20. The restraining and loading conditions were set and consisted in a restriction of the horizontal and vertical movement in the clamped end of the joint (in

the left adherend). The opposite adherend was subjected to a horizontal displacement, and the vertical movement was restricted, closely resembling the loads applied during testing.



Figure 20: Boundary conditions established for the simulation of the SLJ behavior.

It should also be noted that the model does not consider the presence of defects in adhesive layer nor in the interfaces between the adhesive and the substrates.

Moreover, and to create the geometry of the SLJ model, a script in MATLAB © (Mathworks, Massachusetts, USA) was developed, which plots glass spheres with random sizes and locations along the adhesive layer. The coordinates generated by this script were introduced in AutoCAD ©, and the model geometry was imported into Abaqus ©.

In terms of the properties attributed to each material, an elasto plastic isotropic response was considered for the mild steel adherends, since a considerable plastic deformation of the substrates was encountered experimentally. This plastic deformation had a significant influence on the SLJ's performance. For the adhesive and the glass beads, the quadratic stress criterion (QUADS) was used as the crack initiation criterion.

The model was meshed with plane strain solid elements, with QUAD (quadrilateral) structured shapes. The meshes were constructed with bias effects towards the adhesive bond, as these regions are expected to exhibit larger stress gradients, which require finer meshes.

Besides, and although the XFEM methodology implemented in Abaqus © allows to define a pre-crack for damage initiation, the models developed for this study did not use this feature. This approach aimed to allow the crack to freely propagate along the adhesive layer.

All of this information is described in more detail in paper C.

5 Summary of appended papers

Below follow the titles and the corresponding abstracts of the papers that compose this work and present the results and the respective conclusions associated to the developed studies. These papers can be found in the appendices of this document.

Paper	Title: Characterization of the effect of inorganic fillers on adhesives properties
A	Abstract: Adhesives are extensively used in the automotive and the aeronautical industry as they enable durable and light weight joints, with exceptional strength to weight ratios. The constant search for ways of improving the adhesive's mechanical performance led to the use of reinforcements as an attempt of enhancing their properties. This work focuses on the effects caused by the introduction of inorganic fillers in the adhesive layer, in terms of its mechanical properties. To that end, the tensile behavior of the adhesive as well as its fracture properties were thoroughly analysed, by performing bulk and double-cantilever beam tests, at a quasi-static speed. The specimens were manufactured with different amounts of glass beads, an inorganic filler, for two different adhesives. Both adhesives have shown a negligible effect on the tensile strength, a decrease of the strain at failure and the critical energy release rate in mode I, and an increase of the Young's modulus, by increasing the % volume of hollow glass beads. The properties' variation with the amount of glass beads was then analysed, and the corresponding trend lines were obtained.

**Paper
B**

Title: Experimental study of the impact of inorganic fillers on adhesive joint strength and its failure mechanism

Abstract: The use of modern structural adhesives provide a lightweight, practical and high strength joining methodology which is increasingly being adopted in the automotive and aeronautical industries, among many others. However, the strict mechanical performance's standards that must be met in these sectors require a constant search for ways of improving the adhesives' behavior, which has led to the growing use of reinforcements as a way of improving the capabilities of bonded joints. The aim of this work is to analyse how adding inorganic fillers to the adhesive layer affects the joint's strength and its failure mechanism. To this end, single lap joint specimens with mild steel and high strength steel substrates were tested, at quasi-static speeds, for different amounts of glass microspheres reinforcing two different structural adhesives. The experimental results indicate that the addition of glass particles reduces the joint performance for both substrates under study. Furthermore, the failure pattern was found to evolve from adhesive failure to a cohesive type of failure as the amount of glass particles present in the adhesive increased.

Paper C	Title: Numerical model of the damage mechanism in joints bonded using adhesives reinforced with inorganic fillers Abstract: The use of adhesive bonding in the diverse industries, such as the automotive and aerospace sectors has grown considerably. In structural construction, adhesive joints provide a unique combination of low structural weight, high strength and stiffness, combined with a relatively simple and easily automated manufacturing's method, characteristics which are ideal for the development of modern and highly efficient vehicles. In these applications, ensuring that the failure mode of a bonded joint is cohesive rather than adhesive is important, since this failure mode is more controlled, easier to model and to predict. This work presents a technique which enables the precise prediction of the bonded joint's behavior, regarding not only its failure mode but also the joint's strength, when inorganic fillers are added to the adhesive. To that end, hollow glass particles were introduced into an epoxy adhesive's layer in different amounts, and a numerical study was carried out to simulate the behavior of single lap joint specimens. The numerical results were compared with experimentally obtained ones, not only in terms of the joint's strength but also the failure pattern. Both results showed that, by adding glass beads to the adhesive, the crack tends to propagate cohesively, through the middle of the adhesive, and that the joint's performance was generally negatively impacted by the presence of these particles.
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6 Conclusions

The strength, the fracture energy, and the bonded joints' strength and failure mode were established for two different adhesives, and for different amounts of hollow glass beads.

The strength was obtained by performing bulk tensile tests. These tests were carried out for both adhesives under quasi-static conditions, and for different amounts of hollow glass beads.

The fracture energy was defined using the energy release rate in mode I, obtained from double cantilever beam (DCB) tests. These tests were performed for both adhesives under quasi-static conditions, and for different amounts of hollow glass beads.

The bonded joints' strength and failure modes were investigated by manufacturing and testing single lap joint specimens with mild steel and high strength steel substrates. Tests were carried out at quasi-static speed, for different amounts of glass microspheres, and for two different adhesives.

A numerical model was developed, where hollow glass particles were introduced into an epoxy adhesive's layer in different numbers, simulating the behavior of single lap joint specimens. The numerical results were compared with the experimental ones, in terms of the joint's strength and the corresponding failure pattern.

The main conclusions drawn from this work are:

- The tensile strength of the adhesives showed a negligible variation with varying amounts of glass microspheres.
- The strain at failure exhibited a continuous decrease with the growing number of glass beads, while the Young's modulus increased, which showed that these make the adhesive more rigid and more brittle.
- The critical energy release rate, G_{IC} , decreased by increasing the % volume of hollow glass spheres. Therefore, these make the adhesive less tough and reduce its capacity for energy absorption.
- Testing SLJs with high strength steel substrates showed that adding glass beads to the adhesive generally worsens the bonded joint's mechanical performance, for both adhesives. Besides, and for Adhesive B, adding particles to the adhesive layer makes the fracture mechanism to evolve from adhesive to cohesive failure.

- The SLJs specimens with mild steel substrates were found to be more susceptible to the introduction of hollow glass spheres to the adhesive, showing decreased performance. In the case of Adhesive A, doing so causes the crack to propagate in the middle of the adhesive rather than in the interfacial region, due to the formation of local stress fields in the periphery of the particles.
- Generally, the hollow glass beads do not work as tougheners of the adhesive, since they clearly do not establish strengthening or toughening mechanisms within the adhesive. In fact, they weaken the adhesive, since adding these particles has a negative impact on its mechanical performance.
- The numerical models developed show that by adding glass beads to the adhesive, the crack tends to propagate cohesively, close to the middle of the adhesive, and the joint's mechanical performance gets reduced by the presence of the spheres. Furthermore, the numerical models provided satisfactory estimations of the SLJs failure loads, as well as its maximum extension values, showing good agreement with the experimental results. The developed numerical models were also able to simulate the plastic yielding of the substrates accurately.

6.1 Future work

In the future, it would be interesting to determine the impact of hollow glass beads on other adhesive formulations, with very different properties, allowing to assess if the impact of such fillers is the same as the one observed during this study.

Regarding the influence of adding glass beads to adhesives in terms of the strength and failure mode in bonded joints, it would be interesting, again, to perform a similar study for other adhesives, and to use other percent volumes of glass microspheres. Also, testing different joint geometries would be compelling, particularly specimens with thin adhesive layers. This would allow to analyse the effect of the glass beads when these are the ones to establish, through their sizes, the adhesive's thickness.

Additionally, and regarding the numerical simulation, it would be interesting to repeat this study for multiple spheres' distributions, seeking to understand how the location of the spheres influences the crack's propagation path and the bonded joint's performance.

References

- [1] Lucas FM da Silva, Andreas Öchsner, and Robert D Adams. *Handbook of adhesion technology*. Springer Science & Business Media, 2011.
- [2] WA Lees. Adhesive selection. In *Adhesives in Engineering Design*, pages 92–123. Springer, 1984.
- [3] Sina Ebnesajjad and Arthur H Landrock. *Adhesives technology handbook*. William Andrew, 2014.
- [4] MD Banea and Lucas FM da Silva. Adhesively bonded joints in composite materials: an overview. *Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications*, 223(1):1–18, 2009.
- [5] Filipe JP Chaves, Lucas FM da Silva, MFSF De Moura, DA Dillard, and VHC Esteves. Fracture mechanics tests in adhesively bonded joints: a literature review. *The Journal of Adhesion*, 90(12):955–992, 2014.
- [6] David Broek. *Elementary engineering fracture mechanics*. Springer Science & Business Media, 2012.
- [7] MFSF De Moura, Raul DSG Campilho, and JPM Gonçalves. Crack equivalent concept applied to the fracture characterization of bonded joints under pure mode i loading. *Composites Science and Technology*, 68(10-11):2224–2230, 2008.
- [8] P Gálvez, RJC Carbas, Raul DSG Campilho, J Abenojar, MA Martínez, and Lucas FM da Silva. Fracture toughness in mode i (gic) for ductile adhesives. In *Journal of Physics: Conference Series*, volume 843, page 012008. IOP Publishing, 2017.
- [9] M Costa, R Carbas, M Benedita, E Marques, G Viana, Lucas FM da Silva, E Yokoi, S Nakada, and T Furusawa. Static assessment of the mixed-mode behaviour of three epoxy adhesives. *Engineering Fracture Mechanics*, 182:552–565, 2017.
- [10] Ali Nemati Giv, Majid R Ayatollahi, S Hengameh Ghaffari, and Lucas FM da Silva. Effect of reinforcements at different scales on mechanical properties of epoxy adhesives and adhesive joints: a review. *The Journal of Adhesion*, 94(13):1082–1121, 2018.
- [11] AJ Kinloch. Toughening epoxy adhesives to meet today’s challenges. *MRS bulletin*, 28(6):445–448, 2003.

- [12] AQ Barbosa, Lucas FM da Silva, and A Öchsner. Effect of the amount of cork particles on the strength and glass transition temperature of a structural adhesive. *Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications*, 228(4):323–333, 2014.
- [13] AJ Kinloch, JH Lee, AC Taylor, S Sprenger, C Eger, and D Egan. Toughening structural adhesives via nano-and micro-phase inclusions. *The Journal of Adhesion*, 79(8-9):867–873, 2003.
- [14] MS Forte, JM Whitney, and GA Schoeppner. The influence of adhesive reinforcement on the mode-i fracture toughness of a bonded joint. *Composites Science and Technology*, 60(12-13):2389–2405, 2000.
- [15] Qingshi Meng, Sherif Araby, Nasser Saber, Hsu-Chiang Kuan, Jiabin Dai, Lee Luong, Jun Ma, and Chun H Wang. Toughening polymer adhesives using nanosized elastomeric particles. *Journal of Materials Research*, 29(5):665, 2014.
- [16] Hajime Kishi, Kazuhiko Uesawa, Satoshi Matsuda, and Atsushi Murakami. Adhesive strength and mechanisms of epoxy resins toughened with pre-formed thermoplastic polymer particles. *Journal of Adhesion Science and Technology*, 19(15):1277–1290, 2005.
- [17] AQ Barbosa, Lucas FM da Silva, MD Banea, and A Öchsner. Methods to increase the toughness of structural adhesives with micro particles: an overview with focus on cork particles: Methoden zur erhöhung der zähigkeit von strukturklebern mit mikropartikeln: Ein überblick mit dem fokus auf kork. *Materialwissenschaft und Werkstofftechnik*, 47(4):307–325, 2016.
- [18] Aldobenedetto Zotti, Simona Zuppolini, Mauro Zarrelli, and Anna Borriello. Fracture toughening mechanisms in epoxy adhesives. *Adhesives-Applications and Properties*, 1:257, 2016.
- [19] D Verchere, JP Pascault, H Sautereau, SM Moschiar, CC Riccardi, and RJJ Williams. Rubber-modified epoxies. iv. influence of morphology on mechanical properties. *Journal of applied polymer science*, 43(2):293–304, 1991.
- [20] 3M Advanced Materials Division. 3m glass bubbles, 2018.
- [21] VV Budov. Hollow glass microspheres. use, properties, and technology. *Glass and ceramics*, 51(7-8):230–235, 1994.

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- [22] SMR Khalili, A Shokuhfar, SD Hoseini, M Bidkhor, S Khalili, and RK Mittal. Experimental study of the influence of adhesive reinforcement in lap joints for composite structures subjected to mechanical loads. *International Journal of Adhesion and Adhesives*, 28(8):436–444, 2008.
- [23] R Hunter, J Möller, A Vizán, J Pérez, J Molina, and J Leyrer. Experimental study of the effect of microspheres and milled glass in the adhesive on the mechanical adhesion of single lap joints. *The Journal of Adhesion*, 93(11):879–895, 2017.
- [24] Zhen-zhong Du. extended finite element method (xfem) in abaqus. *Simulia: Johnston, RI, USA*, 2009.
- [25] Soheil Mohammadi. *Extended finite element method: for fracture analysis of structures*. John Wiley & Sons, 2008.
- [26] Ali Jenabidehkordi. Computational methods for fracture in rock: a review and recent advances. *Frontiers of Structural and Civil Engineering*, 13(2):273–287, 2019.
- [27] N Sukumar and J-H Prévost. Modeling quasi-static crack growth with the extended finite element method part i: Computer implementation. *International journal of solids and structures*, 40(26):7513–7537, 2003.
- [28] Lucas FM da Silva and Raul DSG Campilho. Advances in numerical modelling of adhesive joints. In *Advances in numerical modeling of adhesive joints*, pages 1–93. Springer, 2012.
- [29] CI da Silva, AQ Barbosa, RJC Carbas, EAS Marques, A Akhavan-Safar, and Lucas FM da Silva. Influence of cork microparticles on the fracture type in single lap joints. *Proceedings of the Institution of Mechanical Engineers, Part C: Journal of Mechanical Engineering Science*, page 0954406220936729, 2020.
- [30] Raul DSG Campilho, MD Banea, FJP Chaves, and Lucas FM da Silva. extended finite element method for fracture characterization of adhesive joints in pure mode i. *Computational Materials Science*, 50(4):1543–1549, 2011.
- [31] NF T 76-142. Méthode de préparation de plaques d’adhésifs structuraux pour la réalisation d’éprouvettes d’essai de caractérisation, 1988.
- [32] BS 2782-0:2011. Methods of testing plastics, part 2011, 2011.
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Appendices

A Paper A

Characterization of the effect of inorganic fillers on adhesive properties

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ABSTRACT

Adhesives are extensively used in the automotive and the aeronautical industries as they enable the creation of durable and light weight joints, with exceptional strength to weight ratios. The constant search for ways of improving the mechanical performance of adhesives has led to the use of reinforcements in an attempt of enhancing their properties. This work focuses on the effects caused by the introduction of inorganic fillers in an adhesive layer, especially with regards to the mechanical properties. To that end, the tensile behavior of the adhesive as well as its fracture properties were thoroughly analysed, by performing bulk and double-cantilever beam tests, at a quasi-static speed. The specimens were manufactured with different amounts of a glass beads, an inorganic filler, for two different adhesives. Both adhesives have shown a negligible effect on the tensile strength, a decrease of the strain at failure and the critical energy release rate in mode I, and an increase of the Young's modulus, with an increase in the % volume of hollow glass beads. The variation of properties with the amount of glass beads was analysed in detail, using fitting functions and curves created using rule of mixtures.

A.1 Introduction

The use of adhesively bonded joints for structural joining instead of using bolts, rivets, and welding is becoming increasingly more frequent. The increased popularity of this technique is related to some very particular and valuable attributes associated with adhesives themselves, such as their low weight, high joint stiffness, uniform stress distributions, the feasibility of automating the joining process, the ability to join parts according to different designs, contours and thicknesses, as well as the unique capability of joining different materials, to mention a few [1, 2].

Due to this emerging use of adhesive joints, particularly in high technological applications, there is also a rising need to find alternative ways to improve their mechanical behaviour and failure mode. Amongst others, some of the methods that have been studied to ameliorate their performance are the combination between adhesive joints with other conventional joining methods, being this combination called hybrid joint, changes in the adherends' surface roughness, as an attempt to improve the adhesion of the joints, and the search for an optimum thickness of the adhesive layer [3].

Nonetheless, some of the methods referred above score poorly in terms of simplicity and expensiveness. Bearing this in mind, the addition of reinforcements to the adhesive layer may come across as a simpler and also more affordable way to enhance the adhesive's performance. Adding reinforcements to the adhesive layer forms a composite material, as it synergistically combines two or more materials [3].

Epoxy adhesives are usually associated with amorphous and highly cross-linked chemistries, which provide valuable structural properties. Despite that, and taking into account that they are slightly brittle materials too, the addition of a second phase of particles, depending on their characteristics, provides a path to improve the adhesive's performance without having an impact on other important properties, such as the rubber particles [4].

In fact, the addition of reinforcements can significantly influence the damage mechanism of adhesives, since it can change the way the crack propagates along the adhesive layer, and there are many damage mechanism that have already been detected in different studies. The bridging mechanism (see Figure 1) is a damage mechanism that can work as a way of enhancing the mechanical properties of the adhesive, and it is highly dependent on the size and geometry of the reinforcements being introduced. Moreover, these can break or pull out from one surface to another depending on the angle formed with the fracture surface and the interfacial bond strength to the adhesive [3, 5]. Besides, the crack deviation mechanism (see Figure 2) is a mechanism which occurs as a consequence

of the presence of particles in the adhesive's matrix, since the crack progression is arrested by the spheres and is forced to change its regular path. This phenomenon can not only increase the fracture toughness but also postpone the failure of the reinforced adhesive [3]. Moreover, the crack pinning and bowing mechanism (see Figure 3) is another damage mechanism which is more relevant in brittle materials than in ductile ones, and it is associated to changes in the velocity and shape of the crack propagation near the second-phase particles, which function as obstacles [3]. This leads to a energy dissipation along the adhesive, since bowing out the crack front between the particles results in improvements in terms of the fracture energy [6]. The microcracking mechanism consists of the formation of multiple cracks ahead of the main crack tip, known as micro cracks, which leads to a decrease of the modulus and the stress concentrations near them, culminating in an improvement of the adhesive's fracture toughness. This mechanism favors the creation of new surfaces and particles debonding, and is dependent on the size of the reinforcements being used [3]. The shear banding mechanism (see Figure 4) derives from local stress concentrations created by the reinforcements and can be provoked by particle debonding and void nucleation. This decreases the stresses next to the particles and facilitates the propagation of the shear bands [3]. Another damage mechanism is the particle debonding and subsequent plastic void growth. In this mechanism, the particle debonding occurs due to local stress concentrations at the particle-matrix interfaces. Then, local plastic void growth may appear as a consequence of the previous conditions. This leads to an increase in terms of the dissipated energy, due to the existing plastic voids [3]. At last, stress fields around the particles are also reported as another damage mechanism, related to the presence of particles in the matrix, such as spherical rubber particles, that introduce local stress concentrations around the equator of the particle. For the case of the rubber particles, and since they have the capability of bearing loads while functioning as stress concentrators, this makes them effective at toughening the adhesive [4, 7].

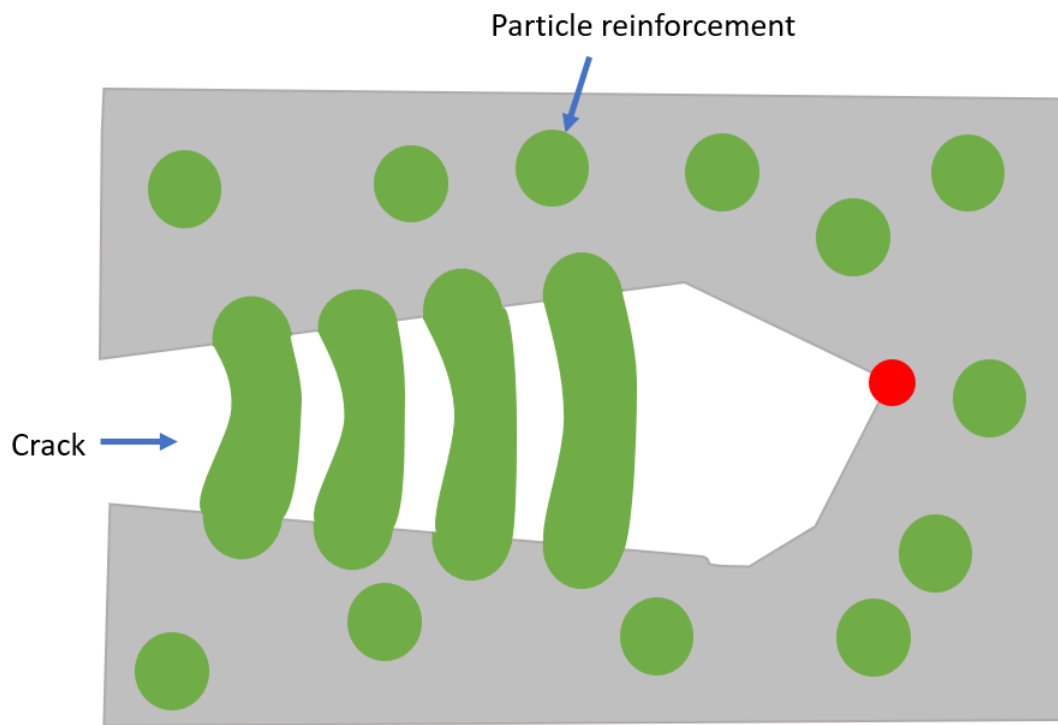


Figure 1: Schematic of bridging mechanism.

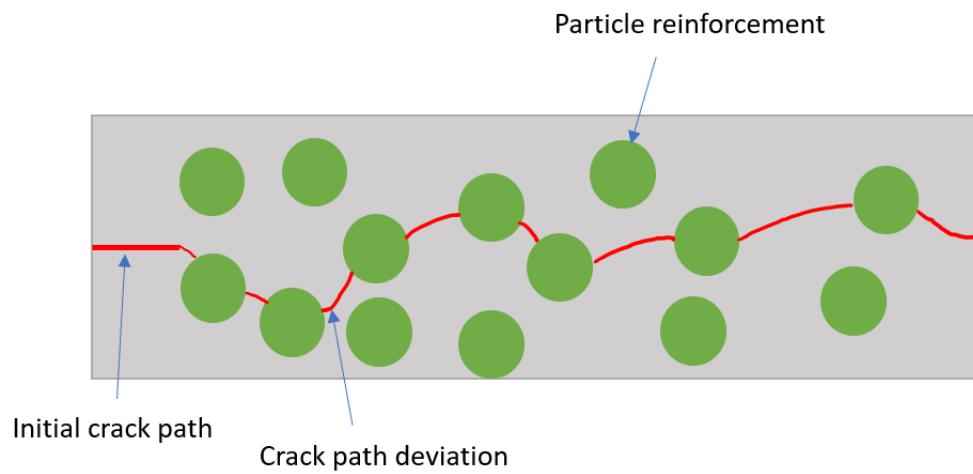


Figure 2: Schematic of crack growth deviation.

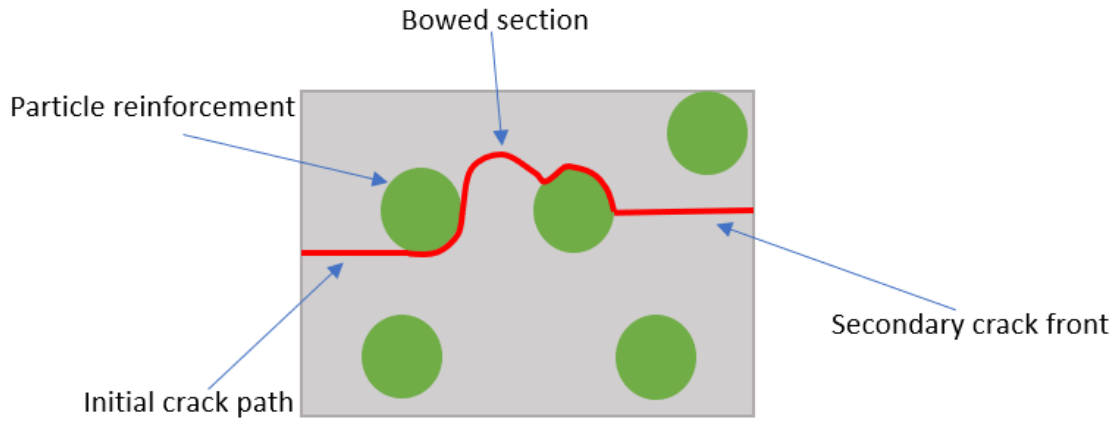


Figure 3: Schematic of crack pinning and bowing mechanism.

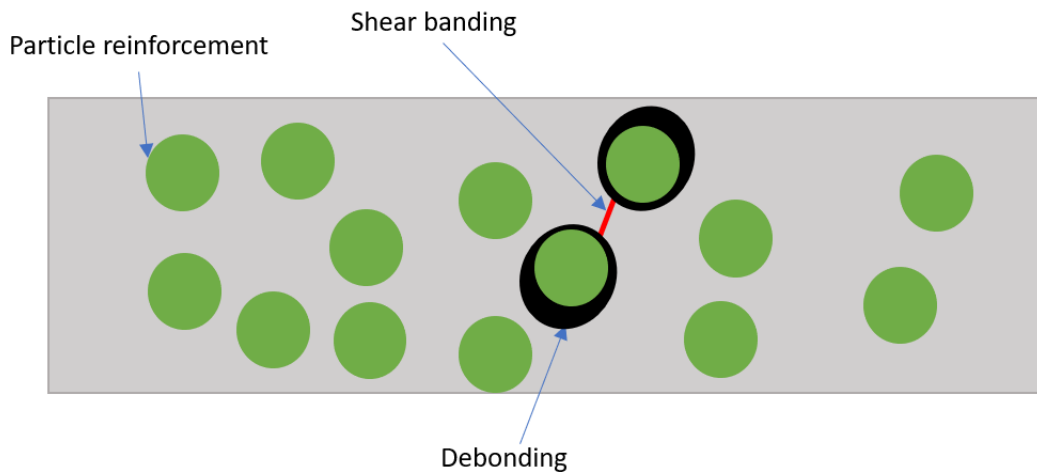


Figure 4: Schematic of shear banding mechanism provoked by particle debonding.

Regarding the influence of reinforcing the adhesive with the adding of particles, there are already some studies that have focused on that. With this in mind, the next paragraphs present some of the experimental works concerning this matter.

Kinloch et al. [8] investigated on how the addition of nanosilica particles to a rubber toughened adhesive influences its toughness. For this purpose, single-lap joint and tapered double-cantilever beam specimens were manufactured and the respective tests were carried in order to assess the evolution of the adhesive's behaviour. Nanosilica particles, which presented an average diameter of 20 nm, proved to guarantee improvements not only in terms of toughness, but also in terms of single-overlap shear strength for amounts between 1 % and 8 % in weight. This happened since the presence of the spheres generates

interactions of the stress field around them and next to the crack tip, which contributes to the plastic deformation of the adhesive's matrix.

Barbosa et al. [9] studied the effect of cork particles on the strength of a structural adhesive. Throughout this experimental study, bulk and single lap joints tests were carried out to evaluate the effects on the adhesive's mechanical properties by introducing different amounts of cork (between 0.5 % and 5%, in weight) to the adhesive layer . The experimental results proved that, for 1 % of cork particles in weight, the adhesive showed an increase in terms of ductility and a higher joint strength. However, by increasing the amount of particles up to 2 %, the mechanical behaviour was negatively affected, as the particles started acting as defects, reducing the adhesive toughness.

Forte et al. [10] showed that the effect of the presence of reinforcements in the adhesive layer is different for the case of fiber-glass fibers. An experimental and numerical study was developed related to the effects of adhesive reinforcement on the mode I fracture toughness of bonded joints. This study focused on comparing the different energy release rates in mode I for different volumes of fiber-glass fibers in the epoxy resin. The experimental results showed that the introduction of glass fibers to the adhesive layer led to a general decrease in terms of the energy release rate, as one can see in Figure 5, when compared to the neat resin. Furthermore, analyses performed through Scanning Electron Microscopy (SEM) demonstrated the existence of fiber breakage for every combination of adhesive and fiber-glass, and a matrix interface failure in most of the analysed specimens.

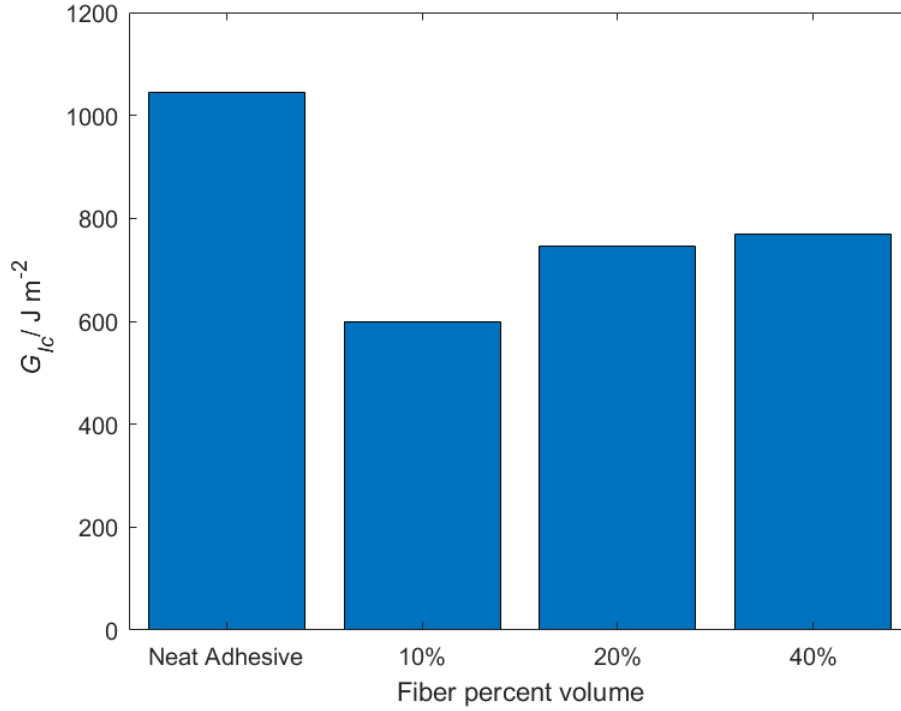


Figure 5: Energy release rate in mode I as a function of the fiber-glass percent volume. Adapted from [10].

Meng et al. [11] presents a research that compares the influence of nanosized and micrometer-sized rubber particles on the fracture toughness and strength of polymeric adhesives, for different adhesive thicknesses. Volume fractions of 2.5 % and 5.0 % were used both for the nanosized and the micrometer-sized particles. Double cantilever beam (DCB), compact tension (CT) and lap shear tests were performed to assess progression of the adhesive's mechanical properties. The experimental results showed that nanoparticles led to considerable improvements in terms of the fracture toughness and shear strength. This is explained by the fact that, for certain volume fraction values, nanosized particles have shorter particle-to-particle distances and provide larger surface areas, which promotes the external energy absorption. Additionally, the presence of microparticles resulted in a modest enhancement of the fracture toughness, depending on the adhesive thickness and the ductility of its matrix.

Kishi et al. [12] compared the differences between using polyamide-12 particles and core-shell rubber particles as reinforcements for an epoxy resin by analysing their impact on the adhesive's properties. This experimental study was built upon the manufacturing and testing of CT specimens, as well as the manufacturing of specimens to evaluate the peel and shear adhesive strength. The results showed that the T-peel adhesive strength

was improved by increasing the amount of polyamide-12 particles. On the other hand, the use of core-shell rubber particles as reinforcements firstly led to an improvement of the adhesive's strength by increasing the particles' amount, until a plateau was attained. Furthermore, the polyamide-12 particles were found to have the capacity to plastically deform before fracture in the peel process. This phenomenon suggested the existence of a bridging mechanism, which enhances the adhesive's toughness, and only occurs for reinforcement particles that have a good adhesion to the adhesive's matrix, the capability of absorbing significant amounts of energy and that have an elasticity's modulus which is lower than that of the adhesive's matrix.

The research presented above proves that the presence of reinforcements in the adhesive layer can significantly influence their mechanical behaviour. Their results and conclusions also confirm that, depending on the type of reinforcements being introduced, properties such as the strength and fracture toughness of the adhesives can either improve or decrease.

With this in mind, and since the impact of hollow glass beads has not yet been deeply researched, this paper focuses on their effects on the strength and fracture toughness on a epoxy adhesive. To do so, the fracture energy in mode I and the tensile strength properties were determined by performing DCB and bulk tensile tests at four different configurations in terms of the percent volume of glass beads: neat adhesive, and adhesive with 5, 10 and 15 % volume of glass beads. Subsequently, the fracture energy in mode I and the maximum tensile strength were defined as a function of the percent volume of hollow glass spheres.

In this study, the effects of reinforcing adhesives with inorganic fillers were analysed. Accordingly, hollow glass beads were introduced into the adhesive layer of two epoxy adhesives in different amounts of particles. With the goal of assessing how the glass beads affect the adhesives' properties, bulk and double-cantilever beam specimens were manufactured and tested. The experimental results were then collected and analysed, namely the stress - strain curves and the data regarding the critical energy release rate in pure mode I. Additionally, SEM analyses were conducted on the fracture surfaces of the bulk specimens, allowing to assess the properties of the materials being studied, and to survey the state of the glass beads and the damage mechanisms that these might trigger. A comparison between the increase amount of glass beads and the strength and fracture properties was then performed.

A.2 Experimental details

This section describes in detail the adhesive and the inorganic filler that was used in this experiment. Moreover, it explains the manufacturing process of the bulk and the DCB specimens, as well as the respective test procedures. Lastly, it also briefly describes the scanning electron microscopy analysis.

A.2.1 Materials

The adhesives studied throughout this work will be referred to as Adhesive A and Adhesive B. These are one component crash-resistant adhesives, which aim to increase durability, crash performance and body stiffness. These two adhesives cure at a temperature of 180 °C during 30 minutes, Figure 6.

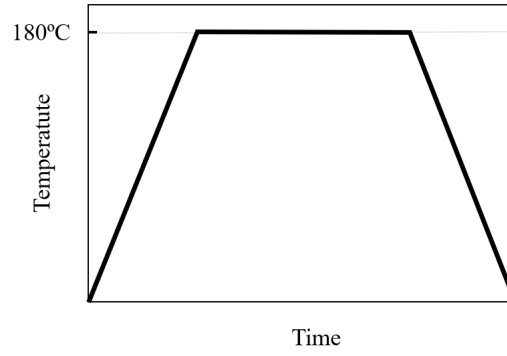


Figure 6: Cure cycle of the adhesives used.

Regarding the hollow glass beads (Figure 7), they consist of thin-walled hollow spheres made of soda-lime-borosilicate glass with a white powder appearance. They can vary in terms of size and density, and are known to be lightweight, to present a high strength-to-density ratio and to have a considerable isostatic crush strength [13,14].

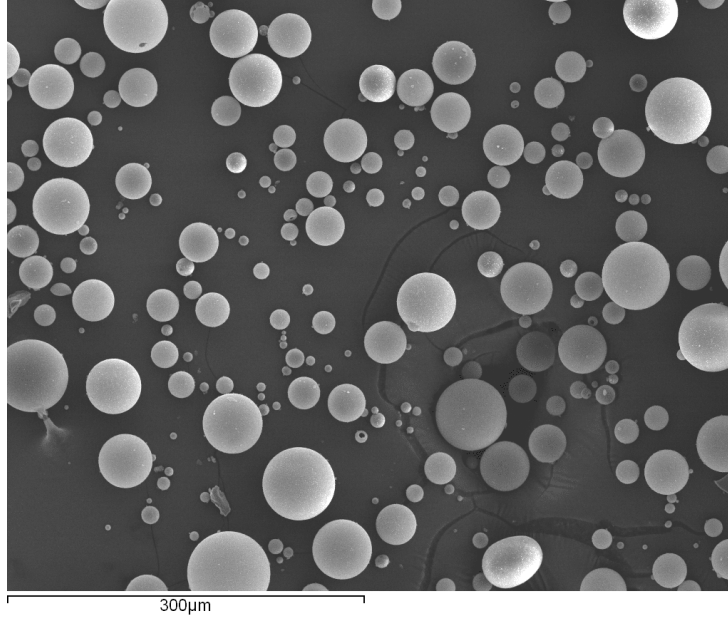


Figure 7: SEM micrograph of glass microspheres.

In Table 1 follow the main properties of the glass beads used.

Table 1: Main properties of the hollow glass beads, provided by the supplier.

Property	-
Density/ $\text{g}\cdot\text{cc}^{-1}$	0.37
Color	<i>White, powdery</i>
Composition	<i>Soda – lime – borosilicate glass</i>
Median particle diameter/ μm	45
Strength/ MPa	20.6

A.2.2 Specimen fabrication

Bulk specimens. The incorporation of the the glass beads in the adhesive has been performed by mixing them directly in the epoxy adhesive. This process was carried out after pre-heating the adhesive, in order to facilitate the distribution of the particles. A centrifuge mixing machine was used in this process, of the type SpeedMixer DAC 150TM (Hauschild, Hamm, Germany). It operated for 40 s, increasing gradually from a 500 r/min to a 3000 r/min rotation speed in the first 10 seconds, and then maintaining the rotation

speed at a 3000 r/min velocity for the remaining time.

Bulk specimens were then obtained by machining cured adhesive plates, which were manufactured by following the French standard NF T 76-142 [15], with a thickness of 2 mm. This manufacturing process involves the use of a steel mould to press the adhesive in the uncured state, and a silicone rubber frame with the aim of delimiting the adhesive and applying hydrostatic pressure to the mixture. By doing so, an acceptable surface finish is assured, as well as the lack of voids. Then, the adhesive was cured by applying the prescribed temperature and pressure.

Finally, the specimens were machined according to the British standard BS 2782 [16].

In Figure 8, the bulk specimens' configuration that was obtained is displayed.

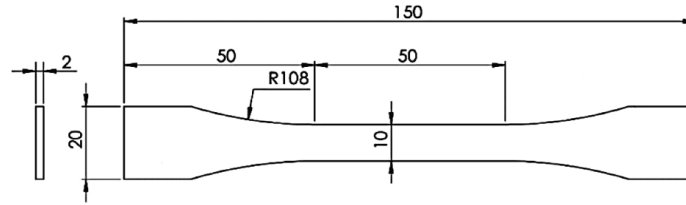


Figure 8: Representation of tensile specimens' geometry, according to BS 2782 [16], dimensions in mm.

DCB specimens. DCB specimens were manufactured as shown in Figure 9.

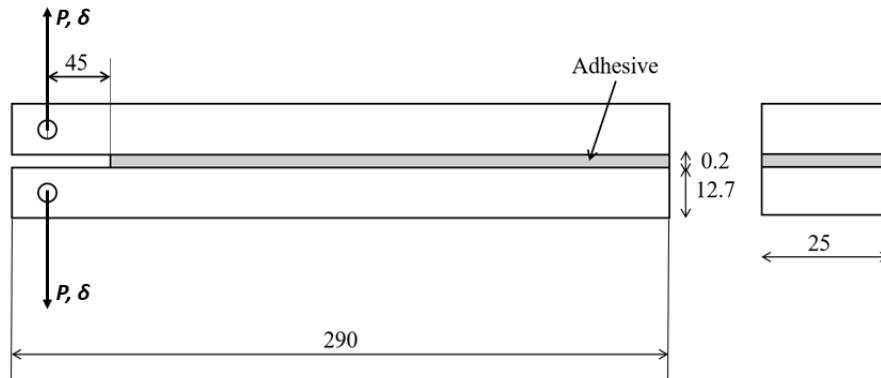


Figure 9: Representation of DCB specimens' geometry and testing, dimensions in mm.

The substrates that are used are made of hard steel, subjected to a grit blasting process to remove oxides and create roughness in the bonded area. Following the grit blasting process, the substrates were degreased with acetone to ensure a good adhesion.

A pre-crack, which had a length of 45 mm, was created by placing a sharp razor blade with 0.1 mm of thickness in one end of the bonded area. A calibrated tape of 0.2 mm

was positioned in the other end of the substrates to ensure the adhesive thickness of 0.2 mm. At last, the adhesive was applied and was then cured by applying the prescribed temperature and pressure.

A.2.3 Testing procedures

Both the tensile and the DCB tests were carried out in an INSTRON 3367 universal testing machine, equipped with a load cell of 30 kN (Norwood, USA). For both cases, specimens with and without glass beads were manufactured according to the test plans represented in Figures 10 and 11.

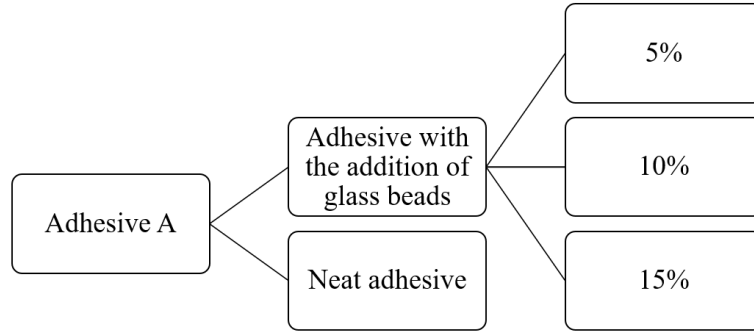


Figure 10: Schematic diagram of bulk and DCB specimens with and without glass beads, for Adhesive A.

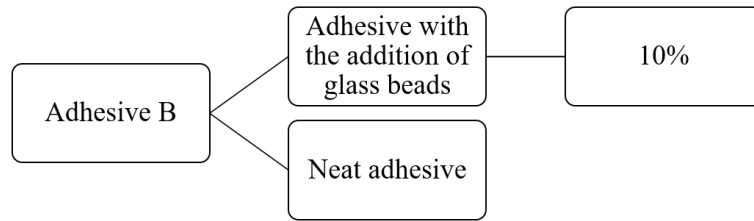


Figure 11: Schematic diagram of bulk and DCB specimens with and without glass beads, for Adhesive B.

At this stage, it is important to state that neat Adhesive A already has 5% volume of hollow glass beads spread along its matrix in its manufacturer supplied state.

Tensile tests. Tensile tests were conducted at a quasi-static speed of $1 \text{ mm} \cdot \text{min}^{-1}$, and the displacement was measured utilizing an extensometer. For each test, the load-displacement ($P - \delta$) curve was registered until failure, and the tensile strength and

Young's Modulus were calculated. Three specimens were tested for each condition.

DCB tests. The aim of the DCB tests was to determine the critical energy release rate in mode I. The tests were conducted at a quasi-static speed of $0.2 \text{ mm} \cdot \text{min}^{-1}$, and the specimens were previously loaded in mode I at low load levels to ensure a stable crack propagation and a sharp pre-crack, for which a new initial crack length, a_0 , was recorded. For each test, the load-displacement ($P - \delta$) curve was registered until failure. Three specimens were tested for each condition.

The compliance based beam method (CBBM) data reduction scheme was used for the determination of the energy release rate in mode I, which was mainly developed by Moura et al. [17] to determine the energy release rate in mode I.

For this method, the crack length is determined based on the specimen's compliance, without requiring to directly measure it. This also allows the fracture process zone effects to be accounted in the calculation of the fracture energy [17], which is defined by the existence of multiple micro cracks and plasticized area ahead of the major crack which absorb certain amounts of energy [17]. Figure 12 schematically represents the fracture process zone.

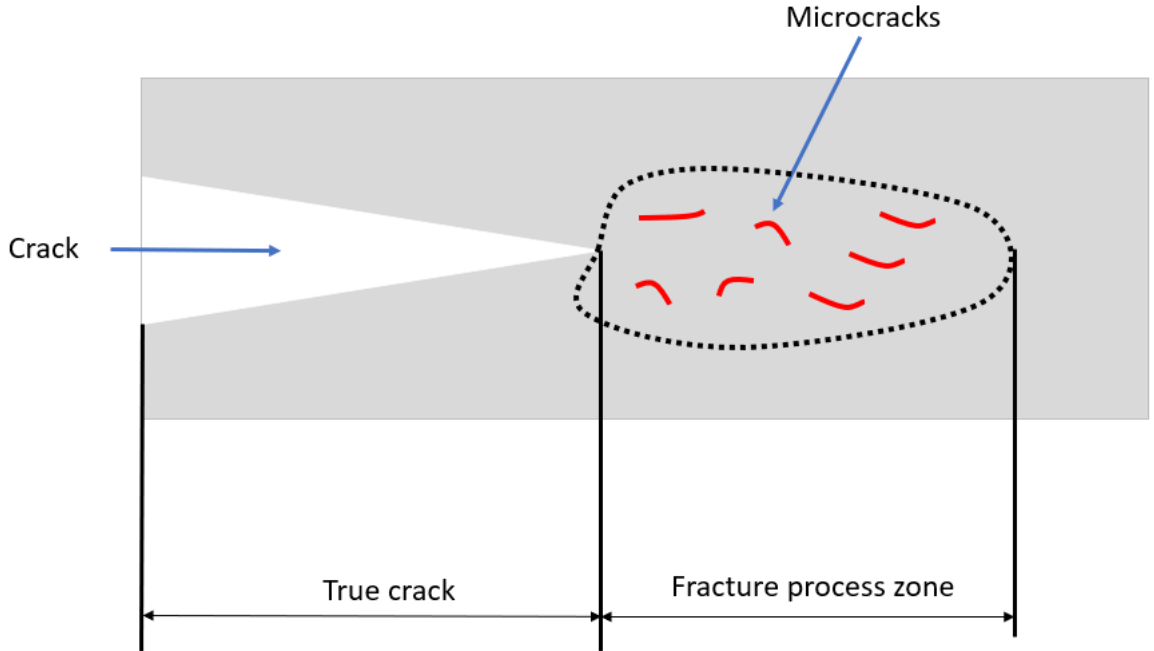


Figure 12: Schematic representation of the fracture process zone (FPZ).

The equation below allows to calculate the fracture energy in mode I using the CBBM methodology [17]:

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

where P represents the load, B is the specimen width, h corresponds to the thickness of the substrates, G_{13} is the shear modulus, a_{eq} is the equivalent crack's length, and E_f corresponds to the corrected flexural modulus, to account for the substrates' rotation near the crack tip [17].

As previously said, the equivalent crack's length, a_{eq} , is calculated based on the specimen's compliance [17]. Figure 13 is a schematic representation of the fracture process zone and the crack equivalent concept, being a the initial crack length, Δ the root rotation correction for the initial crack length and FPZ the correction to take into account the fracture process zone [18].

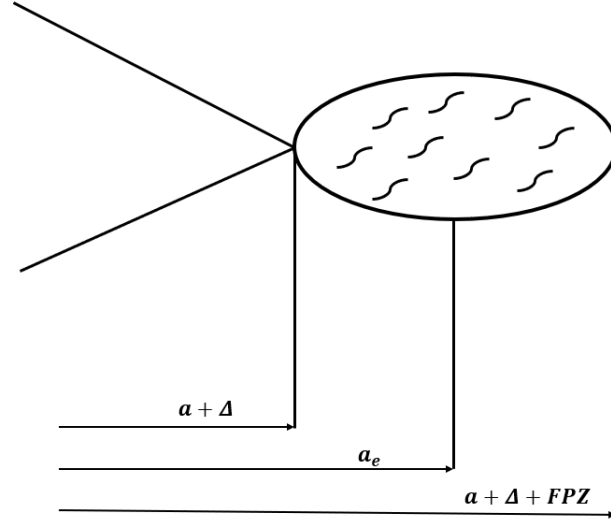


Figure 13: Schematic representation of the fracture process zone and the crack equivalent concept.

A.2.4 Scanning electron microscopy analysis

The scanning electron microscope (SEM) analyses were performed utilizing a JEOL JSM 6301F/Oxford INCA Energy 350/Gatan Alto 2500 microscope (Tokyo, Japan). The use of this equipment allowed to determine the level of damage patent in the glass spheres, their diameter range, the distribution of the beads along the adhesive, and the fracture surfaces. The X-Ray diffraction analysis associated to this equipment also enabled to precisely determine the chemical nature of the materials being tested.

A.3 Experimental results and discussion

A.3.1 Tensile tests

As stated previously, the tensile tests were performed for different amounts of hollow glass microspheres, measured in volume fractions. In Figure 14, the testing procedure that was adopted for this study can be observed.

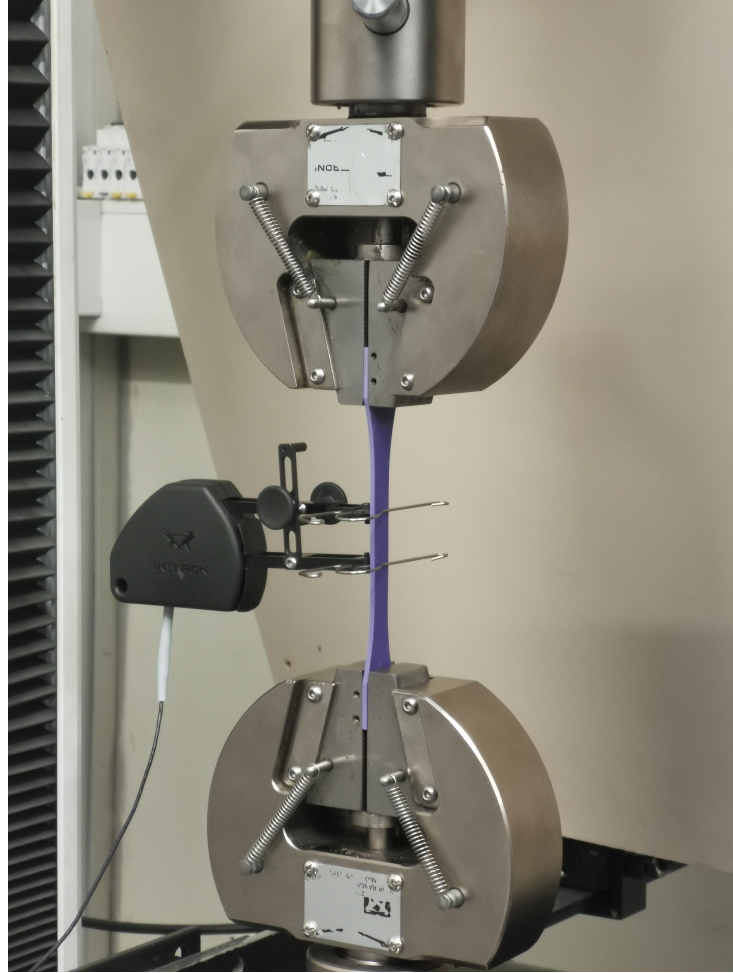


Figure 14: Bulk testing procedure.

The stress, σ , strain, ε , curves obtained for the tensile tests were the ones that follow in Figure 15, for Adhesive A under quasi-static conditions.

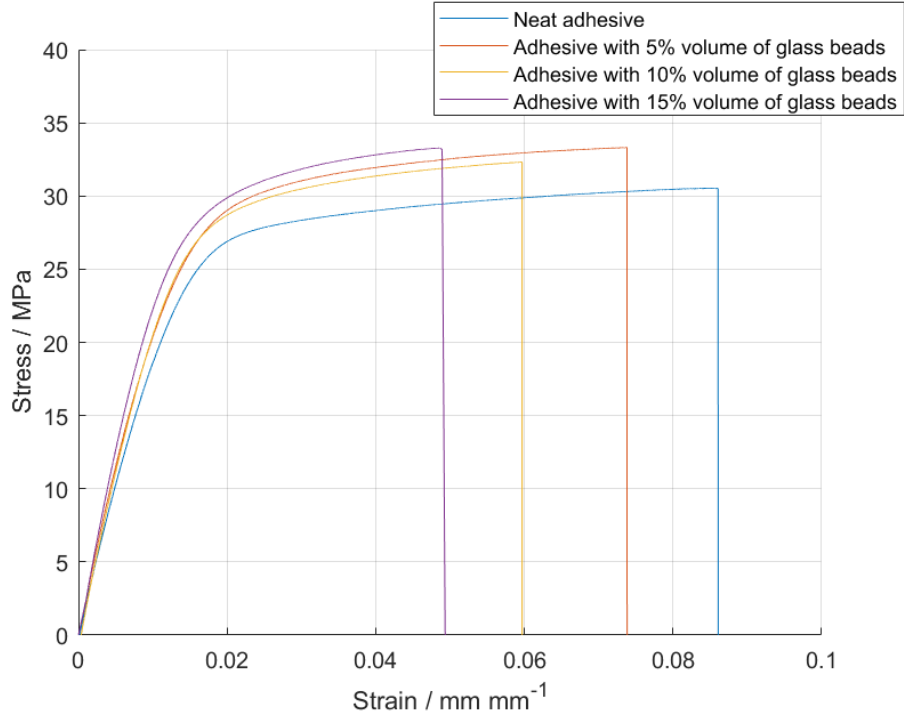


Figure 15: Representative stress-strain curves for the tensile tests of Adhesive A with neat adhesive, and adhesive with the addition of 5, 10, and 15 % volume of hollow glass beads.

The values of the maximum stress, σ_{max} , can be observed in Figure 16, for the different percent volumes of hollow glass beads.

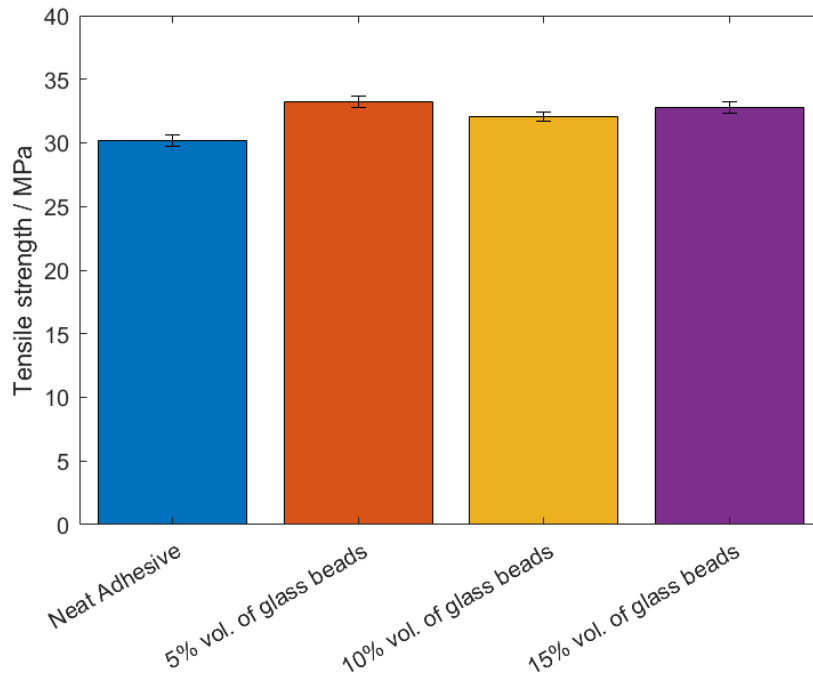


Figure 16: Maximum stress, σ_{max} , of Adhesive A for the four different percent volumes of added glass beads, $\%vol$, tested.

Below, the Young's modulus values for the different amounts of particles can be seen, Figure 17.

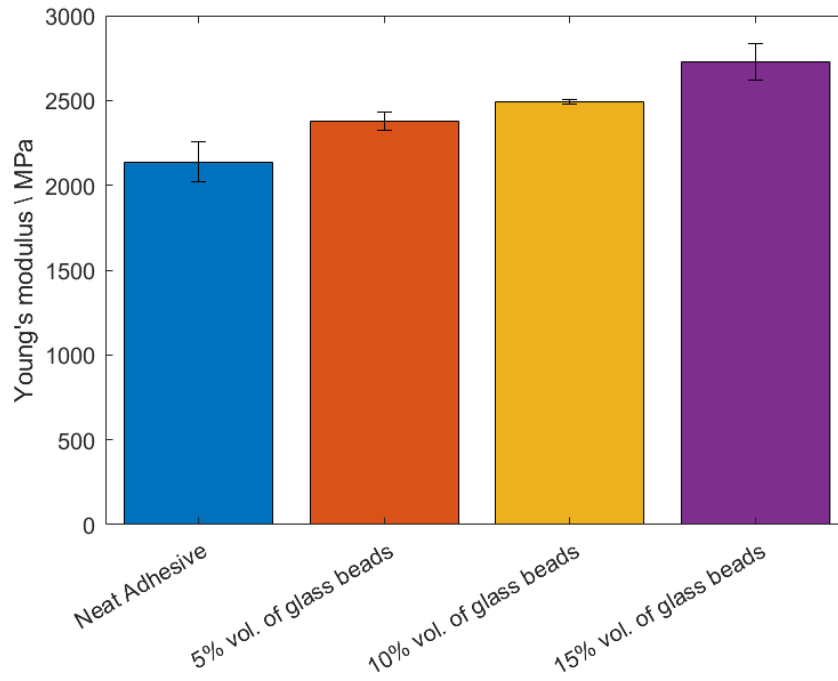


Figure 17: Young's modulus, E , of Adhesive A for the four different percent volumes of added glass beads, $\%vol$, tested.

At last, the maximum strain values for the different amounts of particles are represented in Figure 18

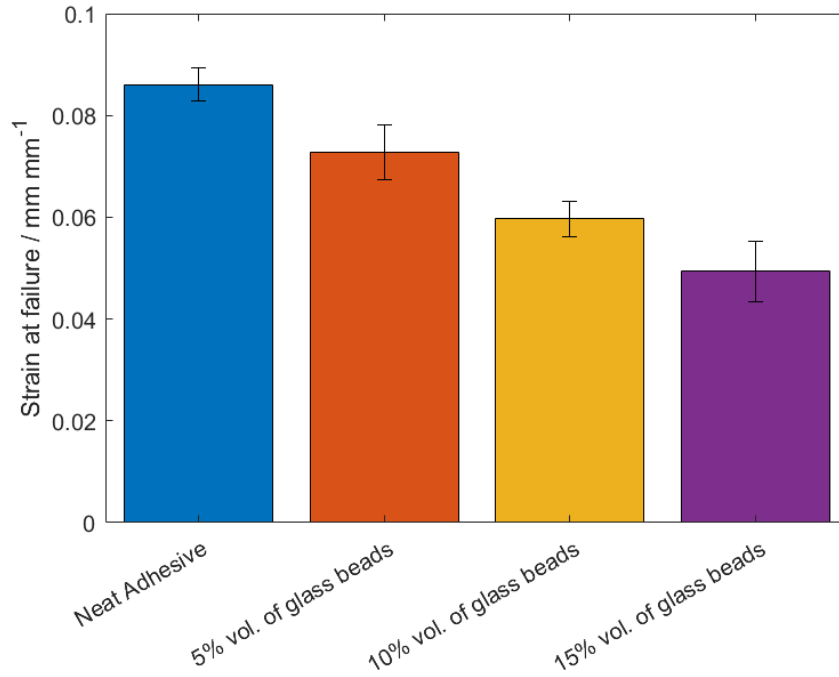


Figure 18: Maximum strain of Adhesive A for the four different percent volumes of added glass beads, %*vol*, tested.

According to the results shown above, one can conclude that, in terms of the maximum stress, the introduction of glass beads does not lead to major variations. Instead, these values remain relatively constant.

However, the evolution of the Young's modulus and the maximum strain values makes it evident that the increase of the particles' amount increases the stiffness of the adhesive. Furthermore, the maximum extension continuously decreases with increasingly larger particle amounts, which translates into a more brittle adhesive.

Tensile tests were also performed for Adhesive B, at quasi-static conditions, in accordance with the diagram displayed in Figure 11.

Figure 19 exhibits the stress σ , strain, ε , curves, for neat adhesive and adhesive with 10 % volume of glass beads.

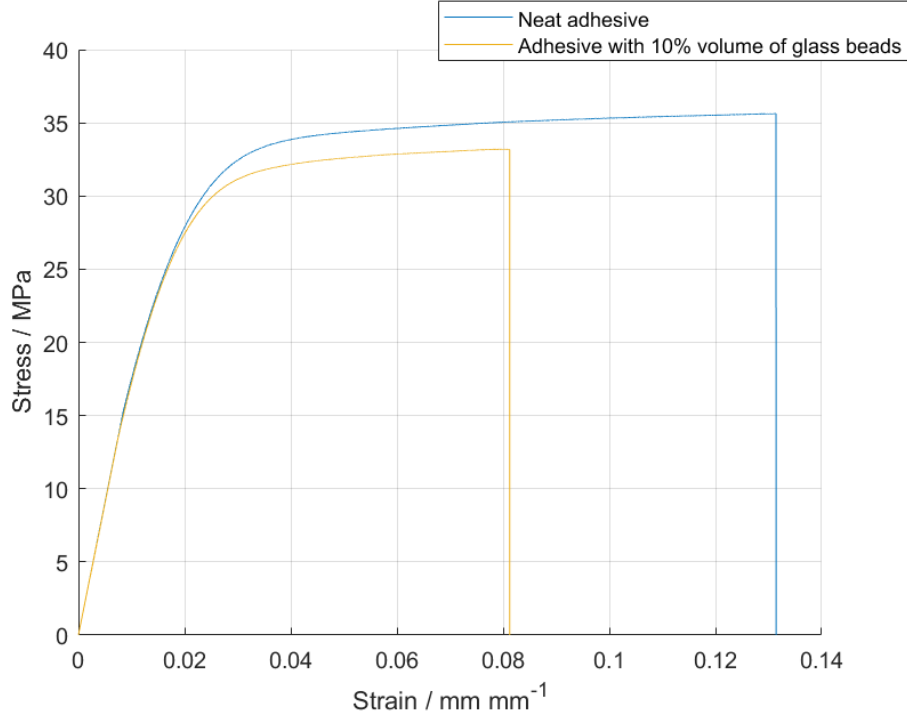


Figure 19: Representative stress-strain curves for the tensile tests of Adhesive B with different hollow glass beads amounts.

The values of the maximum stress, σ_{max} , as well as the Young's modulus and the strain at failure can be observed in Figures 20, 21, and 22 respectively.

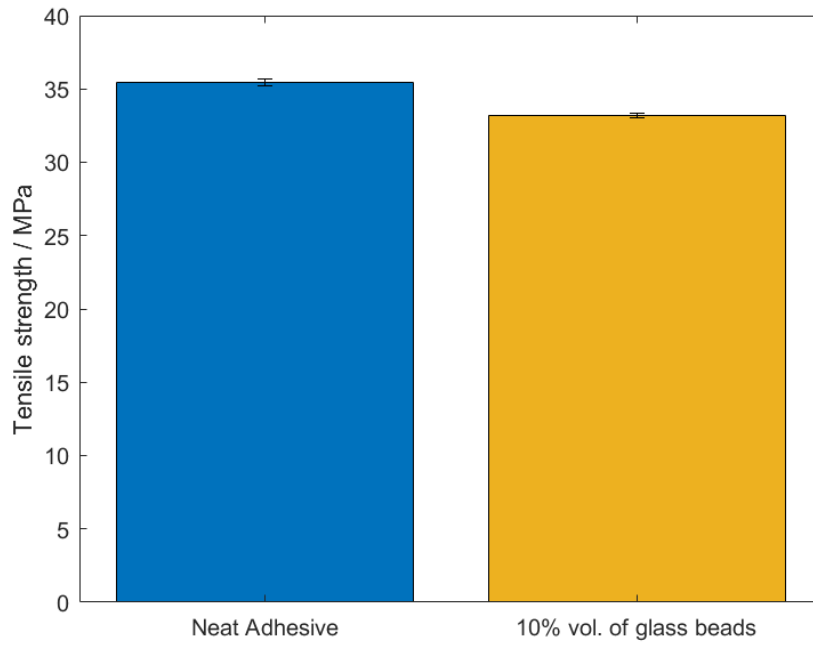


Figure 20: Maximum stress, σ_{max} , of Adhesive B with different hollow glass beads amounts.

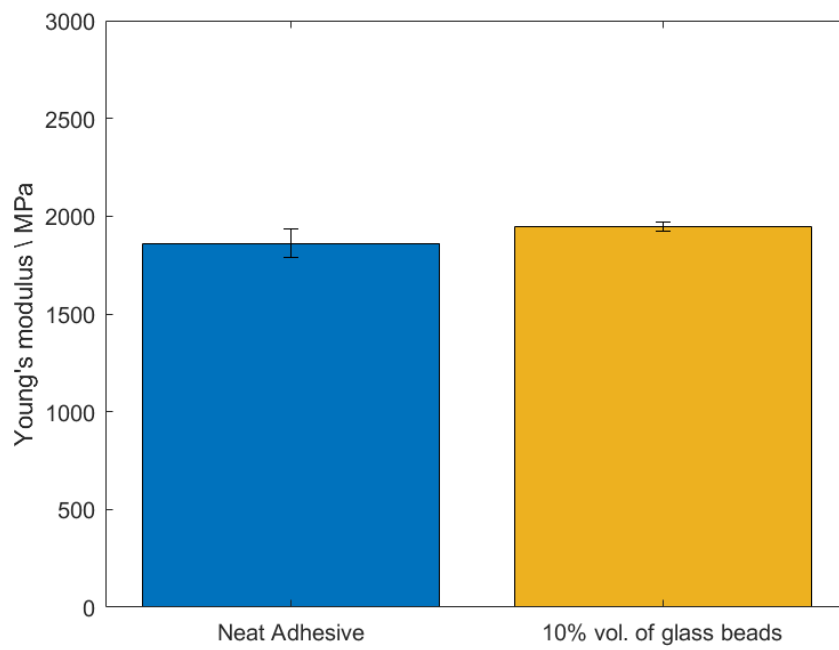


Figure 21: Young's modulus, E , of Adhesive B with different hollow glass beads amounts.

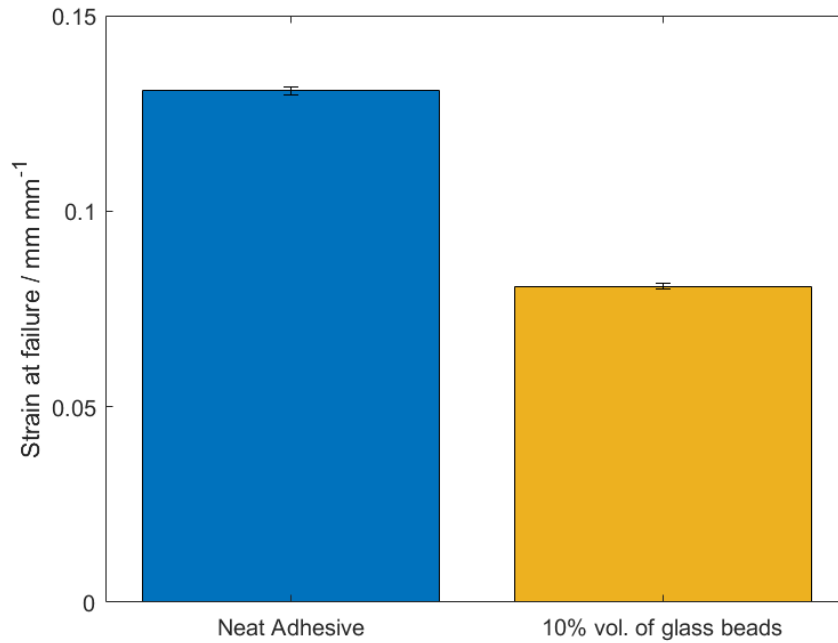


Figure 22: Maximum strain of Adhesive B with different hollow glass beads amounts.

Again, and similarly to what was observed regarding Adhesive A, adding glass microspheres to the adhesive layer does not have a significant impact in the tensile strength of the adhesive. Despite that, the strain at failure is clearly negatively affected by the introduction of glass beads into the adhesive layer, showing a considerable decrease, and the Young's modulus slightly increases for Adhesive B with 10 % volume of glass beads.

The results obtained for both adhesives suggest that, even though the tensile stress does not significantly vary with the different amounts of glass beads, there is a clear impact of adding such particles in terms of the strain at failure and the Young's modulus. The variation of these material properties allows to conclude that hollow glass microspheres damage the adhesive layer and generally turn the adhesive into a more brittle material. Besides, and by taking a look at the stress-strain curves, it is possible to observe that the area below these curves, which can be interpreted as representative of the adhesive's ductility, gradually decreases by introducing rising amounts of particles to the adhesive.

A.3.2 DCB tests

For the DCB tests, different amounts of hollow glass microspheres, in volume, were also used. In Figure 23, it is possible to see the testing procedure that was performed in this experimental study.



Figure 23: DCB testing procedure.

The $P - \delta$ curves obtained for the tensile tests were the ones that are represented in the following figure 24, for Adhesive A.

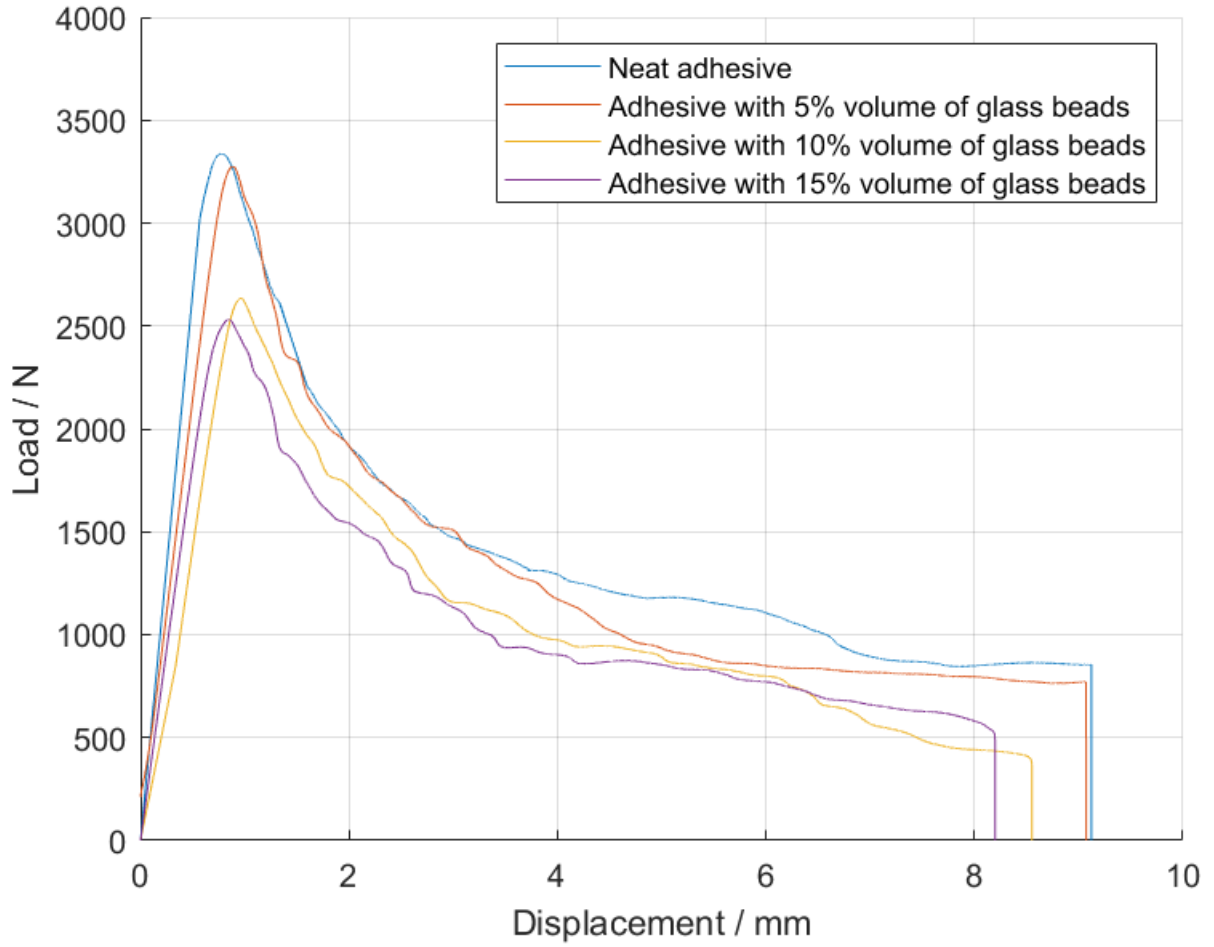


Figure 24: Load displacement curves of Adhesive A recorded using universal testing machines.

Based on the $P - \delta$ curves, and using CBBM, the respective R-curves were determined, and consequently the critical energy release rates in mode I were obtained. The R-curves are represented in Figure 25, for the neat Adhesive A, and with the addition of 5, 10 and 15% volume of glass beads, respectively

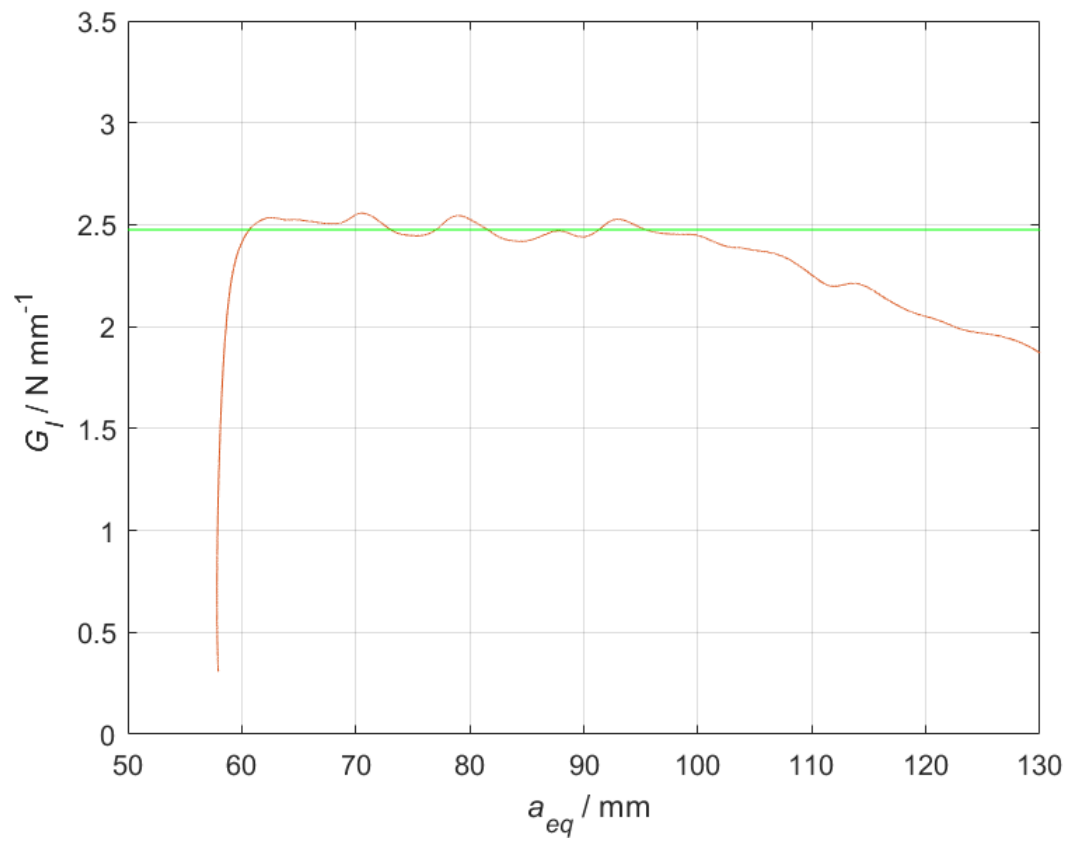
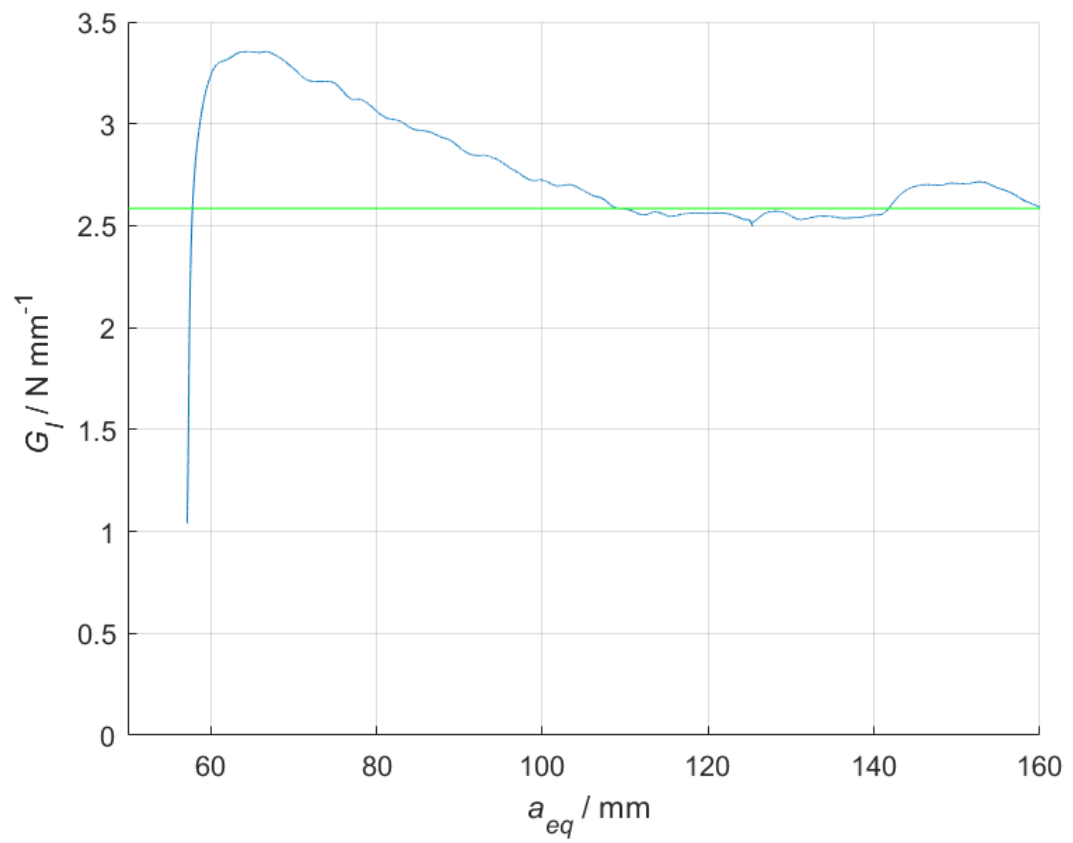


Figure 25: Representative R-curves for Adhesive A.

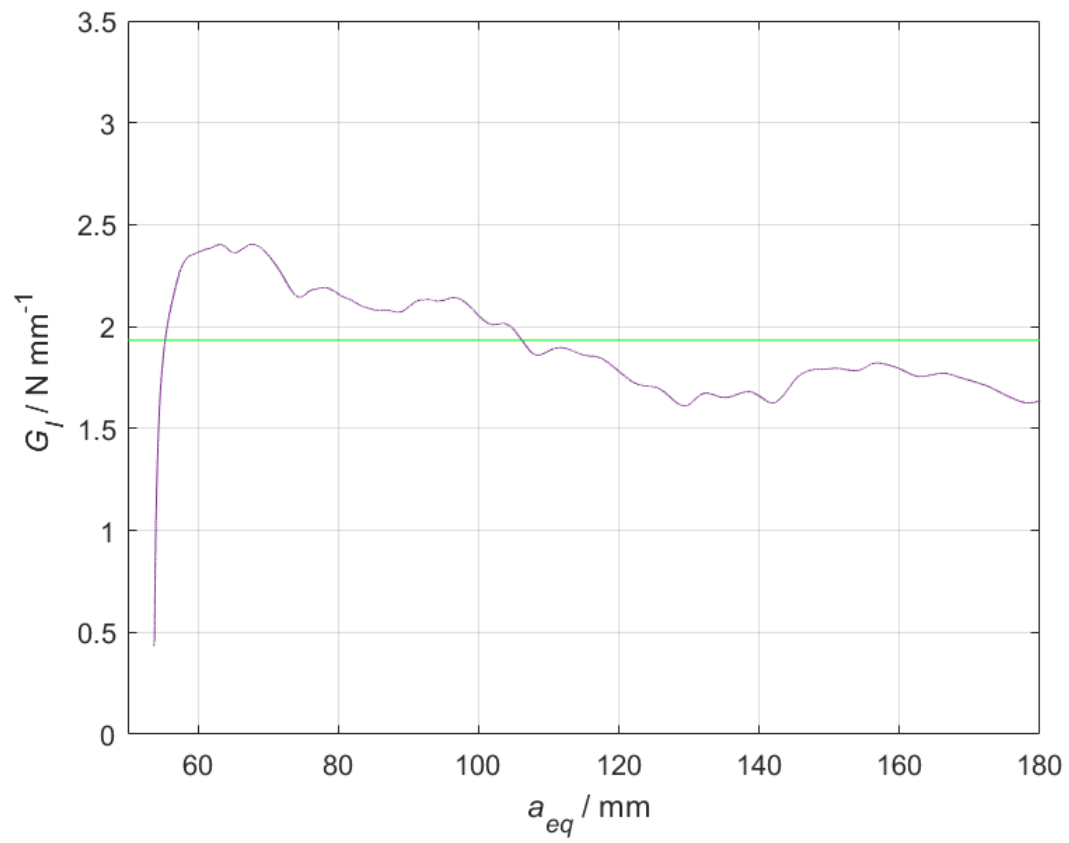
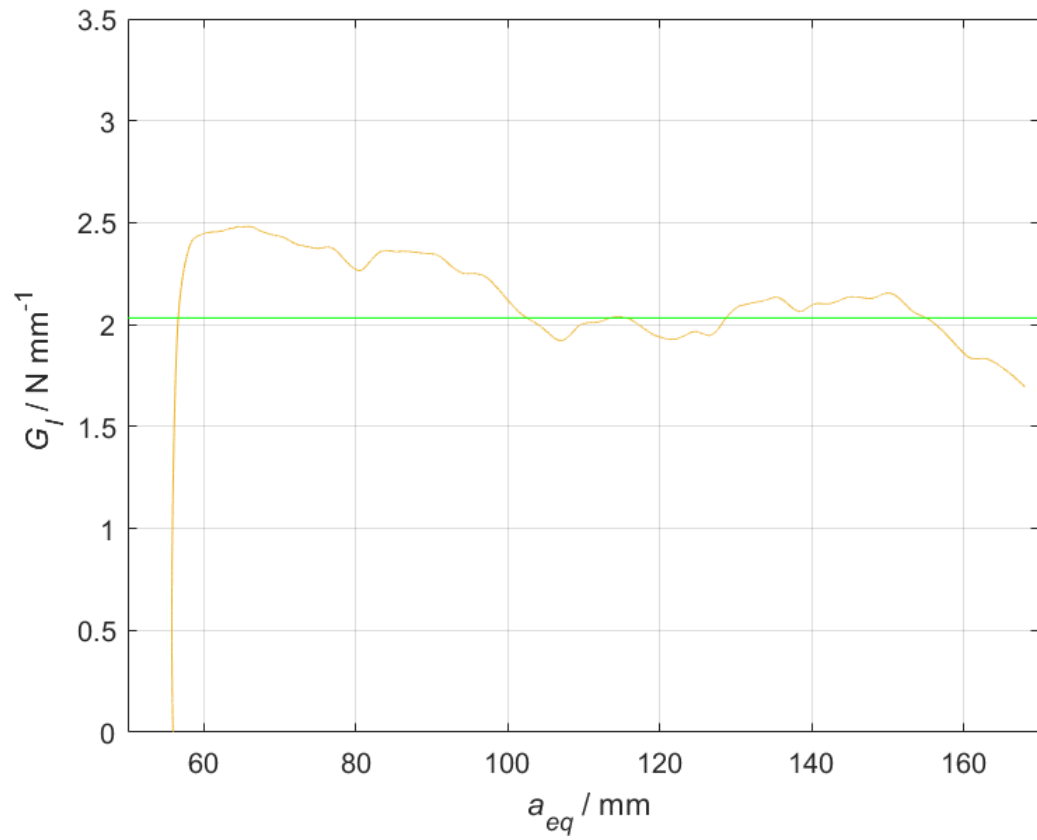


Figure 25: Representative R-curves for Adhesive A (cont.).

The results obtained regarding the critical energy release rate in mode I can be seen in Figure 26.

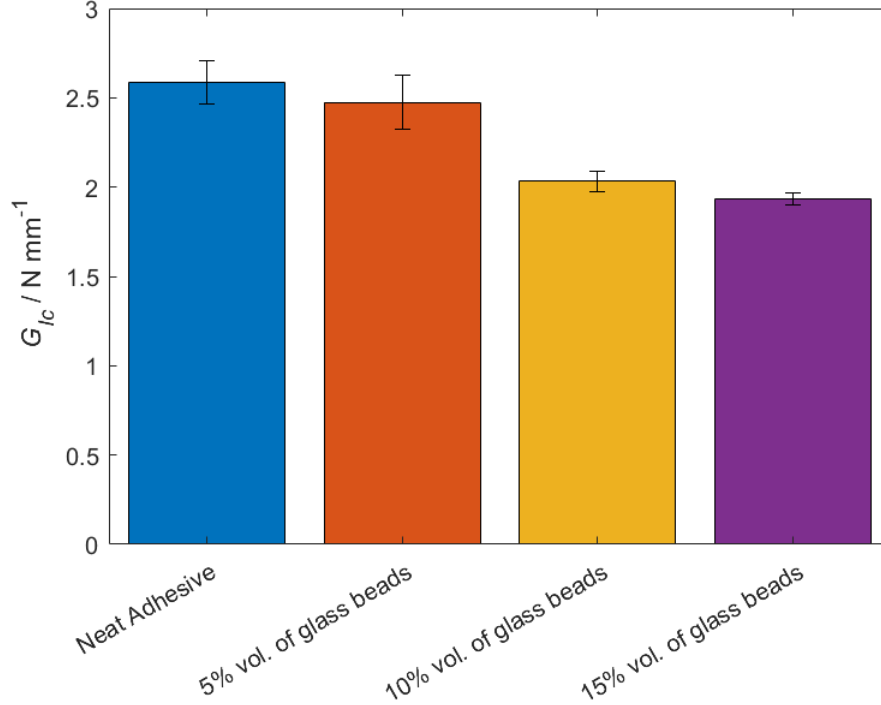


Figure 26: Critical energy release rates in mode I, for Adhesive A.

Similarly to what was reported in the work of Forte et al. [10] regarding the influence of fiber-glass fibers in the adhesive layer, there is an overall decrease of the critical energy release rate in mode-I. However, for this case, the critical energy value seems to decrease progressively with an increasing amounts of glass beads in the adhesive layer. Therefore, and by analysing the previous figure (Figure 26), there seems to be a very significant impact of the glass microspheres' presence in terms of the adhesive's ductility. The introduction of such particles into the adhesive clearly makes it less tough, which further contributes to the conclusion the glass particles work as weakening agents rather than reinforcements. In a nutshell, the presence of the glass beads decreases the adhesive's capacity of absorbing energy, having a negative impact in its mechanical properties.

Furthermore, and according to the schematic diagram represented in Figure 11, experimental tests were performed on DCB specimens with Adhesive B. The associated P - δ curves are shown below, in Figure 27.

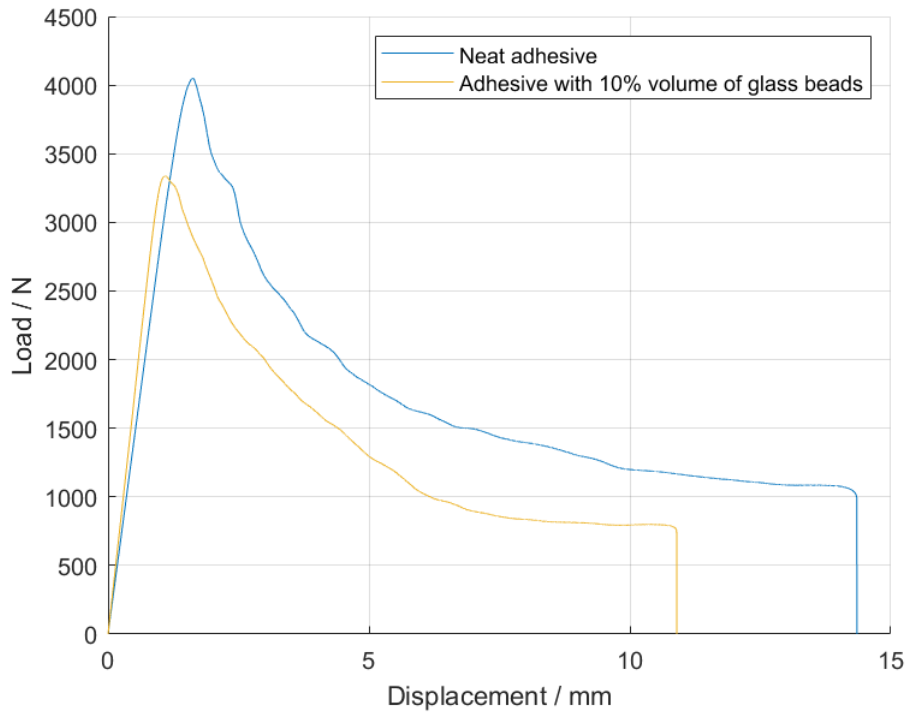


Figure 27: Load displacement curves of Adhesive B recorded using universal testing machines.

Having obtained the $P - \delta$ curves, CBBM was used to determine the corresponding R-curves and the critical energy release rate in mode I, Figure 28.

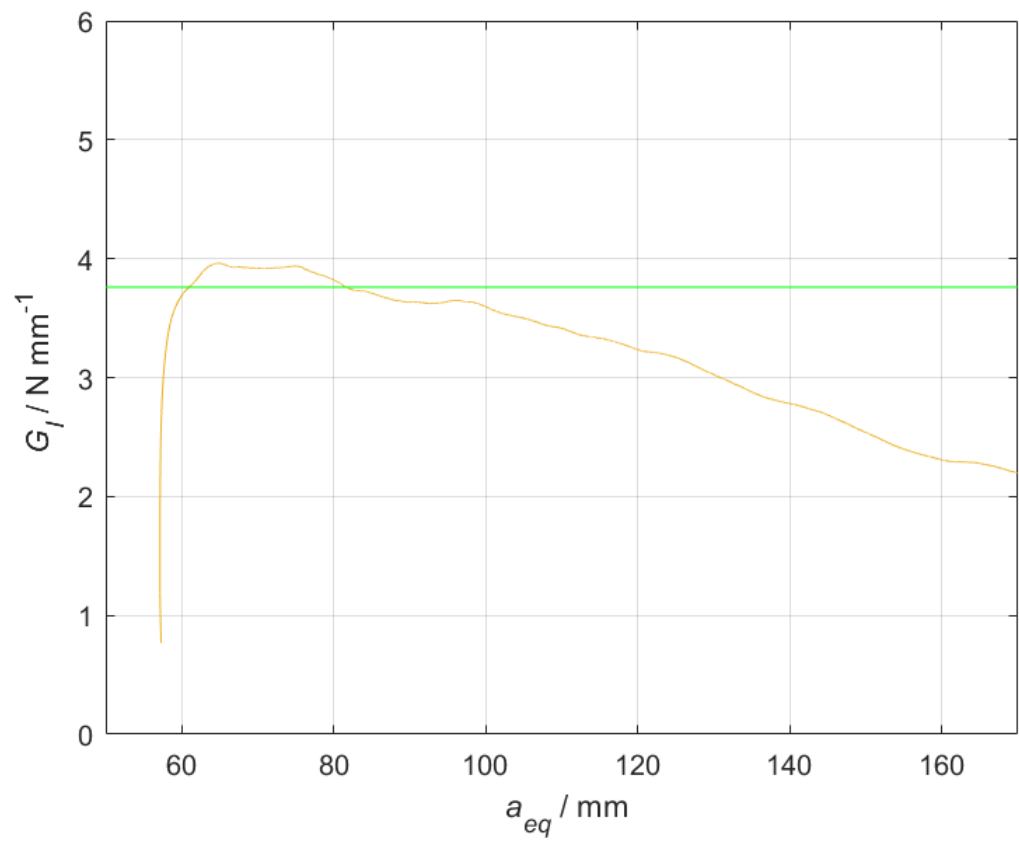
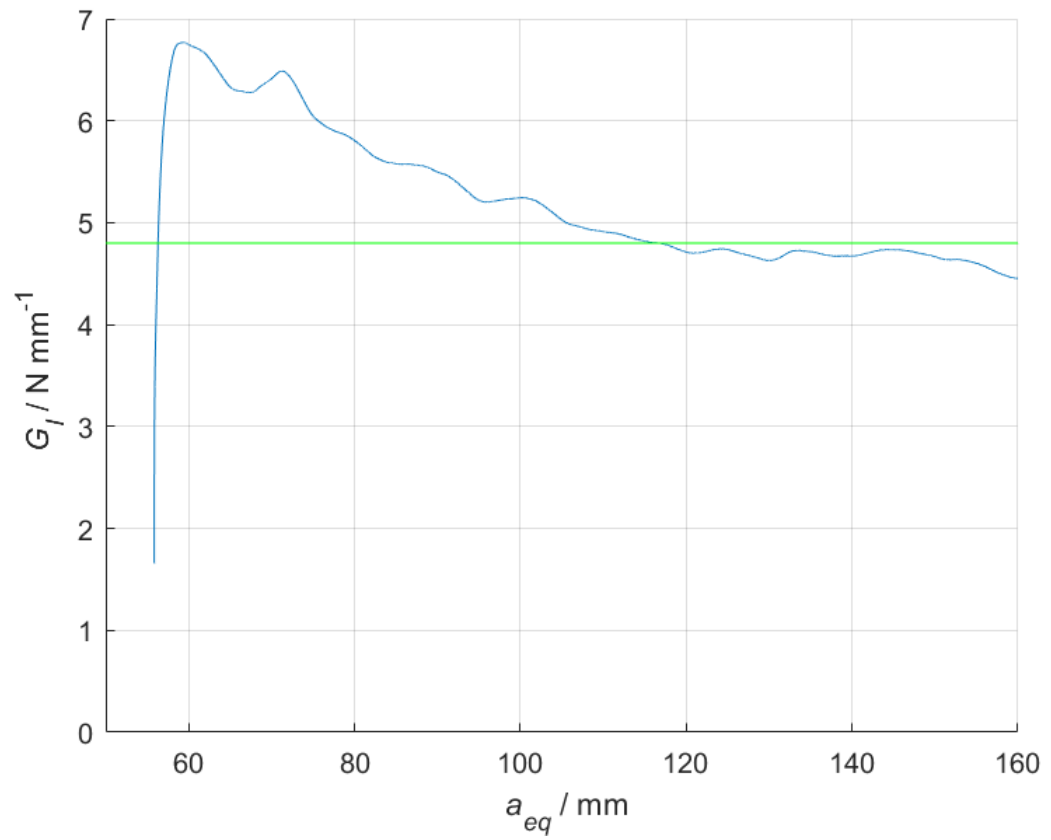


Figure 28: Representative R-curves for Adhesive B.

Below follow the results obtained regarding the critical energy release rate in mode I, Figure 29.

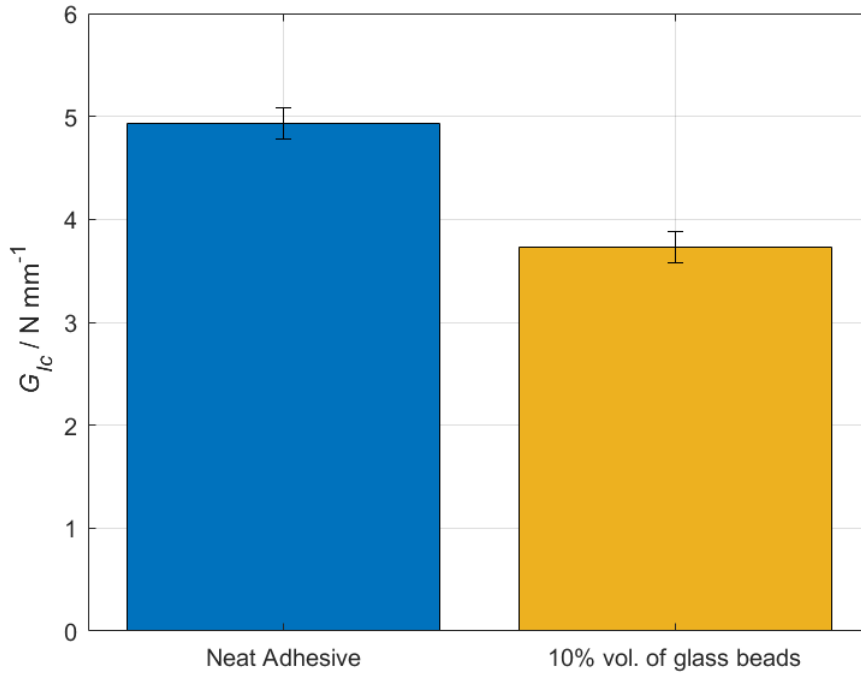


Figure 29: Critical energy release rates in mode I, for Adhesive B.

The previous results reveal that the effect of adding hollow glass spheres into the adhesive layer is rather similar to the one that had been encountered for Adhesive A, since one can surmise that, one more time, the adhesive's fracture toughness, represented by G_{Ic} , worsens with the introduction of glass beads into the adhesive layer, which is consistent with the tensile test results.

A.3.3 SEM analysis

Firstly, the hollow glass beads size and shape was thoroughly scrutinized.

Regarding their shape, and by looking at Figure 30, one can observe that they clearly have an external rounded shape. By measuring their external diameter, the glass beads were found to have a range of diameter's values that vary between 6 and 60 μm .

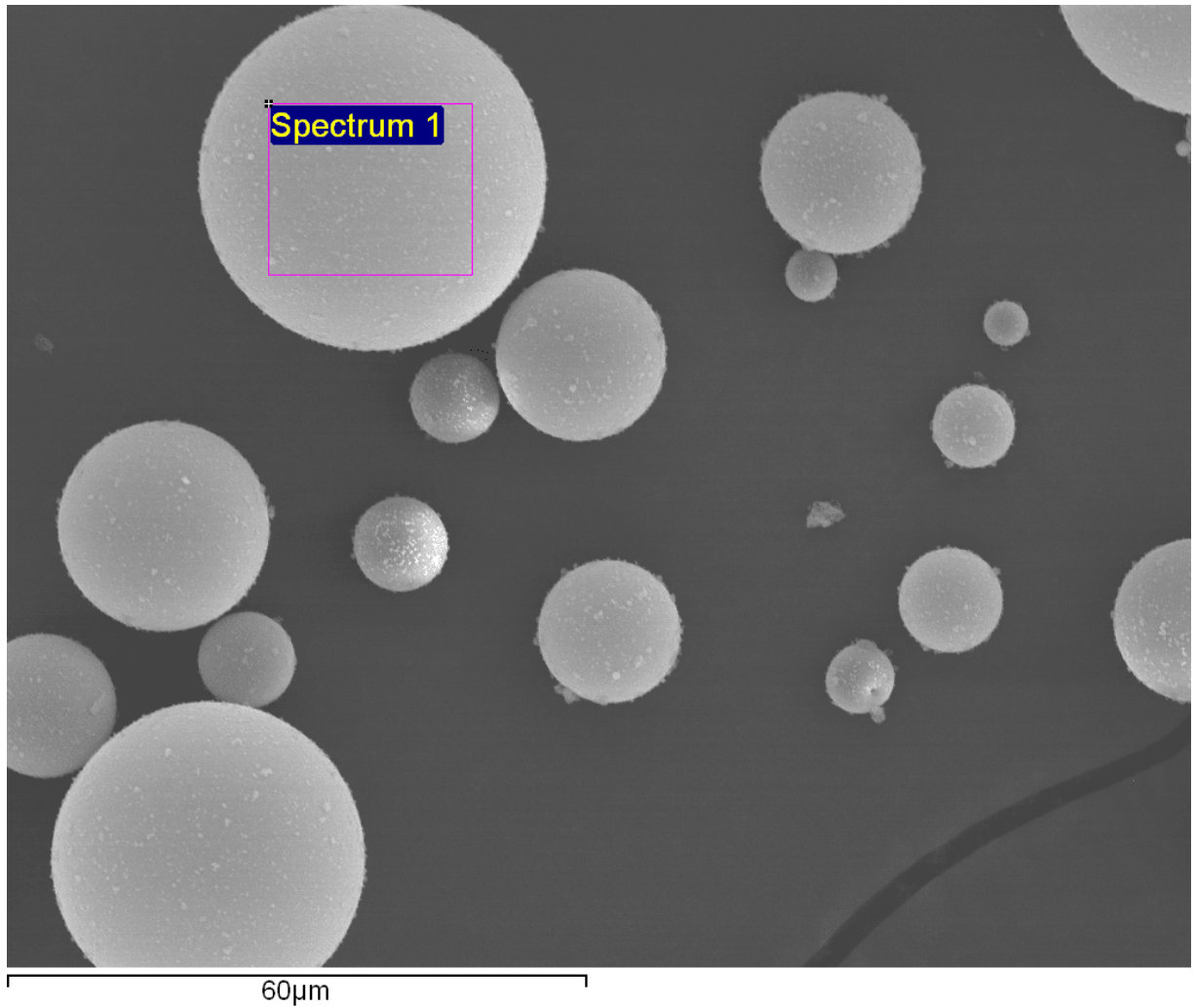


Figure 30: Glass microspheres obtained through backscatter electron analysis.

Also, it was possible to do a spectrum analysis of the hollow glass beads configuration, which provided the composition spectrum that is represented in Figure 31.

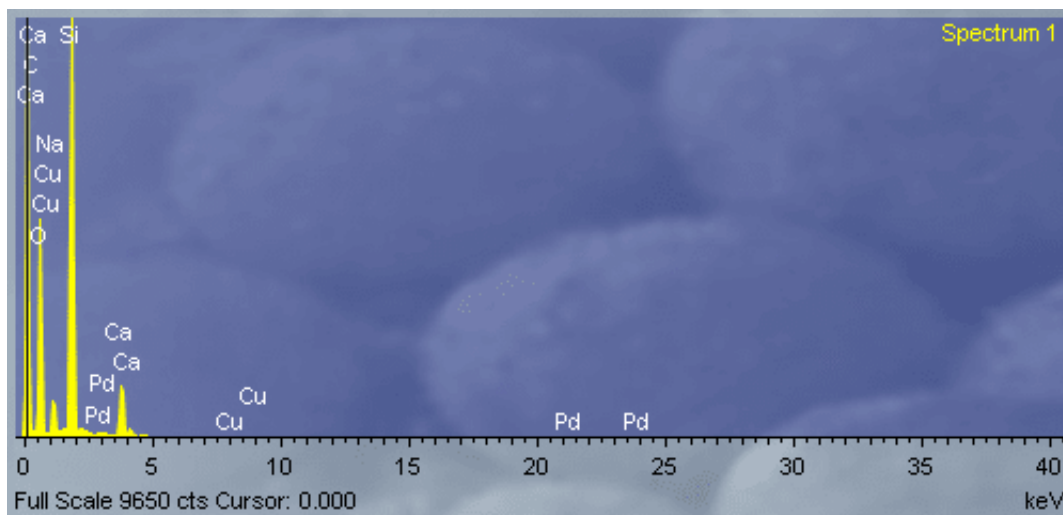


Figure 31: Glass microspheres' composition.

By interpreting the previous spectrum, it can be stated that the particles are mainly composed by Silicate, Sodium and Calcium. Moreover, the particles' coating is composed by Copper and Paladium.

After testing the bulk specimens, their fracture surfaces were analysed through SEM. One of the analysed fracture surfaces is shown below, more specifically one corresponding to the bulk specimens with 10 % volume of glass beads, Figure 32.

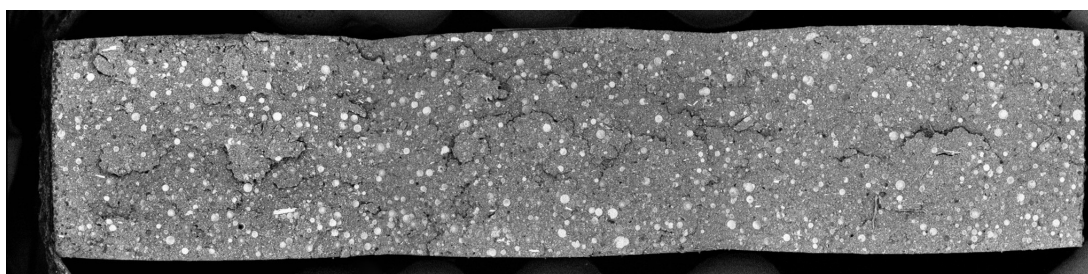


Figure 32: Fracture surface of a bulk specimen of Adhesive A with 10 %volume of glass beads added, obtained through backscatter electron analysis

By doing a more careful observation, specifically at the regions near the glass particles, Figure 33, a very good level of bonding between them and the adhesive's matrix was noticed. Additionally, some of the glass microspheres were completely shattered, and there was no evidence of a particular damage mechanism taking place as a consequence of their presence. Therefore, it is pertinent to conclude that the hollow glass beads do not work as a toughening mechanism. Instead, they weaken the adhesive, since they appear to be in a severely damage state, which is a consequence of their brittleness and lower strength than the adhesive. Without having the capacity of absorbing energy and

without inducing toughening mechanisms as the ones described previously, their presence only contributes to the generation of stress concentrations, representing weak links in the adhesive's matrix.

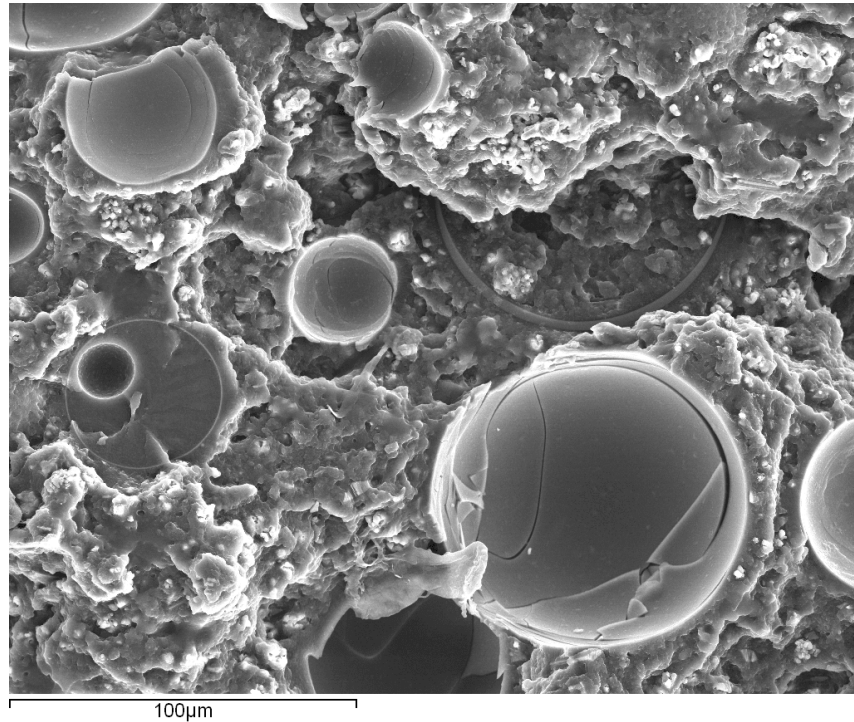


Figure 33: Fracture surface of a bulk specimen with 10 %volume of glass beads added to Adhesive A, with focus on the region around the glass beads, obtained through secondary electron analysis.

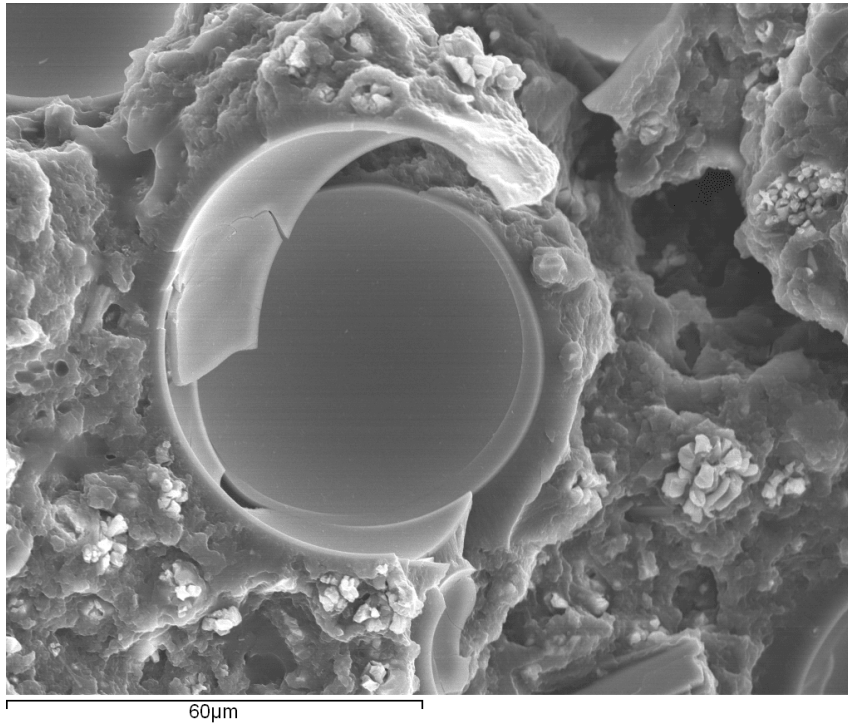


Figure 33: Fracture surface of a bulk specimen with 10 %volume of glass beads, with focus on the region around the glass beads, obtained through secondary electron analysis (cont.).

Additionally, the bulk's fracture surfaces were also analysed for the Adhesive B specimens. In Figure 34, one can observe the fracture surface of a bulk specimen with neat Adhesive B.

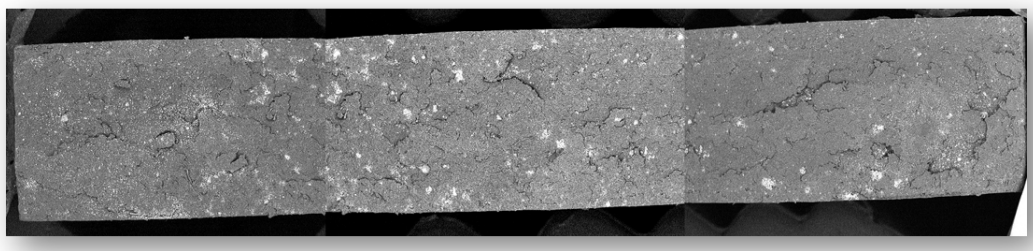


Figure 34: Fracture surface of a bulk specimen with neat Adhesive B.

By using a higher magnification level, it is possible to notice the presence of fillers, uniformly spread along the adhesive's matrix, which can be recognized by their lighter color tone in the backscatter electron images represented in Figure 35.

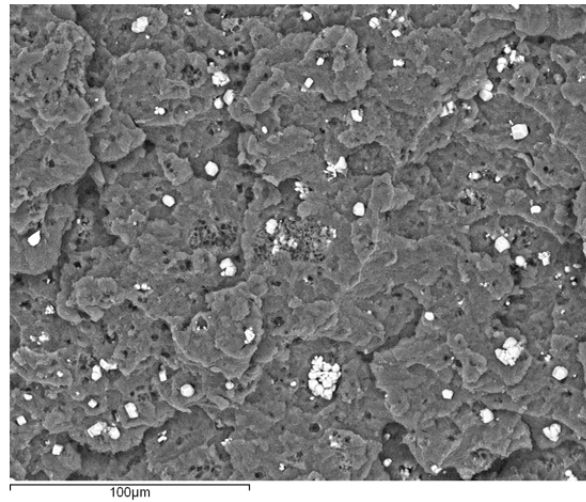


Figure 35: Fracture surface of a bulk specimen with neat Adhesive B, obtained through secondary electron analysis.

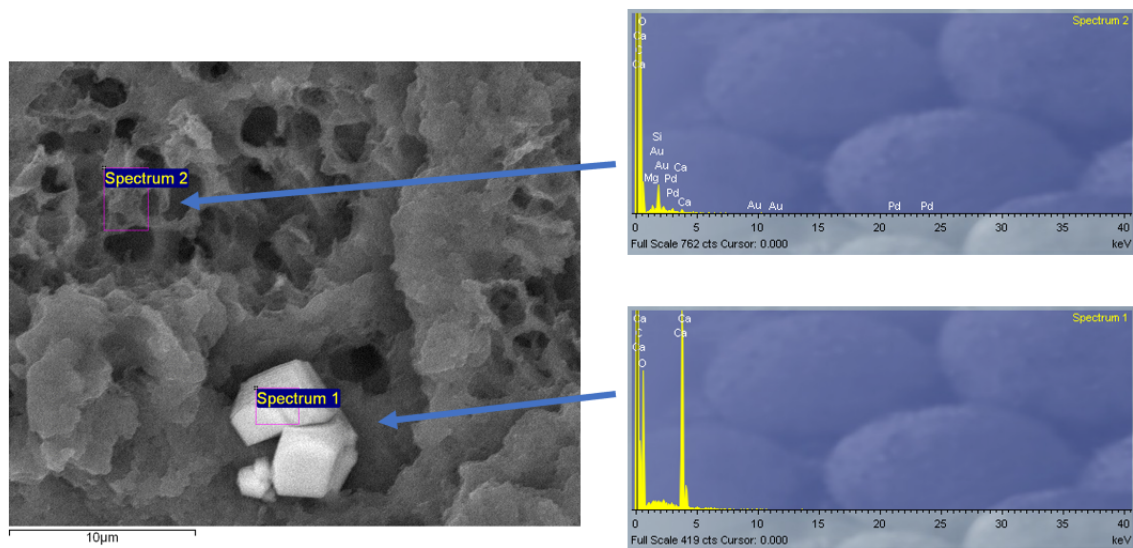


Figure 36: Spectrum analysis of the fracture surface of a bulk specimen with neat Adhesive B, obtained through secondary electron analysis.

The information presented in Figure 36 reveals the presence of fillers composed of calcium carbonate, as one can see in spectrum 1, mixed within the adhesive's matrix, which is mainly composed of carbon, oxygen and silica. Calcium carbonate is one of the most widely used mineral filler used in polymers, since it naturally exists in large amounts, and has very satisfactory properties to be used as a filler, along with a low cost [19].

A.3.4 Summary of the influence of particle content on adhesive properties

Having determined both the strength and the fracture properties for Adhesive A and Adhesive B with the different amounts of hollow glass beads, it becomes possible to display an overall panorama of their evolution. To that end, Figures 37 and 38 represent the variations of those properties as a function of the glass particle content, for Adhesive A and Adhesive B, respectively.

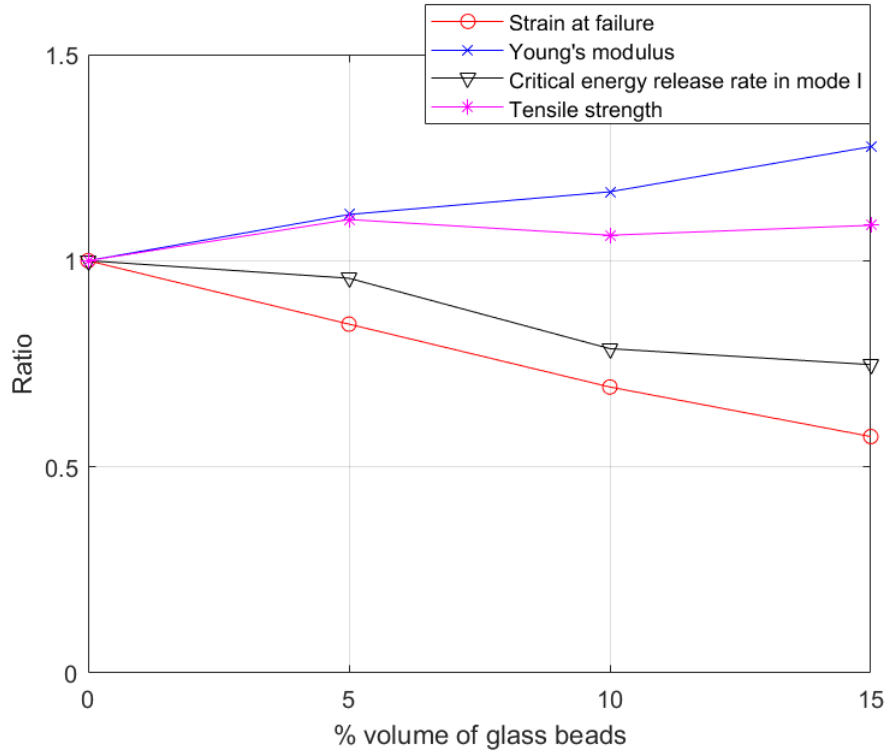


Figure 37: Variation, in ratio, of the fracture and strength properties with the growing % volume of added glass microspheres, for Adhesive A.

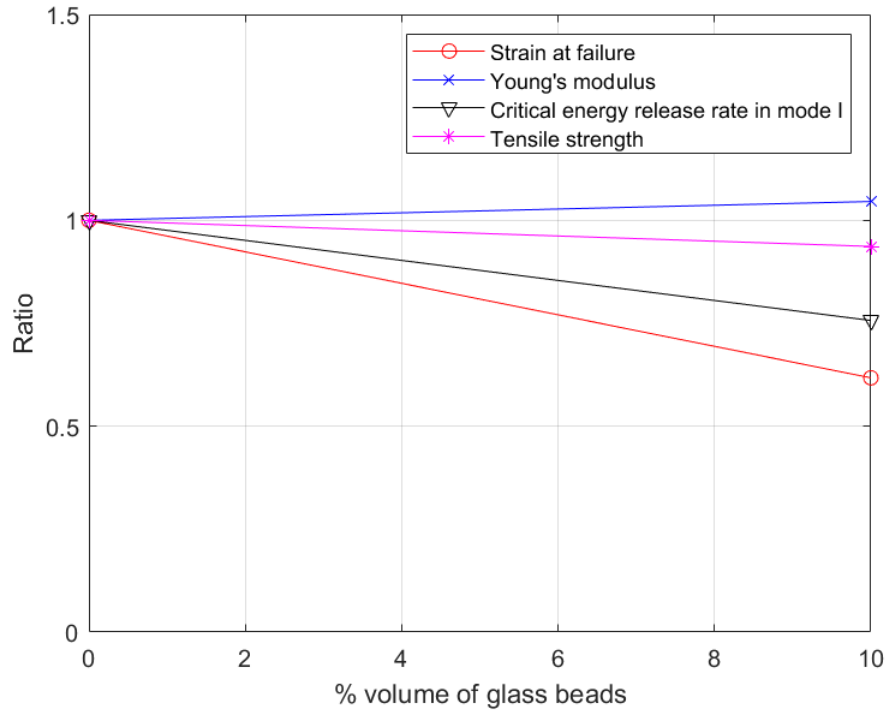


Figure 38: Variation, in ratio, of the fracture and strength properties with the increasing %volume of glass microspheres, for Adhesive B.

As one can conclude by taking into consideration the information reflected in Figures 37 and 38, the introduction of hollow glass beads into the adhesive layer has similar effects on both adhesives.

In terms of the strength properties, the influence of the microspheres on the tensile strength is residual. On the other hand, the strain at failure and the Young's modulus show a clear trend with the increasing amounts of hollow glass beads, being the first subjected to a gradual decrease, while the second generally increases. Therefore, it can be stated that adding glass beads to the adhesive makes it more brittle.

Regarding the fracture properties, there is a consistent decrease of the critical energy release rate in mode I, which translates into a decreasing capacity of the adhesive to absorb energy with increasing amounts of glass particles.

Therefore, it can generally be said that the addition of glass beads to the adhesive weakens it. Instead of working as a toughening mechanism, it reduces the adhesive's capacity of absorbing energy and turns it into a more brittle material.

A.4 Linear approximation of the mechanical property evolution as a function of percent volume of hollow glass beads

To support the prediction of the effect of introducing hollow glass beads into the adhesive layer, the strength and fracture properties were plotted as a function of the percent volume of microspheres, and a trend line was created to fit the evolution of these properties.

A.4.1 Strength properties

As it was concluded in section A.3.1, the maximum stress does not reveal a clear variation with the addition of glass beads to the adhesive. Nonetheless, and when it comes to the strain at failure and the Young's modulus, these evidently change with increasing larger amounts of particles. Therefore, the establishment of a relationship between these two properties and the percent volume of glass beads gains relevance.

As a way of obtaining a versatile equation which enables the prediction of the influence of glass particles for other different adhesives, the obtained functions were plotted in terms of ratios, which portrays the fractional evolution of the maximum strain and the Young's modulus values.

With this in mind, Figures 39 and 40, and Figures 41 and 42 reflect how the maximum strain and the Young's modulus vary with the growing amounts of hollow glass spheres, respectively, for Adhesive A.

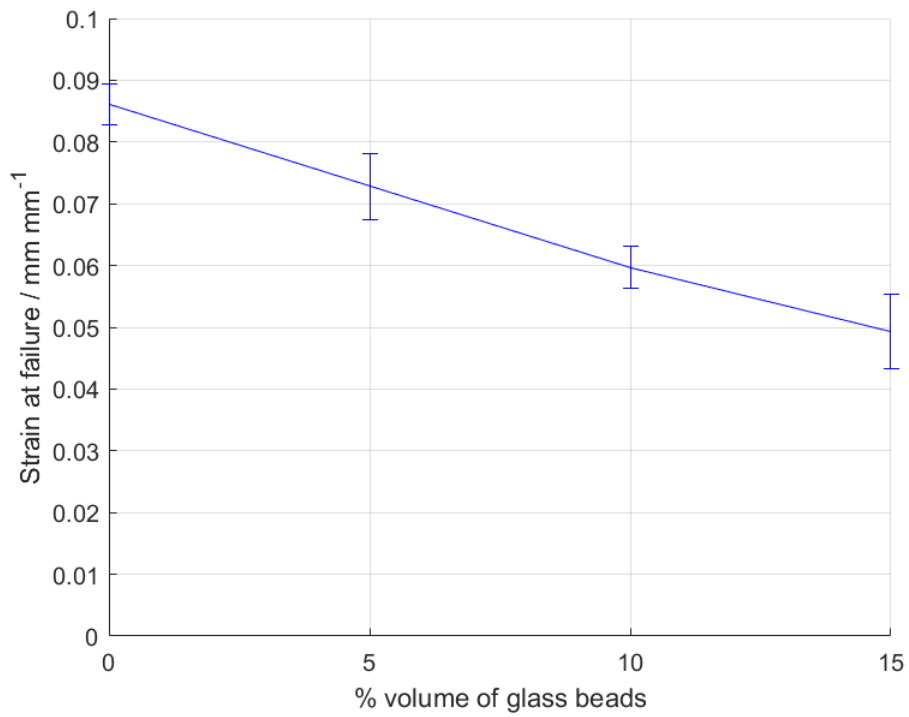


Figure 39: Determination of the maximum strain as a function of the % volume of hollow glass beads.

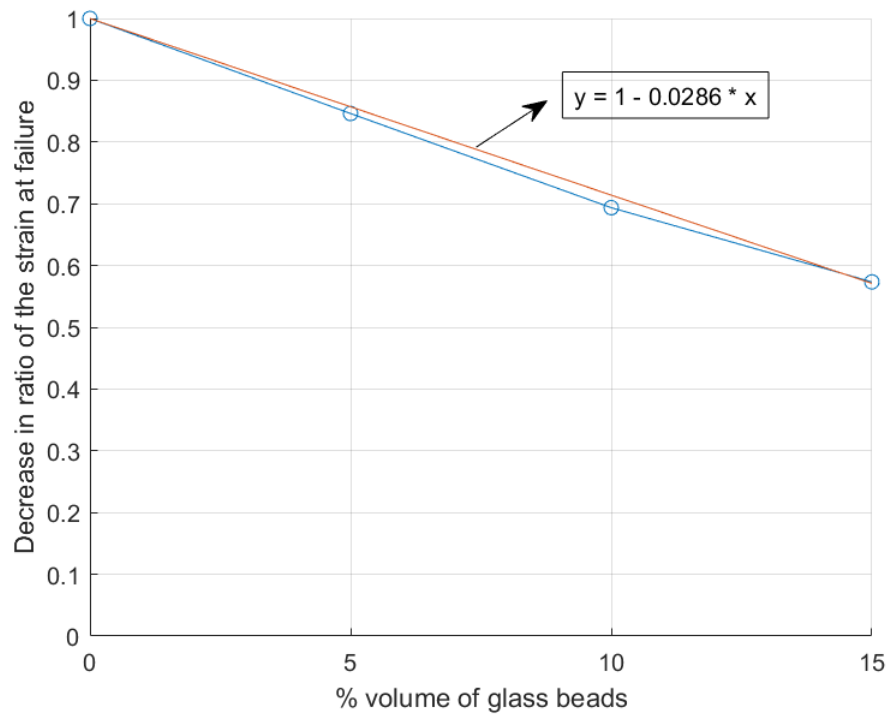


Figure 40: Decrease, in ratio, of the maximum strain as a function of the added % volume of glass beads.

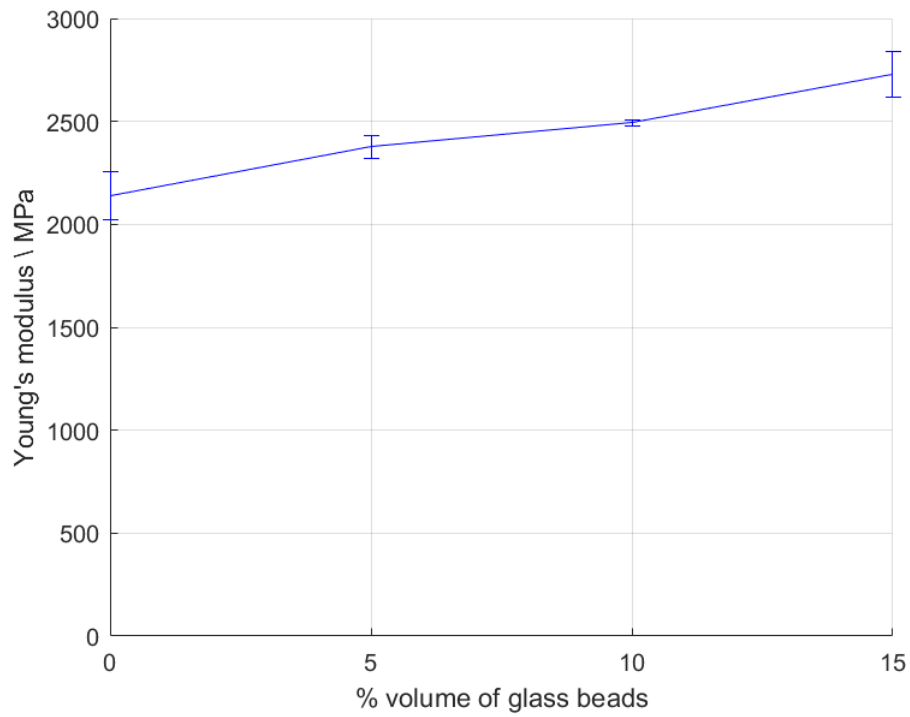


Figure 41: Determination of the Young's modulus as a function of the added % volume of hollow glass beads.

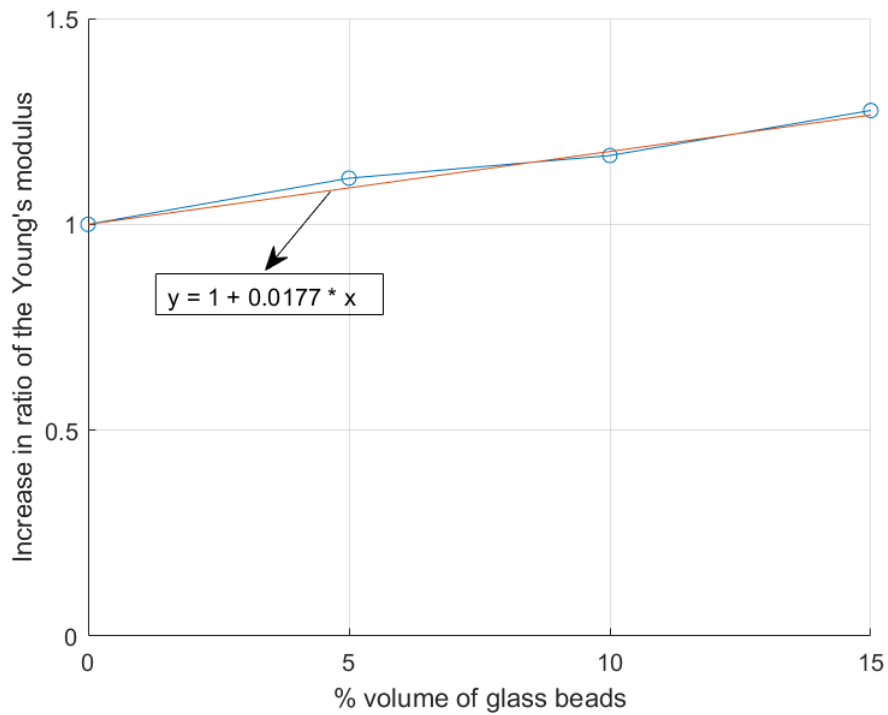


Figure 42: Increase, in ratio, of the Young's modulus as a function of the added % volume of glass beads.

Specifically focusing on Figures 40 and 42, one can state that the trend lines obtained, which are linear approximations of the experimentally obtained results regarding the maximum strain and Young's modulus ratio's evolution, represent significantly accurate estimations of the real values, since the trend lines do not move far away from the experimental values.

As a way of testing the applicability of the trend lines obtained for Adhesive A to predict the strength properties evolution of other adhesives with the introduction of hollow glass beads, Figures 43 and 44 exhibit the comparison between the experimental and the predicted values of the strain at failure and the Young's modulus, respectively, for Adhesive B with 10 % volume of glass beads, and also presents the experimental results for neat Adhesive B.

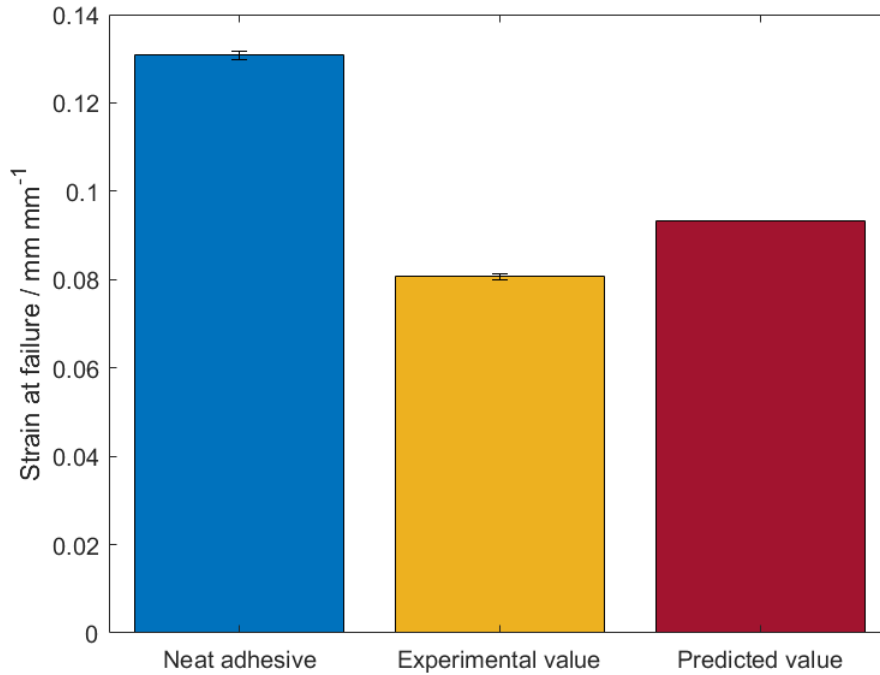


Figure 43: Comparison between the experimental value and the predicted value of the strain at failure for Adhesive B with 10 % volume of glass beads.

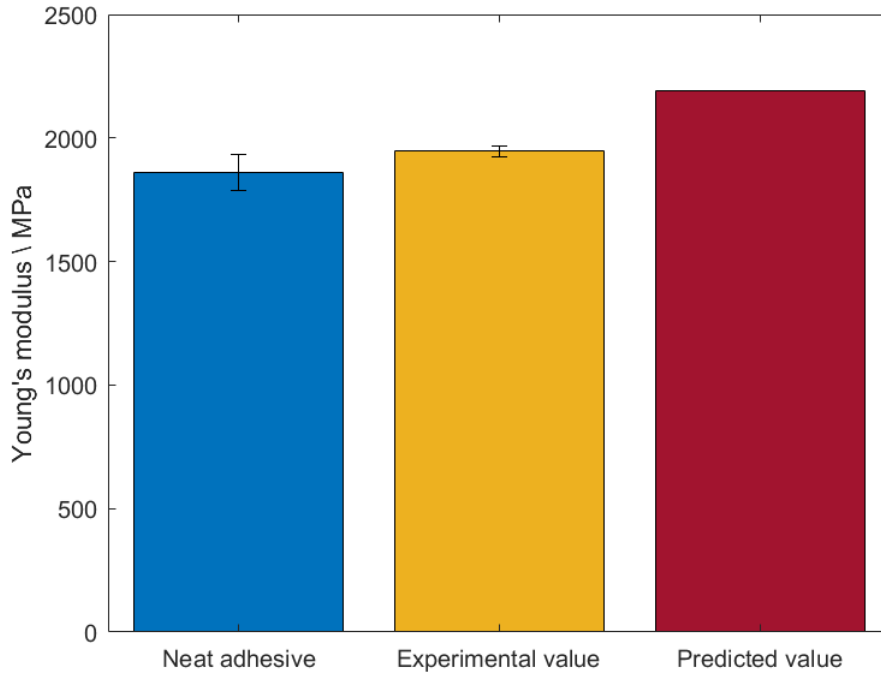


Figure 44: Comparison between the experimental value and the predicted value of the Young's modulus for Adhesive B with 10 % volume of glass beads.

The comparison between the predicted and the experimental values exhibited in Figures 43 and 44 prove that the equations extracted from the experimental data provide reliable approximations of the effects caused by the presence of hollow glass beads in the adhesive layer. The predicted values not only globally reflect the weakening effect caused by their presence, but also have satisfactory low relative errors associated to them, Table 2.

Table 2: Errors associated to the predicted values.

Property	Error (%)
Strain at failure/ mm/mm	13.5
Young's modulus	12.44

A.4.2 Fracture properties

Energy release rate as a function of the percent volume of hollow glass beads. The previously mentioned DCB tests, which provided not only the Load vs

Displacement curves, but also the R-curves, allowed to determine the critical energy release rates in mode I for the different amounts of hollow glass beads. Afterwards, these same experimental values were plotted as a function of the percent volume of hollow glass beads, for Adhesive A, Figure 45.

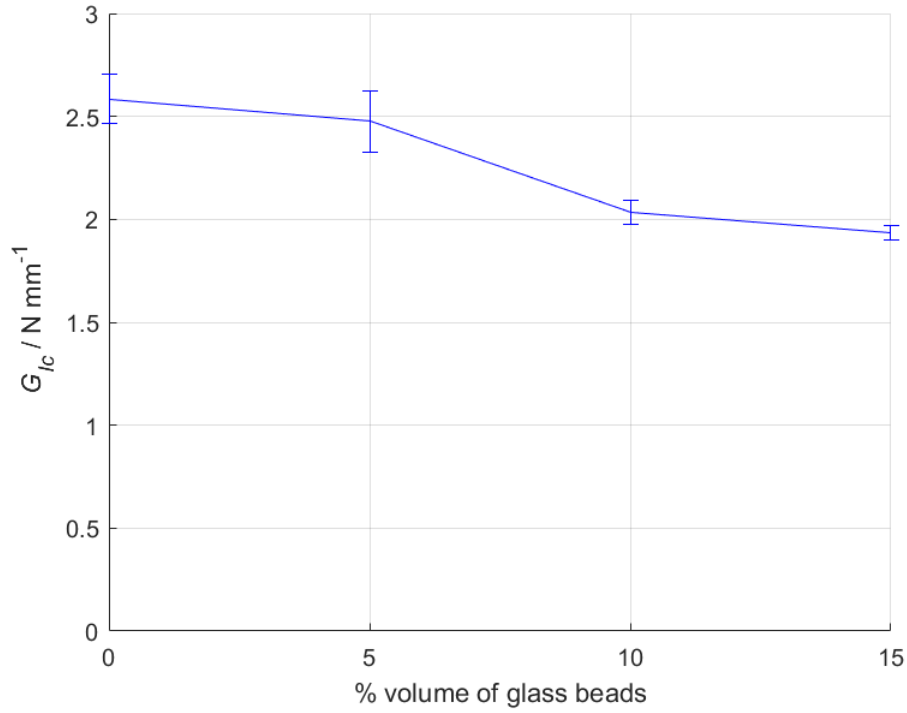


Figure 45: Determination of G_{IC} as a function of the added % volume of hollow glass beads.

Again, and in order to obtain a versatile equation that can be used not only for this adhesive, but also for others, a decrease in ratio of the G_{Ic} values was calculated. Then, these were plotted as a function of the % volume of glass beads, and a trend line was obtained, Figure 46.

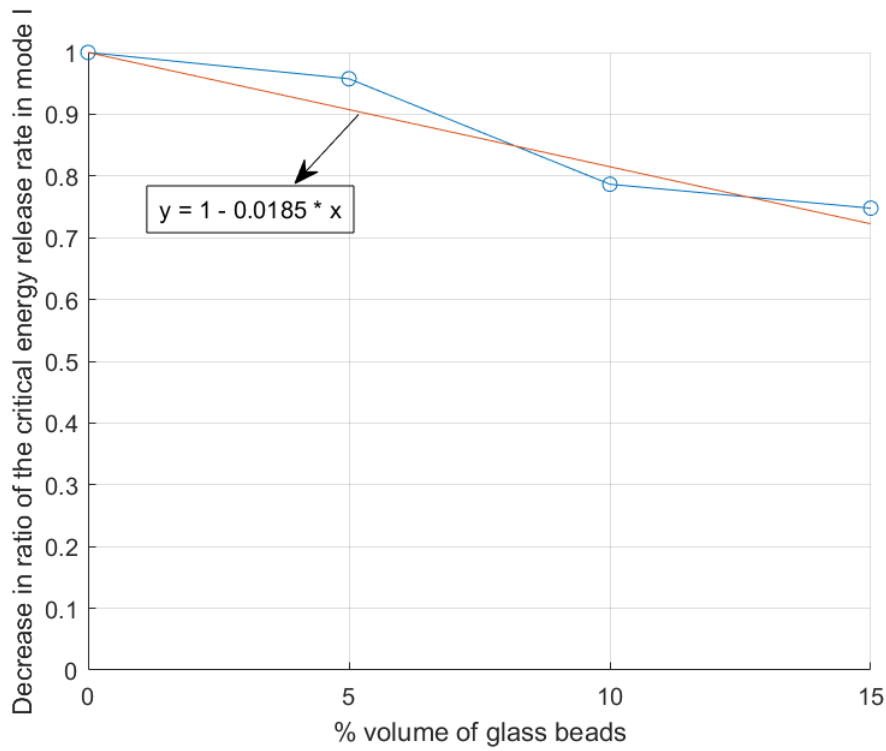


Figure 46: Decrease, in ratio, of the critical energy release rate in mode I as a function of the % volume of glass beads.

The equation that was obtained with respect to the trend line resembles a legitimate approximation of the critical energy release rate's evolution in mode I with the varying amount of glass beads. Nonetheless, and as a way of testing this function's suitability for other cases, the equation was used to predict the G_{Ic} value for Adhesive B with 10 % volume of glass beads. Figure 47 compares the predicted value to the one that was obtained experimentally for Adhesive B with 10 % volume of glass beads, which was already shown in Figures 28 and 29, and also presents the experimental results for neat Adhesive B.

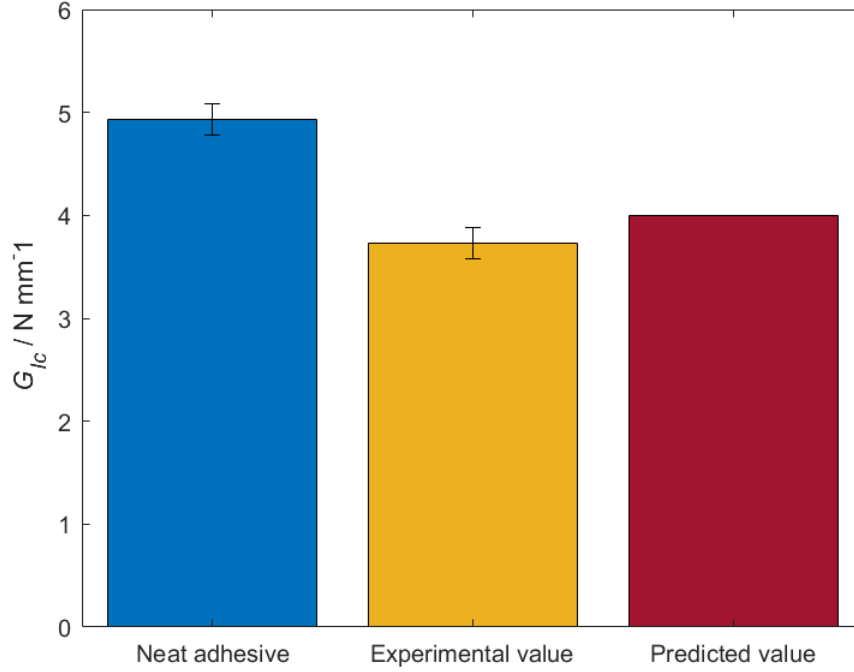


Figure 47: Comparison between the experimental value and the predicted one of G_{Ic} for Adhesive B with 10 % volume of glass beads.

By comparing these two values, it becomes possible to conclude that the G_{Ic} value estimated by the function obtained in Figure 46 provides a good approximation of the experimentally retrieved value. Therefore, one can conclude that, in terms of the critical energy release ratio, the two adhesives show similar response to the introduction of glass beads into the adhesive layer, with the energy absorption's capability of the adhesives negatively impacted by the presence of particles. Besides, the predicted value also exhibits a satisfactory relative error, when compared to the experimental value, Table 3.

Table 3: Error associated to the predicted value.

Property	Error (%)
$G_{Ic} / \text{N mm}^{-1}$	7

A.5 Rule of mixtures

The rule of mixtures was also put into practice during this study, in order to estimate the effective mechanical properties of the adhesive in terms of its constituents, and also as

a way of extracting approximate values of the hollow glass beads properties. This method is a very simple and intuitive one, and the concept that is inherent to it can be applied for composites with particles along their matrix [20].

With this in mind, this rule was applied to Adhesive A, and having extracted the glass beads properties, these were used to estimate the mechanical properties of Adhesive B depending on the particles amount.

A.5.1 Young's modulus

In order to apply the rule of mixtures to the Young's modulus of Adhesive A, Reuss model was used [21], which is ruled by the following equation, adapted for this case:

$$E = \%vol_a \cdot E_a + \%vol_{gp} \cdot E_{gp} \quad (2)$$

where $\%vol_a$ and $\%vol_{gp}$ are the volume fractions of neat adhesive and hollow glass particles, and E_a and E_{gp} are the Young's modulus of neat adhesive and hollow glass particles, respectively.

By fitting this model's equation to the Young's modulus values of Adhesive A with the different configurations, the rule represented in Figure 48 was obtained.

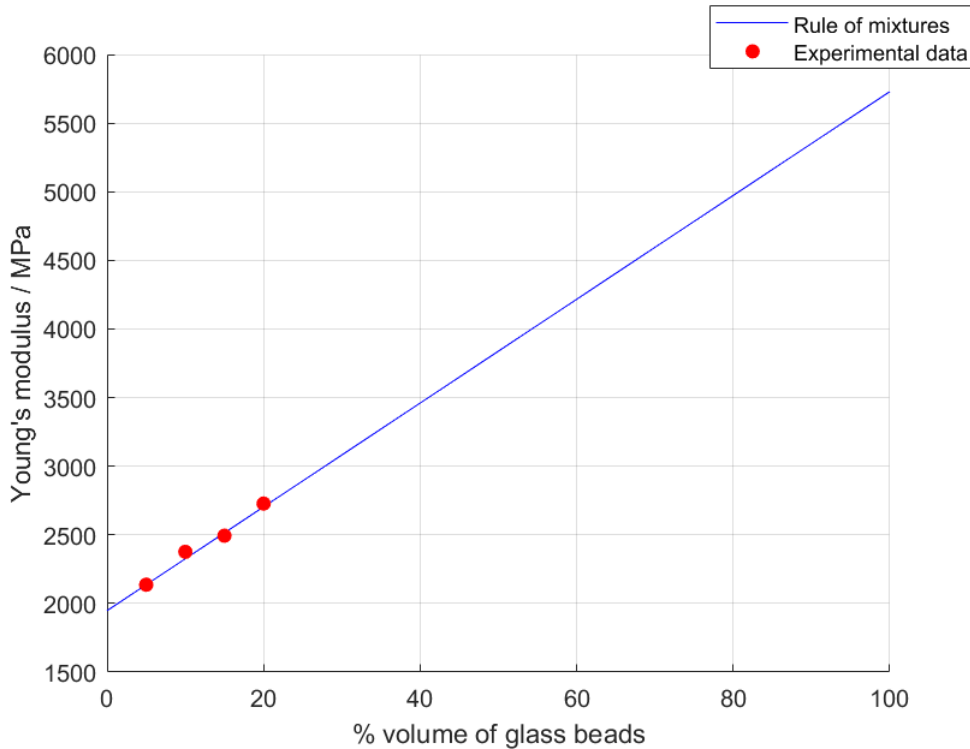


Figure 48: Young's modulus of Adhesive A as a function of the volume fraction of hollow glass beads.

Through this model, and taking into account the fact that the neat Adhesive A already has 5 % volume of glass particles, the following values were obtained regarding the Young's modulus of Adhesive A without glass particles within its matrix, and the Young's modulus of hollow glass beads, 4.

Table 4: Young's modulus of Adhesive A without hollow glass beads within its matrix, and the Young's modulus of hollow glass beads.

Constitute	Young's modulus MPa
Adhesive A without hollow glass beads	1947.5
Hollow glass beads	5729.3

Specifically focusing on the Young's modulus obtained for the hollow glass beads, this value was compared with a previous study, conducted by Verweij et al. [22], which focused on studying the properties of hollow glass particles. Doing so led to the conclusion that the estimation derived by applying the rule of mixtures was within the range of values that had been obtained in this study.

Afterwards, the Young's modulus of the glass beads was used to apply the Reuss model to Adhesive B. Doing so led to the following rule, represented in Figure 49.

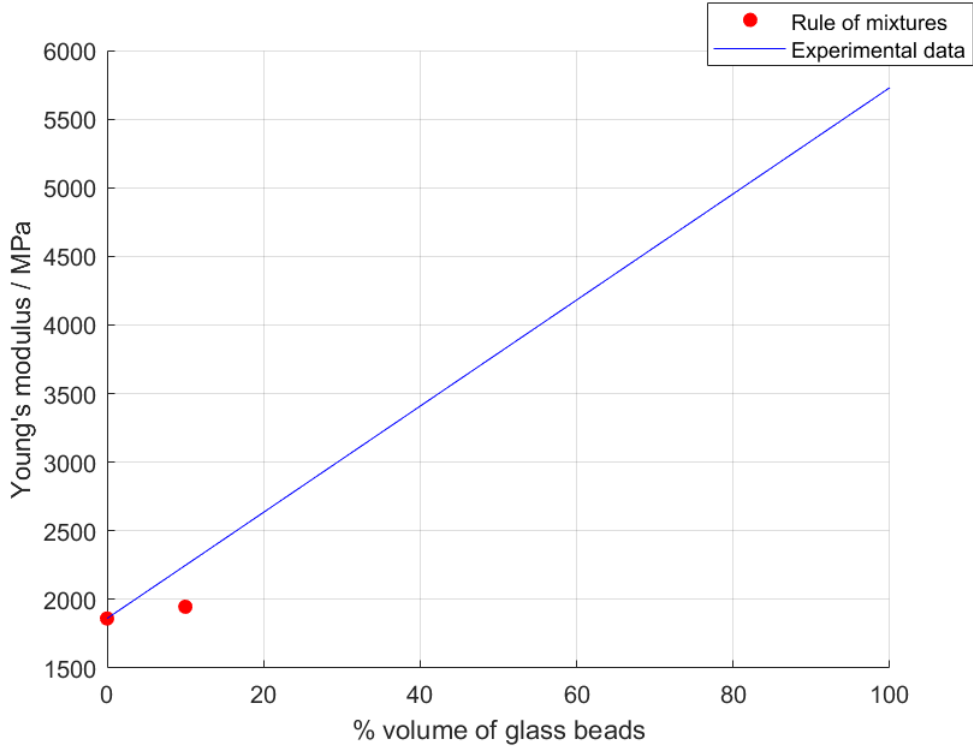


Figure 49: Young's modulus of Adhesive B as a function of the volume fraction of hollow glass beads.

By looking at these results, one can conclude that, even though the Young's modulus of Adhesive B increases with the increasing number of glass particles, this increase is not as pronounced as the one reported for Adhesive A.

A.5.2 Critical energy release rate in mode I

In order to apply the rule of mixtures to the G_{Ic} of Adhesive A, Voigt model was used [23], which is associated to the equation below, adapted for this case:

$$G_{Ic} = \left(\frac{\%vol_a}{G_a} + \frac{\%vol_{gp}}{G_{gp}} \right)^{-1} \quad (3)$$

where G_a and G_{gp} are the critical energy release rate in mode I of neat adhesive and hollow glass particles, respectively.

By fitting this model's equation to the critical energy release rates in mode I of Adhesive A with the different configurations, the rule represented in Figure 50 was obtained.

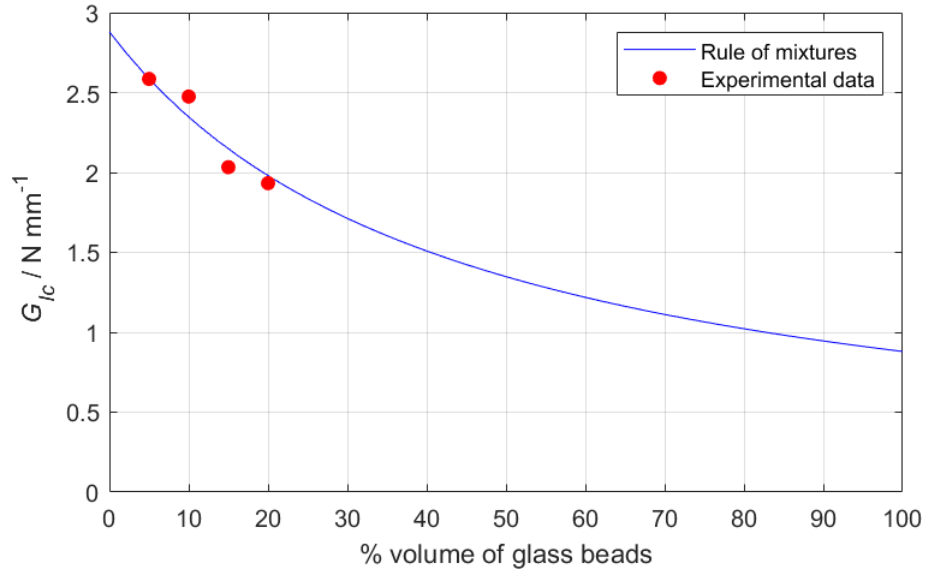


Figure 50: G_{Ic} of Adhesive A as a function of the volume fraction of hollow glass beads.

Having applied this model, the values represented in 5 were obtained regarding the G_{Ic} of Adhesive A without glass particles within its matrix, and the G_{Ic} of hollow glass beads, .

Table 5: Young's modulus of Adhesive A without hollow glass beads within its matrix, and the Young's modulus of hollow glass beads.

Constitute	G_{Ic} N mm ⁻¹
Adhesive A without hollow glass beads	2.88
Hollow glass beads	0.88

Then, the G_{Ic} of the glass beads was used to apply the Voigt model to Adhesive B. Doing so led to the following rule, represented in Figure 51.

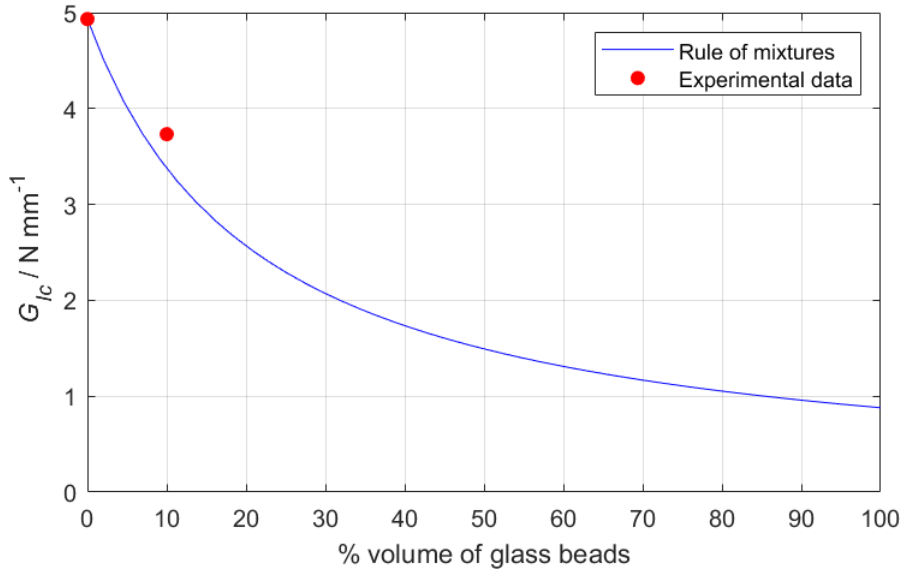


Figure 51: G_{Ic} of Adhesive B as a function of the volume fraction of hollow glass beads.

These results show that the influence of introducing glass particles into Adhesive B leads to a similar decrease tendency of the G_{Ic} as the one observed for Adhesive A, since the experimental result is relatively close to the value predicted by the use of the rule of mixtures.

A.6 Conclusions

The main aim of this work was to determine how do the strength and fracture properties of structural adhesives vary with the introduction of different amounts of hollow glass particles. For that purpose, strength tensile tests and fracture tests were carried out to mechanically characterize the adhesives. The tensile tests allowed to determine the tensile strength, the strain at failure, and the Young's modulus, while the fracture tests in mode I provided the corresponding critical energy release rate. These tests were performed at a quasi-static speed.

The main conclusions which can be extracted from the present work are the following:

- The tensile strength of the adhesives showed a negligible variation with the varying amounts of glass microspheres.
- The strain at failure exhibited a continuous decrease with the growing number of glass beads.
- The Young's modulus increased with the increasing volume occupied by the particles, which suggests that these make the adhesive more brittle.

- The critical energy release rate, G_{IC} , decreased by increasing the % volume of hollow glass spheres. Therefore, these make the adhesive noticeably more brittle and with a decreased capacity of absorbing energy.
- Hollow glass beads generally act as weakening agents of the adhesive, causing it to become a more brittle material, and with a reduced capacity of absorbing energy.
- The linear trend lines extracted from the representative ratio curves of the adhesive's properties were found to provide good approximations of the experimental values.
- The curves obtained with the rule of mixtures curves were found to work satisfactorily as methods for estimating the mechanical properties of the studied adhesives as a function of the volume fraction of hollow glass beads.

References

- [1] Lucas FM da Silva, Andreas Öchsner, and Robert D Adams. *Handbook of adhesion technology*. Springer Science & Business Media, 2011.
- [2] WA Lees. Adhesive selection. In *Adhesives in Engineering Design*, pages 92–123. Springer, 1984.
- [3] Ali Nemati Giv, Majid R Ayatollahi, S Hengameh Ghaffari, and Lucas FM da Silva. Effect of reinforcements at different scales on mechanical properties of epoxy adhesives and adhesive joints: a review. *The Journal of Adhesion*, 94(13):1082–1121, 2018.
- [4] AJ Kinloch. Toughening epoxy adhesives to meet today’s challenges. *MRS bulletin*, 28(6):445–448, 2003.
- [5] AQ Barbosa, Lucas FM da Silva, MD Banea, and A Öchsner. Methods to increase the toughness of structural adhesives with micro particles: an overview with focus on cork particles: Methoden zur erhöhung der zähigkeit von strukturklebern mit mikropartikeln: Ein überblick mit dem fokus auf kork. *Materialwissenschaft und Werkstofftechnik*, 47(4):307–325, 2016.
- [6] Aldobenedetto Zotti, Simona Zuppolini, Mauro Zarrelli, and Anna Borriello. Fracture toughening mechanisms in epoxy adhesives. *Adhesives-Applications and Properties*, 1:257, 2016.
- [7] D Verchere, JP Pascault, H Sautereau, SM Moschiar, CC Riccardi, and RJJ Williams. Rubber-modified epoxies. iv. influence of morphology on mechanical properties. *Journal of applied polymer science*, 43(2):293–304, 1991.
- [8] AJ Kinloch, JH Lee, AC Taylor, S Sprenger, C Eger, and D Egan. Toughening structural adhesives via nano-and micro-phase inclusions. *The Journal of Adhesion*, 79(8-9):867–873, 2003.
- [9] AQ Barbosa, Lucas FM da Silva, and A Öchsner. Effect of the amount of cork particles on the strength and glass transition temperature of a structural adhesive. *Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications*, 228(4):323–333, 2014.

- [10] MS Forte, JM Whitney, and GA Schoeppner. The influence of adhesive reinforcement on the mode-i fracture toughness of a bonded joint. *Composites Science and Technology*, 60(12-13):2389–2405, 2000.
- [11] Qingshi Meng, Sherif Araby, Nasser Saber, Hsu-Chiang Kuan, Jiabin Dai, Lee Lung, Jun Ma, and Chun H Wang. Toughening polymer adhesives using nanosized elastomeric particles. *Journal of Materials Research*, 29(5):665, 2014.
- [12] Hajime Kishi, Kazuhiko Uesawa, Satoshi Matsuda, and Atsushi Murakami. Adhesive strength and mechanisms of epoxy resins toughened with pre-formed thermoplastic polymer particles. *Journal of Adhesion Science and Technology*, 19(15):1277–1290, 2005.
- [13] 3M Advanced Materials Division. 3m glass bubbles, 2018.
- [14] VV Budov. Hollow glass microspheres. use, properties, and technology. *Glass and ceramics*, 51(7-8):230–235, 1994.
- [15] NF T 76-142. Méthode de préparation de plaques d’adhésifs structuraux pour la réalisation d’éprouvettes d’essai de caractérisation, 1988.
- [16] BS 2782-0:2011. Methods of testing plastics, part 2011, 2011.
- [17] MFSF De Moura, RDSG Campilho, and JPM Gonçalves. Crack equivalent concept applied to the fracture characterization of bonded joints under pure mode i loading. *Composites Science and Technology*, 68(10-11):2224–2230, 2008.
- [18] P Gálvez, RJC Carbas, Raul DSG Campilho, J Abenojar, MA Martínez, and Lucas FM da Silva. Fracture toughness in mode i (gic) for ductile adhesives. In *Journal of Physics: Conference Series*, volume 843, page 012008. IOP Publishing, 2017.
- [19] Roger Rethon and Chris Paynter. Calcium carbonate fillers. *Fillers for Polymer Applications; Rethon, R., Ed.; Springer International Publishing: Cham, Switzerland*, pages 149–160, 2017.
- [20] Hyoung Seop Kim. On the rule of mixtures for the hardness of particle reinforced composites. *Materials Science and Engineering: A*, 289(1-2):30–33, 2000.
- [21] András Reuß. Berechnung der fließgrenze von mischkristallen auf grund der plastizitätsbedingung für einkristalle. *ZAMM-Journal of Applied Mathematics and Mechanics/Zeitschrift für Angewandte Mathematik und Mechanik*, 9(1):49–58, 1929.

- [22] H Verweij, D Veeneman, et al. Hollow glass microsphere composites: preparation and properties. *Journal of materials science*, 20(3):1069–1078, 1985.
- [23] Woldemar Voigt. Ueber die beziehung zwischen den beiden elasticitätsconstanten isotroper körper. *Annalen der physik*, 274(12):573–587, 1889.

B Paper B

Experimental study of the impact of inorganic fillers on adhesive joint strength and its failure mechanism

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ABSTRACT

The use of modern structural adhesives provide a lightweight, practical and high strength joining methodology which is increasingly being adopted in the automotive and aeronautical industries, among many others. However, the strict mechanical performance's standards that must be met in these sectors require a constant search for ways of improving the adhesives' behavior, which has led to the growing use of reinforcements as a way of improving the capabilities of bonded joints. The aim of this work is to analyse how adding inorganic fillers to the adhesive layer affects the joint's strength and its failure mechanism. To this end, single lap joint specimens with mild steel and high strength steel substrates were tested, at quasi-static speeds, for different amounts of glass microspheres reinforcing two different structural adhesives. The experimental results indicate that the addition of glass particles reduces the joint performance for both substrates under study. Furthermore, the failure pattern was found to evolve from adhesive failure to a cohesive type of failure as the amount of glass particles present in the adhesive increased.

B.1 Introduction

Adhesive joints are progressively emerging as a valid alternative to the use of bolts, rivets, welding, and other joining options in a wide range of industrial sectors, a result of some of the adhesives' unique properties and advantages. Using adhesive bonding enables the manufacture of joints which combine low weight, high stiffness and uniform stress distributions. Moreover, the application of adhesives to the parts being joined lends itself to being performed by automated joining technologies. Furthermore, the use of adhesives is compatible with complex parts, which may have peculiar contours and designs, as well as different thicknesses, and be composed of highly dissimilar materials [1, 2].

As a result of the rising usage of adhesive joints, especially in high technological industries, such as the automotive and the aeronautical ones, the demands regarding their mechanical performance have greatly increased. Consequently, a set of innovative techniques and solutions have appeared as an attempt to improve the adhesives' performance. Some of these more recent approaches are based on the hybrid joint concept, which consists of a combination between adhesive joints and other conventional joining methods, the search for an optimal adhesive layer's thickness, or even modifications to the adherends' surface roughness to improve their adhesion [3].

However, many of these techniques are complex and expensive, which may discourage their practical use. Taking this into account, the introduction of reinforcements into the adhesive layer has grown as a relevant alternative in the search for a better mechanical performance in adhesive joints. Doing so is more economical and uncomplicated than the methods mentioned previously. It should be noted that the adding of reinforcements to the adhesive forms a composite material, since it turns into a combination of two or more materials [3].

Focusing on epoxy adhesives, these are usually amorphous and highly cross-linked materials, being associated with very favorable structural properties. Nevertheless, these are also particularly brittle materials. For that reason, the introduction of a second phase of particles, fibers, or other kinds of reinforcements, might improve the adhesive's performance, depending on the reinforcements' characteristics and mechanical properties, without negatively affecting other important properties [4].

With respect to the failure modes that can be observed when analysing the fracture surfaces of many types of adhesively joined specimens, there are the four main failure modes, represented in Figure 1. One of these is cohesive failure in the adhesive, which involves the cases where the crack propagates through the adhesive, while the surfaces of

the adherends remain covered in adhesive. This type of failure happens when there is a strong bond between the adhesive and the adherend, and the substrates are stronger than the internal strength of the adhesive. Cohesive failure in the adherend is another possible failure pattern, which occurs when the adhesion between the adherend and adhesive and the adhesive's internal resistance are both stronger than the cohesive strength of the adherend. Interfacial, or adhesive failure is a failure mode which corresponds to the cases where the crack follows a path along the interface between the adhesive and the adherends. This type of failure is often a consequence of bad surface preparation and it is generally not accepted in many applications. Finally, mixed or alternating failure is a failure mode that encompasses the cases where the cracks propagate in an alternating way, varying between the adhesive and the adherend interfaces [1].

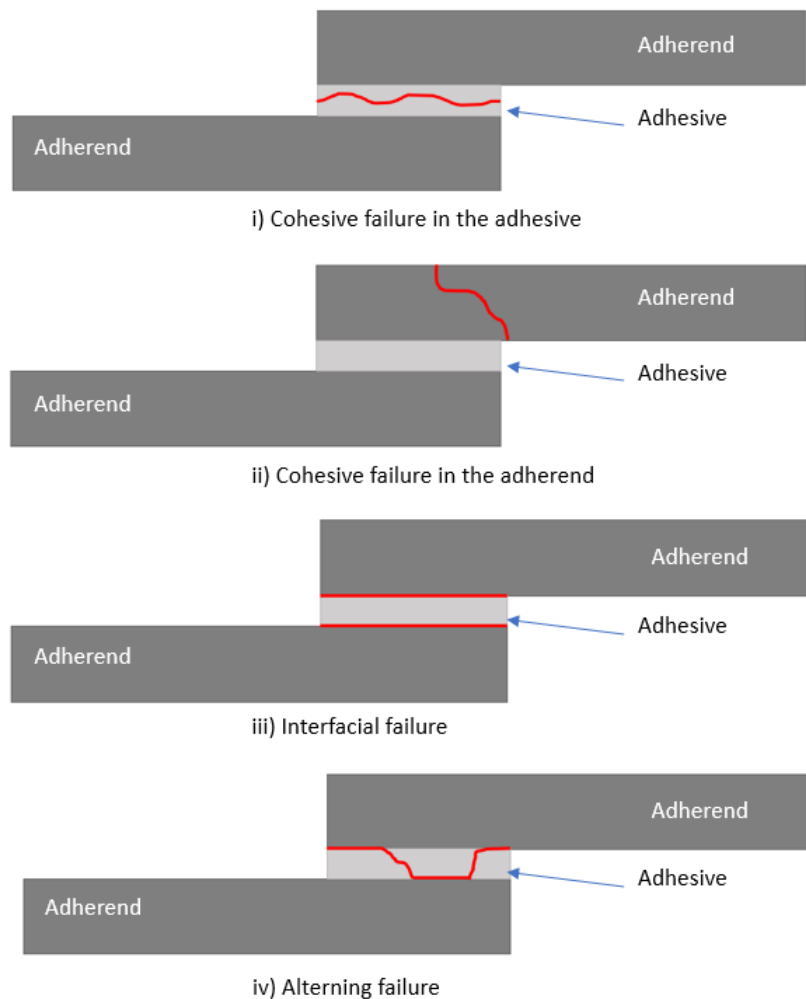


Figure 1: Schematic representation of the different types of failure.

Several important experimental studies have been published on the effects of adding

reinforcements to the adhesive layer, analysing not only the effect on the mechanical performance, but also on the failure modes.

Barbosa et al. [5] worked on studying the impact of cork particles on the strength of a structural adhesive. During this study, multiple joint configurations were tested by introducing different amounts of cork, between 0.5 % and 5%, into the adhesive layer. Afterwards, single lap joints and bulk specimens were tested in order to assess the adhesive's mechanical properties. This experimental study showed that, for 1 % of cork particles, the adhesive became more ductile, and also presented a higher joint strength. However, increasing the amount of particles to 2 % was found to lead to a noticeably worse mechanical behaviour, since the particles start acting as defects.

Kinloch et al. [6] has developed an experimental study with focus on the effects on the adhesive's toughness provoked by the addition of nanosilica particles to a rubber toughened adhesive. Throughout this study, single-lap joints and tapered double-cantilever beam specimens were tested to understand the behaviour of the adhesive as a function of the particles' amount. These particles, which have an average diameter of 20 nm, were found to improve the adhesive's mechanical behaviour in terms of toughness and single-overlap shear strength, for quantities between 1 % and 8 % in weight. The formation of stress fields around the particles and near the crack tip was found to contribute to additional plastic deformation of the adhesive's matrix.

Da Silva et al. [7] worked on an experimental and numerical study to understand the influence of cork microparticles in the failure mode of single-lap joints. Regarding the experimental study, single-lap joint specimens were manufactured and tested, with particle amounts of 0, 1, 2 and 5%, and different particle size ranges (53–75, 75–125, 125–250, 250–500 μm). Subsequently, the P - δ curves were analysed, as well as the corresponding fracture surfaces. With respect to the failure modes obtained experimentally, which were then validated numerically, it has been concluded that, by increasing the size and amount of the cork microparticles, the failure mode changes from an adhesive failure to a cohesive failure. Besides, it has been observed that the failure load only decreases when 5% of 75-125 μm and 250-500 μm particles are introduced, when compared to the neat conditions.

Hunter et al. [8] investigated how the presence of glass microspheres in the adhesive affects the mechanical adhesion of single-lap joints with fiber-reinforced polymers. The authors concluded that the use of these particles has different effects depending on how fast the adhesive is cured. For a slow-curing adhesive, the joints' strength decreased for the 3% and the 10% weight concentrations of glass microspheres, when compared to the neat

adhesive's configuration. For the fast-curing epoxy adhesive, the joint's strength increased for both concentrations, when compared to the joints with neat adhesive. Regarding the failure modes, thin-layer cohesive and fiber tear failure were most commonly found for the slow-curing adhesive, while for the fast-curing adhesive, interfacial failure was also reported.

In the present work, the impact of reinforcing adhesives with inorganic fillers was investigated. To that end, hollow glass beads were added to two epoxy adhesives, considering different amounts of particles. In order to assess the joints' strength, as well as the failure modes, single lap joint specimens were manufactured and then tested. The corresponding fracture surfaces were studied, using SEM analysis to assess the damage state of the glass beads and ascertain the damage mechanisms that took place.

B.2 Experimental details

This section provides data on the adhesive and the inorganic filler which was used in this experimental study. Furthermore, it describes in detail the manufacturing process of the two different single-lap joints configurations that were studied, as well as the respective testing procedures. Moreover, it also briefly describes the scanning electron microscopy analysis which was performed to support the work.

B.2.1 Materials

The adhesives analysed in this article will be referred to as Adhesive A and Adhesive B. These adhesives are both one component crash-resistant adhesives, which are used with the aim of increasing durability, crash performance and body stiffness in automotive construction. Both cure at a temperature stage of 180 °C during 30 minutes, Figure 2.

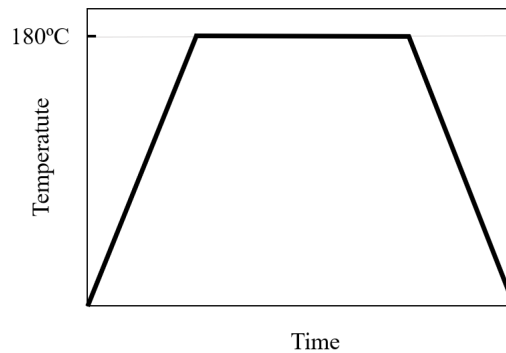


Figure 2: Cure cycle of the adhesives used.

The hollow glass beads used (Figure 3) consist of thin-walled hollow spheres made of soda-lime-borosilicate glass, with a white powder-like appearance. Their size and density can slightly vary, and they are known to be lightweight and have a high strength-to-density ratio and a significant isostatic crush strength [9, 10].

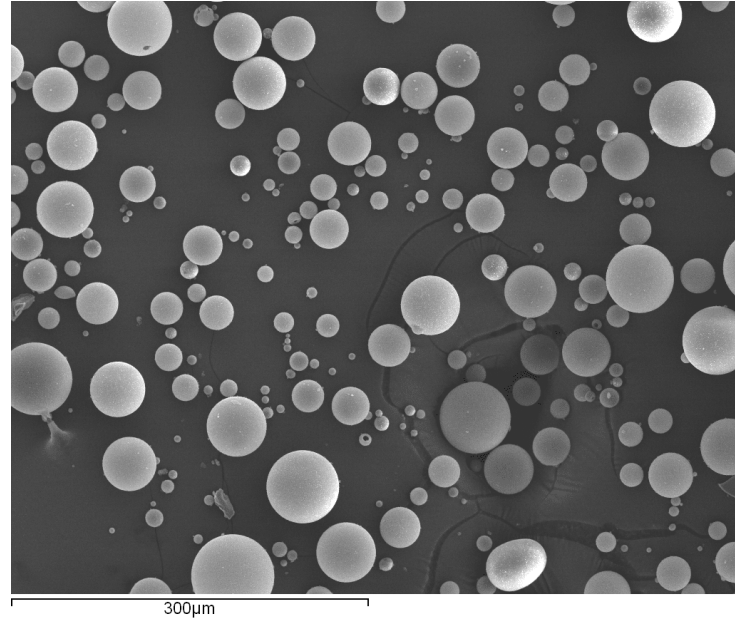


Figure 3: SEM micrograph of glass microspheres.

Table 1 lists the main properties of the glass beads used.

Table 1: Main properties of the hollow glass beads, provided by the supplier.

Property	-
Density/ $\text{g}\cdot\text{cc}^{-1}$	0.37
Color	<i>White, powdery</i>
Composition	<i>Soda – lime – borosilicate glass</i>
Median particle diameter/ μm	45
Strength/ MPa	20.6

B.2.2 Specimen fabrication

Single lap joint specimens. For this experimental study, two different single lap joint geometries were analysed, mainly differing in the material of the adherend and its

dimensions.

One of the geometries that was analysed is represented in Figure 4. For this single lap joint configuration, an adherend made of mild steel was used.

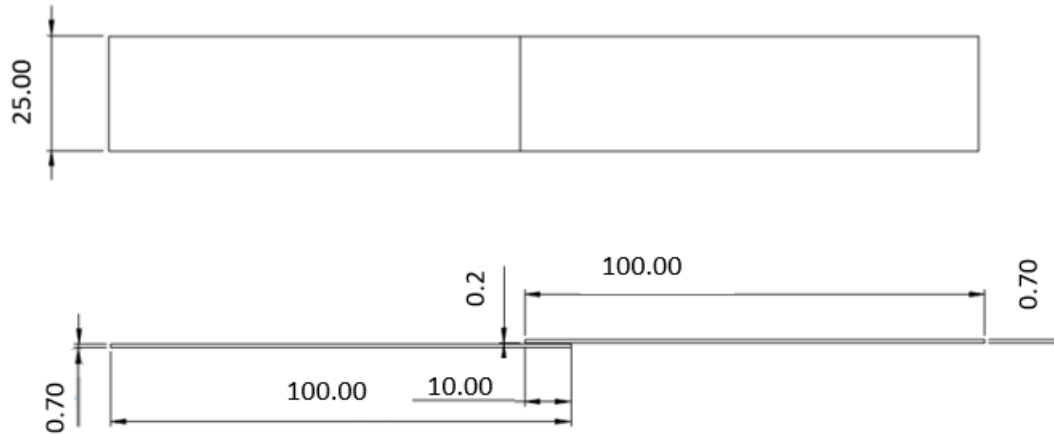


Figure 4: Representation of single lap joint specimens' geometry, dimensions in mm.

The adherends' surfaces to be bonded were degreased with acetone, with the objective of cleaning the surfaces and ensure the proper removal of any kinds of contaminants, dust or oils.

The other SLJ specimen configuration used an adherend manufactured from a high strength steel (DIN C65 heat treated) with a Young's Modulus of 210 GPa and a Poisson's ratio of 0.3. This configuration was selected in order to avoid plastic deformation of the adherend. The geometry and dimensions of the SLJ are detailed in Figure 5.

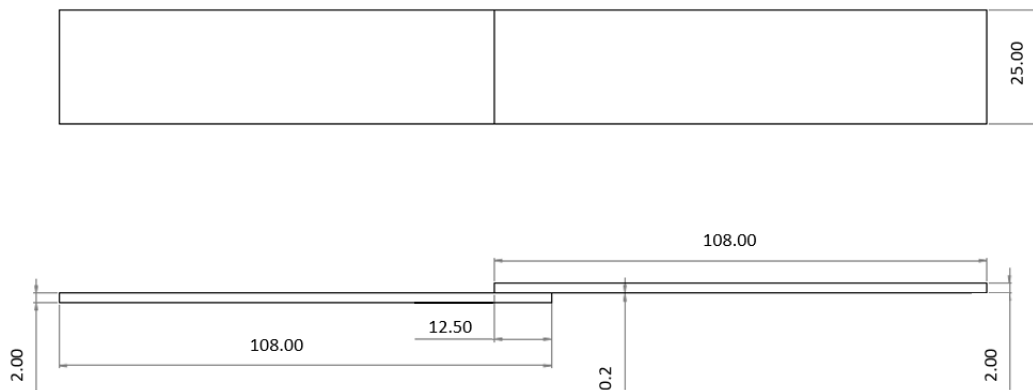


Figure 5: Representation of single lap joint specimens' geometry, dimensions in mm.

The adherends' surfaces to be bonded were firstly subjected to a surface treatment. The surfaces were grit blasted and then degreased with acetone. The main goal of this surface treatment procedure was to clean the surfaces and ensure the proper removal of weak layers, contaminants, dust or oils.

The manufacturing of the single lap joint specimens was achieved with the use of an annealed carbon steel mould, which enables the control of the overlap length, the adhesive's thickness and proper alignment of the substrates. This is possible since the mould has been designed with suitable alignment pins, shims and positioner blocks. Moreover, a silicone based release agent was applied to the mould surfaces, in order to facilitate the release of the specimens after being manufactured.

The addition of the the glass beads into the adhesive layer has been achieved by mixing them in the pre-heated epoxy adhesive, in order to facilitate the distribution of the particles along the adhesive matrix. This was performed using a centrifuge mixing machine, SpeedMixer DAC 150TM (Hauschild, Hamm, Germany) for 40 s, increasing gradually from a 500 r/min to a 3000 r/min rotation speed within the first 10 seconds. The rotation speed was then maintained at 3000 r/min for the remaining time. This procedure was adopted for all the amounts of glass microspheres under study. After curing, the specimens were removed from the mould and the excess of adhesive which flowed away from the bonded area was eliminated manually, with the use of a file.

B.2.3 Testing procedures

For testing the single lap joint, an INSTRON 3367 universal testing machine was used. This machine is equipped with a load cell of 30 kN (Norwood, USA).

Single lap joint tests. Specimens with and without glass microspheres were manufactured. The tests were conducted at a quasi-static speed of $1 \text{ mm} \cdot \text{min}^{-1}$. For each test, the load-displacement ($P - \delta$) curve was registered until failure occurred. Three specimens were tested for each condition.

B.2.4 High speed camera recording

An high speed camera was used in order to record the failure mode of the single lap joints bonded with mild steel adherends. The main aim of this procedure was to visualize the crack's propagation path along the adhesive layer.

B.2.5 Scanning electron microscopy analysis

Scanning electron microscope (SEM) analyses were performed utilizing a JEOL JSM 6301F/Oxford INCA Energy 350/Gatan Alto 2500 microscope (Tokyo, Japan). The utilization of this technology allowed to analyse the single lap joint specimens fracture surfaces, the chemical composition of the particles being studied, as well as their damage level.

B.3 Experimental results and discussion

B.3.1 Single lap joint tests with high strength steel substrates

As stated previously, the single lap joint tests were carried out for different amounts of hollow glass microspheres, in volume, according to the schemes represented in Figures 6 and 7, for Adhesive A and Adhesive B, respectively.

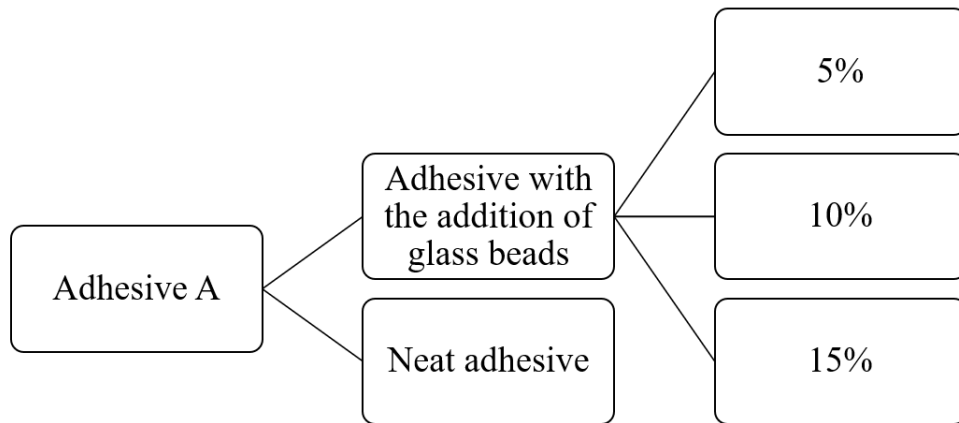


Figure 6: Schematic diagram of SLJ specimens with and without glass beads, for Adhesive A.

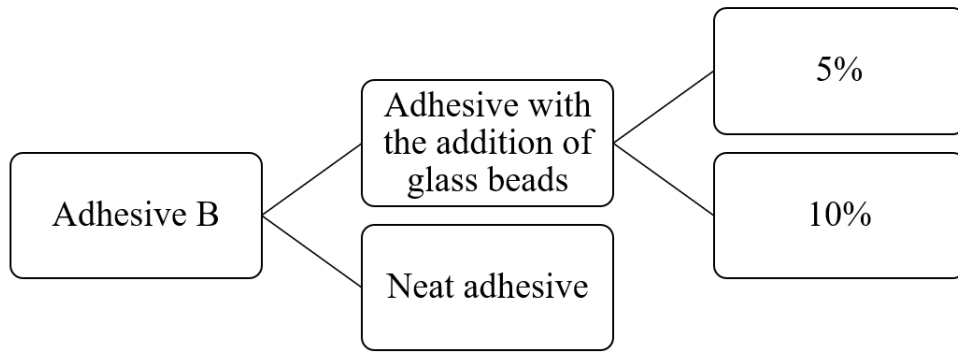


Figure 7: Schematic diagram of SLJ specimens with and without glass beads, for Adhesive B.

It is important to refer that the neat configuration of Adhesive A already has 5% volume of hollow glass beads spread along its matrix, since this is the manufacturer supplied state of this adhesive.

In Figure 8, the testing procedure that was adopted for this study can be observed.



Figure 8: Single lap joint testing procedure.

The $P - \delta$ curves obtained for the single lap joint specimens with high strength steel are shown in Figure 9, for Adhesive A at quasi-static conditions.

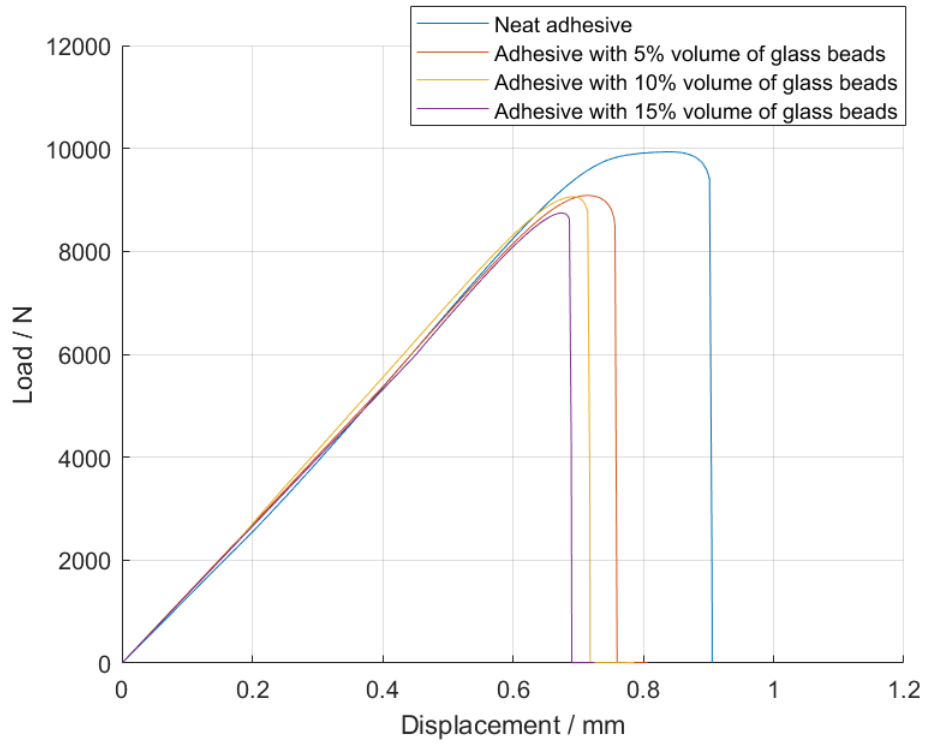


Figure 9: Representative $P - \delta$ curves for the single lap joint tests of Adhesive A with neat adhesive, and the adding of 5, 10 and 15 % volume of hollow glass beads.

The values of the failure load, P_{max} , and the maximum extension, δ_{max} , can be observed in Figures 10 and 11, for the different percent volumes of added hollow glass beads.

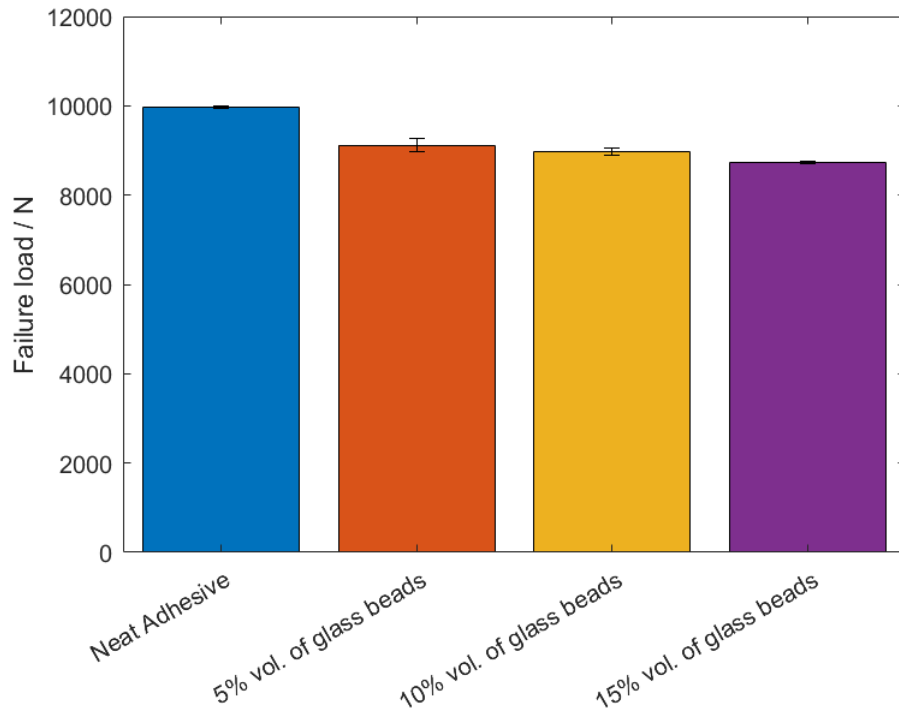


Figure 10: The failure load values, P_{max} , of Adhesive A for the different percent volumes of added hollow glass beads.

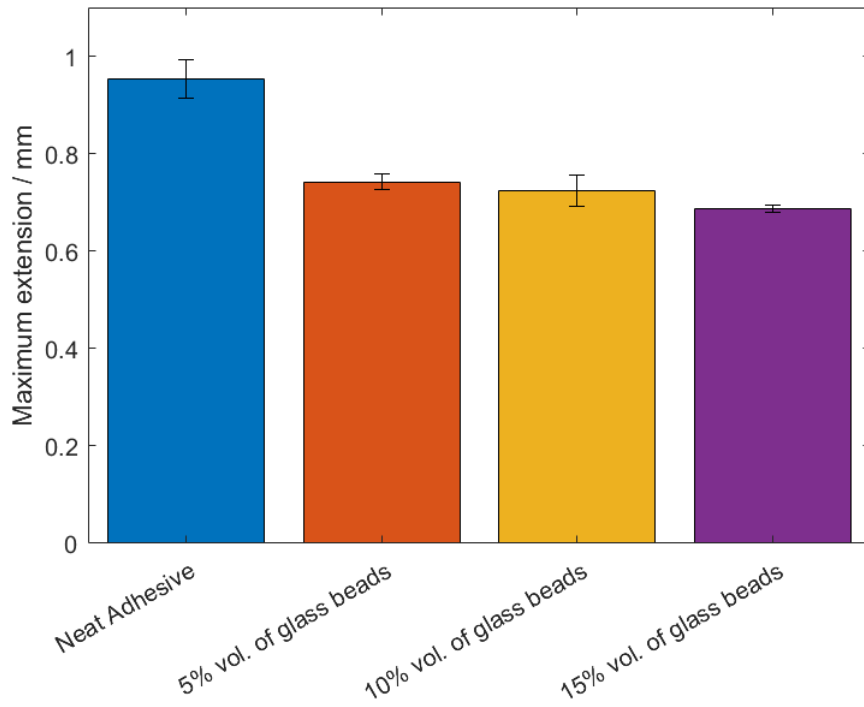


Figure 11: The maximum extension values, δ_{max} , of Adhesive A for the different percent volumes of added hollow glass beads.

Based on the $P - \delta$ curve results and the maximum extension mean values, it can be concluded that adding particles to the adhesive layer has a negative impact on the joint performance, since not only the area below the curves decreases but the maximum extension is also reduced. Therefore, and for the single lap joints with high strength steel substrates, the presence of glass beads has a negative impact on the mechanical behavior of the bonded joints. Regarding the failure load, the effects caused by the presence of the particles can be considered as negligible, since these values do not significantly vary.

With respect to the failure patterns, figure 12 illustrates the fracture surfaces the SLJs for the different percent volumes of added hollow glass beads that were evaluated.

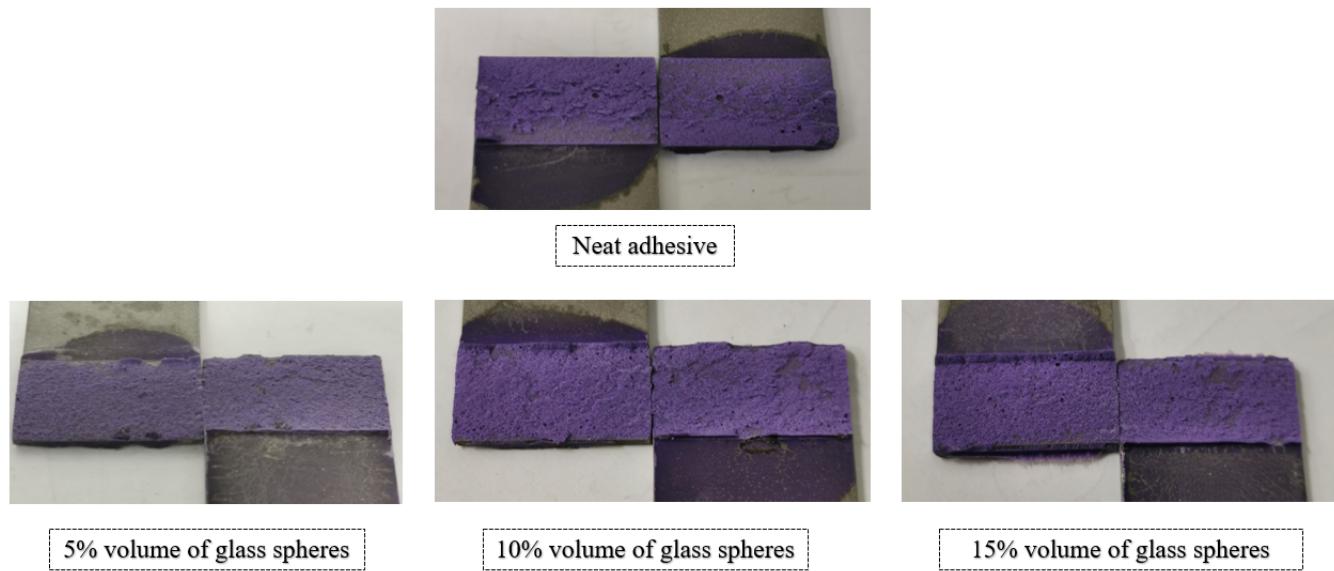


Figure 12: SLJs fracture surfaces of Adhesive A as a function of the added hollow glass beads' amounts that were employed.

These images show that there is clearly a cohesive failure for all the four configurations under study, which means that for this single lap joint's geometry, the presence of glass beads does not affect the failure mode of the bonded joints.

The $P - \delta$ curves associated to the SLJs with Adhesive B are represented in Figure 13.

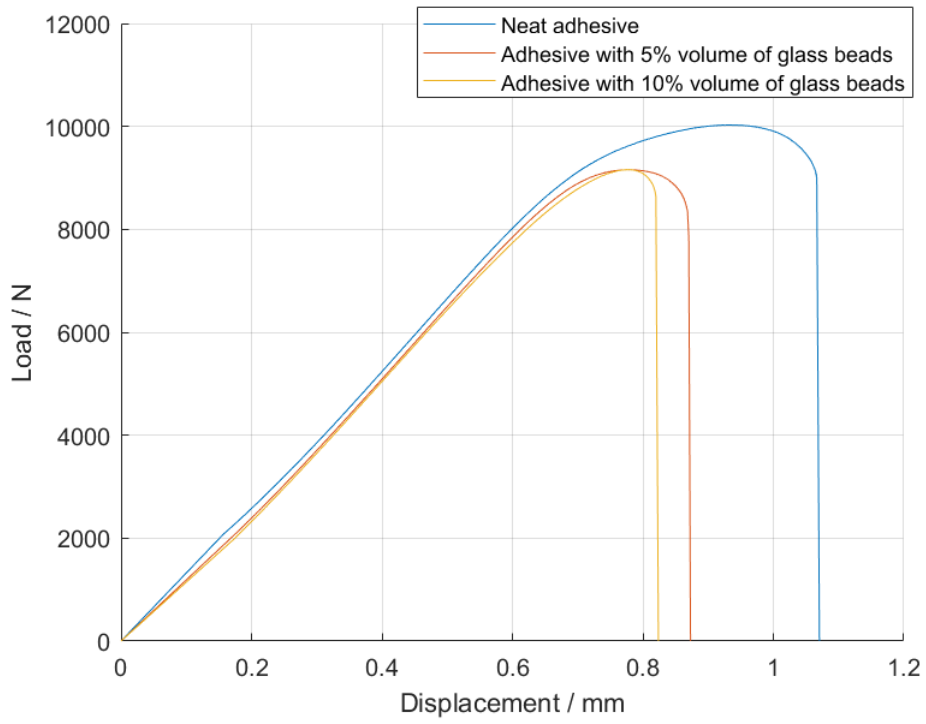


Figure 13: Representative $P - \delta$ curves for the single lap joint tests of Adhesive B with neat adhesive, and the adding of 5, and 10 % volume of hollow glass beads.

The failure load, P_{max} , and the maximum extension, δ_{max} , mean values, as a function of the added amounts of hollow glass beads, are displayed in the figures below, Figures 14 and 15.

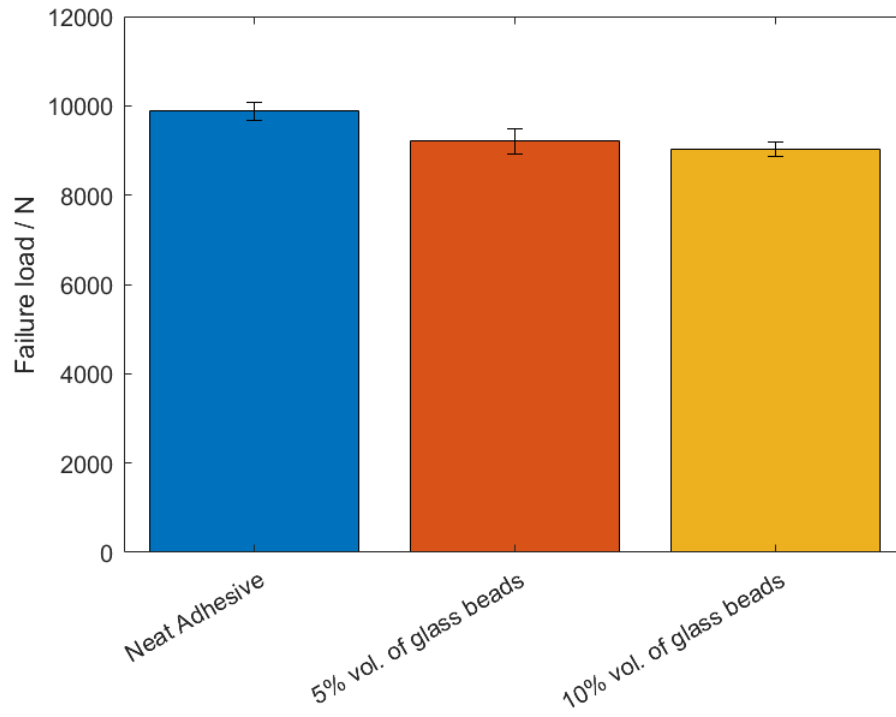


Figure 14: The failure load values, P_{max} , of Adhesive B for the different percent volumes of added hollow glass beads.

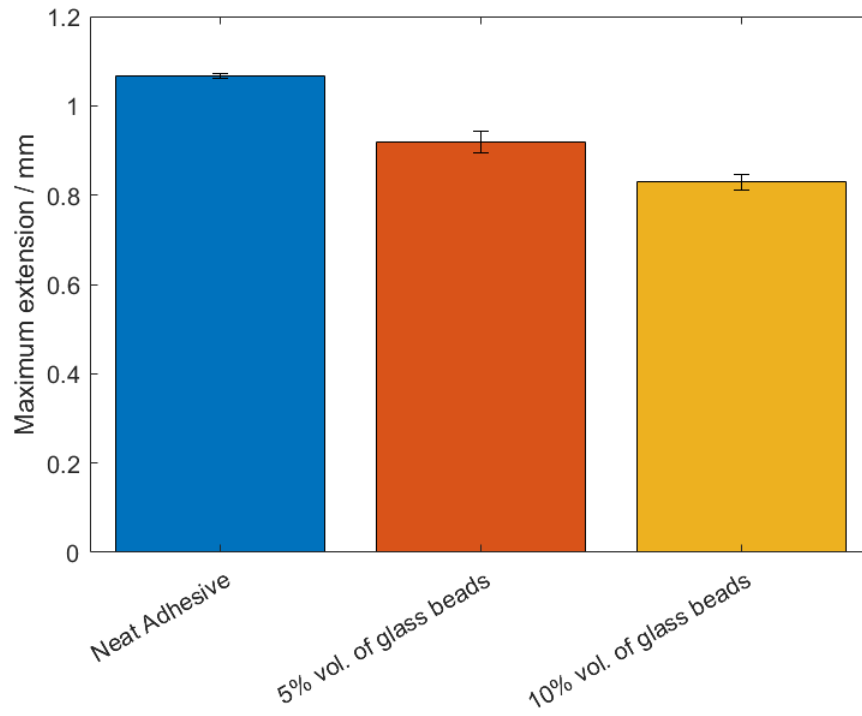


Figure 15: The maximum extension values, δ_{max} , of Adhesive B for the different percent volumes of added hollow glass beads.

For this adhesive, and similarly to what has been reported for Adhesive A, the introduction of glass particles into the adhesive layer worsens the mechanical behavior of the bonded joints, since the area of the curves represented in Figure 13 decreases with an increasing amount of particles. Moreover, the maximum extension, δ_{max} , significantly decreases, and the failure load, P_{max} , does not show a significant correlation to the glass particles' amounts.

Figure 16 illustrates the fracture surfaces the SLJs for the different percent volumes of hollow glass beads that were evaluated.

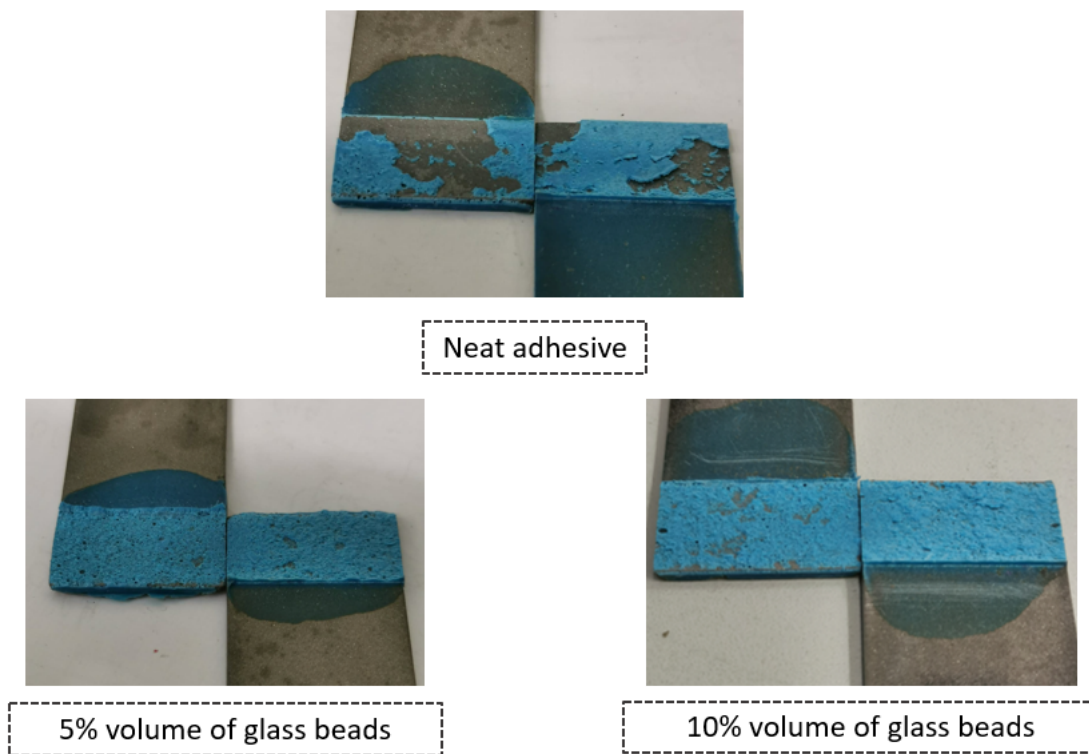


Figure 16: SLJs fracture surfaces of Adhesive B as a function of the added hollow glass beads' amounts that were employed.

The fracture surfaces clearly show that, by introducing glass particles into the adhesive layer, the failure evolves from an adhesive failure towards a cohesive one. This is a consequence of the local stress concentrations formed on the periphery of the glass particles, which constitute the weak links in the adhesive layer. Subsequently, the crack initiates near those points, rather than at the ends of the bonded joints, which would be the most critical regions in terms of stress if there were no glass microspheres. Consequentially, the failure pattern changes from adhesive to cohesive.

At last, and as a way of testing the applicability of simple analytical methods to predict

the joint's failure load, the global yielding criterion was used to do so. This criterion , which considers that the entirety of the overlap is transferring the load, was chosen since it is applicable for short overlaps and when the substrates do not deform plastically, which is the case for the SLJs geometry being studied [11].

To make use of this criterion, the following equation was used [11]:

$$P_{GY} = \tau_y \cdot b \cdot l \quad (1)$$

where P_{GY} is the adhesive's failure load, as a consequence of its global yielding, τ_y is the adhesive's yield strength, b is the bonded joint's width and l corresponds to the overlap length [11].

For the SLJs being studied, the bonded joint's has a width of 25 mm and an overlap length of 12.5 mm. Taking this into account, and knowing that the neat adhesives' yield strength is 30.9 ± 0.9 MPa and 33.9 ± 0.2 MPa, for Adhesive A and Adhesive B, respectively, Table 2 compares the experimental and the analytical values of the failure load for neat Adhesive A and neat Adhesive B.

Table 2: Comparison between the experimental and the analytical values of the failure load for neat Adhesive A and neat Adhesive B.

Adhesive	Experimental result (N)	Analytical result (N)	Error (%)
Adhesive A	9964.0 ± 26.4	9656.3	3.1
Adhesive B	9882.04 ± 207.5	10593.8	7.2

By analysing the results represented in Table 2, it is possible to conclude that the global yielding criterion provides a satisfactory estimation of the bonded joint's failure load, since the errors associated to the values calculated through its equation are relatively small. Furthermore, it is also possible to observe that for Adhesive A, the analytical prediction is higher than the experimental value.

In fact, one can conclude, by calculating the shear strengths of the SLJs for Adhesive A and Adhesive B, that these values are similar to the tensile strengths that have been obtained during the adhesives' characterization process. Furthermore, the shear strengths for some of the SLJs configurations revealed shear strength values which are slightly higher than those associated to the tensile strength. This can be the consequence of the different

strain rates being applied for the SLJs and for the bulk specimens. Even though a quasi-static speed of $1 \text{ mm} \cdot \text{min}^{-1}$ was applied for both cases, the length of adhesive which is being displaced is much smaller during SLJs testing, since the very thin bonded region has an overlap length of 12.5 mm, while the thicker bulk specimen has a length of 150 mm. For that reason, the deformation, in terms of strain, that is being applied to the SLJs is much higher, which results in higher strain rates. Due to the fact that the 2 adhesives are designed for automotive applications, their mechanical performance is optimized for impact applications, showing improved shear strengths for higher strain rates, as Borges et al. [12] also reported in an experimental study of the influence of the loading rate on crash resistant adhesives. Taking this into account, it is expectable for the shear strength to be higher than the tensile strengths obtained during bulk specimen testing.

B.3.2 Single lap joint tests with mild steel substrates

As stated previously, the single lap joint tests were performed for different amounts of hollow glass microspheres, in volume. In Figure 17, the testing procedure that was adopted for this study can be observed.

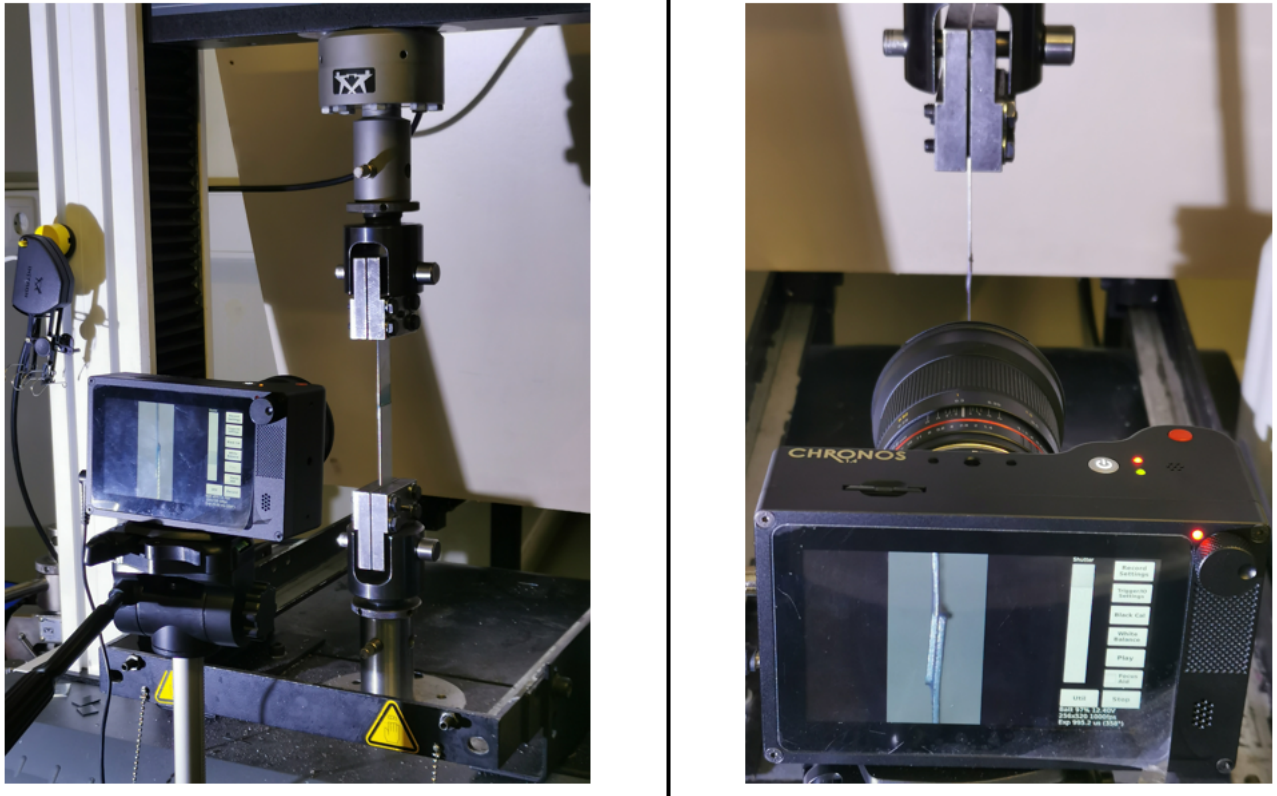


Figure 17: Single lap joint testing procedure.

The $P - \delta$ curves obtained for the single lap joint specimens with mild steel were the

ones that follow in Figure 18, for Adhesive A at quasi-static conditions.

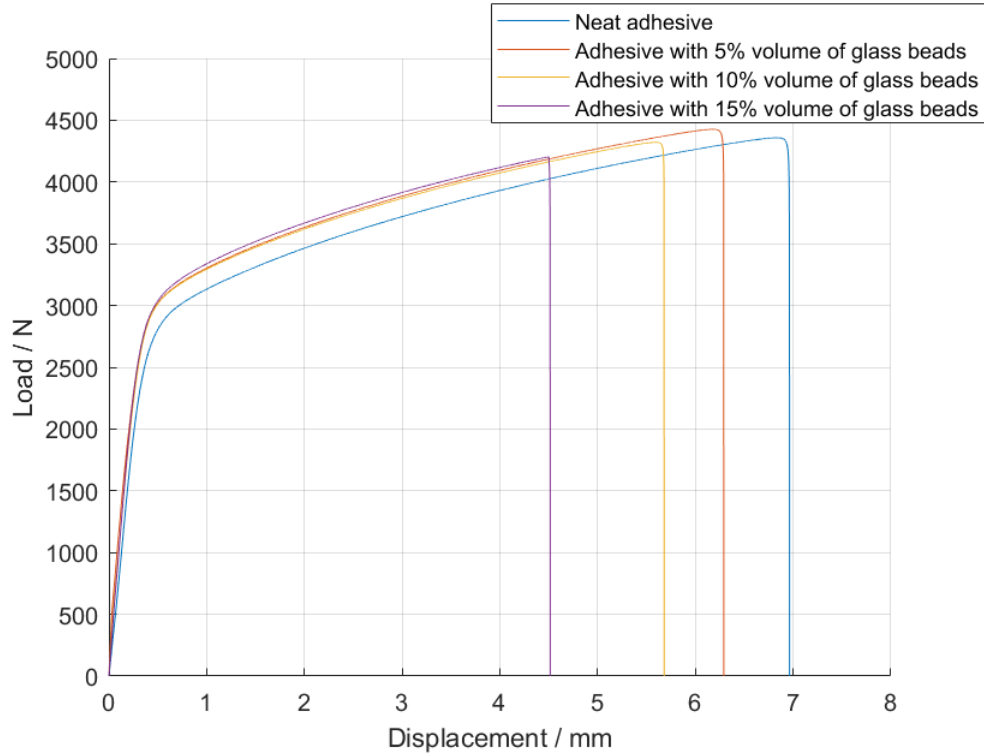


Figure 18: Representative $P - \delta$ curves for the single lap joint tests of Adhesive A with neat adhesive, and the adding of 5, 10 and 15 % volume of hollow glass beads.

The curves represented in Figure 18 clearly show that, after reaching a load value between 2500 and 3000 N, it is the substrate, instead of the adhesive, that is controlling the performance of the single lap joint specimens being tested. This occurs due to the fact that the adherends are not only thin (with a thickness of 0.7 mm), but also made of mild steel, which allows them to deform plastically. This phenomenon can be verified by observing the deformation of the substrates throughout the SLJs tests, as shown in Figure 19.



Figure 19: Representative image of the plastic deformation to which the mild steel substrates are subjected, during the SLJs tests.

By analysing the $P - \delta$ curves, one can notice that the area below them generally decreases with increasing amounts of glass particles in the adhesive. Table 3 displays the decrease in percentage of the area below these curves for the different amounts of glass beads being tested. This trend goes in line with the decrease of the adhesive's ductility with the increasing volume of glass beads that had been observed in paper A "Characterization of the effect of inorganic fillers on adhesives properties."

Table 3: Decrease, in %, of the areas below the $P - \delta$ curves, for Adhesive A.

Added particle content of glass beads, in % volume		Decrease (%)
0		-
5		8.4
10		16.5
15		36.4

The values of the failure load, P_{max} , and the maximum extension, δ_{max} , can be observed in Figures 20 and 21, for the different percent volumes of added hollow glass beads.

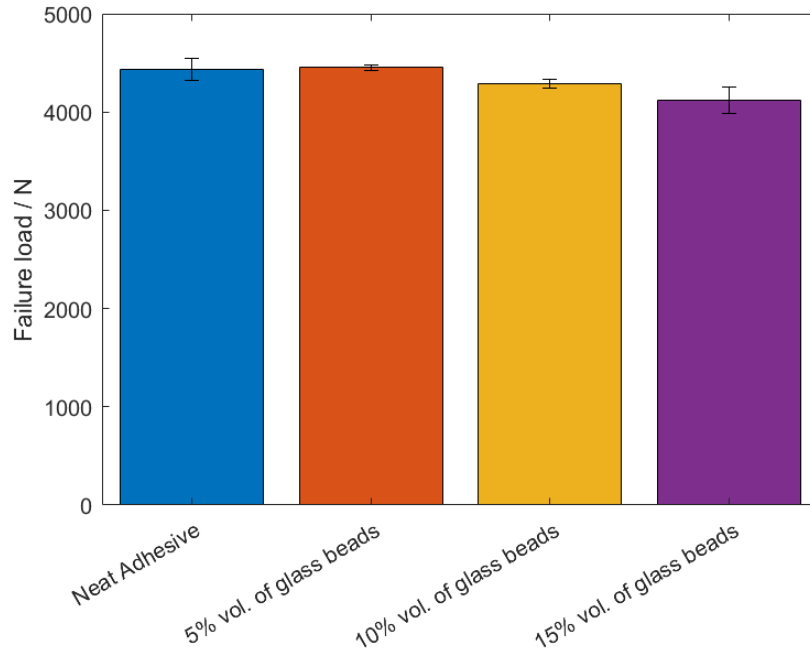


Figure 20: The failure load values, P_{max} , of Adhesive A for the different percent volumes of added hollow glass beads.

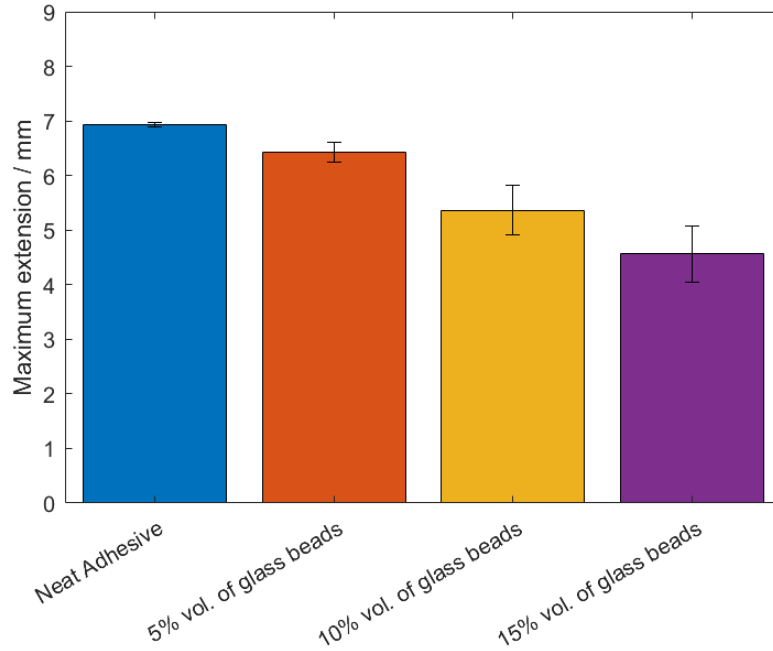


Figure 21: The maximum extension values, δ_{max} , of Adhesive A for the different percent volumes of added hollow glass beads.

By taking into consideration all the experimental results that were obtained, it is possible to generally conclude that the presence of the glass microspheres in Adhesive A

weaken the adhesive (for more detailed information regarding this matter, please consult the paper "Characterization of the effect of inorganic fillers on adhesives properties"), since not only the area below the curves gradually reduces, as shown in 3, but so does the maximum extension, which represents a decrease of the adhesive's mechanical performance.

In terms of the failure load, there is no significant impact caused by the presence of the spheres, even though there is a small reduction of joint strength with the increasing amounts of glass microspheres.

With the intention of comparing the results obtained experimentally with analytical methods, the methodology proposed by Adams et al. [13] to determine the strength of SLJs as function of the overlap length and the adherend thickness was used. Taking into account the fact that, in this particular case, the adherend is yielding, the following equation was used:

$$P_{AY} = \sigma_y \cdot b \cdot t_s / (1 + 3k) \quad (2)$$

where σ_y is the yield strength of the adherend. For low loads and short overlaps, k can be assumed to be 1, resulting in:

$$P_{AY} = \sigma_y \cdot b \cdot t_s / 4 \quad (3)$$

For joints with a length much higher than their thickness, such that $l/t_s \geq 20$, the value of k reduces until tending to zero, which leads to:

$$P_{AY} = \sigma_y \cdot b \cdot t_s \quad (4)$$

Since for this case, the overlap is short, k can be assumed to be 1, which leads to a maximum load, P_{AY} , of 2.54 kN. Analytically, the methodology assumes that when the adherend starts to yield, the $P - \delta$ curve reaches a plateau. However, and as one can observe in Figure 18, this does not happen experimentally, since the substrates, when strained beyond their yield point, require the application of additional stresses to further deform the steel, a phenomenon which is commonly referred to as the strain hardening effect. This can be observed by analysing a schematic representation of the differences between the analytical and the experimental mechanical behavior of the bonded joint, represented in Figure 22.

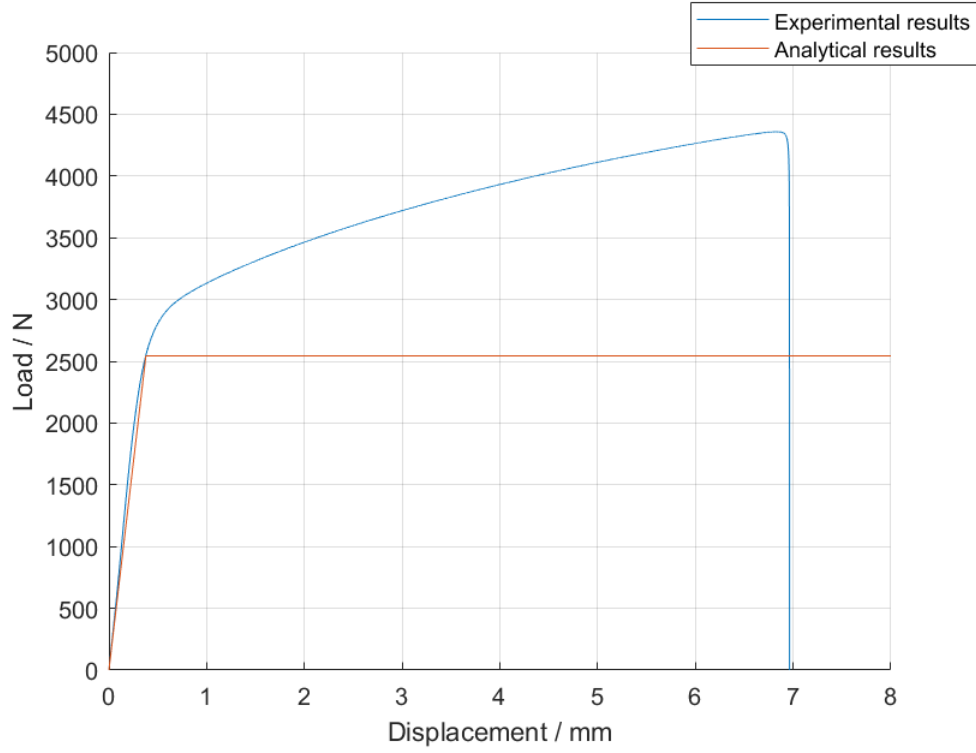


Figure 22: Schematic representation of the differences between the experimental and the analytical results for the single lap joints with mild steel.

In order to investigate on the failure modes associated to each case, it becomes pertinent to exhibit the fracture with respect to each adhesive's configuration, in terms of the amounts of hollow glass beads. Figure 23 represents the different failure patterns that were detected for the four studied cases.

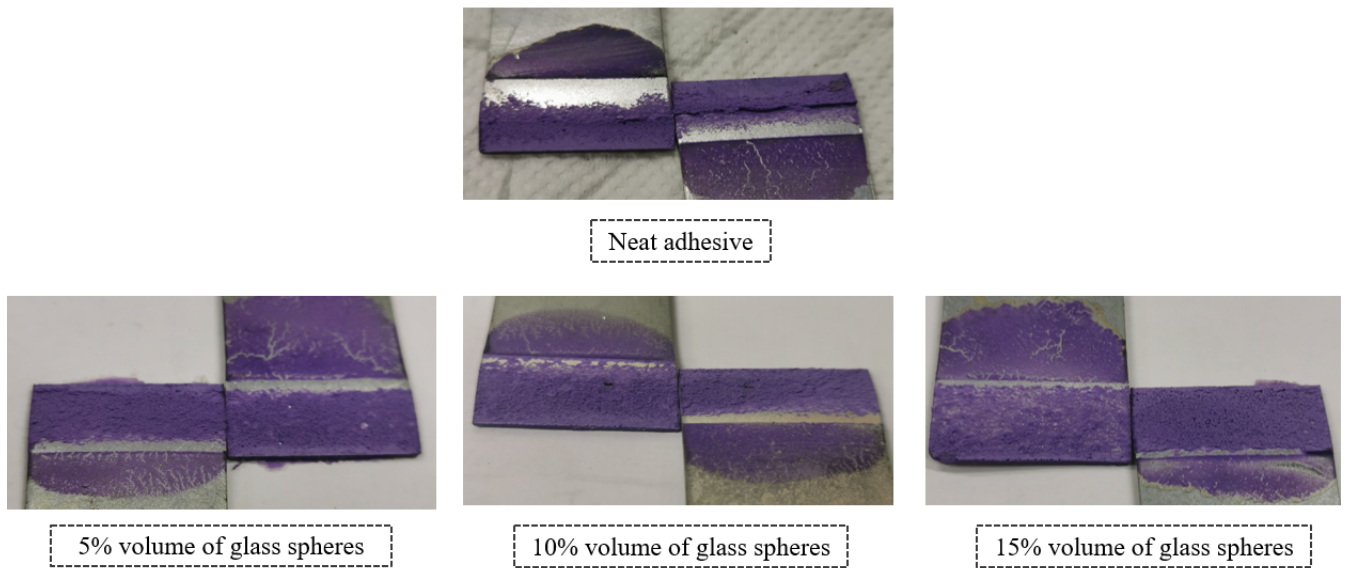


Figure 23: SLJ fracture surfaces of Adhesive A as a function of the added hollow glass beads' amounts that were employed.

By comparing the fracture surfaces exhibited in the figure above with the ones represented in Figure 12, it is possible to observe that, unlike for the single lap joint specimens with high strength steel, failure is not cohesive for the single lap joints with mild steel substrates and neat adhesive. The reason why this happens is associated with the substrates' plastic deformation, which increases the local stress concentrations at the ends of the bonded joint and creates critical peel stresses in those areas. The location of these stress fields causes the crack to propagate closer to the interface, which contributes to adhesive failure.

Nevertheless, the fracture surfaces show a decrease of the bonded regions without adhesive with the increasing amounts of particles. This happens since the glass microspheres introduce local stress concentrations, which are the most highly stressed regions on the adhesive layer. This phenomenon contributes to the change of the crack's path, and consequently to a transition from an adhesive failure to a cohesive one, with the crack propagating close to the middle of the adhesive layer.

In order to minutely comprehend the differences regarding the failure pattern and the crack's propagation path for single lap joints with neat adhesive and with the addition of glass beads, a high speed camera was used to capture these phenomena, as one can see in Figure 17.

Figures 24 and 25 exhibit the different stages associated to the failure of SLJs with neat adhesive and with the addition of glass beads, respectively. Frames I, II, III and IV

correspond to the stages before, in the beginning, during and in the end of the crack's propagation, respectively.

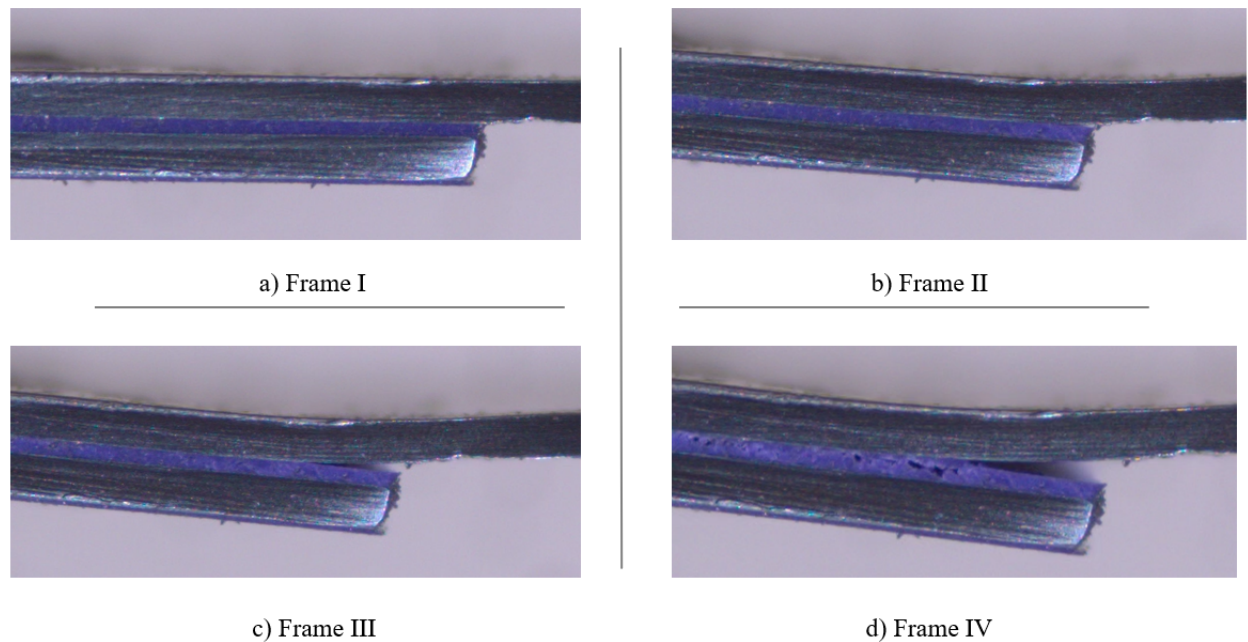


Figure 24: Representative frames of the different stages associated to the failure of a SLJ with neat Adhesive A.

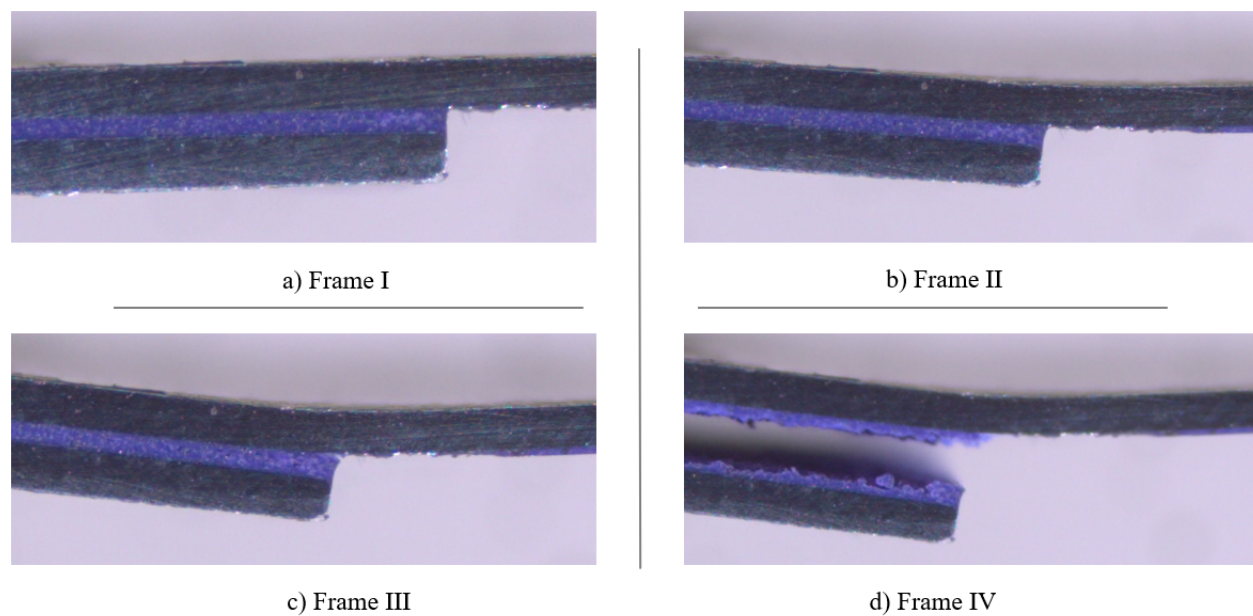


Figure 25: Representative frames of the different stages associated to the failure of a SLJ with the adding of glass beads to Adhesive A.

By observing Figure 24, it is evident that the crack starts to propagate in the interfacial

region, and continuously follows that path as the joint is loaded, as represented in frames II and III. This happens, as mentioned previously, as a consequence of the substrates' plastic deformation, which promotes very significant peel stresses in the ends of the bonded joints, turning those areas into the most critical ones, in terms of stresses. Finally, and as shown in frame IV, the crack moves closer to the middle of the adhesive layer, just before the joint's failure.

Moreover, and with regards to the SLJ with Adhesive A and the addition of glass beads to the adhesive layer, 25 displays a different event to the one described for neat Adhesive A. Even though there is still an interfacial crack in an initial period of its propagation, this happens in a later stage than what had been perceived for neat Adhesive A. The crack does not continue to follow a path within the interfacial region. Instead, the crack path is noticeably shorter and suddenly, just before failing, it moves to the middle of the adhesive layer, as a consequence of the presence of the glass beads, which weakens the adhesive and creates critical stress fields in their periphery that help to change the crack's location.

With the intention of summarizing the two different phenomena which were just described, Figures 26 and 27 exhibit representative schemes of the path followed by the crack with neat adhesive and with the addition of glass beads, respectively.

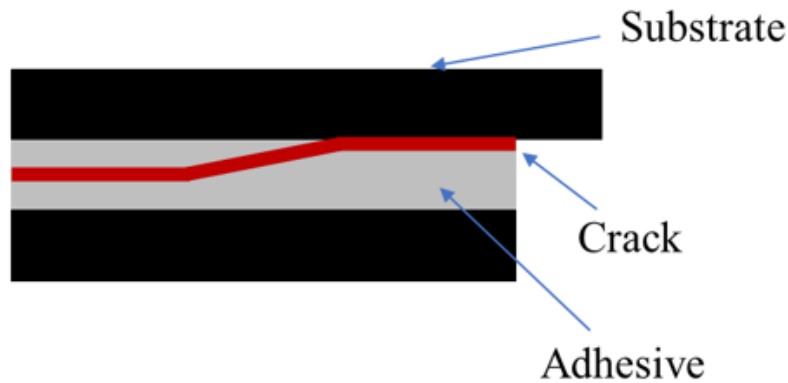


Figure 26: Representative scheme of the crack's propagation path of a SLJ with neat Adhesive A.

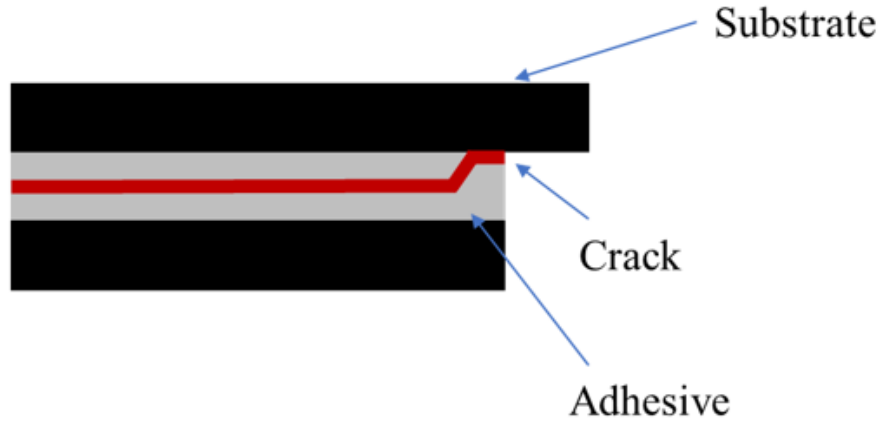


Figure 27: Representative scheme of the crack's propagation path of a SLJ with the adding of glass beads to Adhesive A.

Regarding the SLJs tests for Adhesive B, the $P - \delta$ curves recorded at quasi-static conditions are represented in Figure 28.

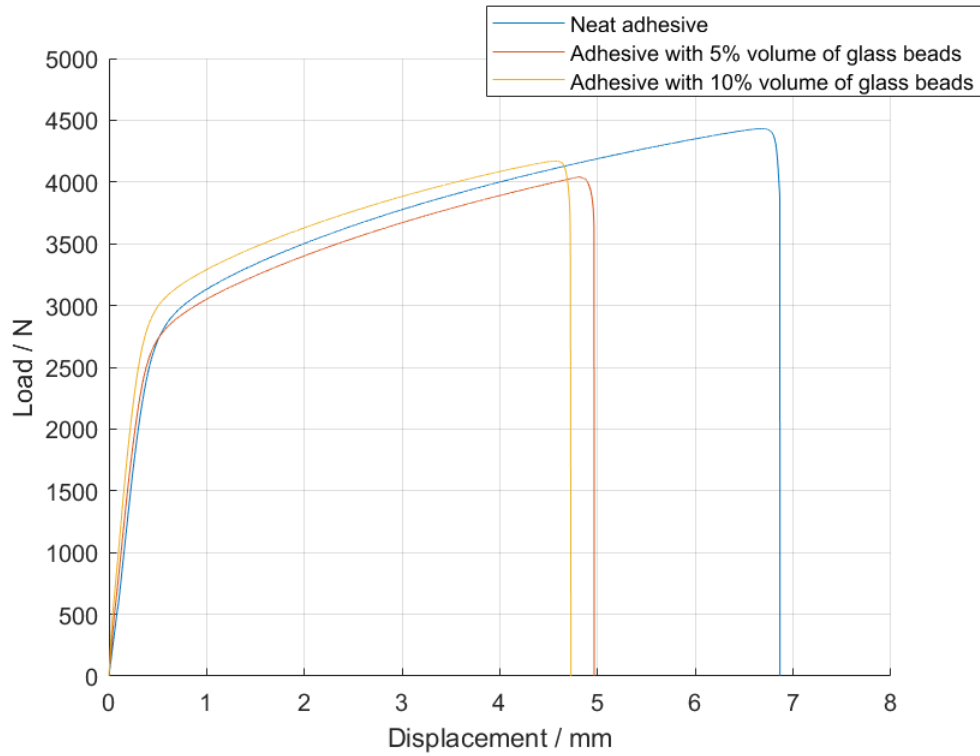


Figure 28: Representative $P - \delta$ curves for the single lap joint tests of Adhesive B with neat adhesive, 5, and 10 % volume of hollow glass beads.

Similarly to what was stated regarding SLJs with Adhesive A, the $P - \delta$ curves represented in Figure 28 show that the substrates also deform plastically. Moreover, it is

evident that the area below these curves decreases by introducing glass particles into the adhesive layer, as one can confirm in Table 4.

Table 4: Decrease, in %, of the areas below the $P - \delta$ curves, for Adhesive B.

Added particle content of glass beads, in % volume		Decrease (%)
0		-
5		32.7
10		33.5

The mean values of the failure load, P_{max} , and the maximum extension, δ_{max} , are shown in Figures 29 and 30, for the different percent volumes of added hollow glass beads.

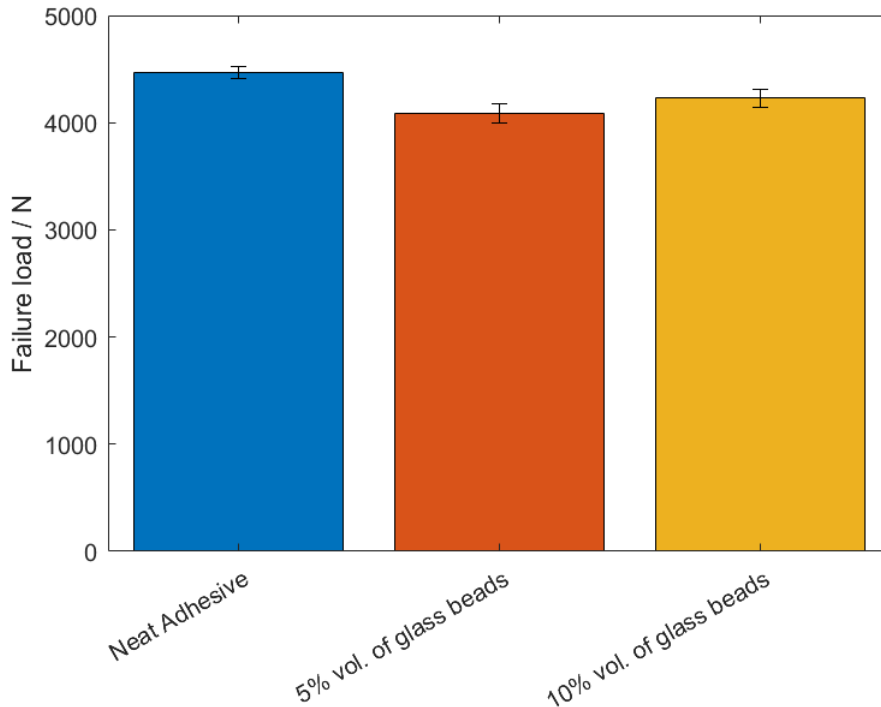


Figure 29: The failure load values, P_{max} , of Adhesive B for the different percent volumes of added hollow glass beads.

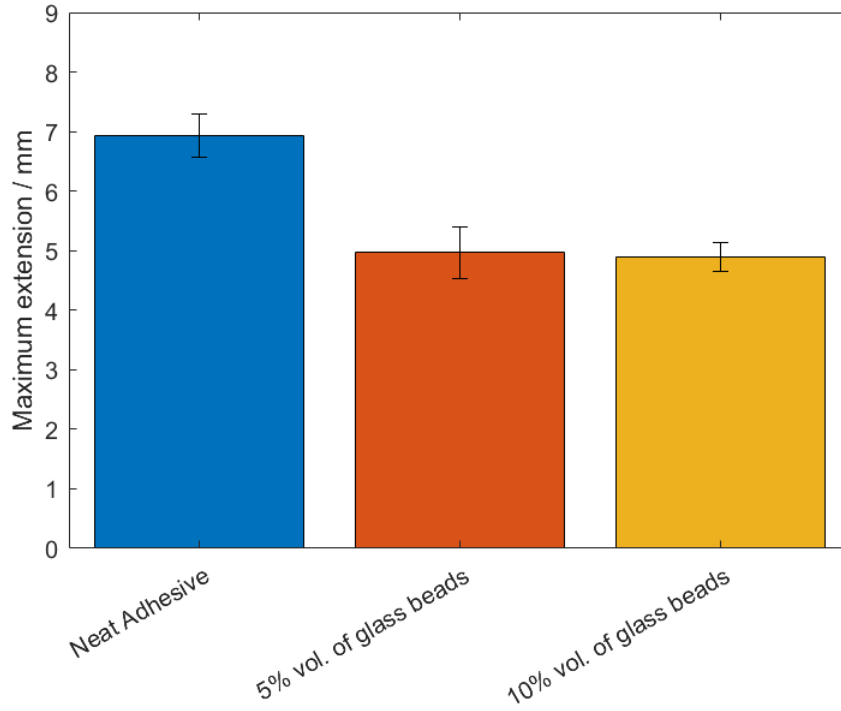


Figure 30: The maximum extension values, δ_{max} , of Adhesive B for the different percent volumes of added hollow glass beads.

Regarding the results of testing of the SLJ specimens with mild steel and Adhesive B, one can conclude that, by adding glass beads to the adhesive, the joint's performance clearly declines. This goes in line with the mechanical characterization described in paper A "Characterization of the effect of inorganic fillers on adhesives properties.", where the adhesive's properties were negatively affected with the introduction of glass microspheres into the adhesive layer. Again, the failure load values do not significantly vary by adding glass beads to the adhesive. Moreover, the maximum extension decreases, while the Young' modulus increases, which indicates that the adhesive becomes more brittle due to the presence of the particles within its matrix.

In order to support the experimental results that were shown previously, it is also relevant to report the failure patterns representative of each of the SLJs configurations. With this in mind, Figure 31 exhibits the fracture surfaces observed for the different volumes of hollow glass beads.

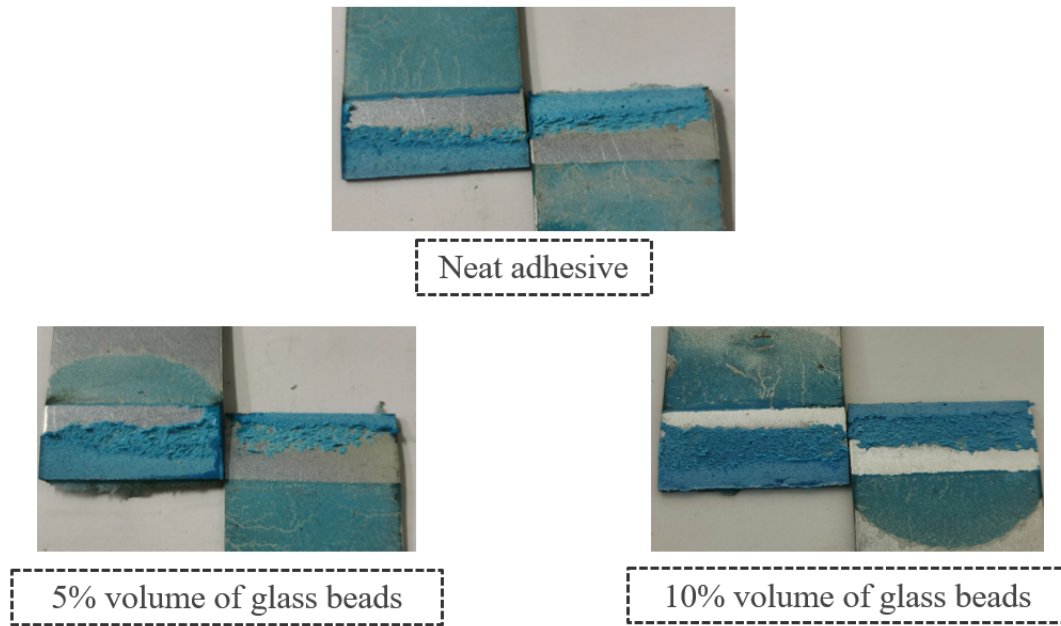


Figure 31: SLJs fracture surfaces of Adhesive B as a function of the added hollow glass beads' amounts that were employed.

Unlike what happened to Adhesive A, the presence of the spheres did not result in a transition from adhesive to cohesive failure. However, the area of the regions without traces of adhesive in the ends of the bonded joints seems to be decreasing. Therefore, it is reasonable to infer that, if the number of glass beads were increased, then the areas without adhesive would gradually decrease until cohesive failure was reached, similarly to what was reported for Adhesive A.

For Adhesive B, and similarly to what had been done for Adhesive A, the SLJs tests were recorded using a high speed camera. In accordance with the fracture surfaces displayed in Figure 31, the addition of glass beads to the adhesive layer did not result in a change of the crack's propagation path. In fact, the failure pattern reported with neat adhesive and with the addition of glass particles was rather similar, as it can be observed in Figure 32. Frames I, II, III and IV correspond to the stages before, in the beginning, during and in the end of the crack's propagation, respectively.

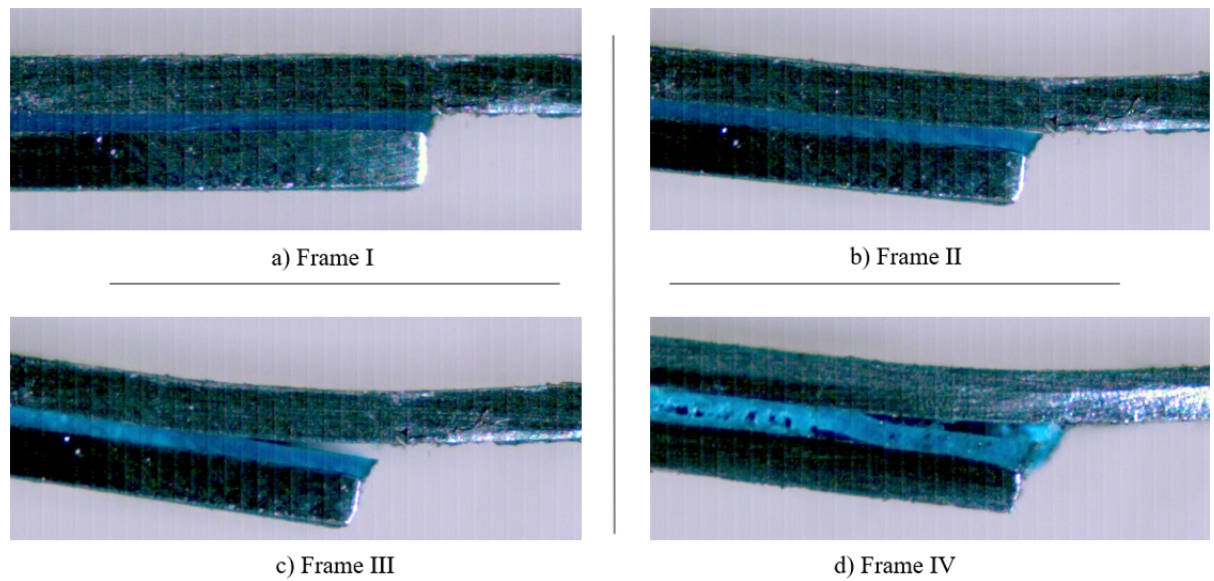


Figure 32: Representative frames of the different stages associated to the failure of a SLJ with Adhesive B.

Observing the representative frames displayed in Figure 32 allows to conclude that the failure pattern of the SLJs with Adhesive B closely resembles the failure pattern observed in the SLJs with neat Adhesive A. Again, the crack starts to propagate interfacially, and continually follows that path. Finally, just before failing, its direction changes, moving closer to the middle of the adhesive. With that said, the representative scheme presented in Figure 26 is also applicable in reproducing the crack's propagation path for the SLJs with Adhesive B.

B.3.3 SEM analysis

Throughout the SEM analysis, the fracture surfaces of the single lap joint specimens with mild steel substrates were thoroughly scrutinized, both for Adhesive A and Adhesive B.

Starting off with the analysis of the fracture surfaces of the SLJs with Adhesive A, Figure 33 displays a comparison between the transition regions (from the adhesive to the end of the bonded joint) in the SLJs with neat adhesive and 15 % volume of glass beads, the two most extreme configurations.

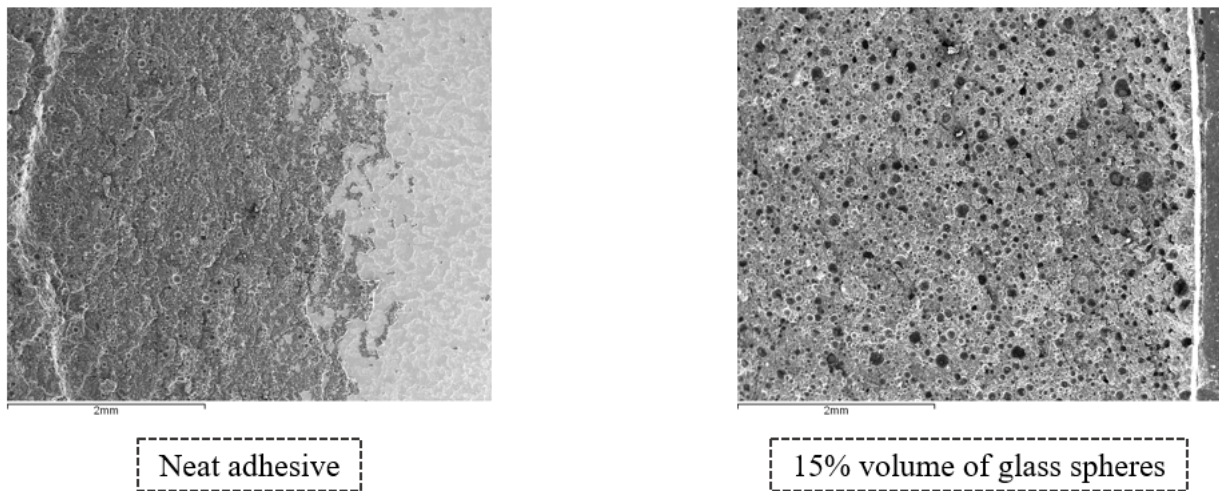


Figure 33: Fracture surfaces of SLJs of Adhesive A with neat adhesive and adhesive with added 15 %volume of glass beads, obtained through secondary electron analysis.

Figure 33 corroborates what had been concluded from visually examining the bonded regions represented in Figure 23, since it is possible to verify that the substrate's areas covered by adhesive do have a clear increase from the neat adhesive's configuration to the one with 15 % volume of glass beads, resulting from an evolution from adhesive to a cohesive failure.

Moreover, and as what had been detected during the SEM analysis in paper "Characterization of the effect of inorganic fillers on adhesives properties.", the hollow glass microspheres being used were found to have a significantly high level of adhesion to the adhesive's matrix, Figure 34.

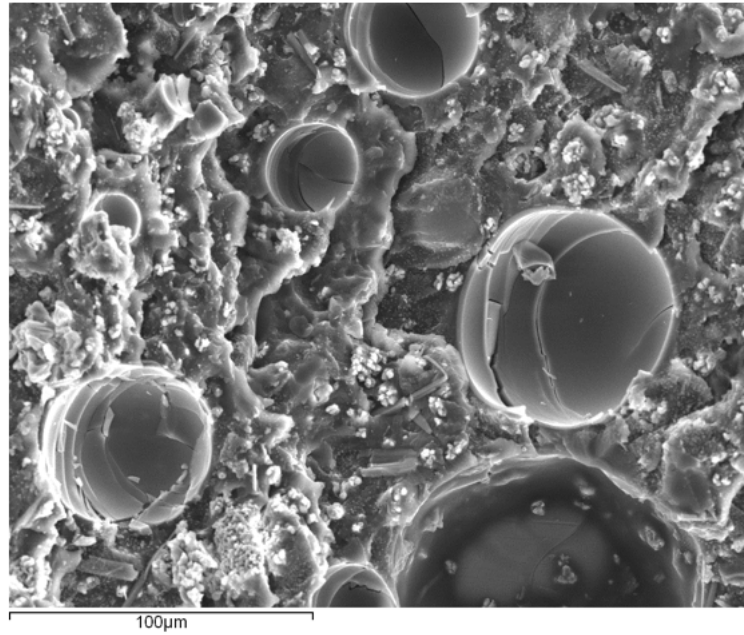


Figure 34: Fracture surface of a SLJ with Adhesive A and added 10 %volume of glass beads, with focus on the region around the glass beads, obtained through secondary electron analysis.

Figure 34 shows that, as a result of the noticeable good bonding between the glass particles and the adhesive's matrix, there is no particular damage or toughening mechanism in place, such as the debonding of the particles or the plastic void growth [3,6]. The nonexistence of such mechanisms also explains why the addition of glass beads does not work as a toughening method, since these mechanisms usually enhance the mechanical behavior of the adhesive, and consequently of the bonded joints.

Furthermore, in Figure 35, it is possible to see that a significant number of glass beads show a high level of damage, which also indicates that these work as weakening links in the adhesive's matrix, concentrating meaningful stress fields in the regions around them, eventually leading to their breakage.

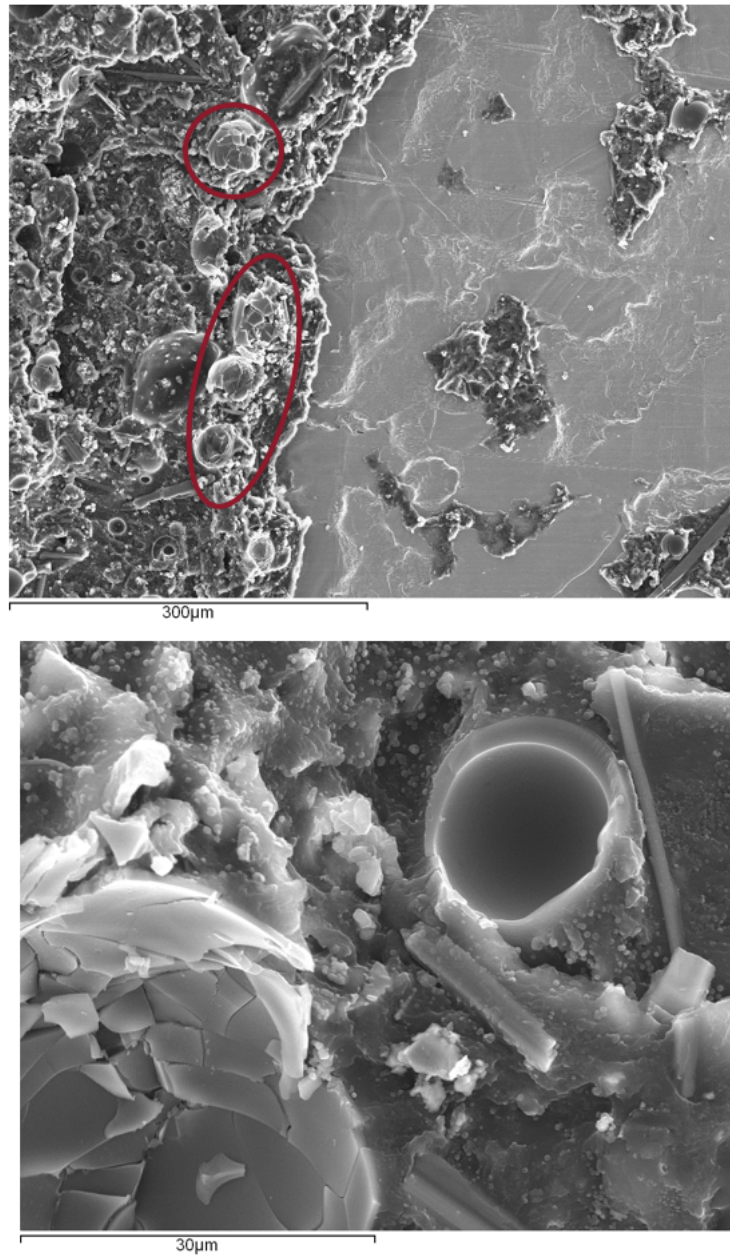


Figure 35: Fracture surface of a SLJ specimen with Adhesive A and added 10 %volume of glass beads, with focus on the region around the glass beads, obtained through secondary electron analysis.

With regards to the SLJs bonded with Adhesive B, Figure 36 represents the transition regions in the SLJs with neat adhesive and 10% volume of glass beads, which are, for this adhesive, the most extreme cases in terms of the amounts of glass particles.

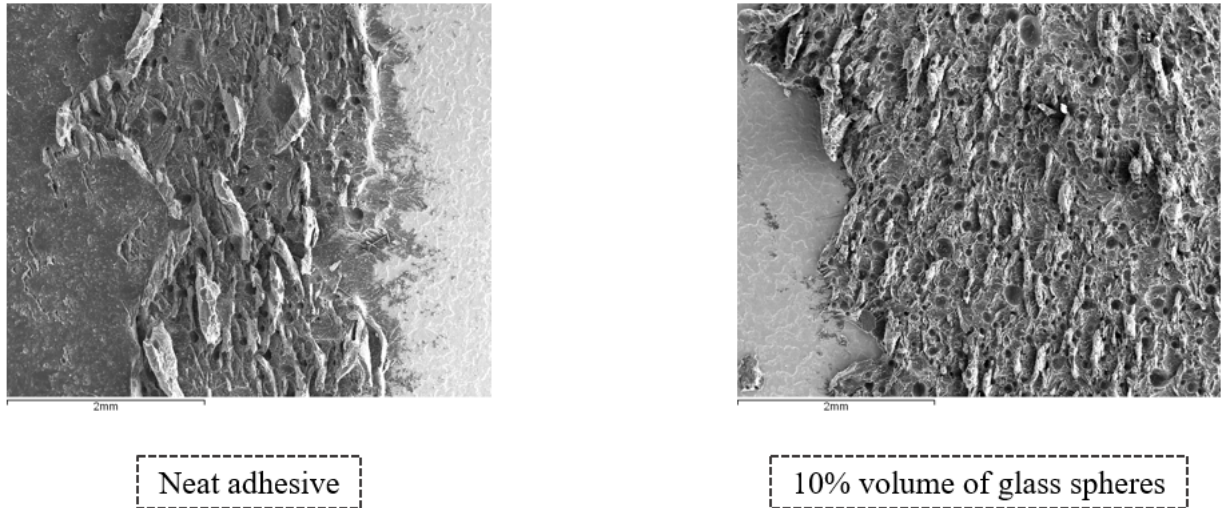


Figure 36: Fracture surfaces of SLJs with neat Adhesive B and adhesive with added 10 %volume of glass beads, obtained through secondary electron analysis.

As one can conclude from observing Figure 36, the addition of glass beads does not lead to a cohesive failure, for Adhesive B. However, the area of the regions which are not covered by the adhesive seems to decrease, in accordance to what had been concluded from Figure 31.

The fracture surface of the SLJ specimen with neat Adhesive B was magnified and a spectrum analysis was realized with further detail to one of its regions, Figure 37.

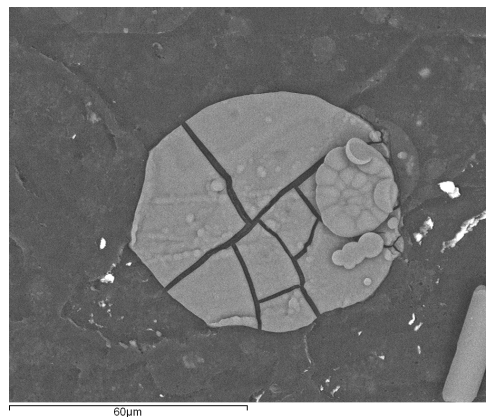


Figure 37: Spectrum analysis to the fracture surface of a SLJ specimen with neat Adhesive B, obtained through backscatter electron analysis.

As one can observe from Figure 37, it is possible to see that some of the calcium carbonate fillers along the adhesive are completely degraded, and again, no specific damage mechanism seems to take place as a consequence of their presence.

At last, for the case of the SLJ specimens with Adhesive B and 10% volume of glass

beads, these were also evaluated in terms of their damage state, Figure 38.

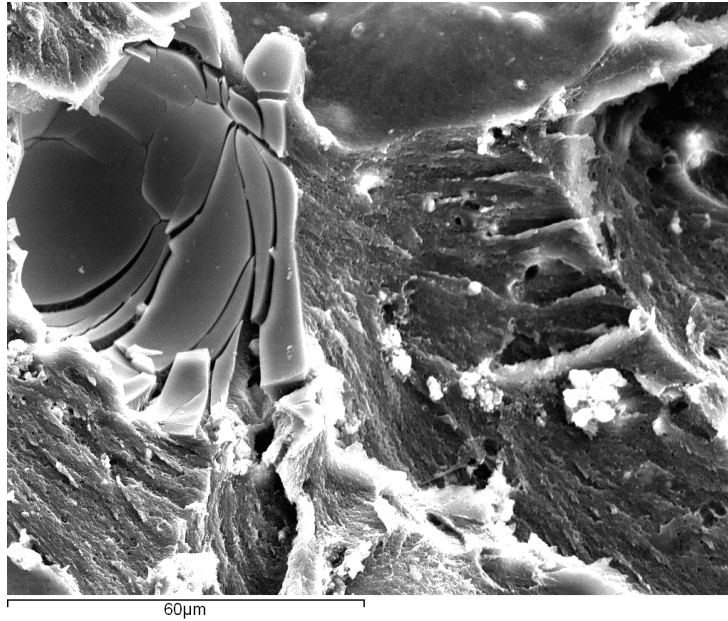


Figure 38: Fracture surface of a SLJ specimen with Adhesive B and added 10 %volume of glass beads, with focus on the region around the glass beads, obtained through secondary electron analysis.

Again, the glass beads reveal a good level of adhesion to the adhesive's matrix, and a significant number of them is completely shattered. This indicates that the glass beads have behaved similarly to what has been reported for the SLJ specimens with Adhesive A., with the formation of local stress concentrations which have the consequence of weakening the adhesive's matrix.

B.4 Comparison between the two adhesives

As a way of comparing the two presented adhesives, the $P - \delta$ curves as well as the respective fracture surfaces associated to the SLJs with high strength steel and mild steel substrates will be displayed.

One should note that, since neat Adhesive A already has 5 % volume of glass beads, the comparisons will be made between neat Adhesive A and Adhesive B with added 5% volume of glass beads, and Adhesive A with added 5% volume of glass beads and Adhesive B with added 10% volume of glass beads, which are the two comparable cases.

B.4.1 Single lap joint tests with high strength steel substrates

For the adhesives with 5% volume of glass beads, the $P - \delta$ curves and the fracture surfaces of each adhesive are presented in Figures 39 and 40.

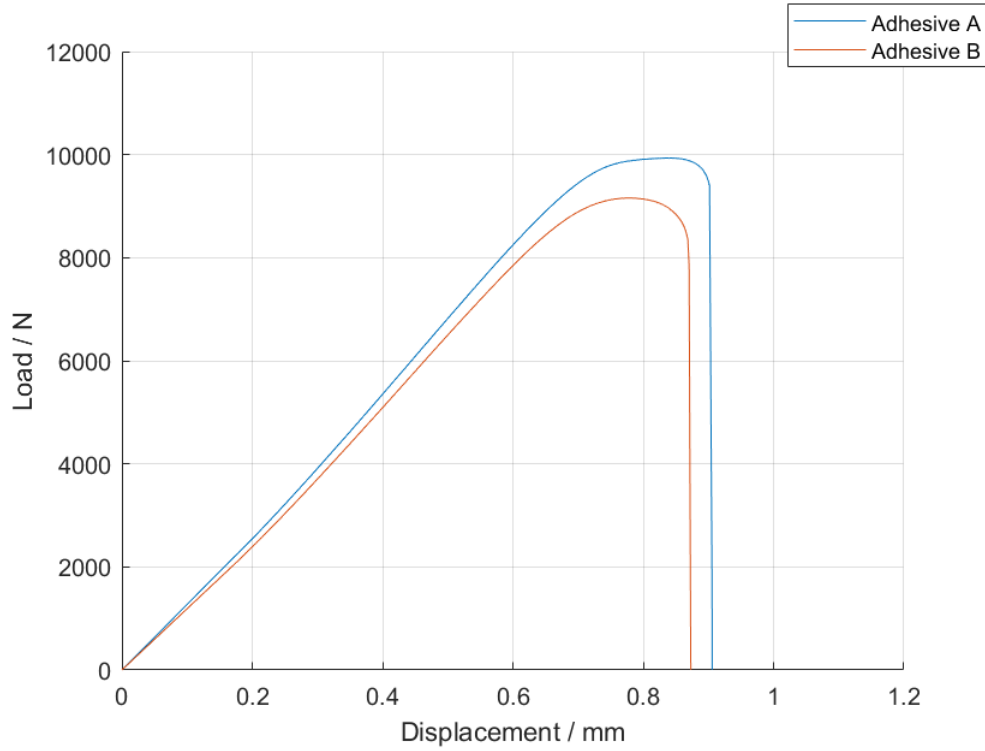


Figure 39: Representative $P - \delta$ curves for the single lap joint tests of adhesives with 5% volume of glass beads.

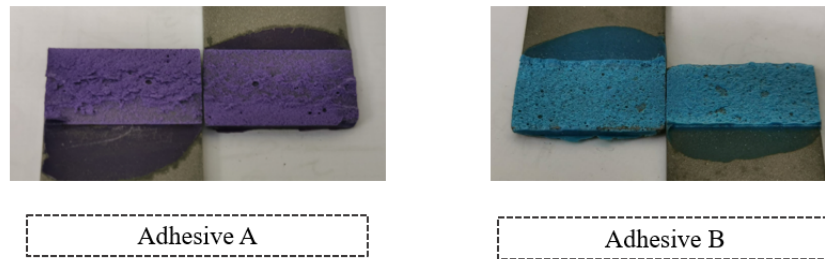


Figure 40: SLJs fracture surfaces of adhesives with 5% volume of glass beads.

For the adhesives with 10% volume of glass beads, the $P - \delta$ curves and the fracture surfaces of each adhesive are presented in Figures 41 and 42.

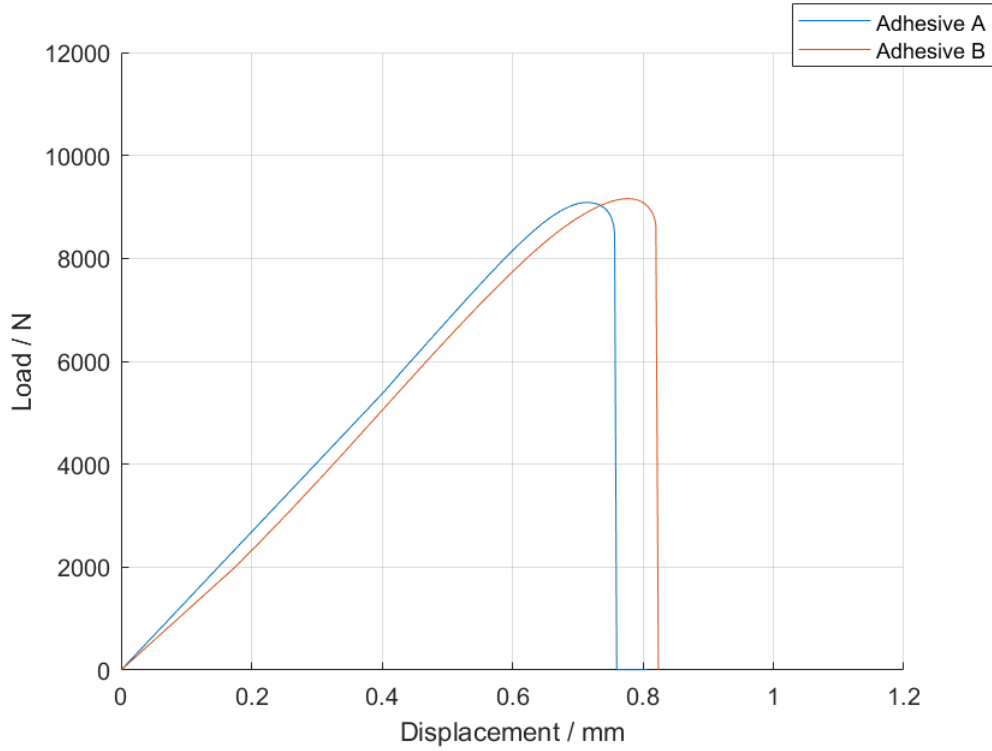


Figure 41: Representative $P - \delta$ curves for the single lap joint tests of adhesives with 10% volume of glass beads.

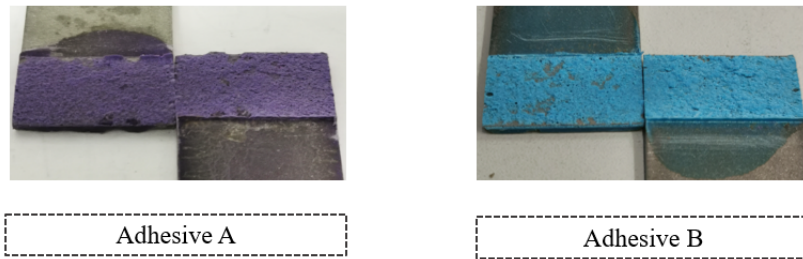


Figure 42: SLJs fracture surfaces of adhesives with 10% volume of glass beads

From an analysis of the results shown above, one can conclude that Adhesive A shows a slightly better mechanical performance than Adhesive B, for 5% volume of glass beads. For 10% volume of glass beads, Adhesive B shows a better mechanical behavior, even though both show very similar results. With regards to the SLJs fracture surfaces, the failure mode is always a fully cohesive one.

B.4.2 Single lap joint tests with mild steel substrates

For the adhesives with 5% volume of glass beads, the $P - \delta$ curves and the fracture surfaces of each adhesive are presented in Figures 43 and 44.

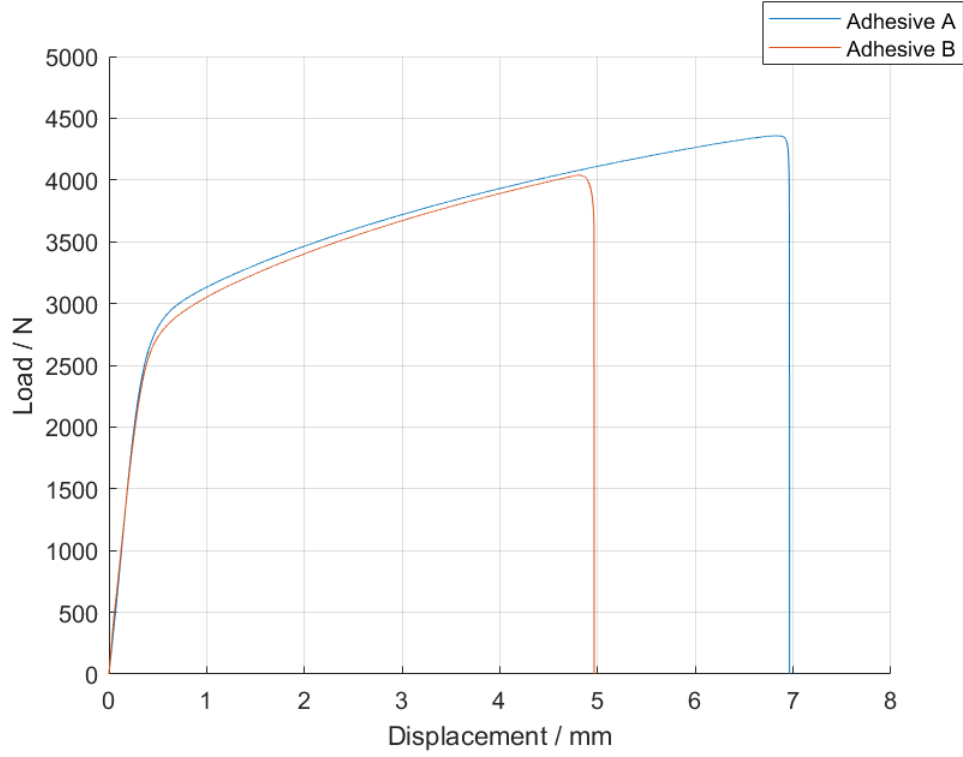


Figure 43: Representative $P - \delta$ curves for the single lap joint tests of adhesives with 5% volume of glass beads.

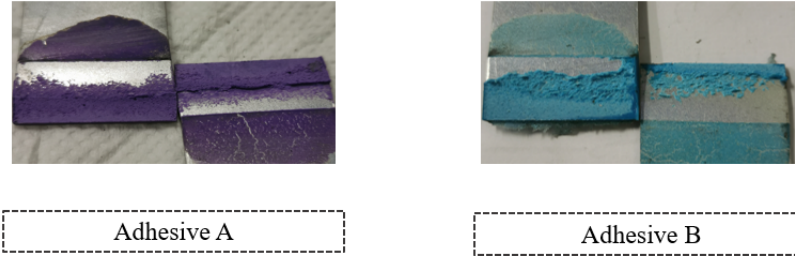


Figure 44: SLJs fracture surfaces of adhesives with 5% volume of glass beads.

For the adhesives with 10% volume of glass beads, the $P - \delta$ curves and the fracture surfaces of each adhesive are presented in Figures 45 and 46.

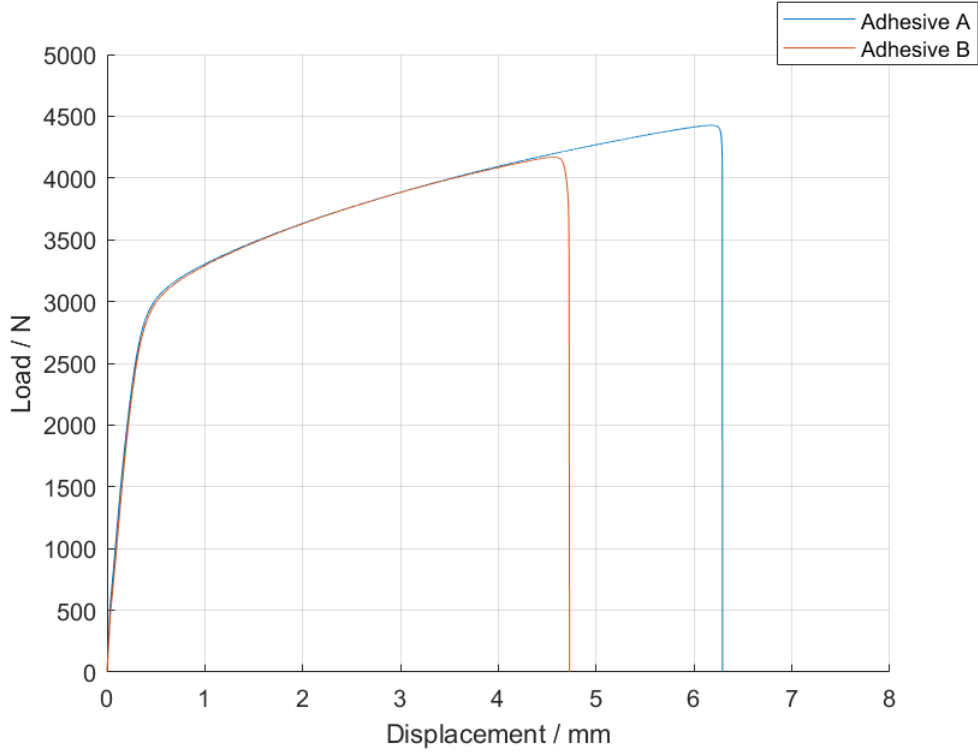


Figure 45: Representative $P - \delta$ curves for the single lap joint tests of adhesives with 10% volume of glass beads.

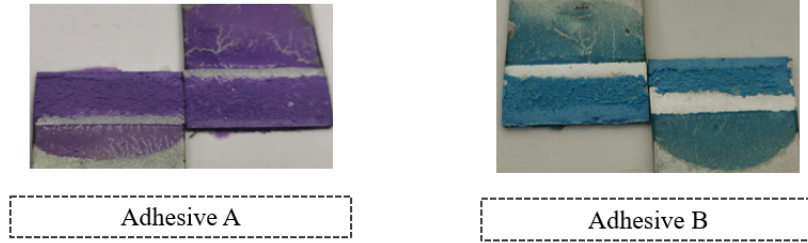


Figure 46: SLJs fracture surfaces of adhesives with 10% volume of glass beads.

The results represented above establish that Adhesive A has a better mechanical performance, both for 5 and 10% volume of glass beads. One can also see that the difference between the two adhesives slightly decreases for a 10% volume of added glass beads.

Regarding the SLJs fracture surfaces, both adhesives show an adhesive failure for the 5% volume of glass beads. However, by adding a further 5% volume of glass beads, the failure transits from an adhesive failure to a cohesive one, for Adhesive A. As stated previously, this happens as a consequence of the presence of the hollow glass microspheres, which introduce local stress fields around them, forcing the crack to propagate closer to the middle of the adhesive layer rather than in an interfacial region. Furthermore, and

for Adhesive B, even though the failure mode is still adhesive in nature, it is possible to observe that the bonded regions without adhesive clearly decrease by increasing the number of glass beads, which happens as a result of the same phenomenon that was just reported.

To sum up, and with regards to the SLJs with mild steel, Adhesive A would be a better solution due to the fact that it not only provides a better mechanical performance than Adhesive B, but also shows a cohesive failure for 10% volume of glass beads, which is an important factor in some industrial sectors, such as the automotive industry, since this is considered to be a safer failure mode and might facilitate a more precise estimate of the joint's strength.

B.5 Conclusions

The addition of hollow glass beads to the adhesive layer was investigated, with a special focus on the way this process affects the joint's strength and the corresponding failure mechanism. To that aim, single lap joint specimens with mild steel and high strength steel substrates were tensile tested, for different amounts of glass beads. The tensile tests allowed to determine the mechanical performance of the joints under study, and the corresponding fracture surfaces were analysed. All tests were performed at a quasi-static test rate.

The main conclusions extracted from this study are the following:

- Testing of the SLJs with high strength steel substrates showed that adding glass beads to the adhesive generally worsens the mechanical performance of the bonded joint's. This was found to be true for both adhesives. Besides, and for Adhesive B, the addition of particles to the adhesive layer forces the fracture mode to evolve from adhesive to cohesive failure.
- For both adhesives, the SLJs specimens with mild steel substrates were found to be more susceptible to the strength reduction as a result of introduction of hollow glass spheres to the adhesive. Doing so leads the crack to propagate in the middle of the adhesive rather than in the interfacial region, which causes the failure mode to evolve from an adhesive failure to a cohesive one.
- For Adhesive A, the addition of hollow glass spheres to the adhesive leads the crack to propagate in the middle of the adhesive layer rather than in the interfacial region, which forces the failure mode to evolve from an adhesive failure to a cohesive one.

For Adhesive B, even though the area of the regions without adhesive in the ends of the bonded joints decrease, the failure does not evolve from adhesive to cohesive.

- The hollow glass beads do not work as tougheners of the adhesive and the joint, since they do not contribute to the creation of damage mechanisms within the adhesive. Oppositely, they create local stress concentration in their periphery, which weaken the adhesive's mechanical performance.

References

- [1] Lucas FM da Silva, Andreas Öchsner, and Robert D Adams. *Handbook of adhesion technology*. Springer Science & Business Media, 2011.
- [2] WA Lees. Adhesive selection. In *Adhesives in Engineering Design*, pages 92–123. Springer, 1984.
- [3] Ali Nemati Giv, Majid R Ayatollahi, S Hengameh Ghaffari, and Lucas FM da Silva. Effect of reinforcements at different scales on mechanical properties of epoxy adhesives and adhesive joints: a review. *The Journal of Adhesion*, 94(13):1082–1121, 2018.
- [4] AJ Kinloch. Toughening epoxy adhesives to meet today’s challenges. *MRS bulletin*, 28(6):445–448, 2003.
- [5] AQ Barbosa, Lucas FM da Silva, and A Öchsner. Effect of the amount of cork particles on the strength and glass transition temperature of a structural adhesive. *Proceedings of the Institution of Mechanical Engineers, Part L: Journal of Materials: Design and Applications*, 228(4):323–333, 2014.
- [6] AJ Kinloch, JH Lee, AC Taylor, S Sprenger, C Eger, and D Egan. Toughening structural adhesives via nano-and micro-phase inclusions. *The Journal of Adhesion*, 79(8-9):867–873, 2003.
- [7] CI da Silva, AQ Barbosa, RJC Carbas, EAS Marques, A Akhavan-Safar, and Lucas FM da Silva. Influence of cork microparticles on the fracture type in single lap joints. *Proceedings of the Institution of Mechanical Engineers, Part C: Journal of Mechanical Engineering Science*, page 0954406220936729, 2020.
- [8] R Hunter, J Möller, A Vizán, J Pérez, J Molina, and J Leyrer. Experimental study of the effect of microspheres and milled glass in the adhesive on the mechanical adhesion of single lap joints. *The Journal of Adhesion*, 93(11):879–895, 2017.
- [9] 3M Advanced Materials Division. 3m glass bubbles, 2018.
- [10] VV Budov. Hollow glass microspheres. use, properties, and technology. *Glass and ceramics*, 51(7-8):230–235, 1994.

- [11] EF Karachalios, RD Adams, and Lucas FM da Silva. Single lap joints loaded in tension with high strength steel adherends. *International Journal of Adhesion and Adhesives*, 43:81–95, 2013.
- [12] CSP Borges, PDP Nunes, A Akhavan, EAS Marques, RJC Carbas, L Alfonso, and Lucas FM da Silva. Influence of mode mixity and loading rate on the fracture behaviour of crash resistant adhesives. *Theoretical and Applied Fracture Mechanics*, 107:102508, 2020.
- [13] Robert D Adams, John Comyn, William Charles Wake, and WC Wake. *Structural adhesive joints in engineering*. Springer Science & Business Media, 1997.

C Paper C

Numerical model of the damage mechanism in joints bonded using adhesives reinforced with inorganic fillers

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ABSTRACT

The use of adhesive bonding in the diverse industries, such as the automotive and aerospace sectors has grown considerably. In structural construction, adhesive joints provide a unique combination of low structural weight, high strength and stiffness, combined with a relatively simple and easily automated manufacturing's method, characteristics which are ideal for the development of modern and highly efficient vehicles. In these applications, ensuring that the failure mode of a bonded joint is cohesive rather than adhesive is important, since this failure mode is more controlled, easier to model and to predict. This work presents a technique which enables the precise prediction of the bonded joint's behavior, regarding not only its failure mode but also the joint's strength, when inorganic fillers are added to the adhesive. To that end, hollow glass particles were introduced into an epoxy adhesive's layer in different amounts, and a numerical study was carried out to simulate the behavior of single lap joint specimens. The numerical results were compared with experimentally obtained ones, not only in terms of the joint's strength but also the failure pattern. Both results showed that, by adding glass beads to the adhesive, the crack tends to propagate cohesively, through the middle of the adhesive, and that the joint's performance was generally negatively impacted by the presence of these particles.

C.1 Introduction

Adhesive bonding is a joining technique which now finds use in diverse structural applications, ranging from the electronics industry to the automotive and aerospace sectors. Bonded joints exhibit generally uniform stress distributions and provide excellent stiffness and strength to weight ratios. In addition, this technique lends itself for industrial implementation in modern structures, since it is well suited for automated processes, can be used to join parts with complex geometries, composed of distinct designs, contours, thicknesses and, perhaps most importantly, it is compatible with a wide range of different materials, with only minimal modifications to the substrates being necessary [1, 2].

Despite this growth, adhesive bonding is still a relatively new technique and there is still a large interest in adopting methodologies and techniques which are suitable for enhancing the performance of bonded joints. The nature of these techniques is varied. Some are based on the application of functionally graded properties, via temperature gradients for example, the combination between adhesive joints and other conventional joining methods, known as hybrid joints, the geometrical optimization of the adherends' configurations [1], adoption of novel surface treatments, among many others [3].

Nevertheless, most of the approaches described provide limited improvements in return for greatly added process complexity. For those reasons, introducing reinforcements into the adhesive layer has become a popular and inexpensive alternative to improve adhesive joint performance [3].

Epoxy adhesives are widely used in structural applications, being amorphous and highly cross-linked materials, with good strength and stiffness but suffering from a generally brittle behavior. Thus, the addition of a second phase of particles, fibers, or other kinds of reinforcements, has a strong potential to enhance the adhesive's mechanical behavior, depending on the reinforcements' properties [4].

In this work, a numerical study, supported with experimental data, was carried out in order to understand how the failure mode of joint bonded with an epoxy adhesive varies with the introduction of inorganic particles into the adhesive layer. There are four main failure modes which are usually observed when analysing the fracture surfaces of adhesively joined specimens, which are represented in Figure 1. One of the main failure patterns is the interfacial or adhesive failure, which includes the failures where the crack follows a path along the interface between the adhesive and the adherends. This failure mode can appear as a consequence of incorrect surface preparation and, due to the low strength it provides, it is totally unacceptable in an industrial context. Cohesive failure in

the adherend corresponds the cases where the adhesive bond and the adhesive's resistance are stronger than the adherend. Moreover, cohesive failure in the adhesive is a failure mode where the crack propagates along the adhesive, with the adherends' surfaces still coated with a layer of adhesive. Such failure usually occurs when the adhesion between the adhesive and the adherend is very strong, and the substrates can support an higher load than the adhesive. Lastly, alternating or mixed failure involves the cases where the crack's propagation path alternates between the adhesive and the adherend's interfaces in the same failure process [1].

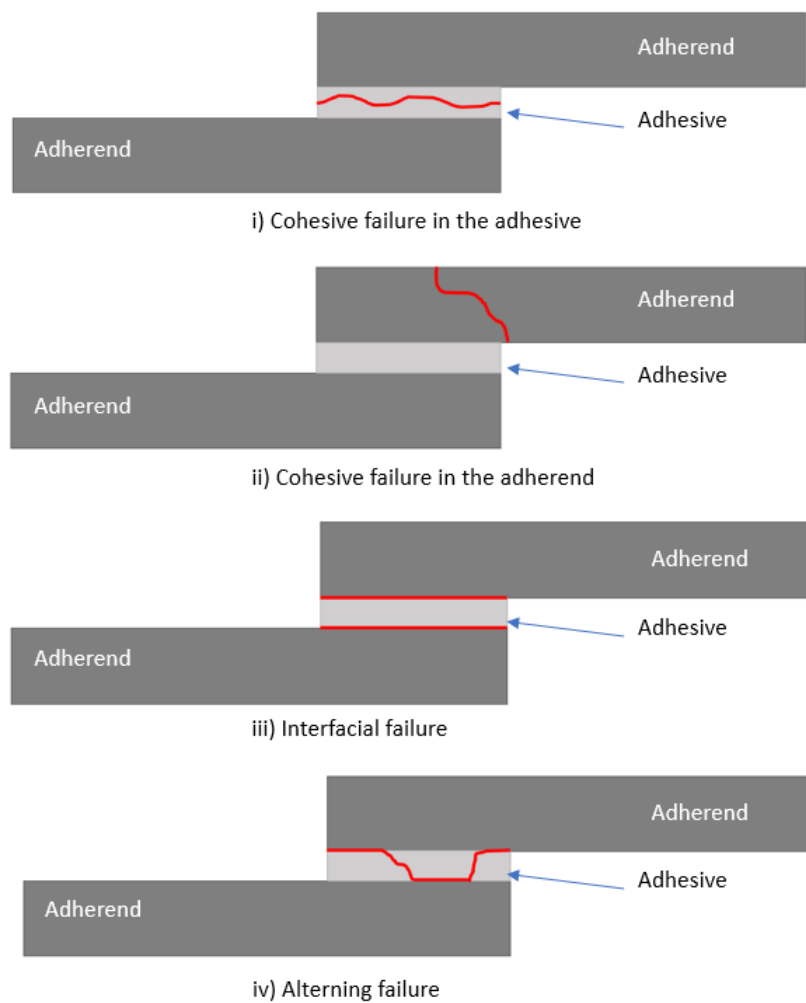


Figure 1: Schematic representation of the different types of failure.

With respect to this subject, and focusing particularly on the failure mode's variation with the addition of glass microspheres, Hunter et al. [5] investigated how the presence of that type of particles influences the mechanical adhesion and behavior of single-lap joints with fiber-reinforced polymers, for adhesives with different curing speed. Throughout

this study, it was concluded that their use has different repercussions depending on the adhesive's curing speed. The slow-curing adhesive showed a joint strength decrease for the 3% and the 10% weight concentrations of glass microspheres, when compared to the neat adhesive's bonded joint. On the other hand, the fast-curing epoxy adhesive revealed a joint's strength increase for all amounts of glass microspheres, when compared to the neat adhesive's configuration. Regarding the failure modes, thin-layer cohesive and fiber tear failure were the failure modes obtained for the slow-curing adhesive, while interfacial failure was also reported for the fast-curing adhesive, as well as the previously mentioned failure modes.

To enable the prediction of the failure modes in bonded joints using adhesives reinforced with glass microspheres, which is the main focus of this paper, the eXtended finite element method (XFEM) was the chosen. This method is an extension of the finite element method (FEM). In fact, XFEM was developed since FEM, which allows to precisely predict the stresses and other mechanical phenomena acting on bonded structures [6, 7], showed difficulties in modelling discontinuities, such as cracks' propagation.

An alternative to XFEM are the cohesive zone models (CZM), also capable of predicting the fracture behavior, the damage initiation and propagation, as well as the joint's strength, by using traction separation-laws that dictate the behavior of its cohesive elements [8]. However, using CZM requires defining the path that the crack will follow beforehand, limiting its ability to predict how a failure process will take place. Taking this into account, and since the aim of this numerical study is to predict the failure mode of the bonded joints, XFEM was the selected method, due to the fact that it allows cracks to grow freely [9, 10].

XFEM is finite element technology which was created in a recent past as an extent to the traditional finite elements' method, having been firstly introduced by Belyschko and Black in 1999. This method allows the crack to be modeled independently of the mesh, without being dependent of any kind of remeshing, and models its path [11]. XFEM can be defined as a minimal remeshing finite element method, and is characterized for introducing discontinuous enrichment functions to the conventional finite element displacement function in order to properly simulate the presence of the crack [9, 12]. When there is damage propagation, phantom nodes are formed to subdivide the elements that are intersected by the crack and enable the existence of discontinuities between the new elements. This allows the crack to propagate along an arbitrary path [11].

In Figure 2, one can observe two representative images of the propagation of a crack in XFEM elements. When a crack propagates through an element, it gets partitioned into

two sub-elements, Ω_A and Ω_B . Initially, the element represented in a) had nodes $n_1 - n_4$, and after being intersected by the crack, four phantom nodes, $\tilde{n}_1 - \tilde{n}_4$, were created and added to allow the existence of discontinuities between the new sub-elements. Therefore the element represented in a), was replaced by two new sub-elements, in b): Ω_A , composed of the nodes $n_1, n_2, \tilde{n}_3$ and $\tilde{n}_4$; and Ω_B composed of the nodes $n_3, n_4, \tilde{n}_1$ and $\tilde{n}_2$ [11].

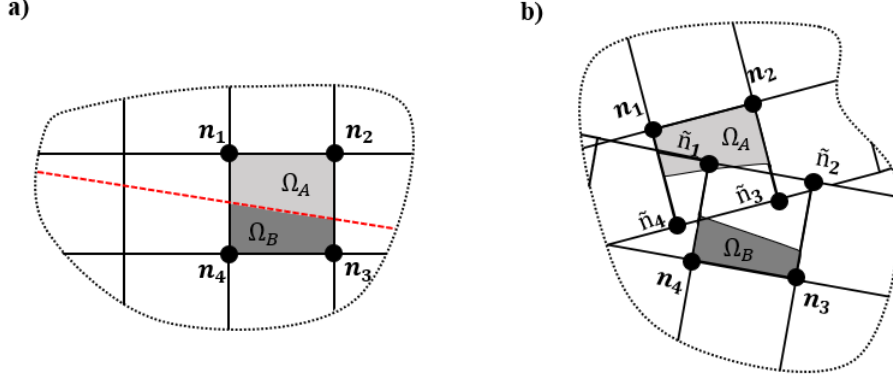


Figure 2: Representative scheme of the partition phenomenon associated to the propagation of a crack in XFEM.

The equation below represents the displacement vector associated to this method [11]:

$$\mathbf{u} = \sum_{I=1}^N N_I(x) \left[\mathbf{u}_I + H(x)\mathbf{a}_I + \sum_{\alpha=1}^4 F_{\alpha}(x)\mathbf{b}_I^{\alpha} \right] \quad (1)$$

where $N_I(x)$ represents the nodal shape function and u_I is the nodal displacement vector of the continuous part of the formulation [11].

Moreover, $H(x)\mathbf{a}_I$, which is the generalized Heaviside enrichment function, and $F_{\alpha}(x)\mathbf{b}_I^{\alpha}$ account for the nodes whose shape function is intersected by the crack. The term $H(x)$ can be generally designated as the jump function or discontinuous shape function, which allows to model the discontinuities across the crack over the points along the crack surface [9].

$$H(x) = \begin{cases} 1 & \text{if } (x - x^*) \cdot n \geq 0 \\ -1 & \text{otherwise} \end{cases} \quad (2)$$

where x is a sample Gauss integration point, x^* is the point of the crack closest to x , and n is the unit vector normal to the crack at x^* [13].

The term n is multiplied by $\mathbf{a}_I$, which is the nodal enriched degree of freedom vector [11]. Another term that also stands for the nodal enriched degree of freedom vector is $\mathbf{b}_I^{\alpha}$.

Moreover, $F_\alpha(x)$ represents the asymptotic crack-tip functions [11].

XFEM can generally follow two damage modelling approaches, which are the cohesive segments approach, and the Linear Elastic Fracture Mechanics approach (LEFM) [11].

The cohesive segment approach is the most widely used for the modelling of bonded joints. This approach is governed by traction-separation-laws, and the damage properties are defined as part of the bulk material definition. Three stress-based and three strain-based damage initiation criteria can be used while following this method, and the crack's initiation depends on the stress/strain value at the center of the enriched elements [11]. The Macaulay Brackets are used in order to specify that a purely compressive stress does not induce damage [14].

- Maximum principal stress (MAXPS) and maximum principal strain (MAXPE), which have the following criteria, respectively:

$$f = \frac{\langle \sigma_n \rangle}{\sigma_{max}^0} \quad (3)$$

where σ_n represents the maximum principal stress at an integration point and σ_{max}^0 is related to the material strength in terms of tension.

$$f = \frac{\langle \varepsilon_n \rangle}{\varepsilon_{max}^0} \quad (4)$$

where ε_n is the maximum principal strain at an integration point and ε_{max}^0 is the material strength in terms of deformation [14].

For these criteria, the crack initiates when $f=1$ and its plane is perpendicular to the direction of the maximum principal stress [11].

- Maximum nominal stress (MAXS) and maximum nominal strain (MAXE), whose criteria are the following, respectively:

$$f = MAX\left\{\frac{\langle \sigma_n \rangle}{N_{max}}, \frac{\sigma_t}{T_{max}}, \frac{\sigma_s}{S_{max}}\right\} \quad \langle \sigma_n \rangle = \begin{cases} \sigma_n & \text{if } \sigma_n > 0 \\ 0 & \text{if } \sigma_n < 0 \end{cases} \quad (5)$$

where σ_n and N_{max} are the maximum nominal stress and the critical stress value in a normal-only mode at an integration point, σ_t and T_{max} are the maximum nominal stress and the critical stress value in a shear-only mode along the first shear direction,

and σ_s and S_{max} are the maximum nominal stress and the critical stress value in a shear-only mode along the second shear direction [11].

$$f = MAX\left\{\frac{\langle \varepsilon_n \rangle}{\varepsilon_n^{max}}, \frac{\varepsilon_t}{\varepsilon_t^{max}}, \frac{\varepsilon_s}{\varepsilon_s^{max}}\right\} \quad \langle \varepsilon_n \rangle = \begin{cases} \varepsilon_n & \text{if } \varepsilon_n > 0 \\ 0 & \text{if } \varepsilon_n < 0 \end{cases} \quad (6)$$

where ε_n and ε_n^{max} are the maximum nominal strain and the critical strain value in a normal-only mode at an integration point, ε_t and ε_t^{max} are the maximum nominal strain and the critical strain value in a shear-only mode along the first shear direction, and ε_s and ε_s^{max} are the maximum nominal strain and the critical strain value in a shear-only mode along the second shear direction [11].

For these cases, the damage initiates when $1.0 \leq f \leq 1.0 + f_{tol}$, being f_{tol} a tolerance value specified by the user, and the user may also define the crack plane normal direction [11].

- Quadratic nominal stress (QUADS) and quadratic nominal strain (QUADE), whose criteria are the following, respectively:

$$\left(\frac{\langle \sigma_n \rangle}{N_{max}}\right)^2 + \left(\frac{\sigma_t}{T_{max}}\right)^2 + \left(\frac{\sigma_s}{S_{max}}\right)^2 = 1 \quad (7)$$

$$\left(\frac{\langle \varepsilon_n \rangle}{\varepsilon_n^{max}}\right)^2 + \left(\frac{\varepsilon_s}{\varepsilon_s^{max}}\right)^2 + \left(\frac{\varepsilon_t}{\varepsilon_t^{max}}\right)^2 = 1 \quad (8)$$

For these criteria, the user also defines the crack plane normal, as for MAXS and MAXE [11].

Regarding the application of XFEM in the numerical study of adhesively bonded joints, da Silva et al. [15] used this method to predict the failure mode of single lap joints reinforced with cork microparticles. This study showed that XFEM can be used as a valid method to predict the failure pattern, as well as the mechanical performance of bonded joints. The failure modes that were experimentally obtained were fully validated numerically and it has been observed that, by increasing the size and the amount of particles, the crack evolves from an adhesive failure to a cohesive one.

Campilho et al. [10] also studied the applicability of XFEM to simulate the mechanical behaviour of DCB specimens, as well as their crack propagation process. The numerical results were compared to the experimental $P - \delta$ curves and the critical strain energy in

pure mode I values. The comparison between these results proved that XFEM provides a reliable simulation of the behavior of adhesively bonded DCB specimens, and can therefore be used to correctly model bonded structures.

In this study, the effects of reinforcing an adhesive with inorganic fillers were studied, supported by a numerical study conducted using XFEM. Hollow glass microspheres were introduced into an epoxy adhesive's layer, and different percent volumes of particles were considered. This allowed to assess not only the crack's path and the corresponding failure mode, but also the strength of the bonded joints. The numerical results were compared to the ones obtained experimentally.

C.2 Experimental details

This section describes the materials and specimen geometries that were used during this experimental work, the corresponding mechanical properties, and the specimens manufacturing and testing procedures.

C.2.1 Materials characterization

In this work, the selected adhesive will be referred to as Adhesive A, which is a one component epoxy and crash-resistant adhesive . This adhesive is used in automotive applications, with the goal of increasing durability, crash performance and body stiffness. Following manufacturer recommendations, it cures at 180 °C for a period of 30 minutes.

The mechanical properties of Adhesive A were determined by tensile testing of bulk specimens, as well as the testing of thick adherend shear test (TAST), double cantilever beam (DCB) and end-notched flexure (ENF) specimens. These were carried out to determine the tensile strength, failure strain and Young's modulus (bulk specimens), the shear strength (TAST specimens), the fracture toughness in mode I (DCB specimens), G_{Ic} , and the fracture toughness in mode II (ENF specimens), G_{IIc} . At least three specimens were tested for each case. Table 1 summarizes the data collected regarding this adhesive's characterization.

Table 1: Adhesive characterization for Adhesive A.

Property	Adhesive A
Young's modulus/ MPa	2136.6 ± 117.0
Poisson's ratio	0.3
Tensile strength/ MPa	30.2 ± 0.4
Shear strength/ MPa	30.9 ± 0.9
G_{Ic} / MPa	2.6 ± 0.1
G_{IIc} / MPa	10.7 ± 0.1

The hollow glass beads used during this work consist of thin-walled hollow spheres made of soda-lime-borosilicate glass, with a white powder-like appearance. Their size and density can vary, and they are characterized by their light weight, high strength-to-density ratio, and a considerable isostatic crush strength [16, 17].

In Table 2 follow the main mechanical properties associated to the glass beads used. These properties were determined indirectly via a reverse analysis.

Table 2: Mechanical properties of the hollow glass beads.

Property	-
Young's modulus/ MPa	3500
Poisson's ratio	0.23
Tensile strength/ MPa	1
Shear strength/ MPa	0.8
G_{Ic} / MPa	0.25
G_{IIc} / MPa	0.1

C.2.2 Single lap joints manufacturing and testing

Single lap joint specimens. For this work, SLJ specimens were manufactured and tested for neat Adhesive A and adhesive with 5,10 and 15% volume of glass beads. For this single lap joint configuration, an adherend composed of mild steel was used, and Figure 3 represented the corresponding geometry.

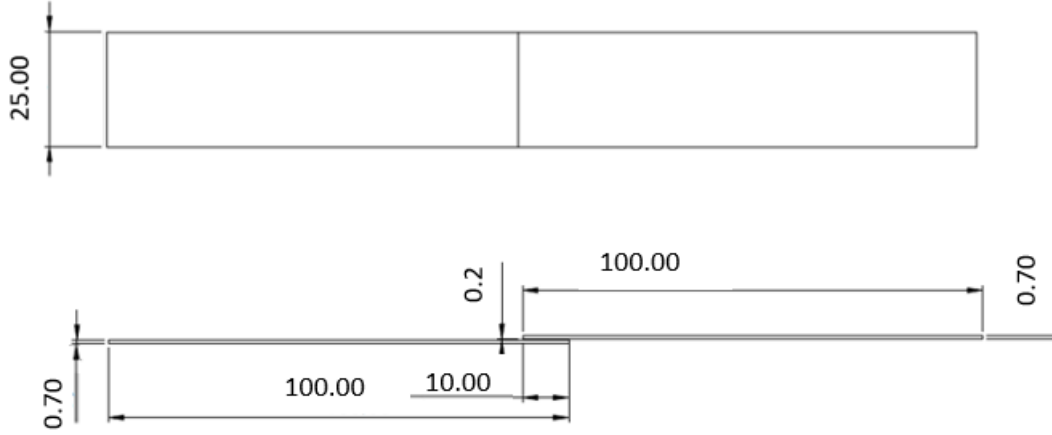


Figure 3: Representation of single lap joint specimens' geometry, dimensions in mm.

The adherends' surfaces to be bonded were degreased with acetone, with the aim of cleaning the surfaces and ensuring the proper removal of any kinds of contaminants, dust or oils.

The SLJ specimens were manufactured using an annealed carbon steel mould that enables to control the overlap length, the adhesive's thickness and the proper alignment of the substrates, which is made possible due to its alignment pins, shims and positioner blocks. Release agent was used to coat the mould, facilitating the removal of the specimens after the adhesive cure is complete.

The introduction of the glass beads into the adhesive layer was achieved by mixing them in the pre-heated epoxy adhesive. The pre-heating process lower the adhesive's viscosity and facilitates an uniform distribution of the particles along the adhesive's matrix. The actual mixing process was carried out using a centrifugal mixing machine, Speed-Mixer DAC 150TM (Hauschild, Hamm, Germany) for a total of 40 s. The mixing speed was increased gradually from 500 r/min to a 3000 r/min rotation speed during the first 10 seconds, followed by a constant velocity of 3000 r/min during the remaining 30 s. This procedure was adopted for all the different amounts of hollow glass beads.

After the curing stage was completed, the specimens were removed from the mould, and the excess adhesive was manually extracted from the specimen using a file.

C.3 Numerical modelling

Numerical simulations were performed in order to predict the bond's strength and the failure pattern of SLJs when tensile tested, at quasi-static conditions. The aim of this numerical study was to clearly understand how the presence of the glass beads affects the failure mode and strength of the bonded joint. To that end, models were created in ABAQUS[®] (Dassault Systems, Providence, USA), adopting elements which support the XFEM technique

Throughout this numerical study, SLJs with neat Adhesive A and with 5 and 10% volume of glass beads were analysed, since experimentally, these SLJs configurations were shown to noticeably alter their failure mode and mechanical performance as a function of the glass bead amounts. The main goal of this work was to comprehend the influence of the glass beads on the crack's propagation path of the SLJ, and consequently on the specimen's failure mode, which should evolve from an adhesive failure to a cohesive one. Moreover, it is also relevant to collect and compare the $P - \delta$ curves to better understand the impact that the glass beads have on the joint's performance.

The SLJ's geometry adopted in the model is the same that was tested in the experimental procedure, Figure 3.

During this study, a triangular traction-separation law was adopted to model the adhesive behavior, similar to the one which is portrayed in Figure 4, and assuming a linear softening. This type of law is created with t_n^0 and t_s^0 , which represent the cohesive strengths in tension and shear, respectively, δ_n^0 and δ_s^0 , which are the corresponding displacements, and δ_n^f and δ_s^f , which refer to the tensile and shear displacements at failure. The triangular traction-separation law considers an initial linear elastic behaviour, until reaching δ_n^0 and δ_s^0 . Then, a linear evolution until damage follows, which is associated to δ_n^f and δ_s^f [14].

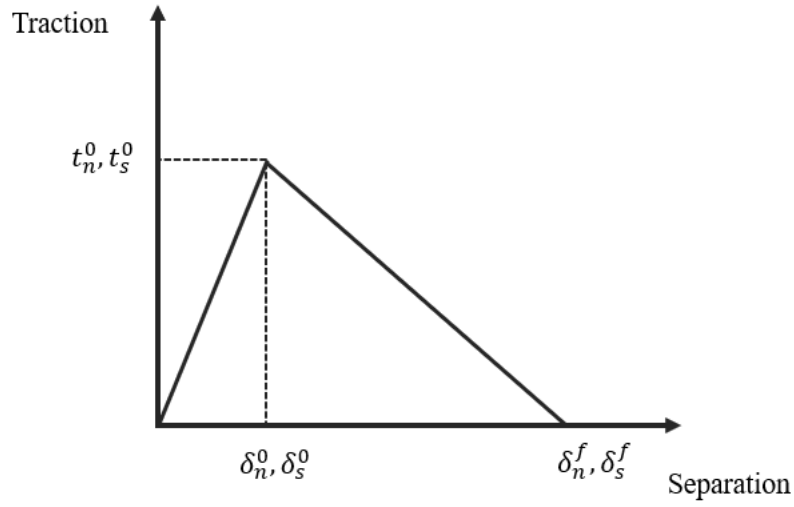


Figure 4: Schematic representation of the triangular traction separation law.

The boundary conditions established during these simulations were the ones that follow in Figure 5. The restraining and loading conditions were set and consisted in a restriction of the horizontal and vertical movement in the clamped end of the joint (in the left adherend). The opposite adherend was subjected to a horizontal displacement, although vertical movement was restricted, closely simulating the loads applied during testing.



Figure 5: Boundary conditions established for the simulation of the SLJ behavior.

It is also relevant to state that the model does not consider the presence of defects in adhesive layer nor in the interfaces between the adhesive and the substrates.

C.3.1 XFEM method

To create the geometry of the SLJ model, a script in MATLAB[®] (Mathworks, Massachusetts, USA) was initially developed, which plots glass spheres along the adhesive layer. The coordinates generated by this script were introduced in AutoCAD[®], and the model geometry was imported into Abaqus[®].

Regarding the properties that were attributed to each material, an elasto plastic isotropic response was considered for the mild steel adherends. This kind of response

was chosen due to the fact that, during the experimental tests, considerable plastic deformation of the substrates was encountered. This plastic deformation had a significant influence on the SLJ's performance. For the adhesive and the glass beads, the crack initiation criterion used was based on the quadratic stress criterion (QUADS). The properties which were assigned for the damage law associated to this criterion were the ones represented in Tables 1 and 2, for the adhesive and the glass beads, respectively.

The model was meshed with plane strain solid elements, with QUAD (quadrilateral) structured shapes. The meshes were created with bias effects towards the adhesive bond, as these regions are expected to exhibit larger stress gradients which were found to require finer meshes. As one can observe in Figure 6, which presents the representative meshes of the SLJs models which were created, the meshes were further refined for the case of SLJs with glass particles, since these call for smaller element sizes in order to properly mesh their geometries. The region around the glass beads were also further refined in these cases, minimizing large element size transitions.

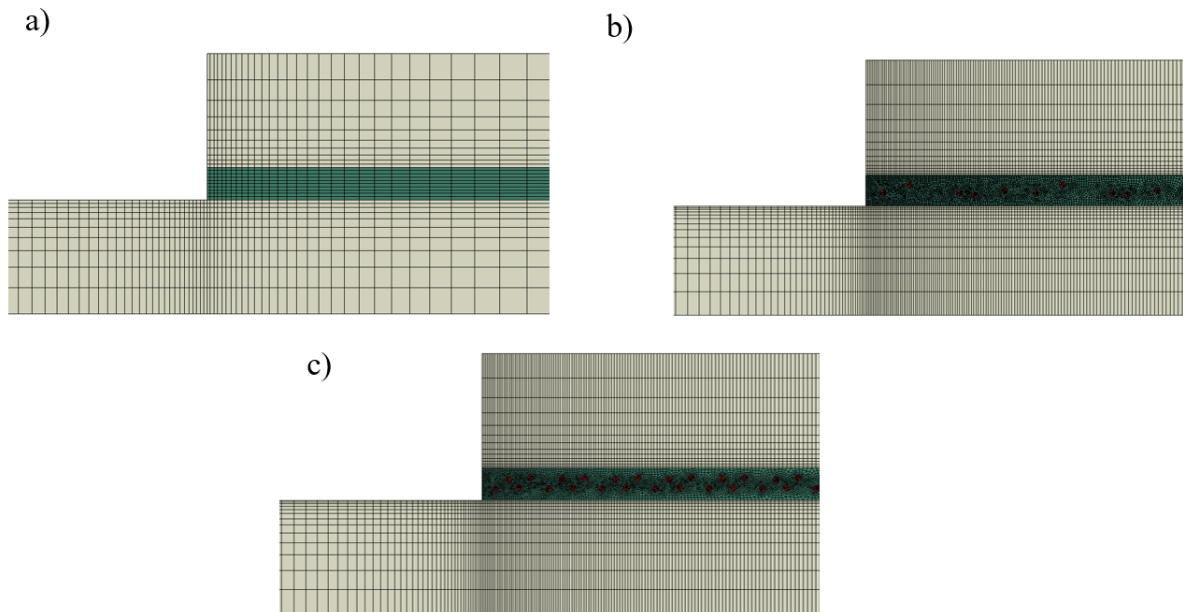


Figure 6: Representative meshes of the SLJs models with a) neat adhesive; b) addition of 5 % volume of glass beads; and c) addition of 10 % volume of glass beads.

Although the XFEM methodology implemented in Abaqus[®] allows to define a pre-crack for damage initiation, the models developed for this study did not employ this feature. This was carried out in order to allow the crack to freely propagate along the adhesive layer.

C.4 Results and discussion

C.4.1 Experimental results

The experimentally determined failure loads and displacements at failure, are represented in Figures 7 and 8.

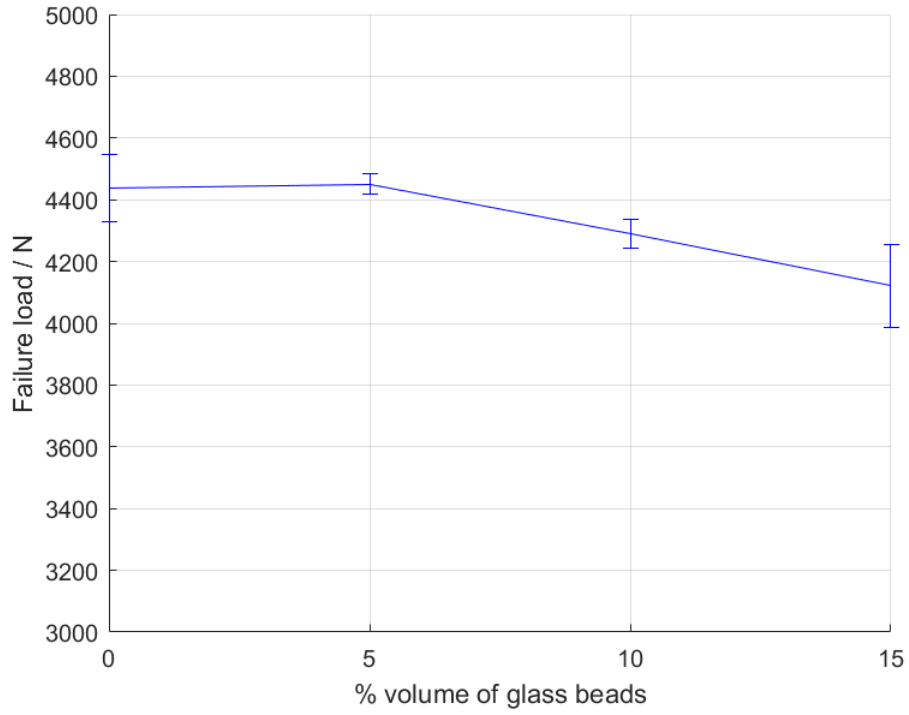


Figure 7: The failure load values, P_{max} , for the different percent volumes of hollow glass beads.

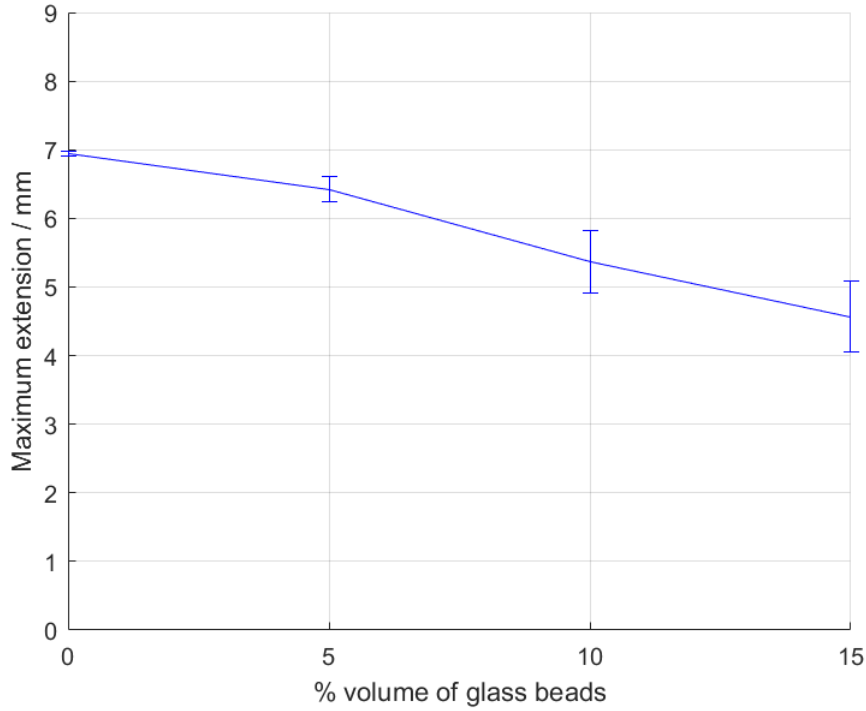


Figure 8: The maximum extension values, δ_{max} , for the different percent volumes of added hollow glass beads.

As one can conclude by looking at the experimental results, the mechanical performance of the bonded joint's tends to decrease with an increasing volume of glass particles. In fact, and even though the failure load values show a global reduction with the rising number of glass beads, the differences are not especially significant, considering that these only vary between 4500 and 4100 N.

However, an analysis of the maximum displacement values as a function of the added % volume of hollow glass beads shows a significantly decrease. This suggests that the adhesive not only becomes more brittle, but also loses some of its energy absorption's capacity.

Figure 9 illustrates the fracture surfaces of the SLJs for each hollow glass beads amount. It is possible to observe that the area of the regions without adhesive in the ends of the bonded joints considerably decreases with the increasing number of glass particles. One can infer that, by going from the neat adhesive system to the one with the addition of glass beads, the failure mode transitions from adhesive to cohesive failure. This happens since the crack initiates its propagation closer to the middle of the adhesive rather than in the interface, going from the neat adhesive configuration to the ones with glass beads added to it, which happens as a consequence of the presence of glass particles

in the adhesive layer. These, being the weaker links in the adhesive's matrix, create local stress fields around them that force the crack to travel closer to the middle of the adhesive layer.

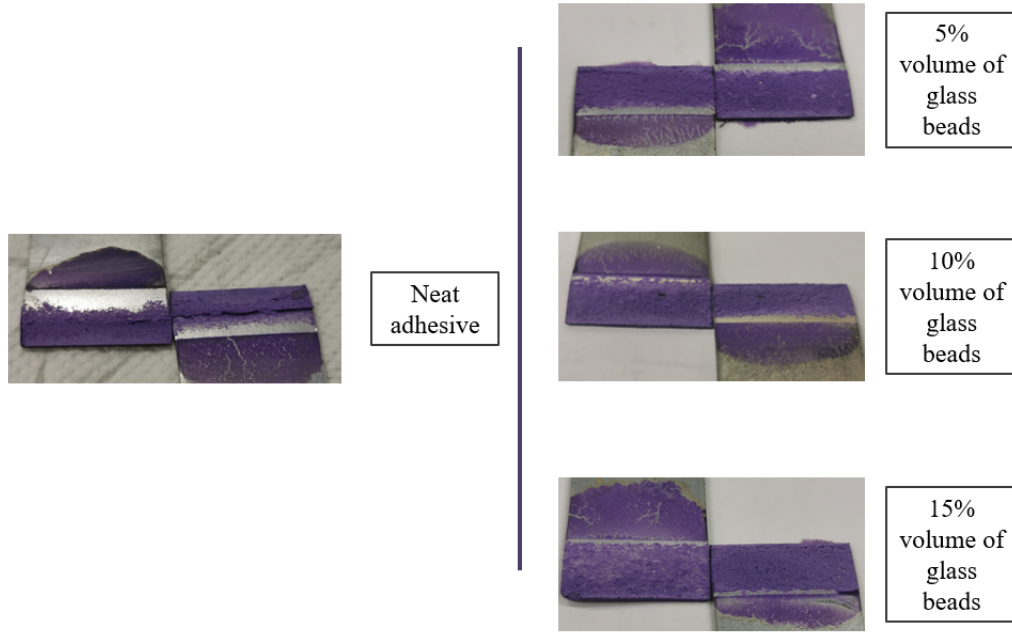


Figure 9: SLJs fracture surfaces as a function of the added hollow glass beads' amounts that were employed.

C.4.2 Numerical results

As stated previously, the single lap joint tests were simulated for different amounts of hollow glass microspheres, in volume.

The $P - \delta$ curves obtained for the SLJs with mild steel were the ones that follow in Figure 10, for Adhesive A at quasi-static conditions.

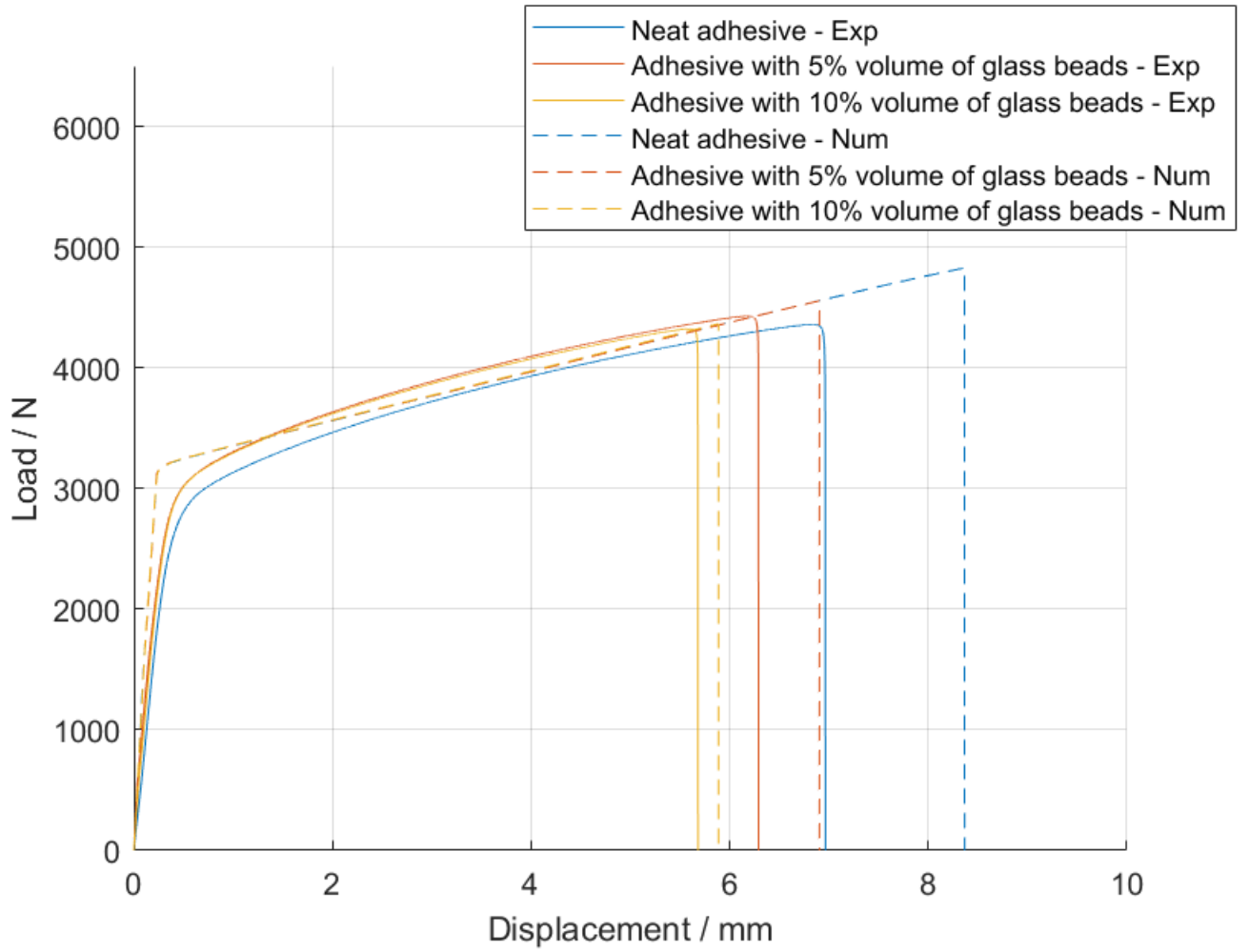


Figure 10: Comparison between the experimental and numerical P - δ curves of SLJs with different adhesive configurations: neat, 5 and 10% volume of added glass beads.

As one can observe in Figure 10, very significant plastic deformation of the substrates is taking place, since the P - δ curves show a transition from an elastic to a plastic behavior after reaching a load value around 3000 N, since it is the mild steel's plastic yielding that is ruling the SLJ's behavior. Comparing the experimental and the numerical results, it becomes possible to conclude that the shift from the elastic to the plastic behavior is more abrupt in the numerical model. This can be explained by the fact that, while developing the numerical models, discrete points are extracted from the stress - strain curve obtained through the mild steel's tensile testing, in order not to minimize the computational times associated to the numerical study.

Figure 11 compares the representative images of the plastic deformation of the substrates, for the experimental and the numerical results. The comparison between the two images allows to confirm that the deformed shape obtained through the numerical

simulation closely resembles the phenomenon that was detected experimentally.

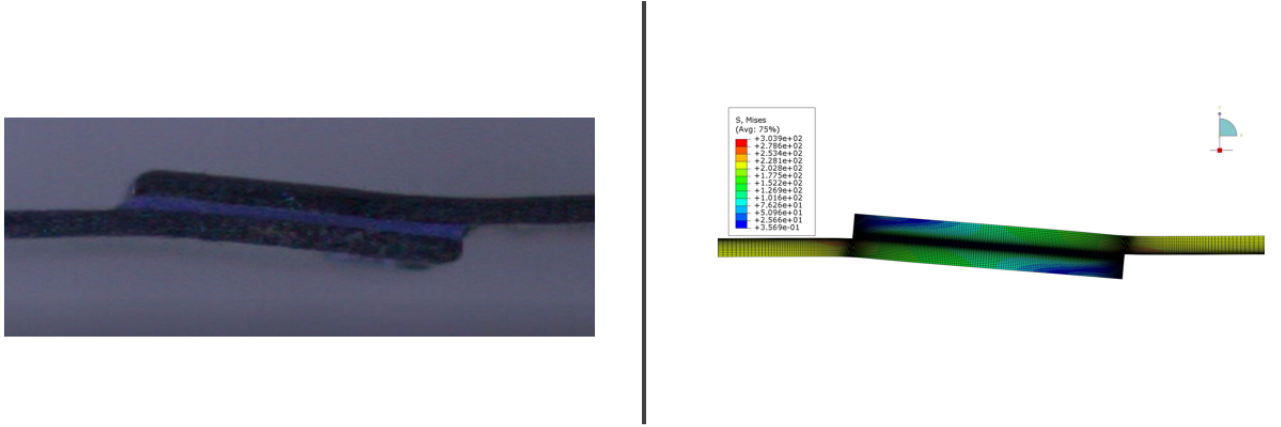


Figure 11: Representative images of the plastic deformations to which the mild steel substrates are subjected, for the experimental and numerical results, respectively.

Regarding the failure loads and maximum displacement, the numerical studies predicted slightly higher values, as one can confirm in Figures 12 and 13, respectively. However, and taking into account the fact that numerical simulations assume the existence of perfect conditions throughout the test, these differences should be considered as expectable. Moreover, and by globally analysing the evolution of the adhesive's mechanical performance, the performance degradation caused by the presence of the glass beads on the adhesive's layer is captured by the numerical models, since not only the maximum displacement but also the area below the P - δ curves gradually decreases with the increasing number of particles.

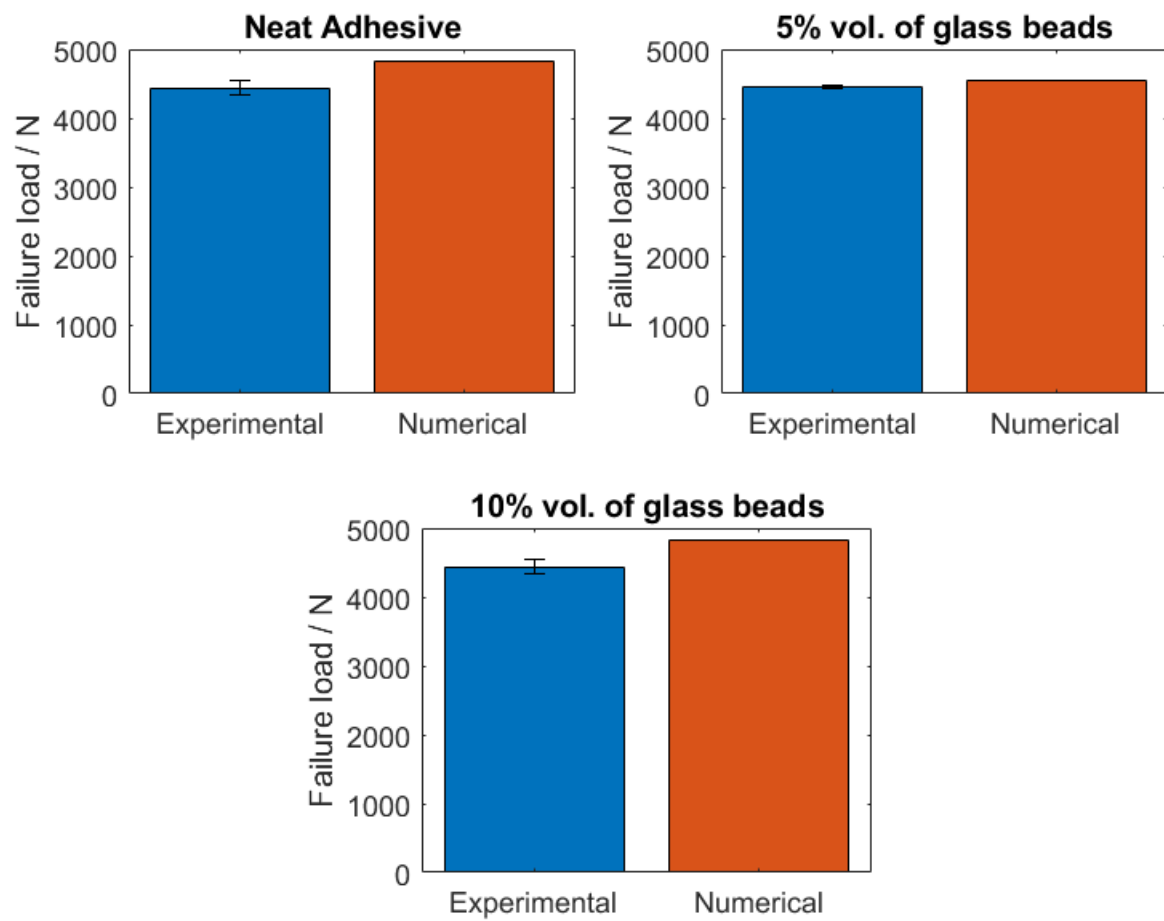


Figure 12: Comparison between the experimental and numerical failure load values of SLJs with different adhesive configurations: neat, 5 and 10% volume of added glass beads.

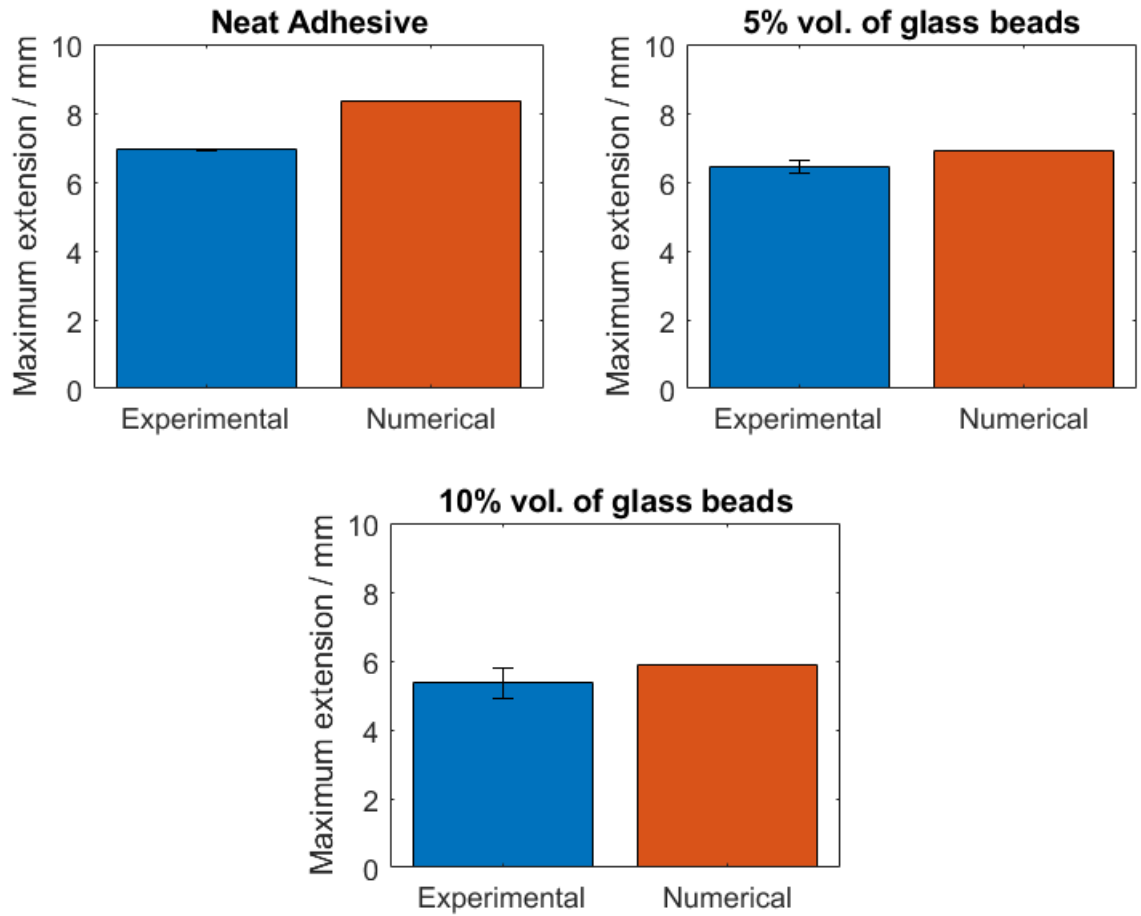


Figure 13: Comparison between the experimental and numerical maximum extension values of SLJs with different adhesive configurations: neat, 5 and 10% volume of added glass beads.

Figure 14 illustrates the crack's propagation path for the SLJs with neat adhesive, and after the addition of 5 and 10 % volume of glass beads. For this matter, the stiffness degradation parameter (SDEG) values are displayed, which represent the level of damage in elements. A value of zero correspond to an undamaged element and an element is considered to be fully damaged whenever this parameter reaches a value of one. The numerical results show that, by introducing glass particles into the adhesive layer, the failure mode evolves from adhesive to a cohesive failure. This is in accordance to what was reported experimentally, where the influence of the glass microspheres clearly changed the crack's propagation path.

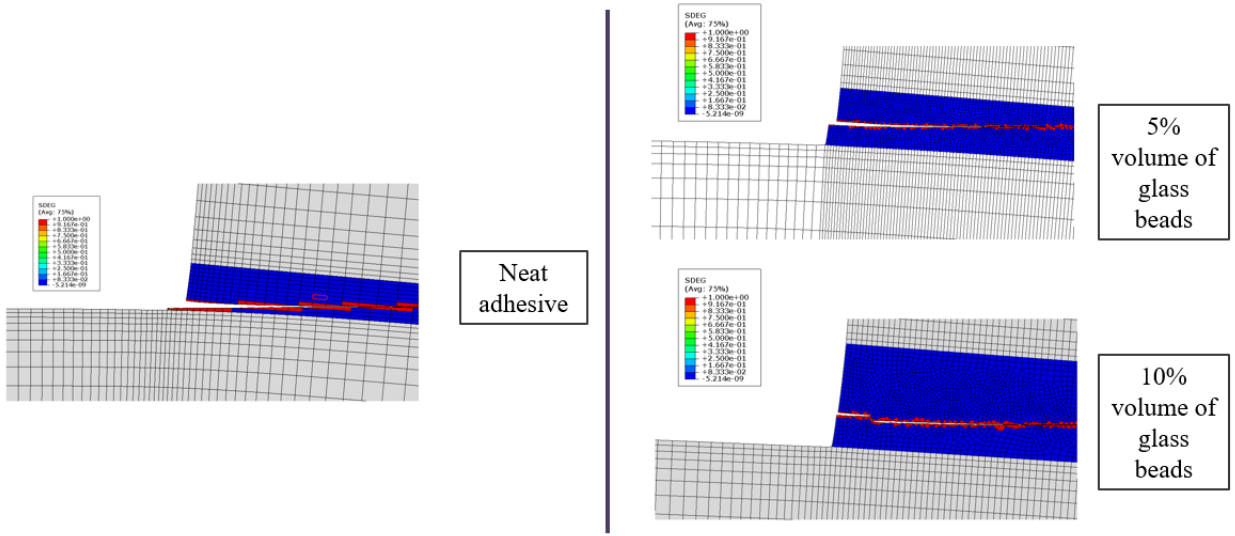


Figure 14: SLJs failure modes with neat adhesive, and with the addition of 5 and 10 % volume of glass beads to it.

Moreover, Figure 15 presents a closer look at the crack's propagation path in SLJ with the addition of 10% volume of glass beads. In this figure, the location of the glass particles is highlighted and the crack is represented, enabling to see that the glass beads presence clearly influences the crack's path. As one can observe, the crack's path follows the regions where the glass beads are present, due to the fact that these create stress concentrations along its periphery. Considering that these represent weaker points in the adhesive matrix, their damage level will be more significant than the one of the adhesive's matrix.

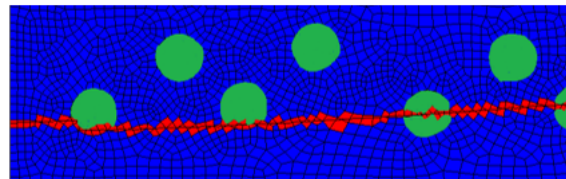


Figure 15: SLJ failure mode with the addition of 10 % volume of glass beads to the adhesive.

Specifically focusing on the initial crack's path, one can observe that, not only experimentally but also numerically, the crack undoubtedly propagates towards the interfacial region. This occurs since the most critical stress regions are located in the ends of the bonded joint. Through the numerical results, this can be confirmed in Figure 16, which

represents a closer look at the critical stress fields in the SLJ with neat adhesive.

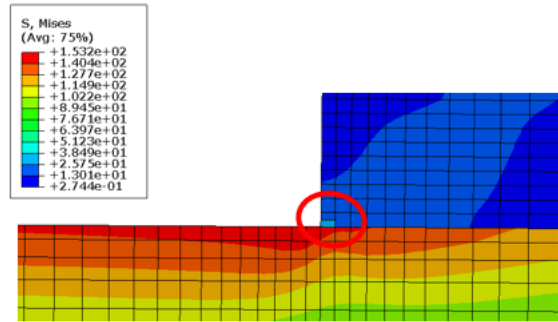


Figure 16: Representative image of the existing local stress fields on the SLJ with neat adhesive, obtained numerically.

However, with regards to the SLJs with added glass particles content, an initial short interfacial crack can be observed experimentally. Numerically, this phenomenon does not occur, since presence of particles within the adhesive layer accumulates more critical stress fields in their periphery than those near the ends of the bonded joints, as one can confirm in Figure 17.

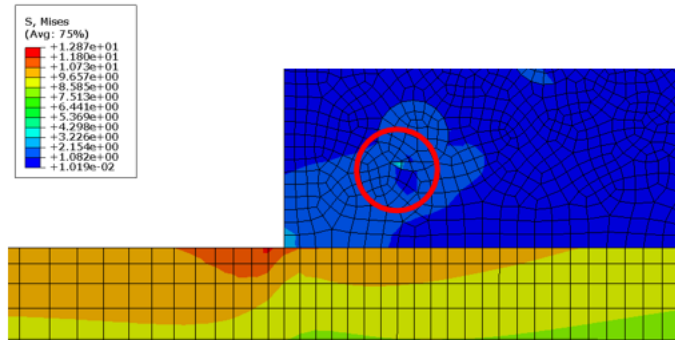


Figure 17: Representative image of the existing local stress fields on the glass particles' periphery, obtained numerically.

Even though a sharp corner geometry, as the one associated to the ends of the bonded joint, is usually more critical than rounded geometries, the glass microspheres distributed along the adhesive layer, generate strength concentrations which greatly lower the load carrying capability of the material and cause it to become brittle. Therefore, the singularities introduced by the presence of glass beads can be considered to be more significant,

in terms of local stress concentrations, than the ones created by the sharp corners of the ends of the bonded joints, as the information presented in Figure 17 confirms.

Nevertheless, the experimental results showed that the crack starts propagating in an interfacial region, due to the fact that experimentally tested SLJs contain additional singularities in the ends of the bonded joints, instead of perfectly shaped sharp corners. This, allied to the existing peel stresses originated from the plastic yielding of the substrates, accumulates significant stress fields in the ends of the bonded joint which force the crack to initially propagate interfacially for a short period. Since the factors described above are not accounted for in the development of the numerical models, this explains the slight differences between the experimental and the numerical results, with regards to the initial crack's propagation path.

C.5 Conclusions

In this work, single lap joints with mild steel substrates and with different adhesive configurations, using different amounts of glass beads added to the adhesive, were studied and compared with regards to the joint's strength and failure mode. To that end, experimental tests were carried out and numerical models were created in order to analyse the evolution of the failure pattern, as well as the impact on the joint's strength.

The main conclusions extracted from this study are the following:

- The introduction of hollow glass beads on the adhesive layer has a negative influence on its mechanical performance since it weakens the adhesive, by decreasing the joint's maximum extension, as well as the adhesive's ability to absorb energy.
- It was observed that adding glass particles to the adhesive clearly influences the joint's failure mode. In fact, their presence creates local stress fields around their periphery, which forces the crack to propagate through those regions. Therefore, one can conclude that adding this type of particles allows the joint to have a cohesive failure rather than an adhesive one.
- XFEM was found to be an effective method for predicting the failure modes of the bonded joints, since the numerical results showed that adding glass particles to the adhesive layer results in a cohesive failure. Moreover, it provided satisfactory estimations of the SLJs failure loads, as well as its maximum extension values. Finally, it is also able to accurately reproduce the experimentally observed plastic yielding of the substrates.

References

- [1] Lucas FM da Silva, Andreas Öchsner, and Robert D Adams. *Handbook of adhesion technology*. Springer Science & Business Media, 2011.
- [2] WA Lees. Adhesive selection. In *Adhesives in Engineering Design*, pages 92–123. Springer, 1984.
- [3] Ali Nemati Giv, Majid R Ayatollahi, S Hengameh Ghaffari, and Lucas FM da Silva. Effect of reinforcements at different scales on mechanical properties of epoxy adhesives and adhesive joints: a review. *The Journal of Adhesion*, 94(13):1082–1121, 2018.
- [4] AJ Kinloch. Toughening epoxy adhesives to meet today’s challenges. *MRS bulletin*, 28(6):445–448, 2003.
- [5] R Hunter, J Möller, A Vizán, J Pérez, J Molina, and J Leyrer. Experimental study of the effect of microspheres and milled glass in the adhesive on the mechanical adhesion of single lap joints. *The Journal of Adhesion*, 93(11):879–895, 2017.
- [6] DA Bigwood and AD Crocombe. Non-linear adhesive bonded joint design analyses. *International journal of adhesion and adhesives*, 10(1):31–41, 1990.
- [7] JA Harris and RA Adams. Strength prediction of bonded single lap joints by non-linear finite element methods. *International journal of adhesion and adhesives*, 4(2):65–78, 1984.
- [8] Raul DSG Campilho, Mariana D Banea, JABP Neto, and Lucas FM da Silva. Modelling adhesive joints with cohesive zone models: effect of the cohesive law shape of the adhesive layer. *International journal of adhesion and adhesives*, 44:48–56, 2013.
- [9] Soheil Mohammadi. *Extended finite element method: for fracture analysis of structures*. John Wiley & Sons, 2008.
- [10] RDSG Campilho, MD Banea, FJP Chaves, and Lucas FM Da Silva. extended finite element method for fracture characterization of adhesive joints in pure mode i. *Computational Materials Science*, 50(4):1543–1549, 2011.
- [11] Zhen-zhong Du. extended finite element method (xfem) in abaqus. *Simulia: Johnston, RI, USA*, 2009.

- [12] Ali Jenabidehkordi. Computational methods for fracture in rock: a review and recent advances. *Frontiers of Structural and Civil Engineering*, 13(2):273–287, 2019.
- [13] N Sukumar and J-H Prévost. Modeling quasi-static crack growth with the extended finite element method part i: Computer implementation. *International journal of solids and structures*, 40(26):7513–7537, 2003.
- [14] Lucas FM da Silva and Raul DSG Campilho. Advances in numerical modelling of adhesive joints. In *Advances in numerical modeling of adhesive joints*, pages 1–93. Springer, 2012.
- [15] CI da Silva, AQ Barbosa, RJC Carbas, EAS Marques, A Akhavan-Safar, and Lucas FM da Silva. Influence of cork microparticles on the fracture type in single lap joints. *Proceedings of the Institution of Mechanical Engineers, Part C: Journal of Mechanical Engineering Science*, page 0954406220936729, 2020.
- [16] 3M Advanced Materials Division. 3m glass bubbles, 2018.
- [17] VV Budov. Hollow glass microspheres. use, properties, and technology. *Glass and ceramics*, 51(7-8):230–235, 1994.