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OVERVIEW OF DISBONDING TECHNOLOGIES FOR ADHESIVE BONDED JOINTS

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Abstract

The ability to separate adhesive bonded assemblies without causing damage to the substrates is clearly very desirable. There are many applications such as in electronics, medical surgery, dentistry, building and general manufacturing where the opportunity to separate assemblies is important. This may be for repositioning in manufacturing, repair in service or recovery of materials at end-of-life. Various methods for adhesive reversibility or *disbonding* have been proposed over the last 40 years but there currently exist no universally accepted solutions for *disbond-on-demand* bonded applications. This paper considers the motivation for *disbonding*, the

requirements and considerations associated with possible methods, and the overall effectiveness of the various mechanisms in the context of non-structural, semi-structural and structural joints. The range of technologies and mechanisms is reviewed, together with the associated methods for activation. The variety of methods is evaluated for their effectiveness in the context of different applications. Particular attention is given to the adverse effects on the performance of bonded assemblies in service, and the ways of mitigating these effects. It is shown that a total materials system approach must be adopted when seeking a debonding technology for a particular set of circumstances.

Keywords: debonding; environmental issues; recycling; applications

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1 INTRODUCTION

The purpose of this paper is to provide an overview of disbonding technologies for adhesive bonded products in the context of sustainable manufacturing and assembly processes. It is now incumbent upon all engineers and technologists to make provision for methods to facilitate dismantling and disassembly of products that they design and specify. The additional imperative is driven by the variety of high value materials that are now commonplace in everyday products.

Contemporary approaches to engineering require whole-of-life thinking whereby materials, design methods and assembly techniques must be selected against many different criteria. From a manufacturing standpoint, assembly processes should be chosen that facilitate re-positioning and re-working. From an operational viewpoint, components should be designed to enable them to be repaired or reconditioned in service. Finally, from an end-of-life perspective, components and structures should be designed with materials recovery in mind (Fig1), to facilitate the reuse and recycling of increasingly valuable resources [1,2].

Many industries have adopted whole life thinking to a greater or lesser extent because it is good practice, it makes sound economic sense, waste streams are reduced, and a supply of materials for future use is guaranteed. Net energy inputs and outputs are reduced, leading to reduced emissions, and the use of primary materials is reduced in line with thinking behind the circular

economy. For example, the EU End of Life Vehicle (ELV) Directive [3] places emphasis on the recovery of materials (up to 95%) and provides a good example of how more sustainable materials supply chains may be obtained by making the producer legally responsible for the product.

Adhesive bonding represents a mature joining technology that is available to engineers and designers. The general advantages of adhesive bonding are well documented, but one of disadvantages is the relative permanence of joints when compared, for example, to mechanical fastening methods. This presents issues for repositioning in manufacturing, for repairs in service and for materials recovery at end of life. Current disassembly practices for an adhesively bonded structure are mostly labour-intensive and inefficient, ranging from mechanical cutting to thermal degradation. Where complex geometries are bonded, knives, scrapers, and wires are used, resulting in high costs and a large amount of adhesive residue still present after separation. This residue makes it difficult to re-use substrates. There is a clear need to address the separation of adhesively bonded structures whilst preserving all salient aspects of adhesive application, performance in service and joint integrity.

Methods to achieve adhesive bond separation, using a combination of heating and bondline susceptors, originated in the shoe industry in the 1970s. A system was devised to work for epoxies, cyanoacrylates and rubber-based adhesives that enabled the bond between a shoe sole and leather upper to be sufficiently weakened to enable re-soling [4].

1.1 Terminology

A detailed consideration of this subject requires some common definitions to be adopted:

- Disbonding – the separation of two or more surfaces that were previously united by an adhesive layer
- Disbond-on-demand – the ability to separate surfaces that were previously bonded together at a time and in a manner that is controllable
- Dismantling – the process of separating the components of a joint
- Disassembly – the opposite of assembly, implying the process of separating components that have previously been joined together
- Reversibility – the ability to reverse a process, such as being able to separate components of a joint and then re-assemble it.

Various methods for adhesive reversibility or *disbonding* have been proposed over the last 40 years but very few of these systems have found a universally marketable application.

1.2 Requirements of methods

A technology is required that is low cost, straightforward to implement and works quickly. Ideally it is something that can be introduced within existing adhesive systems, rather than needing to purchase a bespoke bonding system sourced from a particular manufacturer. It is desirable that clean

separation of substrates can be achieved without adhesive residues remaining on the substrates, leaving at least one clean surface. It is also desirable that minimal damage is caused to the parent materials and to their surroundings, because this allows high value materials to be recovered and retain their intrinsic value. The process of disbonding would ideally involve minimal energy input and be achieved with no hazardous by-products.

Any disbonding technology has to remain stable in the component under the service condition of the component and work, perhaps many years after the bondline was created. The disbonding method should be compatible with the cured/uncured adhesive system and yet have minimal effect on the performance of bonded joints in service. It will be appreciated that it is very difficult to satisfy all of these requirements.

1.3 Considerations

The starting point is whether bonded assemblies are considered to be structural, semi-structural or essentially non-structural. Naturally, this dictates the type and performance requirements of the adhesive system selected. The overall scale of the component parts of the joint is significant because the chosen disbonding system must consider the size, cost and accessibility of the bonded assembly. The type and nature of the substrates represent very important considerations because they may dictate access to bondlines,

physical barriers or may restrict energy transmission to activate a disbonding process.

If a disbonding technology is to be incorporated into an existing system or process, there may be many aspects of compatibility with the chemistry or with the rheology of the mixed materials to appraise prior to application, with associated effects on the adhesive application characteristics. There may be issues with primer layers, and there may be effects on the bulk adhesive properties.

The energy sources that trigger disbonding generally involve heat to stimulate a chemical reaction, or a physical change of some sort, to initiate damage in the adhesive or interface layers that weaken the bond and lead to joint separation. Other triggers include radiation and electrical currents.

Where heat energy is used, the required disbonding temperature is very important. Cure temperatures may also be very significant, because some chemical or disbonding reactions could be initiated during the normal curing process. Likewise, the upper service temperature of the bonded assembly is also important to avoid any unwanted initiation.

A most important consideration is the method of activation of the disbonding process. The normal stimuli are energy sources such as heat or ultra violet light, but electrical conduction also features in some systems. Safety is an important consideration here. On a practical point, bondlines must be

accessible to the activation process and this may dictate the physical design of the joint.

1.4 Effectiveness

The effectiveness of a disbonding procedure is hard to quantify. There is a difference between making a bonded assembly relatively weak, and causing a bonded assembly to separate spontaneously but completely. In the former case, some additional physical force is required to achieve separation and the disbonding effectiveness can be assessed by comparing the joint strength before (τ) and after (τ') activation (Eqn. 1):

$$E_{disb} = \frac{\tau - \tau'}{\tau} \dots\dots\dots (1)$$

Effectiveness may also be considered in the context of time to separation, whether clean separation can be achieved, the amount of material and energy required for separation, or how difficult the process of separation is to achieve.

A further measure of effectiveness is what compromises need to be made. These may be design changes to joints to enable an activation process to take place, they may be acceptable reductions in bondline properties throughout the service life of joints, or they may be adverse effects on the environmental durability of bonded assemblies.

One key question to be answered is whether heat alone would achieve the same effect as the introduction of a sophisticated disbonding technology. This question can only really be answered by practitioners working in different industries.

2. DETAILED CONSIDERATIONS

A primary consideration is the loading and service conditions that a bonded assembly is required to satisfy. Adhesive joint requirements can be broadly classified as structural, semi-structural and non-structural, reflecting the nature of anticipated loading, service environment and design life. These factors are crucial when contemplating changes to performance requirements, such as the need to accept lower joint strengths or reduced durability as a result of a system that incorporates disbonding triggers. Some examples of joint types and requirements are shown in Table 1.

When expandable fillers are incorporated into an adhesive layer, rather than in a primer layer, the volumetric expansion (or pressure) exerted on the bondline must be sufficient to overcome the elastic modulus or toughness of the adhesive [5,6]. The requirements for a structural adhesive are depicted schematically in Fig 2. This shows that, ideally, a viscoelastic material with a suitable glass transition temperature (T_g) is required, whose modulus is sufficiently high for structural application under the working environment and

drops quickly to the rubbery state when heated to a certain temperature to allow expansive pressure to develop.

The considerations related to the mechanical, chemical and thermal stabilities of any additive should also be addressed. In the cases of adverse reactions that may occur between the adhesive and foaming agents, microencapsulation can be considered. A number of physical or chemical encapsulation methods can be used to protect the foaming agent from being reactive with the adhesive matrix or with the service environment. Similarly, the adhesive matrix may need to be protected from the release of chemicals from any additive, such as the acid from expandable graphite.

Other considerations include the substrates' mechanical properties, thermal stability, permeability, ionic conductivity, electrical conductivity and optical transparency. These may, individually or in combination, be important for the selection of particular debonding methods; they are discussed in the context of the different techniques outlined later.

3. DEBONDING TECHNOLOGIES AND MECHANISMS

AVAILABLE

An overview of the different technologies and associated mechanisms available is shown in Fig. 3. A broad classification may be summarized as:

- Destructive approaches
- Tailored adhesive formulations

- Functional additives
- Active substrates and primers.

3.1 Destructive approaches

The most primitive solution for disbonding includes mechanical separation with or without heating and solvent immersion. These conventional methods are cost-effective but inefficient and labour intensive. For windscreen replacement applications in service, it is common practice to use triangular-section wire to cut through the tough polyurethane sealant and replace the glass. In practice some vehicles are designed better than others to facilitate this procedure, to minimise damage to the interior trim. More delicate techniques are required for polymer composite-intensive future vehicles, where the windscreen and other non-opening glazing is present, because of the potential for significant damage to the surface layers of fibre-reinforced plastics. BMW have devised a system for their i-brand vehicles that employs a strong polymeric wire, akin to fishing line, to execute the procedure [7] (Fig.4).

The challenge of automotive glazing replacement using non-destructive means was addressed by Manfre and Vitkala [8], who investigated the incorporation of thermally-expandable microspheres in the primer layer on the metal. Elements of the system were later patented and developed commercially by Autoglass, a commercial windscreen replacement organisation.

Thermally induced disassembly approaches can be achieved through both thermal softening (exceeding the adhesive's T_g) and thermal decomposition (exceeding the temperature of flammability-in-air or auto-ignition point). For the latter, the major concerns are the toxic and irritant emission gases that are produced as a result of chemical decomposition [9].

Certain chemical degrading agents [10], solvent or acid immersion techniques, can also assist the adhesive disassembly process. Polar solvents such as isopropyl alcohol (IPA), methyl ethyl ketone (MEK) and acetone, can be very effective in cleaning and degreasing substrate surfaces and are mainly used in surface treatment processes [11,12,13].

3.2 Tailored adhesive formulations

These are sometimes referred to as re-workable adhesive systems. This generally means that adhesives are formulated to reach their glass transition temperature at a specific point, reducing bond strength and enabling components to be separated with gentle force. The electronics industry has conducted much research on disassembly methods, to enable rapid removal of bonded chips at end of life [11,14,15]. The Battelle Memorial Institute [16] developed a thermally reversible isocyanate-based polymer formulation, which was based on the dissociation of the isocyanate-labile hydrogen-based linkage to the isocyanate and labile-hydrogen starting groups. Upon disassociation, this polymer was found to become a free flowing melt, which

was soluble in acids. A similar approach was developed for different chemistries by Atochem [17] and Yokohama [18].

Sandia National Laboratories [19] developed a thermally removable adhesive by incorporating thermally reversible furan-maleimide Diels-Alder adducts which form below 60°C and dissociate above 90°C in a low modulus epoxy adhesive formulation. The relatively low dissociation temperature and weak mechanical properties exhibited by lap shear joints, of around 4MPa, rule out its use for semi-structural and structural applications. Nevertheless, the reversible Diels-Alder adduct approach has been further investigated by others [20, 21].

DC Polymers [22] adopted electrochemically active cross-linkers, which can be incorporated into the polymer structure. Electrochemical reduction can be triggered by applying an electric current. This leads to scission of the polymer backbone at cross-link sites and thereby polymer degradation. Since crosslinking is quite common to all types of polymers, this technology can theoretically be applied to a wide range of polymeric materials. To date, no mature product or experimental data has been published.

A variation on reworkable adhesive systems is the ability to change the polymer cross-link density through UV irradiation and modest temperature. Sato et al [23] developed an acrylic polymer for use in pressure-sensitive adhesives that responded to these energy stimuli, resulting in modulus increases to the adhesive. When subjected to peel forces, the bonded joints failed rapidly at the adhesive/substrate interface. Akiyama et al [24] induced reversible isothermal phase changes from solid to liquid using UV radiation.

They investigated the hot-melt adhesion between polyamide blends and glass substrates and demonstrated a disbonding mechanism suitable for bonding of glass substrates.

3.3 Functional additives

The use of additives to tailor the properties of an adhesive under certain conditions has led to many different systems being proposed for reversibility. These additives fall into the groups of chemical foaming agents (CFAs) and physical foaming agents (PFAs). CFAs describe any functional additive that exhibits a volumetric expansion from its initial starting material, solely by undergoing a chemical reaction. The generic term PFA describes a foaming agent that undergoes a volumetric expansion due only to a change in state of the initial starting material. The principle in both cases is to expand the bondline and/or chemically change the adhesive material (Fig. 5).

The early work on shoe re-soleing [4] relied upon the incorporation of metallic inclusions into the adhesive to efficiently absorb and emit heat energy when subjected to alternating electro-magnetic field, thereby activating the release of micro-encapsulated solvents, which would further degrade the polymeric structure. Daimler-Chrysler and partners [25] patented similar work and specified examples of encapsulated solvents as organic amines or acids which can cleave an epoxide backbone. Microcapsules were recommended to be made of amino resin or metal halide. Obediat [26] reviewed recent

patented works of different microcapsule preparation methods used in the pharmaceutical industry. The Nissan Motor Company [27] mixed oxidising agents (ammonium perchlorate, potassium permanganate, etc.) into the adhesive matrix which produced a self-burning reaction at elevated temperatures. The decomposition process generated abundant oxygen and caused failures in the bondline.

Henkel [28], IBM [29], the US Army Research Laboratory [30] and Rescoll [31] investigated a series of CFAs including Azo compounds, and hydrazides. The technology developed and patented by Rescoll is called INDAR Inside [32] and a number of practical applications have been documented [33,34].

Different types of foaming agents were incorporated into adhesive systems to promote disbonding at elevated temperatures. It was noted that CFAs would become mobilised at higher temperatures when the adhesive bondline softened and melted [35,36]. Some CFA particles or microspheres would then migrate and finally decompose at the joint interface.

Functional additives, incorporated into adhesive bondlines, have been investigated by many other researchers [37,38,39]. McCurdy et al. [37] investigated the effect of the incorporation of functional additives on the dismantling characteristics of three adhesives: two fracture toughened epoxies and one semi-structural polyurethane. The functional additives selected were four types of chemical foaming agents (CFA): p-toluenesulfonyl hydrazide (pTSH), benzenesulfonyl hydrazide (BSH), azodicarboxamide (ADC) and 5-phenyl-1H-tetrazole (5P1HT). No discernible benefit was observed from the experimental data of CFA incorporated into the semi-

structural adhesive. However, the structural adhesive systems were found to be promising but still showed problems of additive/adhesive incompatibility, which therefore affected the long-term durability. More importantly, additive-matrix incompatibility issues were encountered in this research.

Thermally expandable additives include inorganic materials such as dilated graphite, vermiculite, pearlite or mica. Early work patented by 3M [40] used an expandable additive content of 10 to 50wt% with an initiation temperature between 250°C and 500°C.

Expandable additives have been used for debonding in orthodontic applications. Conventional techniques for the removal of orthodontic attachments carry risks of tooth enamel damage. The use of electrothermal, laser or ultrasonic methods may incur other problems such as expensive equipment cost, potential pulpal damage and mucosal burns [41,42].

Tsuruoka et al. [43] incorporated thermally expandable microspheres (TEMs) into 4-META/MMA-TBB (4-methacryloxyethyl trimellitate anhydride in methyl methacrylate initiated by tri-n-butyl borane) resin adhesive, which is widely used for bonding orthodontic brackets. The microspheres used in this study were Matsumoto F-36D which starts to expand at 80 °C. They identified that an incorporation of 40wt% of these microspheres showed satisfactory bond strength before heating and a reduced bond strength by a third within 8s of heating. The Tokyo Institute of Technology [44, 45] also conducted research on debonding using expandable microspheres manufactured by Matsumoto. These studies additionally looked at its application for construction materials. The weight fraction of microspheres used and the roughness of substrate

surfaces were found to be two major factors in determining the dismantlability of the bonded joints.

PFAs are also known as thermally expandable microspheres (TEMs). They generally consist of a thermoplastic polymer shell encapsulating a hydrocarbon blowing agent with a low boiling temperature. These microspheres are typically 10-50 μ m diameter, expanding to 40-60 times greater than their initial volume (Fig. 6). TEMs were first developed by Dow Chemical Co. and are currently manufactured by many companies such as Polychem Alloy, Sekisui Chemical, Matsumoto Yushi Seiyaku, Akzo Nobel and Sigma Aldrich.

Much research has identified additive-matrix incompatibility and noted that the incorporation of additives affect mechanical properties and durability by acting as weak links in the matrix or, in some cases, reacting with the matrix. Jonsson et al. [46] successfully modified TEMs by grafting glycidyl methacrylate (GMA) on the surface. Atom transfer radical polymerization using activators regenerated by electron transfer (ARGET ATRP) was employed. Special treatment was also used to retain the expansion ratio of the TEMs. A similar approach was taken by Lu [39] who demonstrated the effectiveness of grafting (Fig.7) on the superior strength and durability of epoxy-bonded joints. The ARGET ATRP surface modification treatment given to the TEMs promoted covalent bonding between the adhesive matrix and the thermoplastic surface, successfully solving the environmental durability problem.

De-Bonding Ltd and Evonik focused on thermally-activated disbonding of automotive glazing on by incorporating TEMs in the primer layer of the tough sealant [8,47]. An infra-red lamp was adopted as the heating source. Their research [48] successfully synthesised polypyrrole-coated TEMs; polypyrrole has a broad absorption peak (900-1500 nm) in the infra-red. This provides the polypyrrole-coated TEMs with a very efficient thermal energy absorbance. Experimental results showed great potential for disbonding of automotive glazing and other vehicle panels.

Expandable graphite (or intumescent flake graphite) represents an interesting type of inert, inorganic, layered particle that can be triggered by heat. An acid is intercalated into the crystal layers of the graphite and vaporization of this compound triggers exfoliation of the flake. Expandable graphite (EG) is often used in fire-retardant applications of coatings and sealants. The flake dimensions determine the expansion but this material has the potential to expand many times more than typical thermoplastic microspheres, albeit at a higher temperature (Fig.8).

Some applications of adhesives involve relatively high curing temperatures, e.g. 180°C in the car industry, which rules out the use of CFAs and PFAs. However, inorganic additives are suited to these temperatures. This was recognised by Kishi et al [6,49] and further investigated by Sato [50] and by Pausan et al. [51]. In addition to the higher activation temperature, major advantages are that the additional volumetric expansion enables graphite to be effective at low concentration levels and more effective than TEMs in tough polymeric matrices.

3.4 Active substrates and primers

A series of electrically-reversible amine-cured epoxy adhesives was developed by EIC Laboratories under the trade name of ElectRelease [52]. Disbonding is achieved when a potential difference of 10-50V, and a current of $1-5\text{mAcm}^{-2}$, is applied between electrically conductive substrates (Fig. 9). The disbonding mechanism is achieved through ion conduction along the resin-metal interface, resulting in polarizing of the adhesive boundary layer surface.

The disbonding needs a metal as the positive substrate and another suitable material for the negative substrate. When two metal substrates are bonded using ElectRelease, disbonding at both interfaces can be achieved by reversing the polarity of the disbonding voltage. For non-conductive materials, an ElectRelease foil patch (EFP), which is essentially two sheets of metal foil, is applied on the surface to enable electric conductivity [53]. Leijonmarck et al. [54] investigated the ElectRelease technology and provided further understanding of the disbonding mechanisms. They observed a delamination process at the interface between the aluminium anode and the adhesive layer, detected changes in polymer chemistry using Raman spectroscopy and also analysed the emission of volatile species using mass spectrometry.

Evonik developed Magsilica, a type of adhesive tape [55]. Magsilica is used as a nano susceptor, which is essentially a powder of iron oxide particles embedded in silicon dioxide nanoparticles. These particles behave as nano magnets when subjected to an external magnetic field, which heat up for the purpose of curing and disassembly. This provides two major advantages:

shortened cure times and debonding. A major concern is that conductive or magnetic components can cause electromagnetic shielding, which deactivates the heating mechanism.

Debonding can also be achieved by adding expandable additives in adhesive system primer layers, if present. This is a very effective location for separation. The modified primer expands and separates the adherends upon heating, leaving a clean surface [56].

4. SUMMARY OF EFFECTIVENESS OF DEBONDING TECHNOLOGIES

The research activities of various authors related to different debonding technologies, adhesive/substrate systems and effects are collected in Table 2. Any comparisons are necessarily rather superficial because different materials, adhesive systems and test procedures were involved. The term 'initial strength' refers to the short-term quasi-static joint strength, whilst 'long-term durability' indicates that the authors subjected bonded test joints to some sort of environmental exposure involving water.

There are some data, reported by a succession of researchers at Oxford Brookes University, that do invite comparison because of the commonality of adhesive, additives and similar test procedures. They all used 3M 9323 epoxy adhesive, a commercial two-part formulation, cured at 90°C. They also used single lap joints made with 2mm thick EN AW 6082 T6 aluminium substrates. McCurdy et al. [37] compared different % wt. additions of different types of

CFAs and PFAs (or TEMs). Lu et al. [39] applied a surface modification technique to TEMs to improve adhesive compatibility and matrix stability. Pausan et al. [51] examined the use of expandable graphite (EG) as an alternative functional additive.

For the PFAs, McCurdy and Lu adopted a 20% wt. addition of TEMs, citing this as the optimum addition level to provide effective disbonding. McCurdy et al. [37] employed a 551 DU40 grade whilst Lu et al. [39] employed a 920DU 120 grade, both supplied by Expancel Akzo Nobel. The blowing agent in the former is isobutane and in the latter, isooctane and isobutene. Disbonding effectiveness was assessed by measuring the residual joint strengths following a short period (1-2 minutes) of heating at 250-270°C. Pausan et al. [51] adopted 3% wt. addition of EG and measured the residual joint strengths after heating to 235°C for 1 minute. A detailed discussion of their results can be found in the original papers but a broad comparison of the lap shear behaviour of unaged joints is shown in Fig. 10. The addition of the CFA had an immediate and very detrimental effect on the joint strength. The 20% addition level of PFA was detrimental, but mitigated somewhat by surface modification (retaining 70% of the initial joint strength). The residual strengths after heat activation were around 10% of the original joint strengths. The joints with a 3% wt. addition level of EG retained 87% of their initial strengths and were left with no residual strength following heat activation.

The mechanical performance of adhesives, and of joints made with them, is known to be affected by the presence of water. In the short term, water

absorbed by the adhesive layer will plasticise it and this may lead to a slight increase in joint strength. In the longer term, water is able to displace adhesive bonds which, in turn, leads to joint strength degradation. Pausan et al. [51] used lap shear joints immersed in water for 90 days at 55°C, and wedge cleavage joints immersed in water at 55°C for 7 days. The presence of 3% wt. EG led to a 22% loss of lap shear strength but an increase in the fracture toughness of the joints. Banea et al. [38] reported a similar beneficial toughening effect associated with a small percentage of additive particles. McCurdy et al. [37] used wedge cleavage joints immersed in water for up to 30 days and found that the presence of TEMs severely weakened the bondline and reduced the joint fracture toughness.

Lu et al. [39] examined the effect of ARGET ATRP surface modification of TEMs. It was found that surface modification of the TEMs substantially improved the compatibility with the epoxy adhesive by the formation of strong covalent bonds. Correspondingly, the modified TEMs system had superior toughness and moisture resistance compared to the adhesive containing unmodified TEMs. Most significantly, the bulk tensile strength of the adhesive system containing modified TEMs after environmental conditioning was 8% higher than the strength of adhesive system containing unmodified TEMs before environmental conditioning (Fig. 11).

McCurdy et al.[37] adopted the same test procedures for a particular polyurethane adhesive but he was not able to achieve any significant separation of the joints by incorporating CFAs of PFAs; the weakening that

occurred was attributed to the effect of heat alone on the adhesive material. In essence, the volumetric expansion of the CFAs and PFAs was not sufficient to overcome the modulus of the polyurethane adhesive. On the other hand, Pausan et al. [51] used the same combination of experimental parameters (as for the epoxy) in relation to a polyurethane adhesive with 5% wt. EG. Again, they used lap shear joints for control tests and wedge cleavage joints to examine longer-term durability. Heat alone reduced the strength of the unaged lap joints by 73% but the presence of 3% wt. EG reduced joint strength by 96% and 5% wt. EG led to complete debonding. The fracture toughness of the polyurethane adhesive was unaffected by the presence of 5% wt. and the crack propagation rate was similar, whether or not the adhesive contained EG.

5. CONCLUSIONS

This overview has attempted to improve the reader's appreciation and understanding of the variety of methods available for the dismantling and disassembly of adhesive bonded products. As with all other aspects of adhesive bonding technology, it is essential that bonded products are regarded as systems. This means that the interactions between the combinations of substrate and adhesive (and primers, if present) must be considered carefully, together with the manufacturing aspects and the service requirements. This is a prerequisite to the selection of the most appropriate debonding method.

The over-arching consideration relates to the balance between the adhesive system cure temperature (and any subsequent post-heating operations, such as the paint-bake cycle in the automotive industry) and the disbonding temperature. This automatically dictates the type and nature of an appropriate method.

Heat alone may be sufficient to enable the separation of components bonded with largely thermoplastic-based adhesives, but cross-linked polymers require chemical or physical interventions. In all cases reviewed, heat energy is the dominant trigger mechanism but the energy can also be provided electrically or by photo-irradiation.

Truly re-workable adhesive systems are fairly uncommon, they tend to involve thermoplastic constituents in the adhesive and they are appropriate only for non-structural and some semi-structural applications. There are several examples of methods that are applicable to the electronics industry, particularly for micro-chip bonding. More structurally, research has shown that polymer cross-link density may be varied in a reversible manner. Whilst true disbonding is not an outcome, weakening (or indeed stiffening for peel joints) of the polymer backbone can enable modest mechanical force to complete the disassembly process.

The majority of disbonding systems involve the use of functional additives that are mixed into the adhesive (or primer layer) in quantities of up to 20% by weight. These additives may be chemical foaming agents, physical foaming

agents, metallic particles or expandable inorganic particles. In all such cases these additives generally adversely affect the mechanical properties of the adhesive, both in the short term and in the long term. Thermally expandable thermoplastic microspheres are not bonded chemically to adhesive matrices, unless the surfaces of the microspheres are modified chemically, meaning that severe reductions in adhesive material properties must be accepted. Some researchers have noted that any additive could be a carrier for other functional requirements whilst others have highlighted the beneficial effects of polymer toughening by the incorporation of additives, and this warrants further research. There is no doubt that the greatest amount of flexibility and potential for the development of disbonding methods for structural and semi-structural assemblies lies with functional additives.

Adverse reactions between the adhesive matrix and additives, or between additives and the service environment, can be avoided by microencapsulation techniques applied to the additive particles.

Bonded assemblies may have active substrates which, so far, involve at least one metal or at least a conductive foil patch. A small electrical current is passed across the bondline and ion conduction takes place along the resin-metal interface, triggering disbonding. This method is limited in its applicability currently to non- or semi-structural applications.

It is evident that debonding methods need to be developed for specific applications where all aspects of the manufacturing, materials and service requirements for the bonded product can be considered.

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Table 1. Joint types and debonding requirements

Joint type	Requirements	Example application	Appropriate technology (see Section 3)
Structural	Joints that need to transmit structural loads throughout the service life of a structure	Chassis joints in automotive structures	CFA, PFA, EG
Semi-structural	Joints that need to transmit transient loads and low stresses	Auto windscreens, sandwich panels, road signs, orthodontics	CFA, PFA, EG
Non-structural	Joints that do not need to transmit externally-applied loads	Auto trim applications, electronic circuit boards, glass substrates	Diels–Alder ElectRelease Photoirradiation Hot-melt

CFA=chemical foaming agent; PFA=physical foaming agent; EG=expandable

graphite

Table 2. Summary of research activities in disbonding technologies

Disbonding technology	Substrates	Adhesives	Effect on		Authors
			Initial strength	Long-term durability	
ElectRelease	Metal		√		[53], [54]
Photo-irradiation	Metal glass	Acrylic-based	√		[23], [24]
CFAs and PFAs	Aluminium	Epoxy PU	√	√	[37], [38]
PFAs: Surface-modified TEMs	Aluminium	Epoxy	√	√	[39]
EG	Aluminium	Epoxy PU	√	√	[51]

Figure 1 A virtuous life cycle showing materials flows

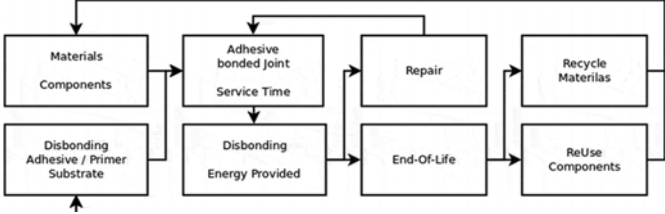


Figure 2. Idealised viscoelastic behaviour of a resin for disbondable adhesives [6]

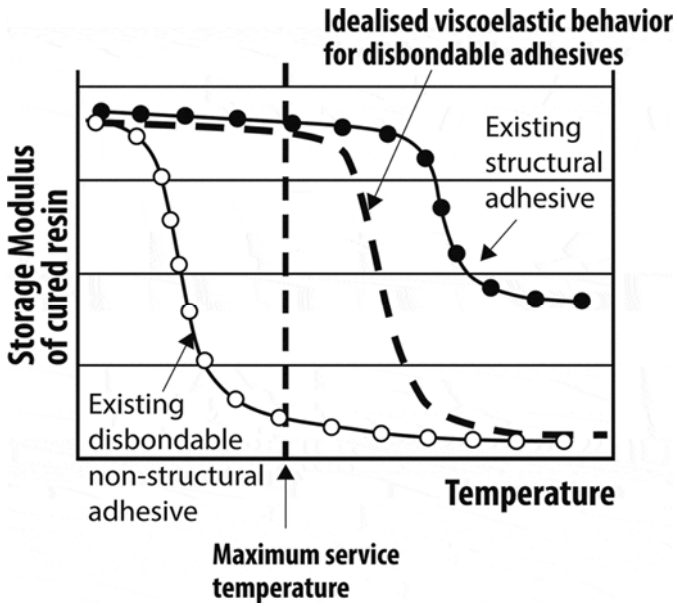


Figure 3. Disbonding technologies and mechanisms

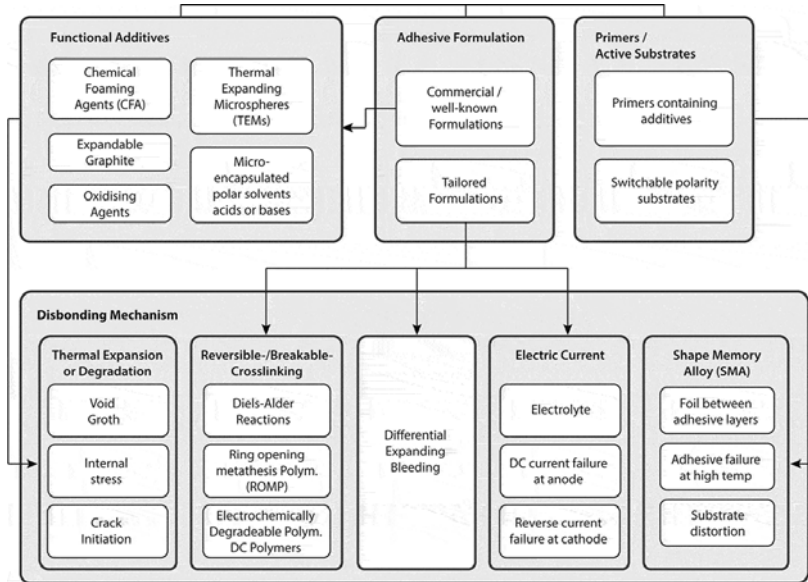


Figure 4. Glass removal tool for the BMW i3 [7]

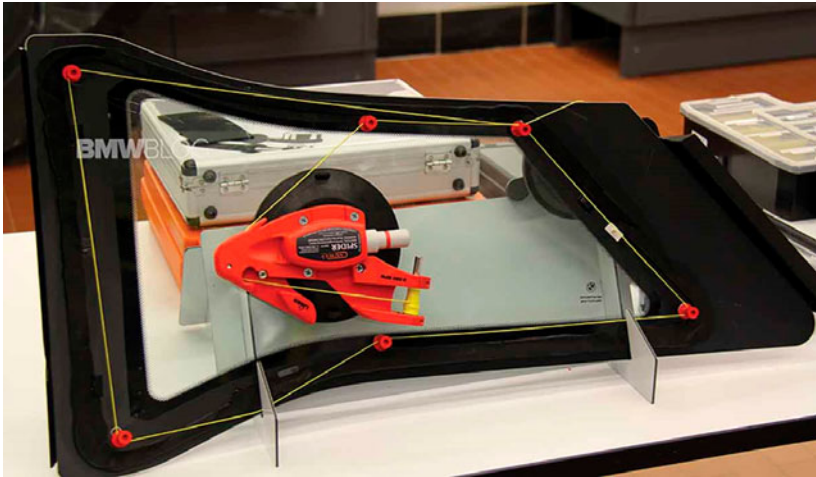


Figure 5. Principle of disbonding with expanding additives

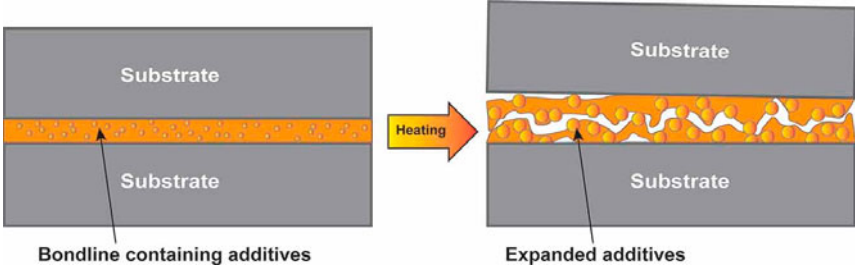


Figure 6. Schematic of TEM expansion mechanism

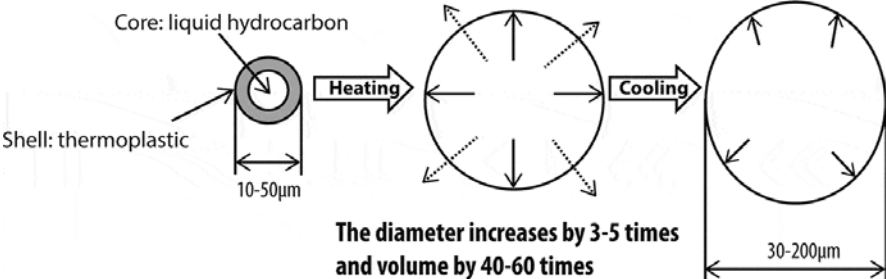


Figure 7. 2-D Schematic of TEM surface modification process

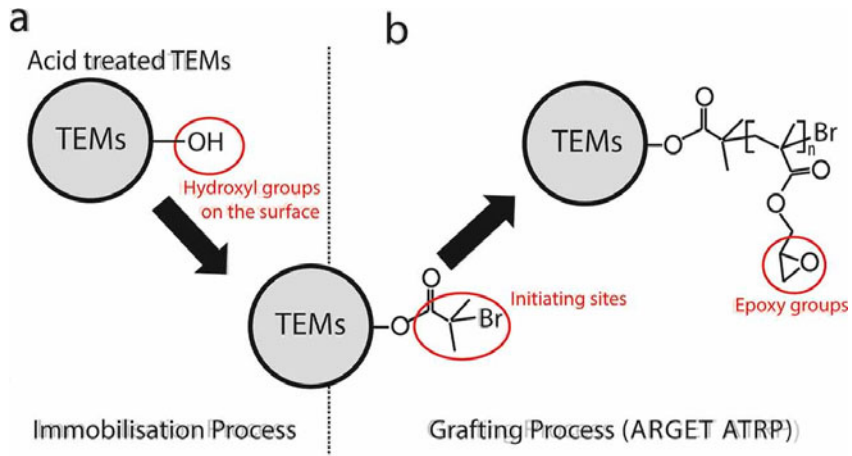


Figure 8. Graphite flakes unexpanded (left) and expanded (right) [51]

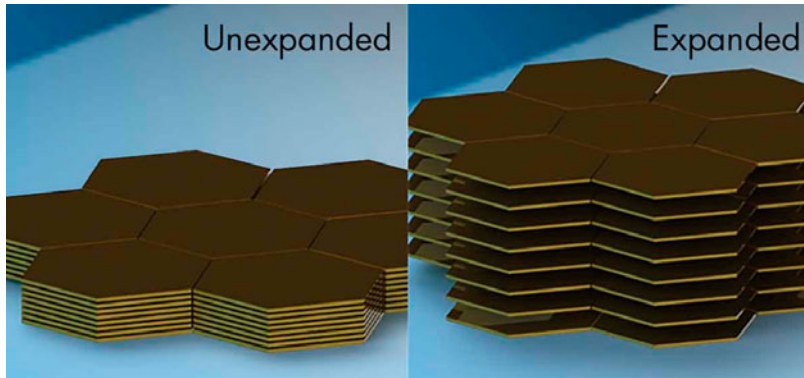


Figure 9. Electrically disbonding adhesive

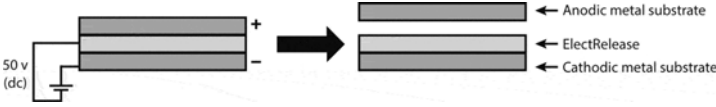


Figure 10. Lap shear joint properties with different functional additives

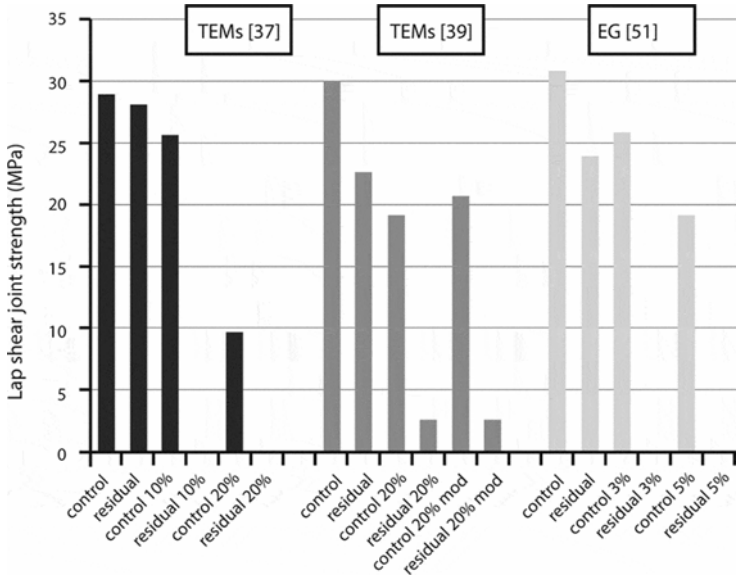


Figure 11. Tensile strength of bulk adhesive specimens containing 20% wt. unmodified and modified TEMs [39]

