Thermomechanical characterization of reinforced and dismountable thermoplastic adhesive joints activated by microwave and induction processes

Thermomechanical characterization of reinforced and dismountable thermoplastic adhesive joints activated by microwave and induction processes

R. Ciardiello[1*], G. Belingardi[1], F. Litterio[1], V. Brunella[2]

[1]Department of Mechanical and Aerospace Engineering, Politecnico di Torino, 10129 Torino, Italy

Email: raffaele.ciardiello@polito.it, giovanni.belingardi@polito.it, fabrizia.litterio@gmail.com.

[2]Department of Chemistry and NIS Research Centre, University of Torino, 10122 Torino, Italy

Email: valentina.brunella@unito.it.

***Corresponding Author**:

R. Ciardiello

E-mail address: raffaele.ciardiello@polito.it

Full postal address:

C.so Duca degli Abruzzi 24,

Department of Mechanical and Aerospace Engineering – Politecnico di Torino,

10129 – Turin,

ITALY

Phone number: +39.011.090.6913

Fax number: +39.011.090.6999

Thermomechanical characterization of reinforced and dismountable thermoplastic adhesive joints activated by microwave and induction processes

R. Ciardiello[1], G. Belingardi[1], F. Litterio[1], V. Brunella[2]

[1]Politecnico di Torino, Department of Mechanical and Aerospace Engineering, Corso Duca degli Abruzzi 24, 10129 Torino, Italy
Email: raffaele.ciardiello@polito.it, giovanni.belingardi@polito.it, fabrizia.litterio@gmail.com.
[2]Department of Chemistry and NIS Research Centre, University of Torino, 10122 Torino, Italy
Email: valentina.brunella@unito.it.

Keywords: Reversible adhesive joint; Thermoplastic adhesive; Automotive applications; Reinforced adhesive; Smart adhesive.

Abstract

In this work, a thermoplastic adhesive used by automotive industries for bonding plastic components has been modified with iron oxide nanoparticles to ease the dismounting process. Two different processes have been considered for the joint dismounting, namely the already studied electromagnetic induction and an innovative microwave heating process. Besides the mentioned heating activation process, a full experimental plan has been designed that includes two different weight percentages (5% and 10%) of the nanoparticles and two alternative mixing methods: hand-mixing and extrusion methods. The mechanical performances and the separation tests have been assessed by using single lap joints. The adhesive joints prepared with the nanomodified adhesives and with the hand-mixing method presents a slightly larger value of the maximum strength due to the presence of small agglomerates that have been observed with scanning electron microscope analysis. On the other hand, the extrusion method gave a very uniform distribution of the particles within the adhesive matrix. Microwave and induction heating tests conducted on both the single lap joint and the adhesive itself showed that the joint separation and the melting of the adhesive are possible by using both adhesives prepared with the two mixing methods.

1. Introduction

In recent years, the automotive industry is facing different challenges related to the weight reduction of the vehicles. On one hand, there is the strengthening of the environmental and safety regulations that suggest decreasing the weight of the vehicles by using lighter and more efficient materials. On the other hand, there are increasing customer demands for higher performances and more luxury and safety features with a consequent weight addition. In this contest, adhesive bonding acquires great importance since it represents a lighter and cheaper solution [1-4] in some cases, with respect to traditional fasteners. Adhesives permit to join components made of materials that are difficult or even impossible to join in other ways and they are able to join substrates made of different materials, such as composite materials with metals [5, 6].

Although they offer some advantages, they cannot be easily separated. Lu et al. [7] reported some of the most traditional methods to separate adhesives joints such as: using chemical solvents, mechanical cutting or heat treatment. The use of heat and chemicals could damage the components (or substrates) because they can be aggressive not only for the adhesive but for the components as well. In this specific case, these techniques cannot be used for reuse but only for recycling.

Mechanical cutting is also complicated and it cannot be applied in many cases because, usually, the bondline of automotive components is included in the inner part of the components that have to be bonded, and therefore not easily accessible. However, even though these techniques can work, in most of the cases, it is very complicated to have a very clean surface in the bonding area of the adherends so that they cannot be re-bonded easily. For these reasons, most of the bonded components in automotive industries need a very complex procedure in order to be dismantled. Banea et al. [8] reported also many complex methods, laborious methods and new technologies that could separate mechanical components but, even in these cases, the resulting surfaces of the adherends are not clean.

The possibility to dismantle components in the automotive industry is very important both for repair and for recycling purposes. In Europe, particularly, the Directive 2000/53/EC [9], even called end-of-life vehicles (ELV) Directive and the Directive 2005/64/EC [10] have set targets aiming the increase of the reusability, recoverability, and recyclability of vehicle materials and components. Because of these directives, automotive industries must produce a detailed report for each model that shows the existing techniques that they can use to dismantle components in order to recycle, reuse or recover those components. Automotive industries are required to achieve the recyclability of materials and components to a minimum of 85% by an average weight per vehicle and the reuse and recovery of components to a minimum of 95%. In this scenario, the development of disassembling technologies is crucial to reach the percentages of recyclability and reuse set by the directive. These Directives encourage automotive companies to find new approaches for the reuse and recycling of automotive vehicles before the adoption of new materials.

In the last decades, innovative technologies have been introduced and studied in automotive industries and research centres to find a feasible solution to these problems. Some studies [11-14] have presented a technology that uses electromagnetic induction systems that activate magneto-sensitive nanoparticles embedded in adhesives. The sensitivity of these particles to the electromagnetic field has been used by [12, 14, 15] for rapidly increase the temperature of thermoplastic adhesive allowing for the separation of joints with greater easiness and without damages. Banea et al. [13] have used the same technology to heat metallic substrate in order to increase, by conduction, the temperature of thermally expandable particles. These particles are able to reduce the resistance section of the adhesive allowing for a separation of the joints. Severijns et al. [16] have used the same technology for curing epoxy adhesives.

In the electromagnetic induction process, an inductor is used to increase the temperature of a workpiece, usually a metallic component. Inductor works as a primary of an electric transformer and the conductive material as a secondary one. The electromagnetic field is generated by a coil that is the final element of the inductor and the shape of the electromagnetic field is given by the shape of the coil. The temperature increase of the particles, in the case of iron oxide nanoparticles, is mainly due to the hysteresis losses and the Neel and Brown relaxation phenomena [17-19]. It is strictly linked to the dimension of the nanoparticles, in fact, particles with a size smaller than 50 nm exhibit superparamagnetic behaviour that leads to a more rapid increase of the temperature, as Ghazanfari et al. [20] have reported.

As shown by [21], this is a promising technology to separate adhesive joints when nanomagnetite particles (Fe_3O_4) are embedded with hot-melt adhesives and coupled with electromagnetic induction systems. The addition of particles in adhesive and polymers in research coupled heating systems have been used for different purposes, such as for fast curing of adhesives and epoxy resins or for increasing the mechanical properties of the polymers [21-24]. A promising technology that can be used for the separation of the adhesive joints by taking advantage of the addition of particles is the use of microwave as heating process. The main sources of microwave heating are two effects: dipolar polarization and conduction. The advantages over induction heating process are a controllable distribution of the electric field, rapid heating and high penetrating radiation and a lower amount of applied external power compared to electromagnetic induction systems [25]. The principle developed in the work presented here is to take advantage of the electrical conductivity properties of iron oxide particles, dispersed into the adhesive matrix, and to use a microwave process to melt the HMA in order to obtain joint separation. According to [26, 27], the presence of these particles also has a beneficial effect on the fracture toughness and mechanical properties of the thermoplastic material.

In this work, microwave technology has been used for the separation of the adhesive joints by taking advantage of the sensitivity of Fe_3O_4 particles to this technology. To the author's best knowledge this technology coupled with the use of these particles has never been used for the separation of adhesive joints. The factors that have been considered for the experimental plane related to the mechanical tests are the mixing methods, hand mixing and extruded, and the particle concentrations, 5% or 10%. Furthermore, separation tests have been carried out by taking into account three different factors of the experimental plan: mixing methods (extruded and hand-mixing), the percentage of particles (5% and 10%) and the separation process (microwave and induction heating). The mentioned factors have been considered for the statistical analysis, namely analysis of variance (ANOVA), in order to assess the factors that significantly influence the responses, maximum mechanical load and separation time.

Although the separation times are short and can meet the industry requirements the mechanical properties need to be investigated because the introduction of particles in an adhesive matrix can eventually lead to a reduction of the mechanical properties [13]. In this work, the mechanical behaviour of a polyolefin thermoplastic adhesive has been studied through the mechanical characterisation of single lap joints. Furthermore, the effect of two weight percentages of nanoparticles (5% and 10% wt.) on the single lap joint performance was evaluated at a fixed overlap length and adhesive layer thickness, while two different mixing methods were adopted: hand mixing method and extrusion method. The first one gave good results in some preliminary activities related to the induction heating system. Here, the performance in the separation process was studied also with respect to the mixing methods. The separation tests carried out with a "traditional" electromagnetic induction system and with the innovative microwave process show that both the technologies are able to separate the adhesive joints. Visual inspection of the separated SLJ specimens both separated by mechanical separation tests display that the separations are completely cohesive for both mechanical properties and separation tests. Field-emission scanning electron microscope (FE-SEM) analysis shows that the extrusion method led to a more uniform dispersion of the particles compared to the hand mixing method.

2. Materials and methods

The adhesive joints used for the experimental tests were obtained by bonding adherents made of a polypropylene copolymer with 10% by weight of talc, (Hostacom CR 1171 G1A G14008, by Lyondell-Basell Industries, Houston, United States). Rectangular adherends, 100 mm long with cross-section 20x3 mm, were used as substrates for the experimental tests.

The substrates were bonded with Prodas, a polyolefin-based HMA by Beardow Adams (Milton Keynes, United Kingdom), a copolymer of polypropylene and polyethylene. A detailed description of the chemical and mechanical properties of this adhesive has been carried out by Koricho et al. [28]. The nanomodified adhesive was prepared by adding two weight concentrations of iron oxide particles 5% and 10%, and by using two different methods: hand-mixing and the extruder method.

The hand-mixed compound was prepared using a procedure commonly adopted in the literature for HMAs [12, 28, 29]. Pellets of HMA were melted together at 190 °C on a hot plate. At 190 °C, the viscosity of this adhesive is low enough to easily mix the particles into the adhesive by means of a glass rod. The iron oxide particles were added gradually and mixed together with the adhesive. Then the modified adhesive was cooled before the use by means of a hot-melt gun.

The extrusion method consists of two different phases:

1. Preparation of a masterbatch of HMA and iron oxide particles with the same technique described for the hand-mixing method.
2. Extrusion of the masterbatch by means of a Haake MiniLab Extruder (Thermo Scientific) at 100 °C and mixed for 10 minutes (time needed to reach a constant value of the mixing torque) at a speed of 120 rpm. The choice of the speed and temperature was aimed to maximize the torque that is acting on the modified adhesive since this is positive for the particle dispersion.

Mechanical tests and separation tests conducted with electromagnetic induction heating were carried out on the Single Lap Joints (SLJ). The joint preparation was performed by using a hot-melt gun and an assembly device, which can control the adhesive layer thickness and the overlap length of the joint, as done in [30]. Single lap joints were prepared by using a fixed overlap length of 15 mm, with a substrate width of 20 mm, as shown in [31]. This configuration was chosen because it led to the relatively high value of the shear strength value and to a totally cohesive failure surface. All the substrates were cleaned with isopropyl alcohol before the joint preparation.

The SLJ tests were conducted at a constant displacement rate of 100 mm/min, as done in [12, 28, 29, 31], using an Instron 8801 servo-hydraulic machine. At least, five bonded joints were tested for both the pristine and modified adhesives, for the statistical validity of the results.

The sensitivity of the nanomodified adhesives to the microwaves was analysed using a CEM Discover Microwave. The magnetron frequency of this microwave is 2450 MHz with a maximum power output of 300 W. This microwave equipment is very useful for the experimental analysis since it has the possibility to work with an open chamber that allows the direct control of the temperature of the specimen by means of an infrared camera (IRtech radiamatic Timage). A preliminary analysis showed that the intensity of the microwaves was not constant in the chamber of the CEM. For this reason, square samples of adhesive modified with iron oxide particles were tested at different positions (heights) in the chamber until the point that led to the most rapid increase of temperature was found at 63 mm from the bottom of the chamber. This height was used for the whole experimental activity carried out in this work. The power of the microwave was set to its maximum (300 W) for all the measurements related to the microwave in order to minimise the heating time.

Separation tests by induction heating were carried out by using the inductor Heasyheat by Ambrell, with a maximum power of 10 kW and a frequency range from 10 to 400 kHz. A more extensive study on the influence of the current and frequency of this system with respect to the modified adhesives used in this work is reported in [31]. In this study, a power of 7.4 kW and a frequency of 270 kHz were used. For each test, a weight of 0.5 N was applied to the lower substrate of the SLJ in order to submit the joint to a constant load and cause joint separation (by part sliding) when the adhesive reaches its melting temperature. This separation system has been used in different works [12, 28, 31].

Unfortunately, the same tests cannot be performed in the same way for the microwave tests, due to the limited size of the chamber and thus a different procedure was found. To this end, preliminary tests at the microwave have been carried out on the modified adhesives alone in order to study the temperature-time curve. Afterward, the separation tests were carried out by cutting the substrates of the SLJ in its transversal direction in order to lean the SLJ on the Teflon stage. The specimen cut was necessary due to the limited size of the microwave chamber. As shown by [28], this adhesive starts to melt at 124 °C and it is completely melted at 155 °C. For this reason, the separation was performed by using the time needed to reach 160 °C.

The temperature was measured by an IR camera. This camera has a high thermal sensitivity (80 mK) and an image acquisition rate of 80 Hz. The Preliminary tests at the microwave instrument were conducted on two different adhesive weights: 0.20±0.05 g and 0.40±0.05 g corresponding to SLJ specimens with 0.5 and 1 mm of thickness, respectively, and with a fixed width and overlap, 20 and 15 mm respectively. The samples were placed in the centre of the chamber on a Teflon stage. The IR camera allows measuring the temperature of the external surfaces of the adhesive or of the adherend. In particular, the temperature of the upper part of the adhesive, in the case of the microwave, while the temperature of the bondline edge in the case of the induction system. The IR camera was calibrated by using a thermocouple and black paint as described in [31]. Three tests were carried out for each sample.

The temperature-time curves obtained by exposing the adhesive alone to microwave were used to establish the time needed to melt the adhesive with 1 mm thickness (160 °C). Subsequently, the SLJs prepared with 10% of iron oxide particles and with the extruded and hand mixing method were exposed to microwaves for the same time. Thus, 10%_HM was exposed for 50 s and 10%_E were exposed for 40 s to microwave. The shape of the specimens and the microwave chamber does not allow for the control of the temperature. For this reason, only the separation surfaces of these joints are reported in Section 3.4.

FE-SEM analysis was carried out with a Zeiss SUPRA40. An accelerating voltage of 10 kV was used together with secondary emission signal. The specimens were properly coated with a gold layer of 10 nm to avoid any charging effect and to have clear images. The energy dispersive X-ray spectrometry (EDS) associated with SUPRA40 was used to establish that the clearer elements visible in SEM images are Fe_3O_4 particles.

3. Results and discussion

In the following sections, HMA refers to the pristine hot-melt adhesive. 5%_HM and 5%_E refer to the adhesive modified 5% wt. of iron oxide particles and prepared with the hand-mixed method and extruded method, respectively. 10%_HM and 10%_E refer to the adhesive modified with 10% wt. of iron oxide particles and with the hand-mixed method and extruded method, respectively.

3.1 SEM Analysis

In this section, pictures from the SEM analysis of the modified adhesives are shown. The iron oxide particles are recognisable by the clearer and rounded spots within the dark matrix. Figure 1 shows two different magnifications of the adhesives modified with 5% wt. for the hand-mixed composition HM (Figure 1a and 1b) and the extruded one E (Figure 1c and 1d). In particular, the first line of the figure shows respectively a lower (left hand) and a higher (right hand) magnification of the hand-mixed adhesives HM, while the second line the same magnifications for the extruded ones E.

SEM images of the compounds prepared with the hand-mixed method HM present distinguishable agglomerates that are visible in both lower and larger magnifications. Blue arrows were used to indicate some of the agglomerates in the Figure 1a, while the Figure 1b shows with a higher magnification a large agglomerate and also a smaller one in order to show how the particles can be recognised in the adhesive matrix. The clearer large spot in the centre of Figure 1b is not an agglomerate but an irregularity (in particular an outward relief) of the adhesive matrix surface. This was verified with the Energy-dispersive X-ray spectroscopy (EDS) associated with SEM, but not reported here. The largest particle size found in 5%_HM is close to 10 μm and it is shown in Figure

1a, as evidenced by the scale of the first image. The higher magnification, Figure 1b, shows that there are some small areas where the dispersion of the particles is not very uniform. On the other hand, the images related to the adhesive compounds prepared with the extruder, Figure 1c, and 1d, display a more homogenous dispersion of iron oxide particles within the adhesive matrix. This behaviour is mainly due to low shear mixing achievable with hand mixing, unable to exfoliate and disperse the iron oxide particles within the polymer matrix [32].

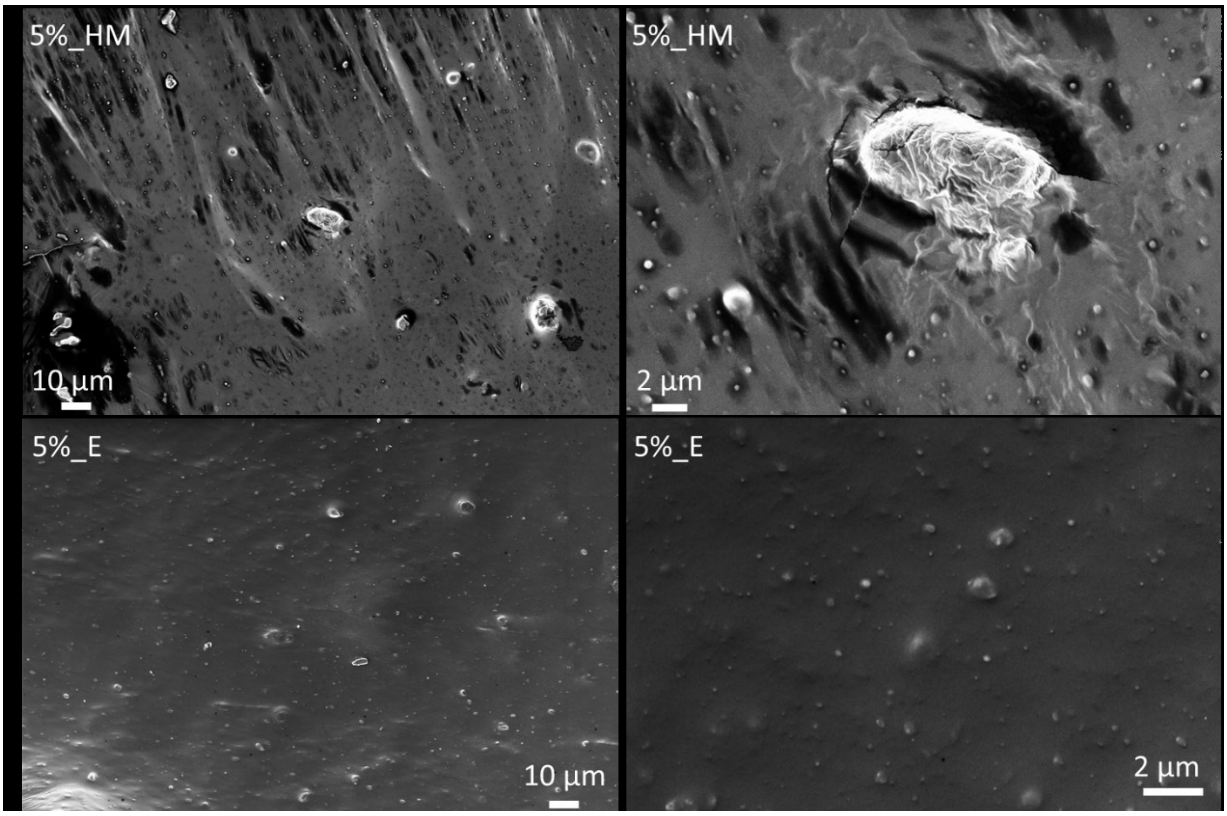


Figure 1: SEM images of 5%_HM and 5%_E, with two different magnifications

Figure 2 shows the SEM analysis conducted on the adhesive compositions 10%_HM and 10%_E. This analysis confirms the results conducted on 5%_HM and 5%_E. 10%_HM presents some agglomerates that are visible in Figures 2a and 2b, some of them are shown with the blue arrows. Furthermore, as showed for 5%_HM, both the images related to the adhesive compounds prepared with the extrusion method display a homogenous dispersion of the iron oxide particles within the adhesive matrix even if there are some small agglomerates, Figure 2d, that are limited compared to the hand mixing method.

Overall, the extrusion method gives a very uniform and good dispersion despite some small agglomerates compared to the hand mixing method. The agglomerates have been measured by using the digital image correlation software of the SEM used. The adhesive 5%_HM present a huge agglomerate that is 10 µm while the other ones were found between 2 and 3 µm. The adhesive 5%_E presents some smaller agglomerates with a size between 0.3 and 0.4 µm. On the other hand, 10%_HM displays some bigger agglomerates that are between 3 and 4 µm. 10%_E presents few agglomerates between 0.2 and 0.3 µm.

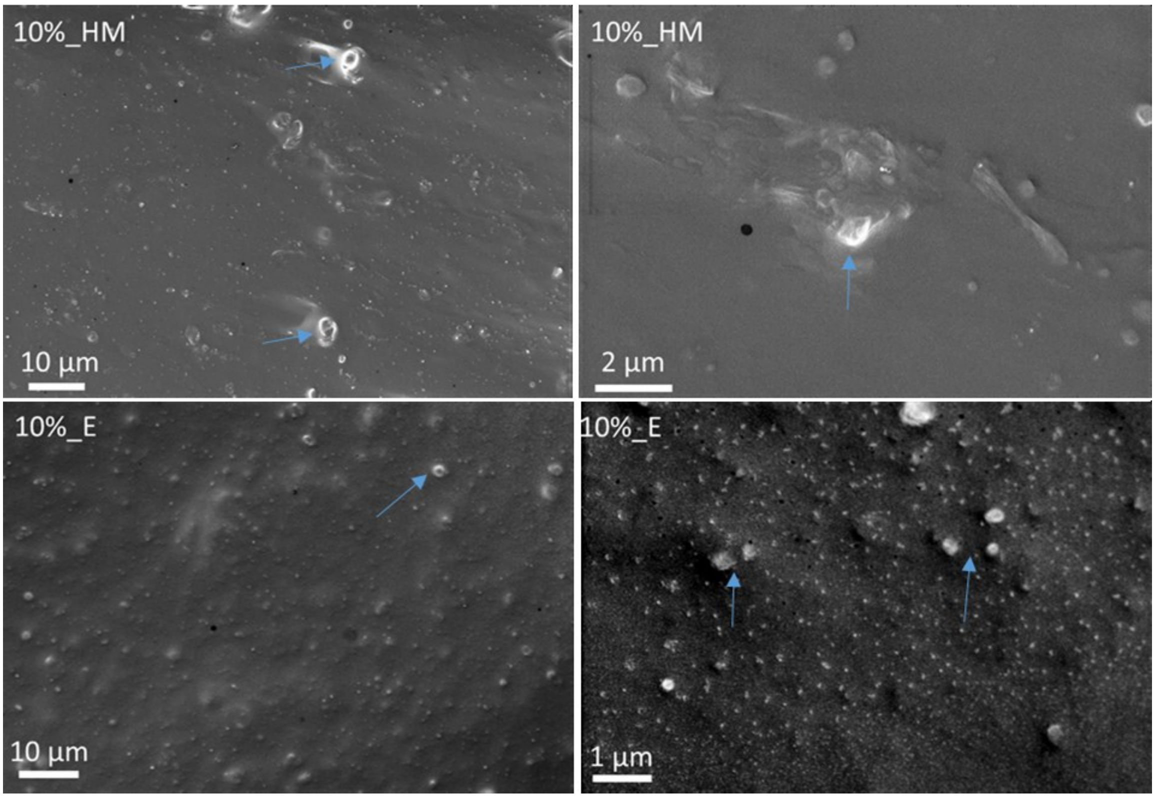


Figure 2: SEM images of 10%_HM and 10%_E, with two different magnifications

3.2 Mechanical tests

Figure 3 reports representative load-displacement curves obtained for the SLJ tests performed with the five different adhesive formulations. In this figure, the mechanical behaviour of SLJ prepared with the pristine HMA is compared with the SLJ prepared with the HMAs modified with hand-mixing method and the extruder. This figure shows that the initial slopes of the modified adhesives, representative of the stiffness of the adhesive joint, do not change significantly. Figure 3 shows also that the values of the maximum load for 5%_HM and 10%_HM are slightly higher than the pristine one while 5%_E and 10%_E present slightly lower values compared to the pristine hot-melt adhesive. The slightly higher load of the joints prepared with the hand mixing method could be due to a toughening effect of the bondline as a consequence of the relatively large presence of particles. Furthermore, all the modified adhesives present a more ductile behaviour compared to the unmodified adhesive.

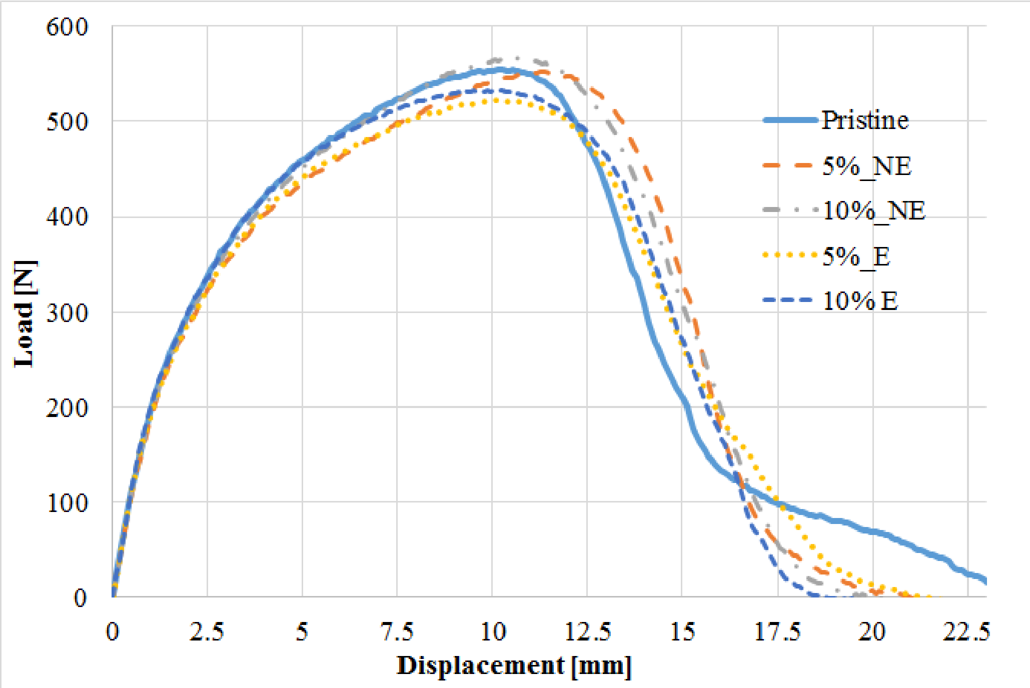


Figure 3: Mechanical test results of the neat and nanomodified adhesives modified with both the manual mixing and extrusion methods

The five repetitions that have been done for each of the performed SLJ tests do not put in evidence significant scatter in the curves so that the ones reported in Figure 3 can be considered fully representative of the structural behaviour.

Figure 4 reports the average maximum loads, the average strengths and their standard deviations for the adhesive joints modified with the hand-mixing and extrusion method. Figure 4 confirms the trend visible in Figure 3. The mean maximum values do not change significantly compared to the pristine adhesive. The largest increase has been found for the joints prepared with 10%_HM that is 5% higher. This could be due to the presence of agglomerates in the adhesive matrix modified with the hand mixing method that could have a toughening effect on the bondline that resulted in a higher value of the maximum load [28-31]. On the other hand, the values of the maximum loads of the joints prepared with 5%_HM, 5%_E, and 10%_E are very close to the pristine one. The standard deviations are reported with the error bars and evidence a very limited scatter.

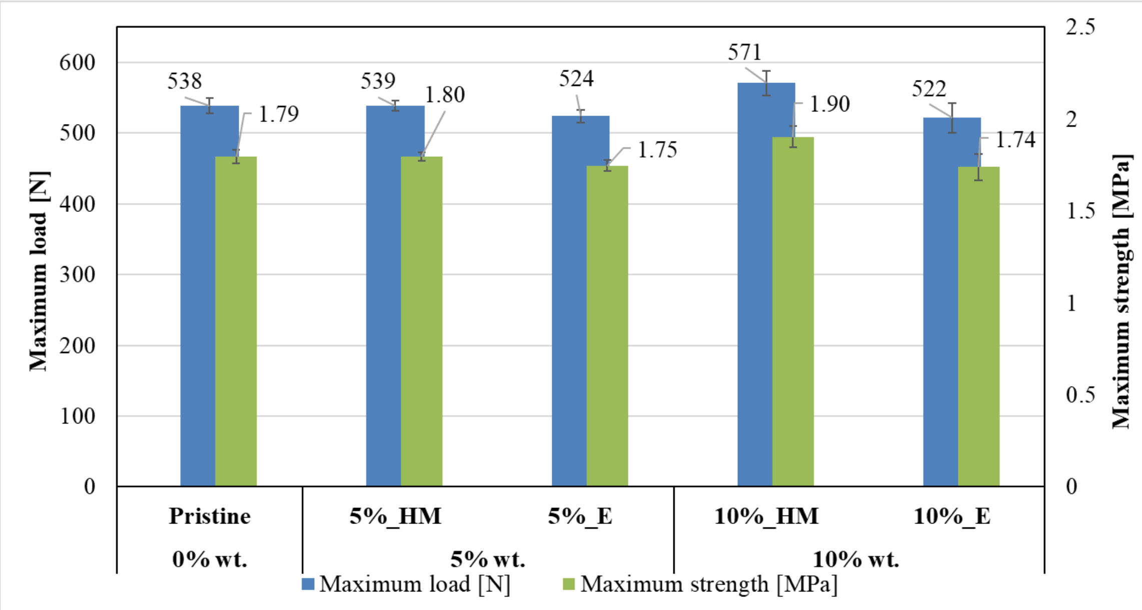


Figure 4: Maximum mean load and strength for all the five adhesive compositions

Statistical analysis was carried out in order to assess whether the compounds or the mixing method significantly influences the maximum loads of the investigated bonded joints. Analysis of variance (ANOVA) was used to statistically compare the data. Generally, the statistics F, computed for each factor and interaction is compared with the considered percentile of the Fisher distribution. F is the ratio between the mean squares of the computed factors, the one related to the compound (C), the one related to the mixing method (M), and their interaction (C-M). If the value F is larger than the percentile calculated with Fisher (i.e. F95% or F90%), then the corresponding factor or interaction significantly influences the investigated response, i.e. maximum force or separation time that has been analysed in this work.

The main parameters used in the calculation of the ANOVA are reported in each table: the sum of squares (SS), degrees of freedom (DOFs), mean squares (MS), and Fisher ratios (F) were computed from the experimental data. F95% and F90% are the 95% and 90% percentiles of the Fisher distribution. The ratio between the experimental SS (sum of squared deviations from a mean value) and the DOF (number of factor levels minus 1) permits the determination of MS (i.e., MS = SS /DOF). The computed MS is used to assess the significance of each factor (factors C and M) and the interaction between factors (Interaction C–M). Significance analysis was performed for each factor and interaction through the statistical hypothesis test proposed by Fisher (F-test). For each factor and interaction, the statistics F (ratio between the MS related to each factor, MS_C and MS_M, and interaction, $MS_{C\text{-}M}$, and the MS related to the Error, MS_E) is compared with the considered percentile of the Fisher distribution.

Table 1 reports the results of the ANOVA for the maximum forces. As it can be seen in the table, for the investigated range of compounds (the different percentage of nanoparticles), the type of compound affects the maximum force when a 95% confidence level is considered. Furthermore, the interaction between compounds and the mixing method affects the maximum force as well when confidence level of 95% is considered. Thus, this significance is mainly related to the higher values obtained with a large percentage of the particles. It is worth to note that the mixing method is not significant neither for F95% nor F90%.

Table 1: ANOVA for the maximum force

Source	*SS*	*DOF*	*MS*	*F*	*F95%*
Compound (C)*	2757	1	MS_C=2757	F_N=11.97	4.42
Mixing method (M)	2757	1	MS_M=2757	F_V=1.94	3.55
Interaction C-M*	2549	2	MS_{C-M}=1274	F_{N-V}=5.54	3.55
Error	4145	18	MS_E=230		
Total	10347	23			

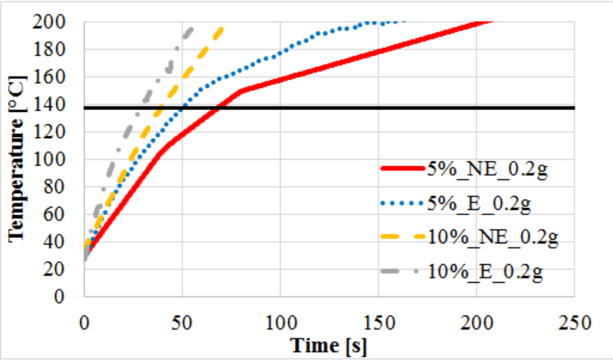
Note: (*) denote the level of confidence: one star for 90% confidence level

3.3 Adhesive heating

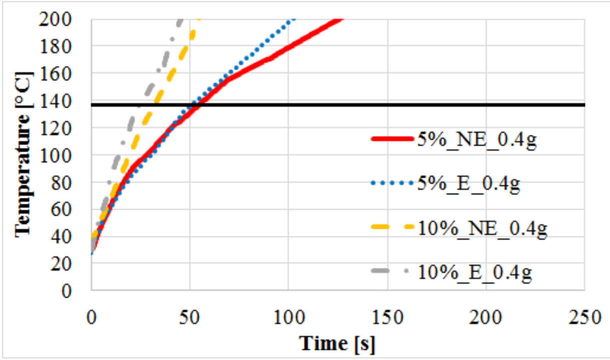
In this section, results related to the heating of the modified adhesive by means of microwave and electromagnetic induction of the modified adhesive have been reported. Figures 5a and 5b display the temperature-time curves of the four different adhesive compositions heated by means of microwave for two different thicknesses that correspond to a weight of 0.2 g (thickness 0.5 mm) and 0.4 g (thickness 1 mm). This extension has been added since Ciardiello et al. [31] showed that the separation tests conducted on different thicknesses do not affect the separation time. For this reason, the effects of microwave on different thickness (and mass) have been added in this study for two different thickness configurations: 0.5 mm (0.2 g) and 1 mm (0.4 g) with fixed width of 20 mm and overlap of 15 mm. The black line in the diagrams of figures 5a, 5b and 5c marks the temperature of 135 °C that is the temperature at which the joint separation occurred in the case of the electromagnetic induction. This temperature has been added in order to obtain a direct comparison between electromagnetic induction and microwave.

As expected, the curve related to the higher percentage of nanoparticles (10%) shows a more rapid temperature increment with respect to the lower percentage (5%), while there is a more rapid temperature increment for the E type specimens with respect to the HM type.

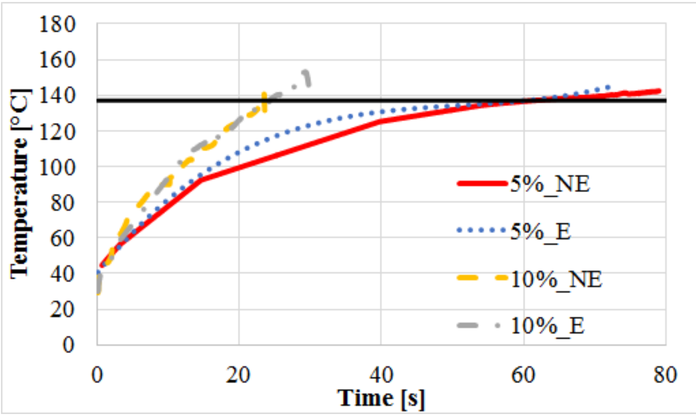
Figures 5a and 5b display that microwave heating does not affect significantly the adhesive heating up to 135 °C. However, the temperature was measured up to 200 °C, that is the point where this adhesive starts to degrade [31]. It is noticeable that the adhesive layer with higher thickness (0.40 g) reaches 200 °C in lower time compared to the adhesive layer of 0.20 g. Figure 5c shows the temperature-time curves measured with the IR camera. Even in this case, the trends related to the overlap of 10%_HM and 10%_E seem to be very similar to the microwave. On the other hand, induction heating of 5%_HM and 5%_E is slower compared to the microwave heating. As showed by Ciardiello et al. [31], the HMA loaded with a lower concentration of particles presents some areas with a lack of particles, as shown also in the SEM section of this work. This lack of particles in some areas for the adhesive prepared with the 5% wt. led to higher values of the separation time (at least 11 s) in the case of the adhesive separated with the induction heating system compared to the one separated with the microwaves. This means that the microwave system is able to heat more efficiently the modified adhesive compared to the induction heating system.



a



b



c

Figure 5: Temperature – time curves of the adhesives prepared with the hand mixing method HM and extruder E. a- Specimen of 0.20 g heated with microwave; b- specimens of 0.40 g heated with microwave; c- specimens of 0.25 g heated with the induction system

The summary of the results related to both induction and microwave heating is displayed in Figure 6. The diagram reports the time to reach the separation temperature, which is 135 °C. Separation tests showed that microwave and induction heating systems are able to heat and separate adhesive joints prepared with the adhesive modified with 5 and 10 weight percent with both mixing methods. That study [31] showed that higher is thickness lower is time to separate substrate. The tests carried out in this activity showed that even in this case higher thicknesses led to lower values of the separation time even when they are heated with microwaves. This could be due to the higher mass involved in the heating process [31]. Overall, microwave tests showed that the sample prepared with higher thickness (0.4 g) can be heated in a lower time for both the adhesive compounds tested in this work. The differences between 0.2 g and 0.4 g are 23% for the adhesive compound 5%_HM 17% for the 5%_E, 8% for the 10%_HM and 5% for 10%_E. Furthermore, it is noticeable that the time to heat the extruded compound is quicker compared to the hand-mixed one, 6% lower in the case of 5% wt. for the 0.4 g samples, 12% for the 5% wt. compound in the case of the 0.2 g sample, 15% lower in the case of 10% wt. compound for the 0.4 g sample and 16% for the same adhesive compound but the 0.2 g sample. This is due to the better distribution of the particles in the adhesive matrix as shown by the SEM analysis in Section 3.1. In fact, HM samples showed some area less rich in particles due to the higher agglomerates that are presented when the hand-mixed method is used for the preparation of the adhesives. On the other hand, the separation time analysed by using the induction heating technology showed that there is no significant change between the separation times with respect to the mixing methods based on the obtained scatter. As expected, and as reported by Ciardiello et al. [31], the 10% wt. compound can be separated in lower time, which is among 2 and 3 times lower compared to 5% wt. It is also important to note that while induction heating and microwaves tests conducted on the 5% wt. samples do not lead to significant differences, the sample prepared with 10% wt. presents lower times when heated with induction heating. This can be related to the higher value of the applied power in the case of the induction heating method and to the higher presence of particles in the case of 10% wt. sample.

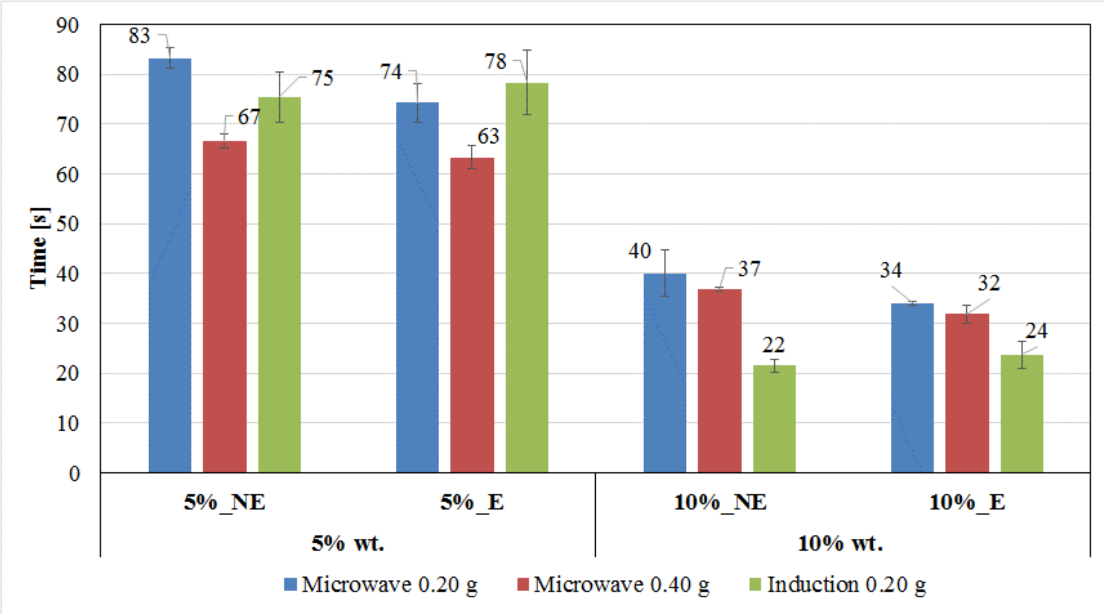


Figure 6: Microwave and induction heating tests conducted on modified adhesives

Table 2 reports the ANOVA of the experimental plane (2^3) related to the separation time. The factors that have been included in this analysis are the two different compounds analysed (5% and 10%), the two different mixing methods (hand-mixed and extruded) and the adopted separation methods (electromagnetic induction and microwave). In this analysis, the values of the separation times related to the microwave are the ones related to 0.4 mm since they are the same as the adhesive joints configuration adopted for the electromagnetic induction. ANOVA shows that the compound (percentage of particles) affects significantly the separation time as well as the interaction between compound and separation method. On the other hand, all the other factors, including both the other two-level and the tri-level interactions, do not affect the separation time neither with F95% nor F90%.

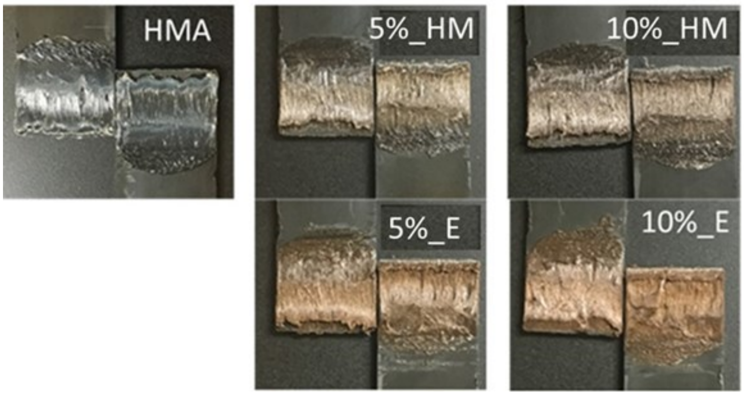
Table 2: ANOVA related to the compound, mixing methods and separation methods

Source	*SS*	*DOF*	*MS*	*F*	*F95%*
Compound (C)*	10584	1	MS_C=10584	F_C=308	4.49
Mixing method (M)	1.5	1	MS_M=1.5	F_M=0.04	4.49
Separation method (S)	0.2	1	MS_S=0.2	F_S=0.00	4.49
Interaction C-M	2.7	1	MS_{C-M}=2.7	F_{C-M}=0.08	4.49
Interaction C-S*	770.7	1	MS_{C-S}=770.7	F_{C-S}=22.45	4.49
Interaction M-S	73.5	1	MS_{M-S}=73.5	F_{M-S}=2.14	4.49
Error	459.3	16	MS_E=34.3		
Total	11981	23			

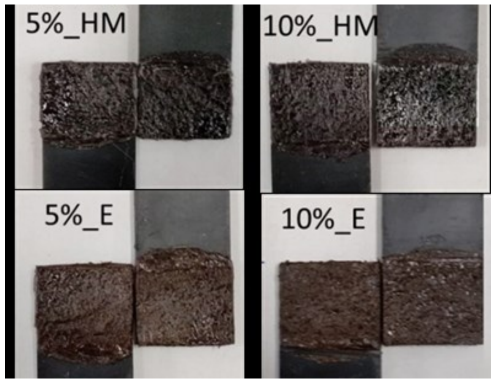
Note: (*) denote the level of confidence: one star for 95% confidence level

3.4 Fracture and separation surfaces

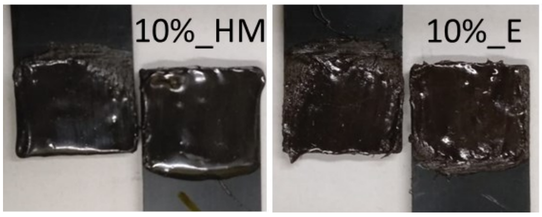
Figure 7 shows representative fracture and separation surfaces of the pristine HMA specimen and the modified ones. Figure 7a) shows the fracture surfaces obtained by the SLJ test. The first row, in Figure 7a), beside the fracture surfaces of the pristine specimen, displays the fracture surfaces of the adhesives modified with the hand-mixing method HM whereas the second row shows the adhesives modified with the extrusion method E. The visual inspection of these fractured surfaces showed that they are totally cohesive and there are no differences among the pristine adhesive and the modified ones, sign of the good adhesion between adhesive and substrates. It is noticeable that the colour of the adhesive modified with the extruder method is clearer than the one modified with the hand-mixing method. This can be attributed to the better mixing in the extruded case. Figure 7b) shows the separation surfaces obtained by the electromagnetic induction system. Even in this case, the separation surfaces are completely cohesive and the colours are clearer for the specimens prepared with the extruded method. Figure 7c) displays the separation surfaces obtained by using the microwave process. Both adhesives look brighter. This is because the adhesive is completely melted while in the case of the electromagnetic induction system the weight that has been used to initiate the slide did not allow the complete melt of the adhesive.



a)



b)



c)

Figure 7: a) Fracture surfaces of the SLJ specimen after the SLJ tests; b) separation surfaces after induction heating; c) separation surfaces after microwave heating

4. Conclusions

In this work, the possibility to dismantle HMA plastic joints by means of iron oxide modified adhesives coupled with microwaves and induction heating is assessed. While the induction heating methods have already been studied by the authors in previously published papers, the microwave heating constitutes, at the author's best knowledge, the relevant novelty of this study. Further two mixing methods, namely extrusion and hand mixing, have been adopted to complete the study. SEM analysis showed the presence of agglomerates within the adhesive prepared with the hand-mixing method while the extrusion method led to a uniform distribution of the particles. The mechanical properties present no detrimental changes in the maximum ultimate load, generally a more ductile behaviour and, for the compound prepared with the hand mixed method, slightly higher ultimate load due to a toughening effect of the bondline.

Separation tests showed that microwave and induction heating systems are able to heat and separate adhesive joints prepared with both the adhesives modified with 5% and 10% in weight and with both mixing methods. Whereas the induction heating system led to a lower separation time in the case of adhesive modified with 10% of iron oxide particles, the separation time of the adhesives prepared with 5% of iron oxide is comparable. However, it can be noted that the microwave process is more efficient, leading to a dismantling time of the joint similar to the one obtained with the traditional induction heating process but with an electric power requirement that is much smaller and thus of large interest for industrial applications.

5. References

[1] Chang B, Shi Y, Dong S. Comparative studies on stresses in weld-bonded, spot-welded and adhesive-bonded joints. J Mater Process Technol 1999;87:230–236.

[2] Belingardi G, Chiandussi G. Stress flow in thin walled box beams obtained by adhesive bonding joining technology. Int J Adhes Adhes 2004;24:423–439.

[3] Rudawska A. Adhesive joint strength of hybrid assemblies: Titanium sheet-composites and aluminium sheet-composites—Experimental and numerical verification. Int J Adhes Adhes 2010;30(7):574-582.

[4] Rudawska A, Worzakowska M, Bociąga E, Olewnik-Kruszkowska E. Investigation of selected properties of adhesive compositions based on epoxy resins. Int J Adhes Adhes 2019;92:23-36.

[5] Belingardi G, Brunella V, Martorana B, Ciardiello R. Thermoplastic adhesive for automotive applications. In: Rudawska A, editor. Adhesive – Application and properties. Rijeka: INTECH, 2016p. 341-362.

[6] Casalegno V, Salvo M, Rizzo S, Goglio L, Damiano O, Ferraris M. Joining of carbon fibre reinforced polymer to Al-Si alloy for space applications. Int J Adhes Adhes 2018;82:146-152.

[7] Lu Y, Broughton J, Winfield P. A review of innovations in disbonding techniques for repair and recycling of automotive vehicles. Int J Adhes Adhes 2014;59:119–127.

[8] Banea MD, da Silva LFM, Campilho RDS. An overview of the technologies for adhesive debonding on command. Weld Equip Technol 2013;24:11-14.

[9] Directive 2000/53/EC of the European Parliament on end-of life vehicles. 18 September 2000.

[10] Directive 2005/64/EC of the European Parliament on the type-approval of motor vehicles with regard to their reusability, recyclability and recoverability. 26 October 2005.

[11] Banea M. Debonding on demand of adhesively bonded joints: a critical review. Rev Adhes Adhes 2019;7(1):33-50.

[12] Verna E, Cannavaro I, Brunella V, Koricho EG, Belingardi G, Roncato D, Martorana B, Lambertini V, Neamtu V, Ciobanu R. Adhesive joining technologies activated by electro-magnetic external trims. Int J Adhes Adhes 2013;46:21–25.

[13] Banea M, da Silva L, Carbas R. Debonding on command of adhesive joints for the automotive industry. Int J Adhes Adhes 2015;59:14–20.

[14] Ciardiello R, Martorana B, Lambertini VG, Brunella V. Iron-based reversible adhesives: Effect of particles size on mechanical properties. Proc Inst Mech Eng Part C 2017;232(8):1446–1455.

[15] Vattathurvalappil SH, Haq M. Thermomechanical characterization of Nano-Fe3O4 reinforced thermoplastic adhesives and single lap-joints. Compos Part B-Eng 2019;175. In press: Article number 107162 (https://doi.org/10.1016/j.compositesb.2019.107162).

[16] Severijns C, Teixeira de Freitas S, Poulis JA. Susceptor-assisted induction curing behaviour of a two component epoxy paste adhesive for aerospace applications. Int J Adhes Adhes 2017;75:155-164.

[17] Banea MD, da Silva LFM, Carbas RJC. Debonding on command of multi-material adhesive joints. J Adhes 2017;93(10):756-770.

[18] Bayerl T, Duhovic M, Mitschang P, Bhattacharyya D. The heating of polymer composites by electromagnetic induction – A review. Compos Part A-Eng 2014;57:27-40.

[19] Suwanwatana W, Yarlagadda S, Gillespie JW. Hysteresis heating based induction bonding of thermoplastic composites. Comp Sci Technol 2006;66:1713–1723.

[20] Ghazanfari M, Kashefi M, Shams S, Jaafari M. Perspective of Fe^3O^4 nanoparticles role in biomedical applications. Biochem Res Int 2016;16:1-32.

[21] Ciardiello R, Belingardi G, Martorana B, Fondacaro D, Brunella V. A study of physical and mechanical properties of a nanomodified thermoplastic adhesive in normal and accelerated ageing conditions. Proceedings of the 17th European Conference on Composite Materials (ECCM16), Munich 26-30 June 2016. p. 1-8.

[22] Ping-Cheng S, Tzu-Huan C, Shih-Chin C. Microwave curing of carbon nanotube/epoxy adhesives. Comp Sci Technol 2014;104:97-103.

[23] Wang C, Chen t, Chang S, Cheng S, Chin T. Strong Carbon-Nanotube–Polymer Bonding by Microwave Irradiation. Adv Funct Mater 2017;17:1979–1983.

[24] Shim H, Kwaka Y, Han C, Kim S. Enhancement of adhesion between carbon nanotubes and polymer substrates using microwave irradiation. Scr Mater 2009;61:32-35.

[25] Galindo B, Benedito A, Ramos F, Gimenez E. Microwave heating of Polymers: influence of carbon nanotubes dispersion on the microwaves susceptor effectiveness. Proceedings of the 6th International Conference On Carbon Nanoparticle Based Composites, Dresden 22-25 September 2013. p. 1-3.

[26] Chong HM, Hinder SJ, Taylor AC. Graphene nanoplatelet-modified epoxy: effect of aspect ratio and surface functionality on mechanical properties and toughening mechanisms. J Mater Sci 2019;51(19):8764–8790.

[27] Rafiee MA, Rafiee J, Wang Z, Song H, Yu Z, Koratkar N. Enhanced mechanical properties of nanocomposites at low graphene content. ACS Nano 2009;3(12):3884–3890.

[28] Koricho E, Verna E, Belingardi G, Martorana B, Brunella V. Parametric study of hot-melt adhesive under accelerated ageing for automotive applications. Int J Adhes Adhes 2016;68:164–181.

[29] Ciardiello R, Belingardi G, Martorana B, Brunella V. Effect of accelerated ageing cycles on the physical and mechanical properties of a reversible thermoplastic adhesive. J Adhes 2018. In press: DOI: 10.1080/00218464.2018.1553714.

[30] Ciardiello R, Tridello A, Brunella V, Martorana B, Paolino DS, Belingardi G. Impact response of adhesive reversible joints made of thermoplastic nanomodified adhesive. J Adhes 2017;94(12):1051–1066.

[31] Ciardiello R, Belingardi G, Martorana B, Brunella V. Physical and mechanical properties of a reversible adhesive for automotive applications. Int J Adhes Adhes 2019;89:117–128.

[32] Atif R, Inam F. Reasons and remedies for the agglomeration of multilayered graphene and carbon nanotubes in polymers. Beilstein J Nanotech 2016;7:1174–1196.