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Mechanical properties of a reversible adhesive used to separate adhesive joints

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Abstract

In this work, the mechanical characterization of adhesive joints made with a thermoplastic adhesive, modified and not modified, has been presented together with the separation tests that are possible by the use of metal nanoparticles embedded within the adhesive. A polyolefin adhesive has been modified with two weight concentrations (5% and 10%) of iron oxide nanoparticles. These particles increase their temperature under electromagnetic field; in this way, they are able to melt the adhesive and, therefore, separate the adhesive joints. The mechanical properties of single lap joints (SLJs) prepared with the neat and modified adhesives have been performed by using different overlap lengths and thicknesses. SLJs prepared with the nanomodified adhesive present higher loads compared with the neat one. Separation tests have been carried out on SLJ specimens to measure the times needed to disassemble the adhesive joints. Scanning electron microscope analysis has been carried out to study the distribution of the particles.

KEYWORDS

adhesive joints, automotive structures, mechanical, mechanical properties, nano mechs

1 | INTRODUCTION

The use of structural and non-structural adhesive has been increasing in the last decades in many industrial sectors due to the ease of manufacturing process, improved stress distribution, and the potentiality to join different materials without introducing holes in the structures. Albeit these are important advantages, the complexity to disassemble adhesive joints to prevent industrial waste, recycle materials, and repair and reuse industrial components can reduce their adoption.¹⁻⁴ In Europe, the directives, 2000/53/EC and 2000/64/EC,^{5,6} require that the reuse and recyclability for automotive vehicles must be respectively 95% and 85% by an average weight per vehicle.

Lu et al.⁷ and Banea et al.^{8,9} reported the most common methods to disassemble adhesive joints with traditional and innovative technologies. However, even though these techniques can work, in some cases, it is very complicated to have a very clean surface in the bonding area of the adherends and they cannot be re-bonded easily. For this reason, most parts of the bonded components in automotive industries need a very complex procedure in order to be disassembled.

In this scenario, the development of disassembling technologies is crucial to reach the percentages of recyclability and reuse set by the directives. These directives encourage automotive companies to find new approaches for the reusing and recycling of automotive vehicles before the adoption of new materials. A promising technology for the separation of plastic joints, bonded with thermoplastic adhesives, uses nanomodified thermoplastic adhesives that are sensitive to electromagnetic fields.¹⁰ In the last decades, technologies that use electromagnetic induction systems have

been studied by researchers in order to find a reliable solution to this problem for structural and non-structural adhesives.⁹⁻¹⁴ Ciardiello et al.¹⁴ showed that this technology can be coupled with thermoplastic adhesive modified with iron oxide particles to increase the temperature of the particles by means of Neel and Brown relaxation effect. Ciardiello et al.¹⁵ showed that this technology is able to separate adhesive joints by using nanomagnetite particles (Fe_3O_4). However, albeit the separation of the adhesive joints can be obtained with the use of this technology, the mechanical properties need to be investigated because the introduction of particles within the adhesive can be detrimental for the mechanical properties.

The mechanical properties of a thermoplastic (polyolefin-based) adhesive have been studied by means of single lap joints (SLJs) together with the separation tests led on the same joints. The mechanical behaviour of these joints has been investigated by considering three different adhesive thicknesses and three overlaps for the joint prepared with the pristine adhesive and with the one modified with 10% wt. only, because they present a larger significant difference compared with the adhesive prepared with 5% wt. Furthermore, the effects of two weight percentages on the mechanical properties and separation time were evaluated at a fixed overlap and thickness on SLJs prepared with two different percentages, 5% and 10% wt.

2 | MATERIALS AND METHODS

The substrates used in this work are made of polypropylene copolymer produced by Lyondell-Basell Industries (Hifax CB 1160 G1). The adhesive is a polyolefin-based hot melt adhesive produced by Beardow Adams (Prodas). Both the substrate material and the adhesive are used in automotive industries to bond automotive components. The physical and mechanical properties of the adhesive and substrates have been studied in several studies.¹⁵⁻¹⁸ Mechanical tests and separation tests were carried out on SLJ specimens prepared with three different adhesive thicknesses, 0.5, 1.0, and 1.5 mm, and three overlaps, 12, 18.5, and 25.

The nanomodified adhesives were prepared by using a hand mixing method already adopted in the literature.¹⁵⁻¹⁹ Tensile tests were carried out on the substrate at 100 mm/min, which is the same speed adopted for the SLJ tests. Separation tests have been carried out by using an inductor (Heayheat by Ambrell) to measure the speed of the dismounting process by using a power of 5.9 kW and a frequency of the magnetic field of 317 kHz. The test was performed by using a weight of 0.5 N that was applied to initiate the sliding of the other substrate when the adhesive reaches its melting temperature as described in Ciardiello et al.¹⁵

3 | RESULTS AND DISCUSSION

In this section, the experimental activity is presented with the following nomenclature. HMA refers to the adhesive joints prepared with the neat adhesive. HMA_5% and HMA_10% refer to the adhesive joints prepared with the same adhesive and modified with 5% and 10% wt. of iron oxide particles, respectively.

3.1 | SLJ tests

Figure 1A presents the representative load-displacement curves of SLJ tests for the three adopted adhesive formulations, namely HMA, HMA_5%, and HMA_10% with a fixed overlap of 25 mm and adhesive thickness of 1 mm. The curves related to SLJ bonded with these three adhesive compositions present a similar trend. The first part of the curves is superimposed for the adhesive compositions while the maximum loads and the ultimate displacements are different. The adhesive joints bonded with the HMA led to the lowest values of the maximum load and of the ultimate displacements. The addition of the nanoparticles had a beneficial effect on the mechanical properties because it led to an increase of the maximum bearing load of the joints and to an increase of the ultimate displacement that means higher elongation of the adhesives. The increase of the maximum load is around 5% for HMA_5% and 7% for HMA_10% over the neat adhesive. Ciardiello et al.¹⁵ connected this behaviour to the presence of the micro agglomerates^{15,16} that lead to a toughening effect of the bondline that resulted in an increase of the maximum load and shear strength. The scanning electron microscope analysis that shows the presence of micro agglomerates prepared with the mixing technique is not presented in this work because it has been widely discussed in other studies.¹⁵⁻¹⁷ Figures 1B to 1D illustrate the

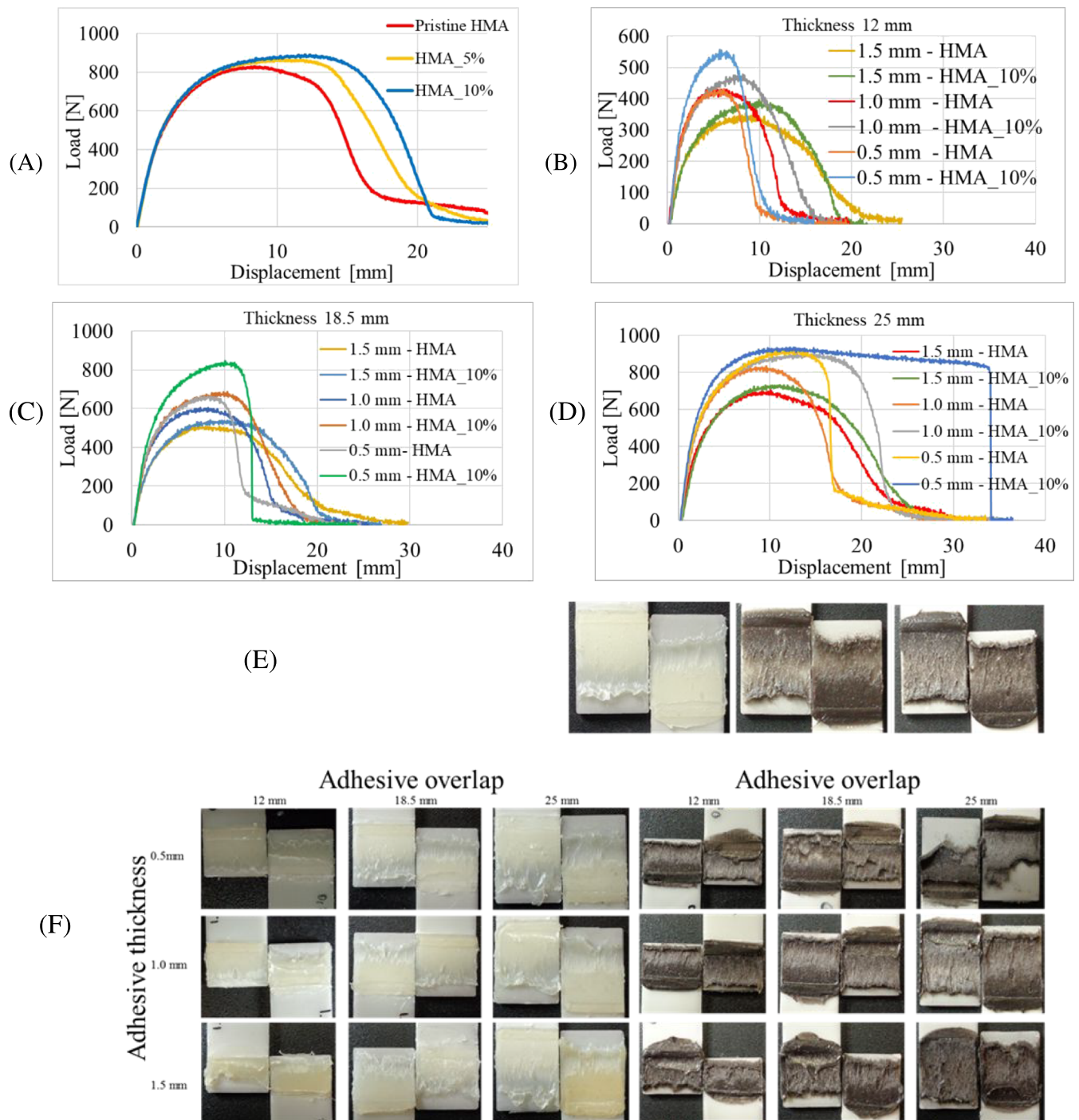


FIGURE 1 (A) Representative curves of single lap joint (SLJ) tests for the three adopted adhesive compositions. Representative curves of SLJ tests for the three adopted thicknesses and overlaps: (B) 12 mm, (C) 18.5 mm, and (D) 25 mm. (E) Representative fracture surfaces of three different adhesive compositions and (F) representative fracture surfaces for all the adopted joint configurations

representative load-displacement curves of the SLJ tests prepared with neat HMA and HMA_10% for the three adhesive thicknesses that are 0.5, 1.0, and 1.5 mm and for the three different overlaps, 12 (Figure 1B), 18.5 (Figure 1C), and 25 mm (Figure 1D). These three figures show that the curves are very similar for all the joint configurations. In all the cases, the values of the maximum loads related to the joints prepared with HMA_10% are higher than the ones prepared with HMA for SLJ prepared with the same configuration. Generally, the values of the maximum bearing load increase for the larger overlaps and for the smaller thicknesses, as expected and as shown also in Koricho et al.¹⁹ Furthermore, these curves show that the increase of the adhesive thickness leads to lower loads and higher displacements.

It is noticeable that all the fracture surfaces obtained for the curve presented in Figures 1B to 1D were mostly cohesive, except for the SLJ prepared with a thickness of 0.5 mm and an overlap of 25 mm. In this case, the lower thickness and the high overlap led to a break of the substrate. For this reason, the curve presents the largest displacement that is due to the deformation of the substrate.

Figure 1E illustrates the fracture surfaces of the SLJ specimens after the test. This figure displays representative fracture surfaces of the joints prepared with HMA, HMA_5%, and HMA_10% at a thickness of 1 mm and an overlap of 25 mm. The cohesive zones are recognizable by the colours that are slightly clearer when compared with the zones where the separation was adhesive. However, this figure shows that the addition of nanoparticles increases the cohesive fracture zone and it is worth to note that the size of cohesive fracture areas increases with the particle weight concentration.

Figure 1E illustrates the representative fracture surfaces obtained by the SLJ tests for the six configurations of both HMA and HMA_10%. This figure shows that the cohesive areas for the joints prepared with HMA_10% are higher than the ones prepared with the neat HMA. As expected, the lower is the overlap length, the higher is the cohesive area that the fracture surfaces present. Figure 1F depicts that the larger cohesive zone is obtained for the joints prepared with the overlaps of 12 and 18.5 mm and the thicknesses of 0.5 and 1.0 mm. The SLJ specimen prepared with HMA_10% and with a thickness and overlap of 0.5 and 25 mm, respectively, presented a deformation of the substrate, as already written, that led to an adhesive failure as can be detected by Figure 1E.

Figure 2A reports the maximum mean loads for all the configuration of the adhesive joints prepared with HMA and HMA_10%. As anticipated, the maximum bearing load increases for larger overlaps and it decreases for lower adhesive thicknesses. This figure illustrates that the maximum loads experienced with the SLJ prepared with HMA_10% are larger than the ones prepared with HMA for all the analysed cases. Figure 2B shows the percentage increase of the joints prepared with HMA_10% over the ones prepared with HMA. The values of the standard deviations are reported in the error bars of Figure 2A, and it is possible to note that the scatter is very limited. This figure shows that the increase of the maximum load of HMA_10% over HMA is more evident for the adhesive joints prepared with a lower overlap and thickness, 20%. This is related again to the presence of the small agglomerates, which led to a more evident toughening effect for the joints prepared with smaller thicknesses.^{20,21}

3.2 | Separation tests

The separation tests were carried out by using all the parameters that have been found by Ciardiello et al.¹⁵ to minimize the separation time, which are the highest power found in that work (5.9 kW), the highest frequency (317 kHz), and a solenoidal coil. The SLJs prepared with nanomodified adhesive were placed in the centre of the solenoidal coil where the electromagnetic field is maximum. Table 1 shows that it is possible to separate both HMA_5% and HMA_10% within 54 and 12.5 seconds, respectively. Of course, the higher presence of particles led to a lower separation time because the presence of the particles is the heating source of this technology.

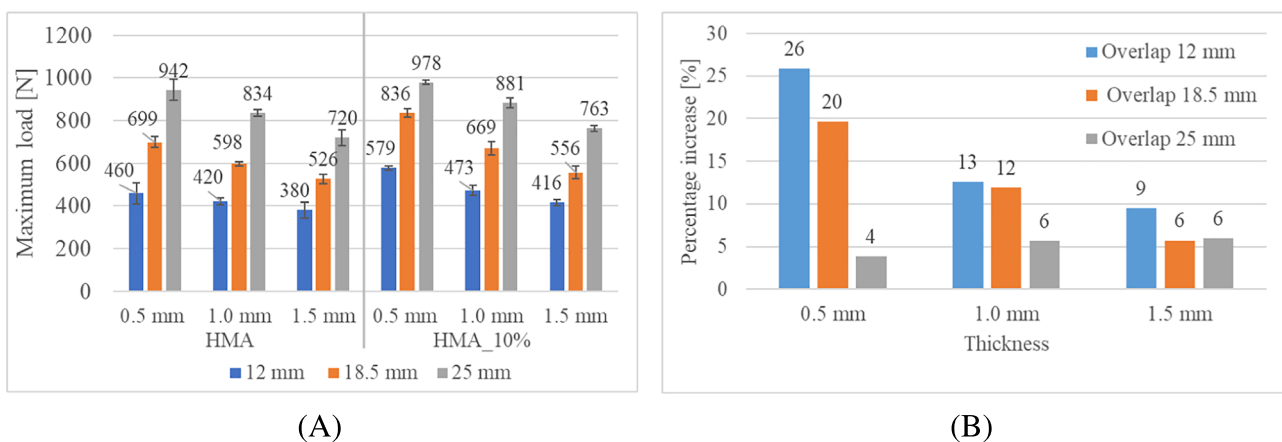


FIGURE 2 (A) Effect of the adhesive thicknesses and overlaps on the maximum load; (B) percentage increase of HMA_10% over HMA

TABLE 1 Separation time for HMA_5% and HMA_10%

| | HMA_5% | HMA_10% |
|---------------------|------------|------------|
| Separation time [t] | 53.8 (3.5) | 12.5 (1.2) |

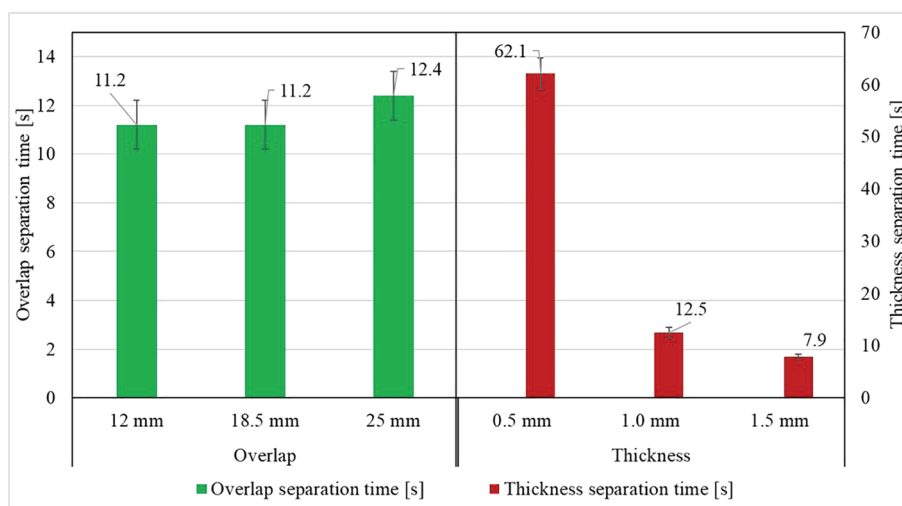
FIGURE 3 Separation time for different overlaps and thicknesses

Figure 3 reports the separation times that were obtained with the three different adhesive overlaps and thicknesses considered in this work. On the left part of the graph, the thickness was kept constant at 1 mm and the overlap was changed, whereas on the right part, the overlap was kept constant at 25 mm and the thickness was varied to evaluate both the effects. The standard deviations of the obtained values are reported by the error bars in the figure. Figure 3 shows that the separation time seems to be influenced by the adhesive thickness. In fact, the separation time of the joints prepared with a thickness of 0.5 mm is 400% higher than the case with a 1.0-mm thickness. Ciardiello et al.¹⁵ explained that this larger separation time is due to the higher interfacial strength of the adhesive joints prepared with a thickness of 0.5 mm compared with the ones of 1.0 and 1.5 mm, as shown in the mechanical properties section. This can be understood by the analysis conducted with a thermal camera and reported in their study.¹⁵ They have shown that the temperature of the substrate of the joints prepared with a thickness of 0.5 mm before the separation was very high compared with 1.0 and 1.5 mm. This behaviour is due to the higher interfacial strength of the joint prepared with a thickness of 0.5 mm. In fact, in this case, the weight that has been used for initiating the joint separation is too small compared to the improved interfacial strength.

4 | CONCLUSION

This paper presents a comprehensive study of the mechanical properties of reversible plastic joints prepared with two different weight concentrations of iron oxide particles. The work shows that the adhesive joints prepared with the modified adhesive led to higher maximum bearing loads and higher displacements. Furthermore, the separation of the adhesive joints was possible and shown for both the adhesives considered, HMA_5% and 10%. Within 12.5 seconds, it is shown that it is possible to fully melt and separate adhesive joints free from damage, which is of key importance to the automotive sector.

CONFLICT OF INTEREST

I approve the submission and declare that there is no conflict of interest regarding the publication of this paper.

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