Disbonding Technology for Adhesive Reversible Assembly in the Automotive Industry

D.D. Rodrigues[a], P.H. Winfield[b], D. Morrey[c]

Mechanical Engineering and Mathematical Sciences, Oxford Brookes University, OX33 1HX, UK

[a]david.rodrigues@brookes.ac.uk, [b]phwinfield@brookes.ac.uk, [c]dmorrey@brookes.ac.uk

Abstract. Development of the automotive industry is currently driven by three fundamental considerations, i.e. environment, safety and cost, within a strong legislative framework. The reduction of material waste, production stages and weight have become key factors within this scope in the design of vehicles. Therefore, it is important to make greater use of non-conventional materials to take advantage of their recyclability, light weight and mechanical properties, for example new alloys and reinforced polymeric matrix composites (PMC). The dissimilar nature of the materials makes adhesive bonding the principal assembly technique for structural and semi-structural applications. Despite the enhanced performance and durability provided by the use of adhesives compared to that of more conventional joining technologies, bonded materials are very difficult to separate for recycling or reusing components at end of life. Currently, disassembly of adhesive bonded structures is conducted ineffectively by mechanical force, heat, and solvent or acid immersion. Previous research, to overcome these limitations has been mostly for applications other than automotive. Normally, reversible adhesive bonding is obtained through the development of engineered thermoplastic and/or thermosetting resins or incorporation of functional additives into commercial formulations. These technologies generally result in adhesive bonded joints with limited reliability, decreased adhesion strength and reduced resistance to higher temperature. Therefore, no effective disbonding technology has been developed for structural and semi-structural applications for the automotive industry. A comprehensive review will be presented on the adhesive disbonding technology which is currently or intended to be used by industry. This will highlight the advantages and limitations of the various techniques in order to develop an effective disbonding method for the next generation of vehicles at the end of life cycle (ELC).

Introduction

Adhesive bonding has gradually increased over the last 10 years as result of providing faster production, enhanced performance, higher durability, and lower costs compared to those of more conventional joining technologies. However, bonded materials are very difficult to separate for recycling or reusing components from end of life vehicles. This fact has limited adhesive usage in the automotive industry, corresponding to only 5.9% of the global market in 2010 [1]. Adhesives have been used by the OEMs only for non-structural application or as a complementary structural joining technique to welding or mechanical fastening. Epoxy (EP) resins are probably the most valued structural adhesive in industry due to their exceptional mechanical strength and adhesion to a wide range of materials, although these same properties make it difficult to disbond joints on demand.

Current Techniques

Current disbonding techniques for permanent adhesive bonded materials, which can tolerate some damage, are based on mechanical force and heat. These techniques are time consuming, labour-intensive and pose health risks to operators.

Mechanical separation can be conducted in easy access, low modulus/strength adhesive joints by cutting through the bondline using knives, wires or other tools. This approach is generally used to replace windscreens in vehicles. Nevertheless, adhesive residue remains on the substrate after

separation, requiring its removal before reuse or recycling. Mechanical vibration or ultrasound can also be used for disbonding, whereby vibration receptors, ultrasound resonators or piezoelectric additives are incorporated into the bondline [2]. This technique can be optimised by varying the frequency and amplitude of the vibration. A similar approach was successfully conducted using ultrasound harmonics to dissociate carbon-oxygen bonds in an alkoxylated acrylate based adhesive [3]. Another concept is the use of a continuous ply (rubber based material) placed across the bondline centre to acts as a release mechanism by applying a lateral force [4].

Thermo-mechanical disbonding can be attained through heat-conduction, infrared radiation, induction-heating, and microwaves and ultrasonic radiation, followed by mechanical force on the bondline [2]. Disbonding is normally easily achievable with most non-structural adhesive joints by reaching melting temperature (T_m) [5]. Whereas, disbonding of structural adhesive assemblies is more difficult attain through relaxation (T_g) or even thermal decomposition ($>> T_g$) [6], presenting a health risk to the operator [7]. Furthermore, long-term exposure to elevated temperature was found to reduce strength due to loss of dangling chains in the adhesive network [8]. Induction-heating has been used effectively with thermally sensitive substrates or adhesives incorporating ferromagnetic susceptors, through an electromagnetic field. The effectiveness depends on the process (work coil geometry, induction frequency, power density and time), susceptor (electromagnetic nature, geometry, orientation and concentration) and contact between materials. Nevertheless, disbonding depends mostly on the adhesive intolerance to elevated temperatures. Advanced nano-susceptors composed of a single super-paramagnetic iron oxide crystal coated with a thermo-chemically stable amorphous silica matrix (ex: MagSilica) were developed [9] and incorporated in adhesives or primers to achieve separation of bonded joints [5,10]. Joints without heat-induction exhibited strengths comparable to that without nano-susceptors. Similar work done by Oxford Brookes University (OBU) using MagSilica or iron oxides in a toughened EP adhesive was unable to achieve disbonding of GFRP joints even exposing to short-time at temperatures above T_g [2].

Recent Research

Research has been conducted with tailored adhesive formulations to achieve disbonding or loss of strength, despite mostly having been done for applications other than automotive, in particular for small assemblies and specific situations such as electronics. Some EP adhesives have been tailored to retain properties at high temperatures but allowing rework of the joints when exposed to weak acid medium [11]. A study was done on reversible crosslinking EP adhesive using disulfide-containing aromatic and aliphatic amine curing agents [12]. Other work was conducted with an EP adhesive containing a thermoplastic resin possessing full functionality under service conditions but allowing disbonding at elevated temperatures [13]. Various thermo-reversible crosslinking adhesives using dissociation mechanisms were developed, such as, polymer structures with isocyanate-labile hydrogen bonds or divinyl ether and ester groups, elastomer with acid anhydride and hydroxyl reactive groups, diamine cured low modulus EP adhesive containing Diels-Alder adducts, and a hard layered polymer structure comprising branched long carbon backbone polymers with hydrogen or fluorine atoms [2]. Dissociation temperatures depend on the polymer chemistry but can be tailored by modifying the ratio and molecular structure of formula components. Unfortunately, all previous applications are not rigorous for structural applications, although an exception was a specially formulated structural EP adhesive that allows ionic current to pass through the bondline. The use of a potential difference promotes electro-chemical reactions at the interface and consequent disbonding between the conductive substrates [14,15]. The efficiency depends on ionic conductivity of the adhesive, which in the case of EP resins was enhanced with addition of plasticisers. In order to recover the strength losses from the addition of the plasticiser and to improve even further the ionic conductivity, a nano-scale block copolymer was incorporated into the formulation to form a co-continuous network within the EP structure [16]. This mechanism was found to be dependent on the applied voltage and exposure time, temperature, moisture content and pH of the bondline, and surface chemistry, morphology and electrical conductivity of the substrate. Some applications of the technology were patented as release/closing systems and for

recycling. A later study successfully used this technology (ElectRelease) showing delamination, decrease in pH and formation of oxides to sulfur-containing organic film at the anodic interface during voltage exposure [14]. Several debonding mechanisms were considered with a corrosion concept of anodic undermining and mechanical stress from the reaction products at the interface being the most convincing.

Active disbonding can also be achieved independently of adhesive type, using a commercial product loaded with functional additives sensitive to electromagnetic fields, temperature and hot-water immersion. Research has shown micro-encapsulated solvents, acids or bases could be released in the matrix through heat-induction by dissolving slightly polar derivatives, decreasing T_g, promoting bond cleavage and cracks in the matrix [17]. A different approach used expanding inorganic additives for disbonding or loss of strength [18]. Also, thermally labile agents incorporated into commercial adhesives have gained some interest for material separation by generation of a gas, depolymerisation and volume increase at elevated temperatures. Oxidising agents were exploited to obtain self-burning adhesive bondlines through combustion with the oxygen excess formed at elevated temperatures [19]. Other agents crosslinked with EP resin that have also been used to breakdown the polymer network are known as chemical foaming agents (CFAs). Their effectiveness depends on chemical composition, concentration, purity, size and distribution, and exposed time, temperature and surface condition of the substrates [20]. A study done by OBU has successfully achieved interfacial disbonding by incorporating malonic acid into a structural EP adhesive [2]. This is probably as a result of a loss of effective bonded area and/or gases/acids from the thermal decomposition of the additive. Limited durability of the joints was observed when immersed in warm water due to the dissolution of the malonic acid which led to the formation of a porous bondline, reduced effective bond area, lower pH and corrosion of the substrates. This is in agreement with the disbonding mechanism proposed in other work, in which the CFA melts and migrates to the interface before decomposing to gas [21]. Therefore, work was done to encapsulate a CFA in a wax membrane [22]. Physical foaming agents (PFAs) have also been incorporated into adhesives for disbonding to replace/recycle glazing and panels in vehicles [23]. PFAs are microspheres that expand by heat, in which an encapsulated low boiling point hydrocarbon suffers a change in state from liquid to gas inside a polymeric shell. The polymer wall softens as internal pressure of the shell increases during expansion. Commercial products exhibit a tailored expansion ratio and temperature by varying the liquid composition, and the polymer, geometry, size and wall thickness of the shell. Research has mainly been conducted with PUR adhesives bonded to glass and metals, using different grades and concentrations of PFA, heating methods, and testing conditions [23,24]. Moreover, improved effectiveness and speed of disbonding was achieved by coating the PFAs with a thin layer of polypyrrole which provides higher thermal energy absorbance [23]. However, the coating effect on PFA-matrix interactions and the expansion process was unclear, despite some hypothesis and models proposed in confidential reports [25]. Also, internal stress exerted by the PFA was estimated as insufficient for disbonding, suggesting that the thermal energy is also required to soften the adhesive [24].

Methodology

Several approaches will be conducted using incumbent products with additives possessing different functionality and concentration or novel resin adhesive formulations, with the combination of one or more external factors to synergistically initiate disbonding. The requirements will be defined according to the manufacturing processes and materials combinations used and being developed for recycling in vehicles. Initially, the reliability of the disbonding mechanisms will be addressed by evaluating the practicality and optimal conditions. Chemical and mechanical bulk properties of the adhesives will also be obtained. Adhesion strength will be evaluated on the most relevant alloys and composites. Additional mechanical tests will be done after artificial ageing in order to predict durability of the reversible adhesive joints. Furthermore, disbonding mechanisms will be evaluated through the failure surfaces of the joints before and after ageing (Fig. 1).
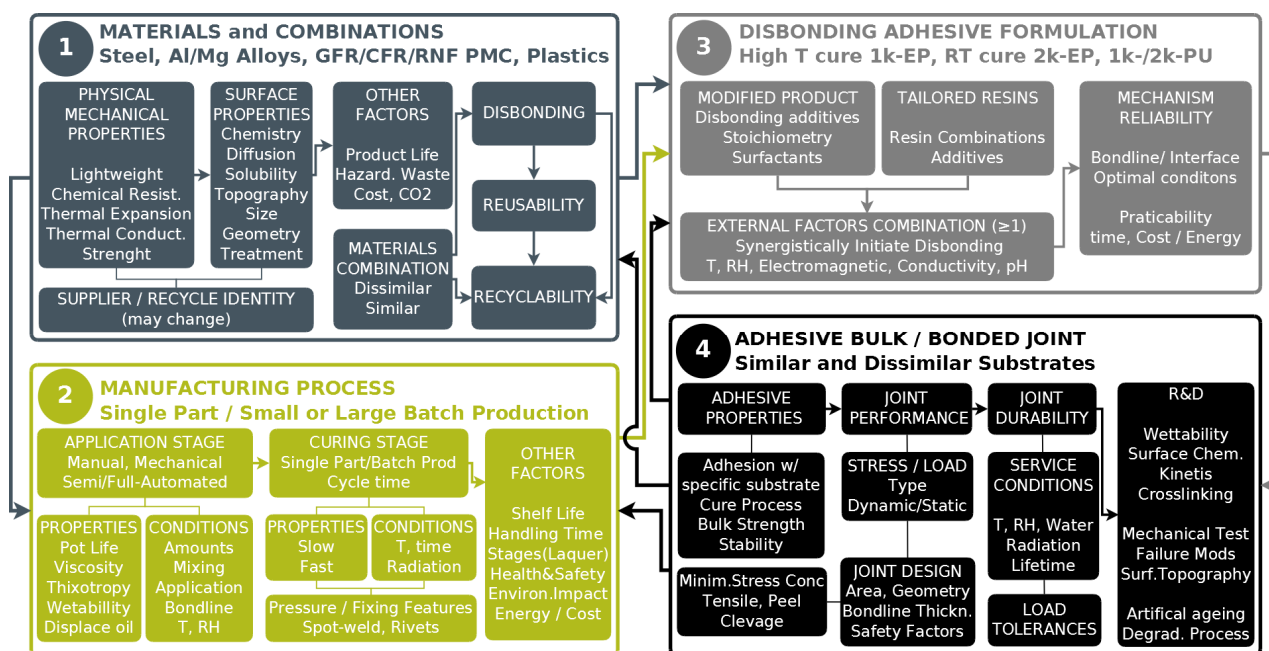
Fig. 1. Methodology to be consider in the whole disbonding system design for Automotive.

Summary of Findings

The technologies described in this paper generally exhibit limited reliability or are not strictly for structural applications, resulting generally in joints with low modulus, decreased strength and low resistance to elevated temperatures. Furthermore, the literature available consists mostly of patents or internal reports with limited information to support the mechanism, such as, failure modes, joint strengths or trigger processes. Non-standard methodologies are also evident which do not allow comparison of the disbonding techniques. Some studies used heat to activate disbonding which led to the formation of toxic compounds or unintentional heating of materials sensitive to elevated temperatures (plastics, PMC or even aluminium). Thus, thermal energy or other external factors may be used only as a complementary technique for disbonding according to the nature of the substrates and adhesive. Also, CFAs seem to have limited application due to their volatile or solubile nature, which can result in a decreased long-term strength or even disbonding of the joints under service conditions. Furthermore, PFAs have restricted application to unfilled and low-modulus adhesives and service temperatures. In the case of the tailored formulations, all are non- to semi-structural adhesives with the exception of the ionic conductive EP adhesive that is confined to metal substrates or simple geometry joints with a two-aluminium foil EP laminate in bondline centre. However, despite none of the technologies described fulfilling all the requirements for disbonding of structural and semi-structural assemblies in the automotive industry, the tailored EP formulations or coated CFAs incorporated into commercial adhesives show some potential for material separation on demand of the next generation of vehicles at the ELC.

References

[1] Markets and Markets, Global Adhesives and Sealants Market by Type, Applications, Trends, and Forecasts, Report Code CH 1638 (2011).

[2] R. McCurdy, Disassembly Methods for Automotive Structures, PhD Thesis, Oxford Brookes University (2011).

[3] D.A. Brown, H.L. Kaufman, U.S. Patent US20050159521 A1 (2005).

[4] E.A. Lehner, F. Stellberger, DE Patent DE10254096 B3 (2004).

[5] T. Sekine, in: IECON Proceedings, Industrial Electronics Conference, 2007, pp. 1357-1362.

[6] C. Kirsten, U.S. Patent US20050039848A1 (2005).

[7] N. Grassie, M. Guy, N. Tennent, Polym. Degrad. Stab. 12 (1985) 65-91.

[8] J. Unsworth, Y. Li, J. Appl. Polym. Sci. 46 (2003) 1375-1379.

[9] M. Kröll, VDI Berichte. 1839 (2004) 3-6.

[10] J. Kolbe, T. Kowalik, M. Popp, M. Sebald, O. Schorsch, S. Heberer, M. Pridohl, G. Zimmermann, A. Hartwig, E. Born, U.S. Patent US0249037A1 (2004).

[11] C. Wong, J. Wu, U.S. Patent US006380322 (2002).

[12] G. Tesoro, V. Sastri, J. Appl. Polym. Sci. 39 (2003) 1425-1437.

[13] J. Licari, G. Bakhit, U.S. Patent US5002818 (1989).

[14] S. Leijonmarck, A. Cornell, C. Danielsson, T. Åkermark, B. Brandner, G. Lindbergh, Int. J. Adhes. Adhes. 32 (2012) 39-45.

[15] M.D. Gillbert, U.S. Patent US7465492B2 (2008).

[16] C. Sato, H. Shiote, M. Ohe, in: 11th Triennial Conf. Sci. Technol. of Adhes. Adhes., 2011, pp. 103-106.

[17] P. Becher et al, U.S. Patent US0067645659B1 (2004).

[18] K. Kawate, T. Kanki, INT Patent WO 00/40468 (2000).

[19] Y. Okada, K. Shirota, U.S. Patent US003993524 (1976).

[20] E. Langkabel, M. Bauer, G. Wachinger, A. Concord, VDI Berichte (2008) 405-406.

[21] M. Olive, P. Dalet, E. Papon, European Space Agency, ESA SP. (2006) 616.

[22] J. Alcorta, E. Papon, P. Dalet, INT Patent WO/2006/048585 (2006).

[23] A. Schmid, L.R. Sutton, S.P. Armes, P.S. Bain, G.Manfre, J. Soft. Matter. 5 (2009) 407-412.

[24] G. Manfre, Phenomenological View, Hypothesis and Models, Classified Rep. by De–Bonding Ltd (2005).

[25] C. Sato, Y. Uratani, in: 4th Congress Adhes. & Relat. Phenom. Abstract Booklet (2010) 137.

Light Metals Technology 2013
10.4028/www.scientific.net/MSF.765

Disbonding Technology for Adhesive Reversible Assembly in the Automotive Industry
10.4028/www.scientific.net/MSF.765.766

DOI References

[7] N. Grassie, M. Guy, N. Tennent, Polym. Degrad. Stab. 12 (1985) 65-91.

http://dx.doi.org/10.1016/0141-3910(85)90057-6

[8] J. Unsworth, Y. Li, J. Appl. Polym. Sci. 46 (2003) 1375-1379.

http://dx.doi.org/10.1002/app.1992.070460806

[12] G. Tesoro, V. Sastri, J. Appl. Polym. Sci. 39 (2003) 1425-1437.

http://dx.doi.org/10.1002/app.1990.070390702

[14] S. Leijonmarck, A. Cornell, C. Danielsson, T. Åkermark, B. Brandner, G. Lindbergh, Int. J. Adhes. Adhes. 32 (2012) 39-45.

http://dx.doi.org/10.1016/j.ijadhadh.2011.09.003

[23] A. Schmid, L.R. Sutton, S.P. Armes, P.S. Bain, G. Manfre, J. Soft. Matter. 5 (2009) 407-412.

http://dx.doi.org/10.1039/b811246k