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DEBONDING OF ADHESIVE JOINTS: EXISTING AND EMERGING TECHNOLOGIES

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Abstract. *Adhesive bonding has found applications in various areas from high technology industries such as aeronautics, aerospace, electronics, and automotive to traditional industries such as construction, sports and packaging. Adhesives can be used to join metals, polymers, ceramics, cork, rubber, and combinations of any of these materials. Moreover, nowadays, new products consist more and more of a combination of special new materials, which needs to be joined according to their specific characteristics. These materials (i.e. ceramics, carbon fibre reinforced polymers (CFRP), etc.) are usually expensive which create an increasing demand for recyclability, driven by economic as well as environmental reasons. Thus, the development of new technologies and processes for easy recycle and repair of bonded structures are becoming of great interest for the industry. If bonds can be broken without damage of the components, recycling is easier. Also for an environmental friendly disassembly of bonded structures, it is necessary to separate the joint between the bonded components so that the different materials can be reused on a qualitatively high level. In this paper, an overview of the existing technologies of debonding adhesive joints is presented. Further, an innovative technique for adhesives debonding by combining the inductive heating method and the use of Thermally Expandable Particles (TEPs) is presented.*

Keywords: *Adhesive joints, thermally expandable particles (TEP), inductive heating, additives, debonding*

1. INTRODUCTION

Adhesive bonding has found applications in various areas from high technology industries such as aeronautics, aerospace, electronics, and automotive to traditional industries such as construction, sports and packaging (Banea and da Silva, 2009). Adhesives can be used to join metals, polymers, ceramics, cork, rubber, and combinations of any of these materials. Moreover, nowadays, new products consist more and more of a combination of special new materials, which needs to be joined according to their specific characteristics. These materials (i.e. ceramics, carbon fibre reinforced polymers (CFRP), etc.) are usually expensive which create an increasing demand for recyclability, driven by economic as well as environmental reasons. Thus, the development of new technologies and processes for easy recycle and repair of bonded structures are becoming of great interest for the industry. If bonds can be broken without damage of the components, recycling is easier. Also for an environmental friendly disassembly of bonded structures, it is necessary to separate the joint between the bonded components so that the different materials can be reused on a qualitatively high level (Banea, *et. al*, 2014a).

For the automotive industry, recent mandatory targets state that all vehicles must be 95% recyclable by the year 2015 (ELV, 2000). Consequently, there is a growing need to develop adhesives that debond on command in order to easy disassembly the bonded structures and to separate parts for reuse or recycling. Currently, debonding of structural adhesive joints is mainly based on mechanical destruction (thermal degradation of the adhesive, cutting of the adhesive or a combination of these methods), which can damage the substrates. Solvents may also be used, but there are health and safety concerns which arise. For example, in the case of disassembly of windscreens and frames in the automotive industry, electric knives are used. This produce significant residue, wastage, and breakage and also raises serious safety issues.

Another demand for adhesives that can debond on command is in applications that need repair or maintenance. This requirement is generally important with products that have a very high degree of complexity or cost such as integrated electronic components, aerospace structures or wind turbine blade components. With adhesives that can be easily debonded, repairs can be made easier and parts can be upgradable.

In this paper, an overview of the existing technologies of debonding adhesive joints is presented. Further, an innovative technique for adhesives debonding by combining the inductive heating method and the use of Thermally Expandable Particles (TEPs) is described.

2. TECHNOLOGIES FOR ADHESIVE DEBONDING ON COMMAND

Two main approaches were investigated in order to obtain adhesives that can be debonded on command: one is to add nanoparticles, micro particles or additives to the adhesives and the second one is to modify the resins with intrinsic backbone features.

2.1 Thermally expandable particles (TEPs) and additives used for adhesive debonding

One of the technologies developed for adhesive debonding on command involves imbedding thermally expandable particles in the adhesive layer. Thermally expandable particles, microcapsules or microsphere, were developed by Dow Chemical Co in the early 1970´s and are particles made up of a thermoplastic shell filled with liquid hydrocarbon. On heating thermally expandable particles, two transformations will occur (see Fig. 1). One is the softening of shell material and the other is the gasification of the hydrocarbon liquid inside it. As a consequence, the shell will expand as the gas inside it will push the softened shell from inside out causing it to grow in size. When fully expanded, the growth in volume of the particle can be from 50 to 100 times. When heat is removed, the shell stiffens and the particle remains in its expanded form. Expansion temperatures range from 70°C to 285°C depending on particle and grade (Matsumoto, 2014). They are commercialized worldwide by the companies Expancel Nobel Industries (Sweden) (Expancel, 2014), and Matsumoto Yushi Seiyaku (Japan) (Matsumoto, 2014).

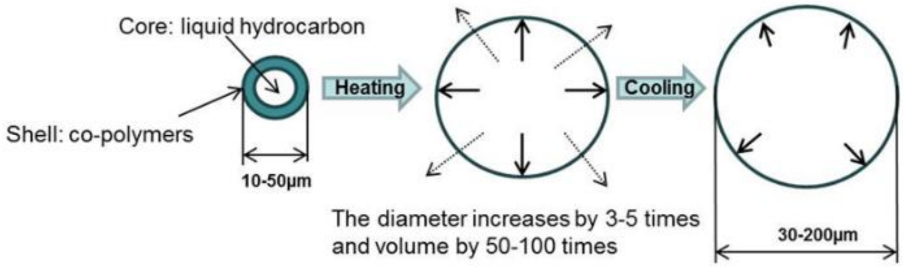

Figure 1. Model of expansion mechanism (Banea, *et. al*, 2014b).

Owing to this unique behavior, TEPs are used by the industry in a wide variety of applications. However, the use of this technique applied to adhesives started in Japan by Sakurai *et al*., 1998 to bond plywood boards. Later, Ishikawa *et al*., 2005 modified and improved the technique for bonding wallpaper on plywood or plasterboards in construction fields. This innovative idea has been extended to structural adhesives for recycling purposes by Nishiyama *et al*. 2003. The simple heating of the joint over 100ºC leads to an easy separation of the bonded materials. The mechanism of separation can be seen in Fig. 2. The adhesive expansion may be up to 400% according to the study of Nishiyama and Sato, 2005. However the high temperature performance was poor because the particles began to expand around 60 °C and the glass transition temperature (T_g) of the adhesive was lower than the expansion temperature of the particles. High temperature performance is vital for structural applications such as automotive and aerospace. To overpass this problem, Kishi *et al*. 2006, used expandable graphite which begins to expand at 200º C with an epoxy adhesive to study de-bond of CFRP and steel plates adhesive joints and found a good dismantlability at 250 °C. However, presently TEPs producers develop particles with higher expansion temperatures. More recently, Kim *et al*., 2012 used TEPs with a polyurethane adhesive and found that the dismantlement of the joint was possible with microwave treatment for 4 min.

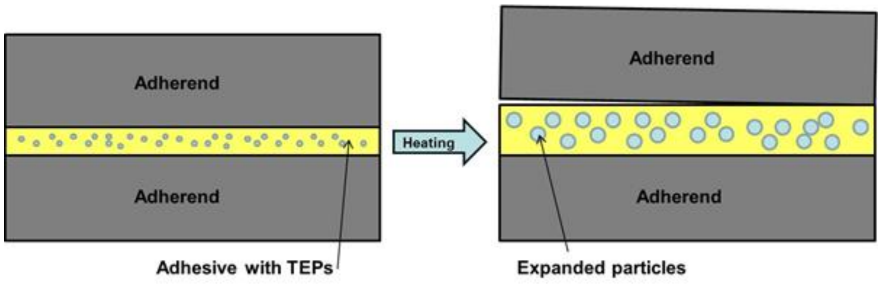

Figure 2. Model of expansion mechanism.

2.2 Nanoparticles used in adhesive debonding

Nanoparticles can also be used in adhesive debonding (Kirsten, 2008). The bonded joint has at least one primer layer which contains nanoscale particles which have ferromagnetic, ferrimagnetic, superparamagnetic or piezoelectric properties. As a result of the alternating electromagnetic fields, large amounts of localised heat are produced in the primer layer of the bonded joint. This localised heat input causes, in the case of adjacent thermoplastic adhesive layers, softening of the thermoplastic binder. If thermoset adhesives are adjacent to the boundary layer of the primer, then the cross-linked structure in the binder matrix is broken up due to the high degree of localised warming. In both cases, quasi-adhesive substrate separation with a low energy input is enabled as a result of the high localised warming of the boundary layer. A similar system have been presented by Kolbe *et al*., 2009 and generally follow the principle of an alternating magnetic field inducing heat to adhesive. However, this approach can be used for any application where at least one non-metal substrate is bonded. Preferably, both substrates should be non-metallic as metal by itself is strongly heated in the magnetic filled and shield the field.

2.3 Electrically disassembly of adhesive joints

Electrically induced debonding of adhesives is a technique in which adhesives can be released at command with help of an applied potential (Haydon, 2002, Leijonmarck, *et al*, 2011, 2012). Depending on the applied potential (10-50 volts recommended) to the adhesive joint, the substrates can be fall apart in seconds. This technology called ElectRelease™ was developed by EIC Laboratories. The joint surfaces must be electrically conductive and kissing bonds must be avoided in order to prevent any short circuit between substrates. However, the technology can be applied to non-conductive or coated substrates using an aluminium foil patch pre-bonded with ElectRelease™. There were patented a number of adhesive formulations (Norberg *et al*, 2008, Gilbert, 2004). Two of these, epoxy-based, are on the market, as E4 and M4 (ElectRelease™, 2014).

3. ADHESIVE DEBONDING BY COMBINING TEPS AND INDUCTION HEATING

In this section, a method to assure easy debonding of adhesive joints by combining the inductive heating method and the use of TEPs is presented. First, single lap joints (SLJs) were fabricated and tested to assess the influence of TEPs content on the lap-shear strength of the adhesive joints. Further, the ability of the TEPs-modified joints to support temperature controlled debonding is evaluated.

3.1 Materials and methods

A two-component structural polyurethane adhesive SikaForce®7888, supplied by Sika (Sika, Portugal) was selected for this study. Expancel 031 DU 40 particles supplied by Expancel Nobel Industries (Sweden) were selected. The diameter of these particles ranges mainly from 10 to 16 µm.

Hard steel substrates characterized by a high tensile strength with a 25 mm width were used. The joint surfaces were grit blasted and degreased with acetone prior to the application of the adhesive. The substrates were bonded and then the specimens were cured at room temperature (RT) for 7 days. A mould with spacers for correct alignment of the substrates was used (Banea *et al*, 2012). The bondline thickness was controlled using packing shims. The bondline thickness was 0.2 mm and the length of the overlap was 25 mm. The geometry of the lap-shear joint specimens used is shown in Fig. 3.

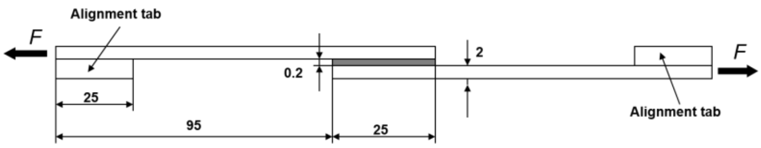

Figure 3. SLJ specimens' geometry.

The TEPs-modified adhesive was mixed with a Speed Mixer (DAC 150.1 FVZ Speedmixer, Hauschild, Germany) for 60 s at 2500 rev/min. This allows an efficient and homogeneous dispersion, creating visibly bubble-free mixing. The TEPs-modified adhesive cured matrix structure was checked through a scanning electron microscopy (SEM) analysis, in a previous study, and a uniform dispersion of particles within the adhesive matrix was observed. Single lap joints with different TEPs concentration by percentage weight were produced.

Testing was conducted at RT at a constant displacement rate of 1 mm/min using a MTS 312.31 servo-hydraulic machine. At least three joints were tested to failure for each wt% TEPs. For each joint tested, a load-displacement curve was produced.

3.2 Evaluation of debonding

The evaluation of debonding was performed by exposing the SLJs' adhesive layer to an electromagnetic field (the SLJs specimens were locally heated by an electromagnetic induction method). The induction heating process relies on induced electrical currents within the material to produce heat. It works directly only with conductive materials, normally metals. The main advantage of the induction heating technique is that heat can be focused at or near the adhesive bondline (see Fig. 4).

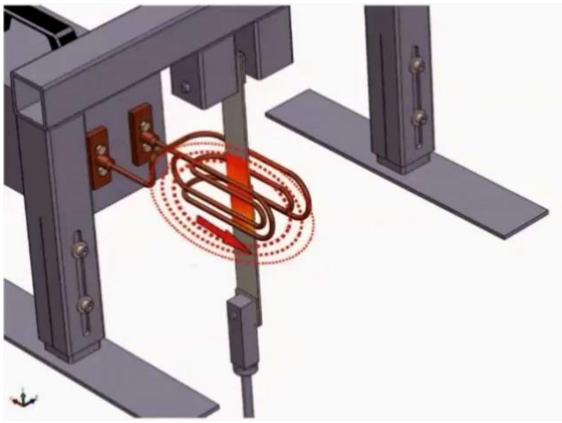

Figure 4. Induction heating process scheme.

3.3 Results and discussion

First, the influence of TEPs content on the lap-shear strength of the TEPs-modified single lap joints was investigated (TEPs-modified SLJs were tested and the results were compared to unmodified SLJs).

Average lap shear strength and failure displacements of SLJs as a function of wt% TEPs are presented in Fig. 5.

The average lap-shear strength is given by:

$$\sigma = P/bL \quad (1)$$

Where P is the maximum load, b is the joint width and L is the joint overlap length.

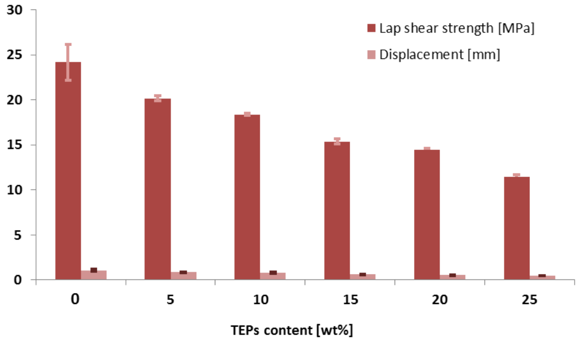

Figure 5. Average lap shear strength and displacement of the SLJs

It can be seen that the lap-shear strength of the hard steel TEPs-modified joints with 5 wt% TEPs decreased by approximately 16 % compared to the unmodified specimens, for SLJs modified with 10 wt% TEPs decreased by approximately 24 %, while tests of SLJs modified with 15 wt% TEPs showed a reduction of the lap-shear strength by approximately 36%. Finally, for 20 and 25 wt% the decrease was by approximately 40 % and 53 %, respectively.

The debonding temperatures as a function of TEPs contents are summarized in Table 1 (debonding temperatures are mean average values, while values in parentheses are standard deviations). It can be seen that for unmodified SLJs, debonding of the adhesive joints is possible only by induction heating, at 230ºC.

Table 1. Debonding temperatures as a function of TEPs contents.

	0 wt%	5 wt%	10 wt%	15 wt%	20 wt%	25 wt%
$T_{deb.}$ (ºC)	230 ± 0	228 ± 2.8	226 ± 3.6	168.3 ± 2.8	143.3 ± 5.7	142.3± 2.5

Furthermore, the introduction of 5 and 10 wt% TEPs into the adhesive matrix shows an insignificant effect on the SLJs debonding temperature, which remained almost the same as of the unmodified adhesive SLJs (~230 °C). Consequently, the debonding also occurred due to the adhesive thermal degradation. For 15 wt% TEPs-modified SLJs the debonding temperature decreased to approx. 170 ºC. Finally, for 20 wt% and 25 wt% TEPs-modified SLJs the debonding occurred at the same temperature (~140 °C) within 80 s. Therefore, it can be concluded that for this particular adhesive system (SikaForce®7888), the optimum conditions for easy debonding are: TEPs content: > = 20 wt% and $T_{deb.}$: > = 140 ºC.

To summarize, if only the induction heating method is used for debonding, the debonding temperature is the temperature at which the thermal decomposition of the adhesive occurs. However, the joint debonding requires high temperatures which can overheat the joint components and even damage or destroy them. Besides, toxic and irritant emission gases are produced due to chemical decomposition. Furthermore, by addition of TEPs the debonding temperature can be lowered by approximately 40% for SikaForce®7888 adhesive TEPs-modified joints.

4. CONCLUSIONS

Structural adhesives that can be debonded or released on command are of great interest for the industry for economic as well as environmental reasons. Easily removable adhesive joints would allow for flexible assembly, easier and cheaper repair, easy upgrading as new technology becomes available and simplified dismantlement and recycling. Some of the technologies presented in this paper are commercially available and are of interest in several markets. However, there is a great amount of developing innovative technology that need to prove its efficiency.

Newly developed smart adhesive technologies and focus from industry on new adhesive products that can debond on command open up new exciting opportunities of development in this field and offer a promising potential in the future.

Debonding of adhesive joints bonded with commercially adhesive systems used in the automotive industry is possible. The weight fraction of TEPs used and the temperature were found to be the major factors in determining the debondability of the joints.

The mechanical properties (tensile lap-shear strength) depends on the TEPs content, decreasing with increasing the TEPs content. However, the joints retain an adequate amount of strength (i.e. more than 60 % of strength (approx. 14.5 MPa)), which can meet, depending on the application, the tensile lap-shear strength requirements of the automotive industry.

The debonding temperature is a function of TEPs content and can be lowered by increasing the TEPs content (i.e. by approximately 40%).

5. REFERENCES

Banea, M.D. and L.F.M. da Silva, 2009. "Adhesively bonded joints in composite materials: an overview". *Proceedings of the Institution of Mechanical Engineers Part L-Journal of Materials-Design and Applications*, vol. 223, no. 1, p. 1-18.

Banea M.D., da Silva L.F.M., Campilho R.D.S.G., Sato C., 2014a. "Smart Adhesive Joints: An Overview of Recent Developments", *Journal of Adhesion*, 90(1), p. 16-40.

Banea M.D., da Silva L.F.M., Campilho R.D.S.G., 2012. "Moulds design for adhesive bulk and joint specimens manufacturing", *Assembly Automation*, 32(3), p. 284 – 292.

Banea M.D., da Silva L.F.M., Carbas R.J.S., Campilho R.D.S.G, 2014b."Mechanical and Thermal Characterization of a Structural Polyurethane Adhesive Modified with Thermal Expandable Particles", *International Journal of Adhesion and Adhesives*, 54, p. 191-199.

Concil Directive (EC) 2000/53/EC of the European Parliament & Council, on ELV, 2000.

Expancel Home page. http://www.akzonobel.com/expancel/, 2013.

ElectRelease™ Homepage. http://www.eiclabs.com/ERhome.htm, 2013.

Gilbert, M.D., 2011. "Electrically disbonding materials". United States EIC Laboratories, Inc. (Norwood, MA, US), US patent no 7968188.

Haydon, D.W., 2002. "ElectRelease - Electrically disbonding epoxy adhesive". *Assembly Automation*, vol. 22, no. 4, p. 326-329.

Ishikawa, H., et al., 2005. "Bond strength and disbonding behavior of elastomer and emulsion-type dismantlable adhesives used for building materials". *International Journal of Adhesion and Adhesives*, vol. 25, no. 3, p. 193-199.

Kishi, H., et al., 2006. "Design of dismantlable structural adhesives with high temperature performance". J. *of the Adhesion Society of Japan*, vol. 42, no. 9, p. 356-363.

Kim, D., I. Chung, and G. Kim, 2012. "Dismantlement studies of dismantlable polyurethane adhesive by controlling thermal property". *Journal of Adhesion Science and Technology*, p. 1-19.

Kirsten, C., 2008. "Process for adhesive separation of bonded joints". Henkel KGAa (Duesseldorf, DE) US patent no. 7407704.

Kolbe, J., et al., 2009. "Curable bonded assemblies capable of being dissociated", Degussa AG (Duesseldorf, DE), Fraunhofer-Gesellschaft zur Foerderung der Angewandten Forschung E.V. (Munich, DE): United States.

Leijonmarck, S., et al., 2011. "Electrochemical Characterization of Electrically Induced Adhesive Debonding". *Journal of the Electrochemical Society*, vol.158, no.10, p. P109-P114.

Leijonmarck, S., et al., 2012. "Electrolytically assisted debonding of adhesives: An experimental investigation". *International Journal of Adhesion and Adhesives*, vol. 32, p. 39-45.

Morehouse, D.S.J. and R.J. Tetreault, 1971. "Expansible thermoplastic polymer particles containing volatile fluid foaming agent and method of foaming the same", US Patent 3615972.

Matsumoto Yushi-Seiyaku Co., http://www.mtmtys.co.jp, 2013.

Nishiyama, Y., et al., 2003. "Dismantlement behavior and strength of dismantlable adhesive including thermally expansive particles". *International Journal of Adhesion and Adhesives*, vol. 23, no. 5, p. 377-382.

Nishiyama, Y. and C. Sato, 2005. "Behavior of dismantlable adhesives including thermally expansive microcapsules, in Adhesion – Current research and application", W. Possart, Editor, Wiley-VCH Verlag GmbH & Co. KGaA: Weinheim.

Norberg P., et al., "An electrochemically weakable adhesive and a laminate structure". 2008, WO/2008/150227.

Sakurai, H. et. al., 1998. "Evaluation of adhesive properties of elastomeric adhesive. The development and utilization of the removable adhesive". *Bulletin of Shizuoka Industrial Technology Research Center*, vol. 43, p. 11-16.

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