Adhesive joining technologies activated by electro-magnetic external trims

ADHESIVE JOINING TECHNOLOGIES ACTIVATED BY ELECTRO-MAGNETIC EXTERNAL TRIMS

Elena Verna [b,c*], Irene Cannavaro[a], Valentina Brunella[c], Ermias Gebrekidan Koricho[b], Giovanni Belingardi[b], Davide Roncato[a], Brunetto Martorana[a], Vito Lambertini[a], Vasilica Alina Neamtu[d], Romeo Ciobanu[d]

[a]*Centro Ricerche FIAT, Strada Torino, 50 – 10043 Orbassano (TO), Italy*
[b]*Politecnico di Torino, Dipartimento di Meccanica, Corso Duca degli Abruzzi, 24 – 10129 Torino,*
[c]*Dipartimento di Chimica I.F.M, Università degli Studi di Torino, Via P. Giuria, 7 – 10125 Torino*
[d]*Technical University from Iasi, Faculty of Electrical and Energetic Engineering and Applied Informatics, Prof. Dr. Dimitrie Mangeron Street, No 67, Iasi, Romania*

e-mail: elena.verna@gmail.com

Keywords: reversible adhesive joint, electromagnetic field, hot-melt adhesive, ferrite nanoparticles.

Abstract

Joining is a key and fundamental aspect of vehicle design and manufacturing process. The development of efficient, simple, inexpensive and reversible adhesive bonding technologies offers low cost, improved life cycle and recycling solutions for a vehicle. This innovative technology is based on magneto sensitive nanoparticles dispersed into the adhesive. Different typologies of susceptors, embedded in thermoplastic hot-melt adhesives, have been tested to investigate the thermal behaviour of these innovative materials when exposed to electromagnetic fields. These innovative technologies are intended to optimize the bonding process offering new opportunities connected with cost reduction, improved resistance to applied loads, easy and rapid dismantling and smart recycling. Experimental procedures to optimize the electromagnetic process have been performed. Results about both shear strength and sliding temperature of the modified adhesives are presented.

1 Introduction - Electromagnetic welding process

Due to continuous rising in demand of implementation of recyclable polymeric materials on vehicle structures, it is very essential that appropriate joining techniques, that have already proved to be capable to offer excellent structural solutions in terms of fatigue and impact strength, can facilitate easy of disassembly and optimize the recycling process. One of promising adhesive types that can be taken as environmental friendly and cost effective is hot-melt thermoplastic adhesives. In light load application area these kinds of adhesives are used more and more due to their short curing time and nature of reversibility at high temperature. However, one of critical problems related to adhesive joint in automotive application is that it doesn't allow disassembling components during maintenance and recycling process.

Taking into consideration inherent property of thermoplastic substrates and hot-melt adhesives, reversibility at high temperature, the process of electromagnetic bonding has gained a considerable acknowledgment to join composite and plastic materials using different ferromagnetic nanoparticles embedded inside polymer matrix.

The process of electromagnetic bonding is based on the principle of electromagnetic heating: magnetic field sensitive materials increase their temperature when subjected to a high frequency, alternating current (AC) field [1].

Electromagnetic radiation absorbed by ferromagnetic particles embedded in the adhesive matrix causes the susceptor particles to rapidly heat. Conductive heat transfer from the ferromagnetic particles into the polymer matrix material causes creeping flow of the implanted material [2].

The physics of electromagnetic induction applies to both Joule heating (eddy current losses) and magnetic heating (hysteresis losses). Benatar [3] has described the heating of ferromagnetic susceptor materials as the combination of both of these effects. He also described that magnetic and eddy current intensity decreases rapidly with increasing distance of the penetration depth. At each cycle of the applied radio frequency (RF) field, the susceptor responds by completing the full cycle. The area bounded by the hysteresis curve is proportional to the energy converted into heat. High frequencies are required because the incremental temperature risen for each hysteresis cycle is very small [4, 5].

Nanoparticles, are being used in automotive industries to foreshorten the curing time of thermosetting adhesives and ultimately to achieve comparatively minimum assembly time. For instant, the curing time of a commercially available epoxy based adhesive is in the range of several hours; however with embedded susceptible nanoparticles inside, when it is subjected to electromagnetic field, its curing time is reduced to around 17 minutes. Actually the curing time is dependent on the power generated by induction coil that generate eddy current around the matrix modified with metallic nanoparticles, by the amount of nanoparticles percentage and by permissibility value that characterizes the considered nanoparticle type. Appropriate controlled power should be supplied to avoid material degradation because of too high temperature.

In general, nanoparticles are believed to enable novel adhesive with unsurpassed properties [6, 7]. With controlled mixing mechanisms nanoparticles play a great role towards an improved mechanical behaviour and rheological control.

In this study, five metallic particles were chosen and embedded in the hot-melt adhesive to investigate their thermal response while they were subjected to external electromagnetic field using induction heating process. Ranges of frequency of AC were chosen for the selected materials. It is not within this present technical capability to directly measure the proportion to which these heating effects contribute to the total heating of the susceptor material, nor able to measure effective penetration depth. In this work, certain fundamental relationships, that regulate the electromagnetic welding process, have been explored through an experimental approach. The considered parameters associated with the modified adhesive and with the process are: typology and amount of nanoparticles in the adhesive, intensity and frequency of the electromagnetic field. There are some other parameters worthy of investigation, such as the shape of the coil, the coupling distance, but they are not evaluated within this present investigation. Based on the found result, a best type of nanoparticle that generates elevated temperature, was chosen and proposed for further future work.

2 Materials and testing methods

2.1 Materials

This work employed five types of iron based nanoparticles: magnetite Fe_3O_4 (Sigma-Aldrich), hematite α-Fe_2O_3 (Nanophase), cobalt-ferrite $CoFe_2O_4$ (Nanotesla), manganese-zinc ferrite $(Mn,Zn)Fe_2O_4$ (Nanotesla), and maghemite γ-Fe_2O_3 (Nanotesla).

These ferrites were selected to evaluate their effects when they are embedded into a hot-melt adhesive, characterized by the formulation listed in Table 1.

Table 1. Qualitative and quantitative composition of the hot-melt adhesive.

In order to investigate the mechanical properties of modified and unmodified adhesives, joints in homopolymer polypropylene (PP) 10% talc filled were prepared. This compound is largely used in automotive internal and external applications.

2.2 Testing methods

Each typology of iron powder was ultrasonically dispersed in isopropyl alcohol prior to depositing it onto a plate. Tapping mode-atomic force microscopy (AFM) was used to study the size and shape of the particles. A Veeco Digital AFM Instrument was used for these studies (scan area 1 µm x 1 µm).

In order to test the mechanical properties of modified and unmodified adhesives, standard PP 10% filled talcum tensile specimens of size 100x20x3 mm were used. The adherends were bonded for single lap joint specimens (SLJ) with constant adhesive thickness of 1 mm and an overlap length of 25 mm.

For surface preparation, the substrates were cleaned with isopropyl alcohol. The adhesive was dispensed by a Nordson Durablue adhesive melter and the excessive adhesive at the overlap edges was removed in all joints. A compressive force of 20 N was applied to the lap joints during the cooling period (two minutes). The single lap joints were assembled by means of a specially manufactured apparatus, that allows the standardized joint preparation technique to be used repeatedly with constant adhesive film thickness. Then, the specimens were exposed at room temperature for 24 hours prior to testing.

Five tests were carried out for each adhesive Single-lap joint, SLJ. Specimens were subjected to tensile loading using a Instron 5544 Series dynamometer at room temperature under displacement control (100 mm/min, as suggested by automotive standards); tabs at the extremities of the SLJs were bonded to assure a correct alignment into the dynamometer cell. Throughout this work, the average shear strength was used to characterize the joint strength, and was calculated as the maximum applied load of each test divided by the measured bonded area. The failure mode was determined by visual inspection.

To identify the slipping temperature, i.e. the temperature at which SLJ is deboned due to softening of the hot melt adhesive, SLJs were loaded with the weight force given by a mass of 500 g and subjected to gradually increasing temperature (50°C/h) inside a temperature controlled chamber.

Nanoparticles were manually blended with hot melt adhesive at a temperature of 180°C. Mechanical tests have been used to investigate samples.

SEM analysis was carried out on the surface and cross-section of modified adhesive film to verify the distribution of nanoparticles and obtain clear images of their morphology, respectively. As usual, the samples were coated with a thin film of Au in order to increase their conductivity.

For the electromagnetic test, a tape of modified adhesive (17 x 110 mm) was placed inside the inductive coil and AC current was supplied through the coil. Heating profile was measured using a opSens fiber optical sensor model OTG-M170, placed in the middle of the adhesive sample. Different work coils were used on the generator throughout the experimental study.

In order to manufacture joints by new assisted assembling technology, a new manufacturing methodology was set: modified adhesive was cut to obtain a tape with the proper size of the joint overlapping (20 x 25 mm). This sample was placed on a polypropylene substrate and the second substrate was laid upon that. Finally the joints were tacked on by a PTFE wire.

In order to perform assembling tests by use of the electromagnetic field, SLJs were hanged inside the coil, placing the overlapping area in the middle of the coil. In order to measure the heating temperature, the fiber optical sensor was put inside the adhesive.

To perform disassembling tests a similar setup was used: one extremity of SLJs was loaded with the constant weight force given by a mass of 500 g, and inserted inside the coil. Placing the joint inside the coil, the RF field activates the modified adhesive allowing the joint dismantling.

3 Experimental results and discussion

3.1 Nanoparticles characterization

Atomic Force Microscopy (AFM) revealed for each typology of iron particles, agglomerates of particles in the range from 10 to 60 nm, with an average radius smaller than the superparamagnetic limit. Figure 1 shows magnetite nanoparticles morphology. All the ferrite nanoparticles resulted typically spherical in shape.

Figure 1. Atomic force image of magnetite Fe_3O_4 particles.

3.2 Modified and unmodified adhesive characterization.

"Figure 2a shows SEM image, taken on the surface of adhesive modified adding 5 weight percent of magnetite nanoparticles. As a result of the above described mixing process, the nanoparticles tend to agglomerate and form clusters of micrometric dimensions. Figure 2b ~~3b~~ was taken on the modified adhesive cross section and shows the morphology of nanoparticle clusters. It is not possible to determine if aggregates of smaller size are present within the adhesive, since they are difficult to be distinguished from the adhesive matrix because of their low size, and the insufficient resolution obtained at higher magnifications."

Figure 2. Scanning electron micrograph of the adhesive filled with 5% by weight of magnetite nanoparticles. (a) surface analysis 200x, (b) cross-section analysis 5000x.

Figure 3 represents the typical applied force/displacement curve obtained for three SLJ samples with different adhesive formulations: unmodified and modified adhesives (5 and 10 weight percents of magnetite). Modified adhesive SLJs exhibit an increase in maximum shear strength against unmodified SLJs.

The average shear strength was obtained on 5 samples for each type of adhesive formulations and the results are listed in Table 2. Although the difference in strength between the 'as prepared' and the '5%' system does indeed appear to be significant, the difference between 5% and 10% appears close, with overlap of dispersion bars when taking standard deviation values into account. This implies that because of nanoparticles inclusions inside the adhesive matrix, the damage initiation is delayed and gives higher load carrying capacity of SLJ specimens. This phenomenon can be explained in other way: the interfacial interactions between the nanoparticles and the adhesive matrix allow the transfer of stress to the reinforcing particles which results in an increase in material strength. It was observed that the interfacial bond strength between the modified adhesive and polypropylene substrate is not altered compared with the unmodified adhesive and leads to consistent cohesively dominated failures, as shown in Figure 4.

Figure 3. Applied force/displacement curve for SLJ prepared with unmodified and modified adhesive (5 and 10% weight percent of magnetite).

Table 2. SLJs results of unmodified and modified adhesive (5 and 10 weight percent of magnetite).

Figure 4. Failure mode of SLJ (a) unmodified adhesive, (b) 5% weight percent of magnetite, (c) 10% weight percent of magnetite.

The introduction of ferrite nanoparticles into the adhesive matrix shows insignificant effect on SLJ slipping temperature, which remained almost the same of the unmodified adhesive SLJs (105°C).

3.3 Bonding process optimization.

The temperature response as a function of time for each modified adhesive, 5 weight percent of different nanoparticles, is shown in Figure 5. Table 3 presents temperature response at a representative value of 40 seconds for the different modified adhesives.

Figure 5. Thermal profile of modified adhesives exposed to electromagnetic fields (172 kHz, 3.3 kW).

Table 3. Adhesive heating results at 40 s – 172 kHz, 3.3 kW.

The adhesive which was modified by magnetite shows the best heating capability in electromagnetic field. This adhesive was able to reach in 40 seconds a temperature of 151°C (adhesive melting point).

Modified adhesive (5 weight percent of magnetite) was tested at different intensity of the electromagnetic field, in order to evaluate the temperature response as a function of time. Results are shown in Figure 6: the x-axis shows the power supplied by the voltage source, relatable to the intensity of the electromagnetic field, while y-axis is referred to the temperature reached within 40 s.

Figure 6. Temperature reached in 40 s vs electric power for magnetite modified adhesive (5 wt%, 172 kHz).

For tests performed at 1 kW the heating rate was quite lower than at the other tested powers. After 40 seconds, the temperature was still below 100°C (below the softening temperature of the adhesive).
For the remaining samples tested at more elevated powers, the temperature reached after 40 seconds was much greater. The adhesive samples tested at 3.3, 4.8, 6.6 and 9.9 kW show a similar behavior and the reached temperature after 40 seconds didn't change a lot.
By analyzing these test results it could be reasonable the choice of 3.3 kW as optimized power to be used for future characterization. The temperature reached with this power was slightly lower than the temperature reached with higher powers, but in any case satisfying for the innovative process. Therefore, the use of 3.3 kW could be advantageous in terms of limiting the spent energy and consequently dismantling cost.

Figure 6 shows that the modified adhesive increases its temperature logarithmically with the power supplied by the alternating voltage source.

From the electromagnetic equations, one can expect that the amount of heat generated by induction heating is directly proportional to the frequency of the electromagnetic field. Modified adhesive (5 weight percent of magnetite) were submitted to a 3.3 kW electromagnetic field, for different frequencies. Temperature increased with increasing frequency of the electromagnetic field in a linearly dependence manner.

The last test was carried out in order to understand the influence of the amount of magnetite nanoparticles, on adhesive matrix heating.

Figure 7. Temperature reached in 40 s vs weight percent of magnetite for modified adhesive (172 kHz, 3.3 kW).

Figure 7 demonstrates that heating rate varied with the increasing amount of magnetite nanoparticles into the adhesive as a 3rd order polynomial function.
Small amounts of nanoparticles (1, 3 weight percent) embedded into the adhesive matrix weren't enough to cause adhesive melting, while bigger amounts, from 5 to 10 weight percent, brought the modified adhesive to higher temperature and rapid heating.

3.4 Disassembling of joints by innovative RF technology

SLJs were tested by exposing them to electromagnetic field. In table 4 test results of modified adhesive based SLJ, with different amount of magnetite, are summarized.

Table 4. Disassembling time for modified adhesive (5-10-30 weight percent of magnetite), exposed to electromagnetic field: 172 kHz, 15.8 kW

Increment in the amount of embedded magnetite in the adhesive matrix leads to a decrement of the time needed to reach the complete disassembly of the joints in electromagnetic field.

4 Conclusions

Adhesive joint made with adhesives nanomodified by adding magnetic field sensitive particles have been studied. Different typologies of susceptors, embedded in thermoplastic hot-melt adhesives, have been tested in order to investigate the thermal behavior of the innovative materials exposed to electromagnetic fields.
An increase of magnetite content into the adhesive matrix affects mechanical properties: in particular the shear strength increases significantly.
In this first study of bonding behavior of ferrite modified adhesives, .the procedure followed to optimize the high frequency bonding process has been essentially based on direct experimental tests performed on standard single lap joint specimens. Five different types of susceptors have been considerd, magnetite powder showed the best performance for induction bonding.
The performed tests allow to point out that heating rate varied with the increasing amount of nanoparticles dispersed into the adhesive as a 3rd order polynomial function.
Other variables influenced the bonding process: heating rate increased by increasing frequency of the electromagnetic field in a linearly dependence manner, while it increased logarithmically with the electromagnetic field power.

Finally an increment of the amount of magnetite inserted into the adhesive matrix, also decreased the time it takes to reach the disassembly of the joints, when submitted to electromagnetic field.

5 Acknowledgements

The research activities were performed in the frame of the project “MANTA” (RBIP065YCL) funded by the M.I.U.R.

The international cooperation between Italian research group and the University of Iasi (Romania) research group was supported by the project PERFORM-ERA "Postdoctoral Performance for Integration in the European Research Area" (ID-57649), financed by the European Social Fund and the Romanian Government.

References

[1] J. Rotheiser, Induction/Electromagnetic Welding in: Joining of plastics Handbook for Designers and Engineers”, Hanser Publishers, Munich, 2004, pp. 331-332.
[2] R. Nichols, D. LaMarca, B. Agosto, Performance of Susceptor Materials in High Frequency Magnetic Fields, in Proc. Of ANTEC, SPE (2006).
[3] D. A. Grewell, A. Benatar, J. B. Park, Plastics and Composites Welding Handbook, Hanser Publishers, Munich, 2003, pp. 110-112.
[4] J. Park, B. Au, J. Esposito, D. Lewis, Weldability Evaluation on Effects of Filler Content of Polypropylene Copolymers for Electromagnetic Welding Process, in Proc. of ANTEC, SPE (2007).
[5] Pilar Rivera Gi, Loretta L. del Mercato, Pablo del_Pino, Almudena Muñoz_Javier, and Wolfgang J. Parak, Nanoparticle-modified polyelectrolyte capsules, Nano Today 3 (2008) 12-21.
[6] Michaeal Gleiche, Observatory Nano, VUI Technologiezentrum GMBH, (April 2010).
[7] http://www.patentstorm.us/patents/7147742/description.html.

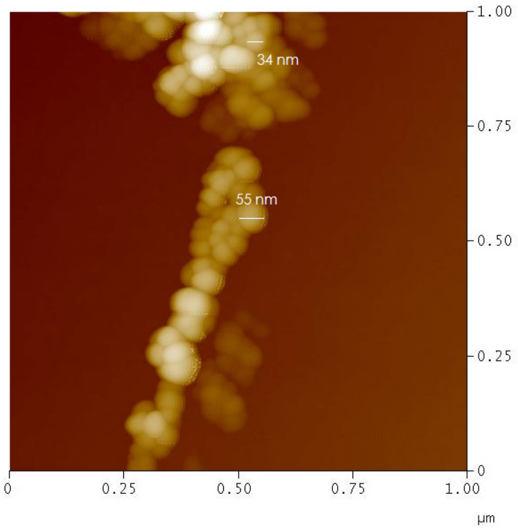

Figure 1. Atomic force image of magnetite Fe_3O_4 particles.

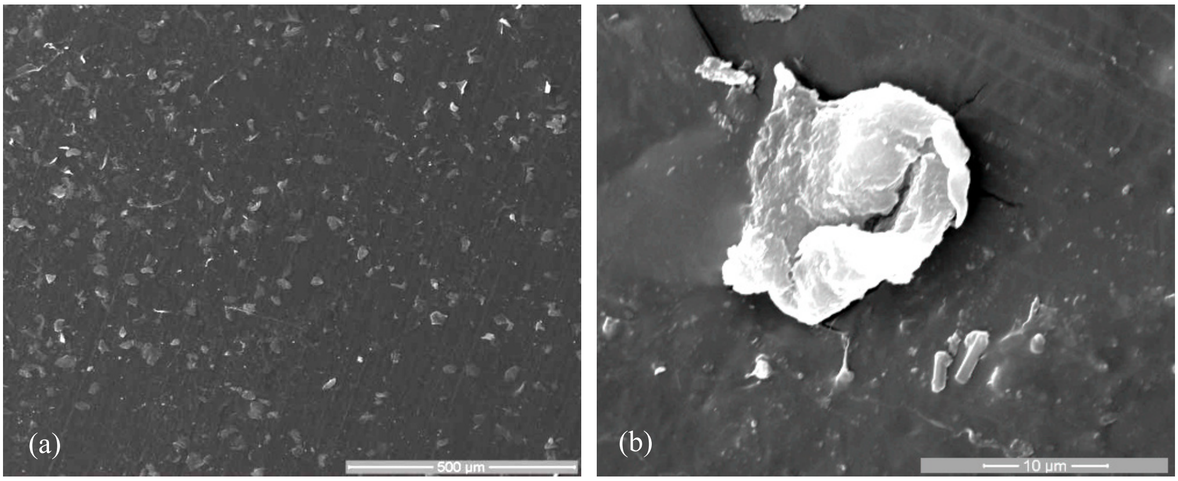

Figure 2. Scanning electron micrograph of the adhesive filled with 5% by weight of magnetite nanoparticles. (a) surface analysis 200x, (b) cross-section analysis 5000x.

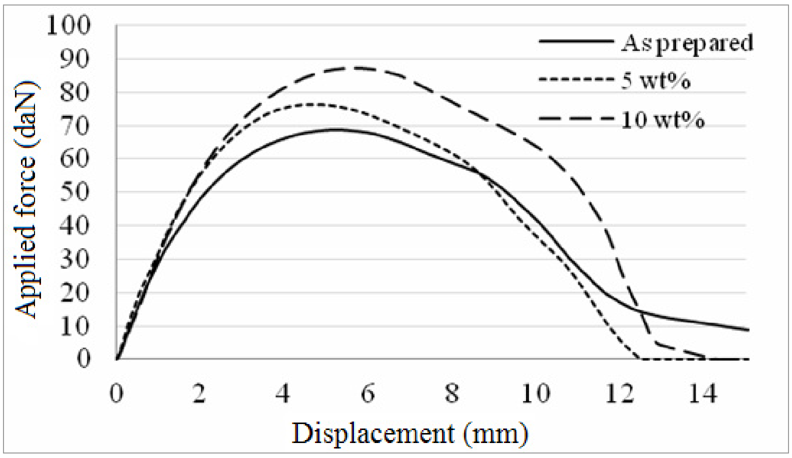

Figure 3. Applied force/displacement curve for SLJ prepared with unmodified and modified adhesive (5 and 10% weight percent of magnetite).

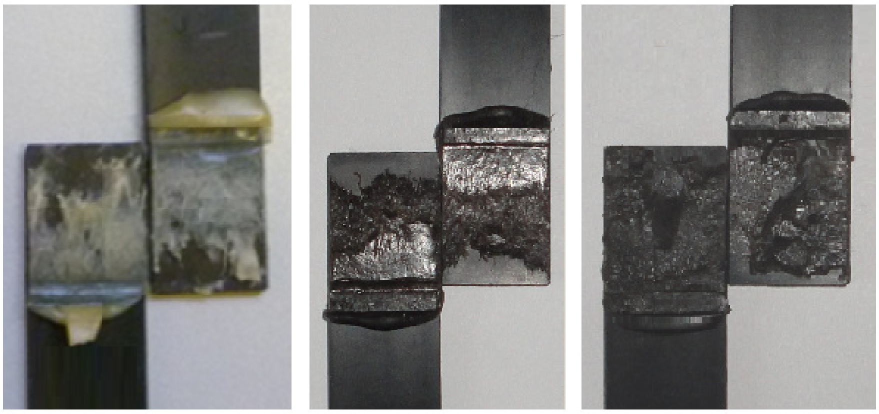

Figure 4. Failure mode of SLJ (a) unmodified adhesive, (b) 5% weight percent of magnetite, (c) 10% weight percent of magnetite.

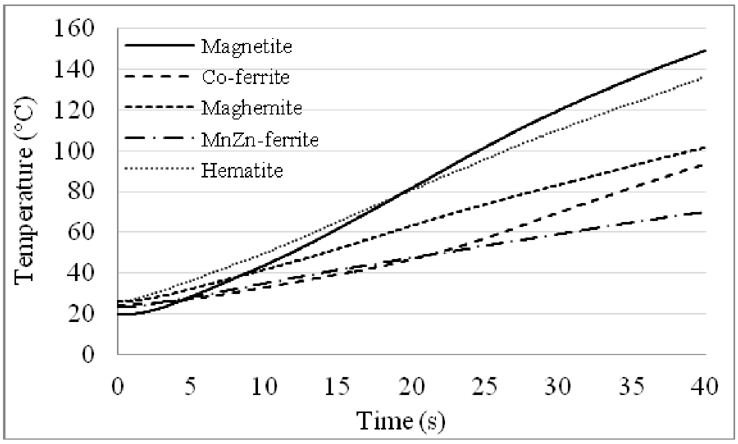

Figure 5. Thermal profile of modified adhesives exposed to electromagnetic fields (172 kHz, 3.3 kW).

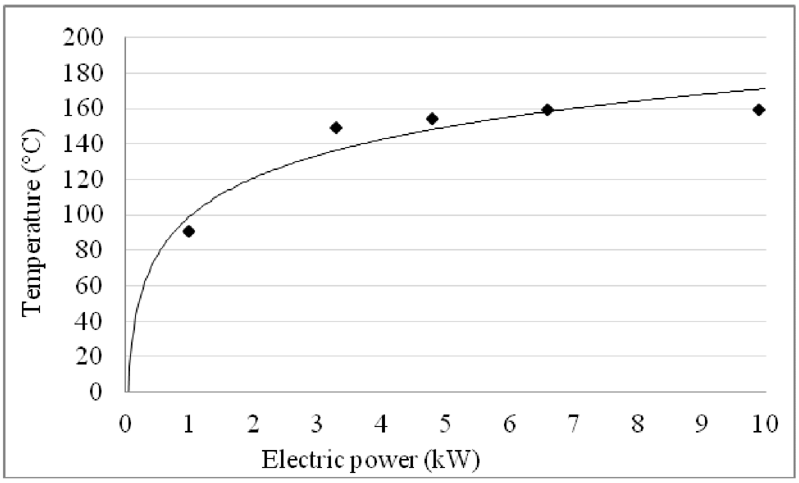

Figure 6. Temperature reached in 40 s vs electric power for magnetite modified adhesive (5 wt%, 172 kHz).

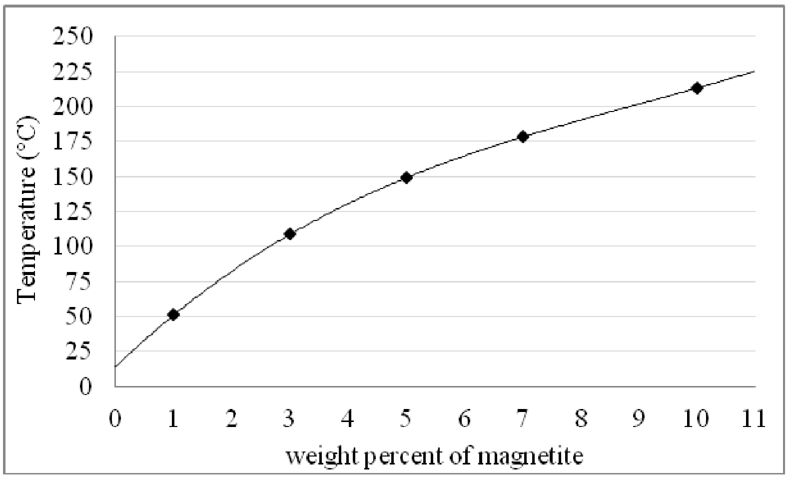

Figure 7. Temperature reached in 40 s vs weight percent of magnetite for modified adhesive (172 kHz, 3.3 kW).

Components	**% (w/w)**
Amorphous –poly-alpha-olefin (*APOA*)	53.20
Styrene-isoprene-styrene block polymer (SIS)	6.60
Polyolefin ester	38.13
Polybutene tackifier	1.73
Antioxidants	0.34

Table 1. Qualitative and quantitative composition of the hot-melt adhesive.

	Shear strenght (MPa)	Standard deviation (MPa)	Shear modulus 0.5-1% (MPa)	Standard deviation (MPa)
As prepared	**1.34**	**0.06**	**17.4**	**1.2**
5 wt % magnetite	**1.54**	**0.04**	**18.4**	**1.7**
10 wt % magnetite	**1.63**	**0.14**	**17.2**	**2.2**

Table 2. SLJs results of unmodified and modified adhesive (5 and 10 weight percent of magnetite).

	magnetite	**maghemite**	**Co-ferrite**	**ematite**	**MnZn-ferrite**
Temperature (°C)	151	138	98	86	67

Table 3. Adhesive heating results at 40 s – 172 kHz, 3.3 kW.

	Disassembling time (s)
5 wt% magnetite	63
10 wt% magnetite	20
30 wt% magnetite	5

Table 4. Disassembling time for modified adhesive (5-10-30 weight percent of magnetite), exposed to electromagnetic field: 172 kHz, 15.8 kW