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Dehybridisation of fibre-metal laminates via blowing agents in the thermosetting adhesive layer

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ABSTRACT

The experiments conducted in this study examine Debonding on Demand strategies for fibre–metal laminates by using blowing agents embedded in the epoxy adhesive layer, with motivation rooted in recycling and efficient end-of-life separation of material compounds. Four blowing agents—two chemical (OBSH and ADC based) and two physical (thermally expandable microspheres) are tested through thermal analysis, mass/volume changes and microscopic evaluation focusing on their interaction with an epoxy matrix and their separation mechanisms after thermal activation. The experimental results demonstrated that the utilised chemical blowing agents induce pronounced cracking and enable targeted weakening of the adhesive interface, while the physical agents used in this study primarily lead to volumetric expansion and changes in colour after activation without sufficient macroscopic damage. Edge shear testing of manufactured fibre–metal laminates confirmed that ADC-based chemical blowing agent exhibited nearly complete delamination after activation, while the utilised physical blowing agent showed a significant but insufficient reduction of 29.0% in shear strength. The systematic analysis of thermal behaviour, interfacial shear strength, and mechanical degradation mechanisms demonstrates the feasibility of tailoring FML architectures for separation via targeted weakening of the adhesive interface, supporting improved recycling and material reuse.

1. Introduction and motivation

Fibre-metal-laminates (FMLs) are hybrid materials that combine the properties of different components to achieve a customised property spectrum [1]. Especially hybrid composite materials, consisting of several layers of metal and fibre-reinforced plastic (e.g. GLARE [1] and ARALL [2]), have become particularly well established in the aerospace and automotive industries. The benefits of metallic materials, including their high formability and impact strength, are thus integrated with the advantages of fibre-reinforced plastics, such as their high specific stiffness and high specific strength [1,3]. This results in lightweight, high-strength or high-stiffness structural components with good energy absorption capacity [1], which exhibit improved crack initiation and crack propagation behaviour compared to neat metallic structures under cyclic mechanical loading conditions [4].

Traditional FMLs use adhesively hybridised interfaces, which complicate the separation and subsequent recycling of the constituent materials. It is apparent that the sustainable utilisation of these materials has been overlooked in the present situation.

Mechanical separation is only feasible for adhesive joints with low bond strength; otherwise, the process becomes time-intensive and labour-demanding, limiting its practical applicability [5]. While traditional thermal recycling, like pyrolysis or solvolysis, require or release harmful chemicals and results in the irreversible loss of the polymeric component and degradation of the fibres. [5]. Consequently, alternative methods have been developed. A mechanical separation under temperature is primarily applied to thermoplastic adhesive systems, as these can be re-melted through heat input, even after solidification, allowing for mechanical separation with minimal residual force. Furthermore, fillers in thermoplastic polymers can be used, that expand or generate gas [6–8], creating internal pressure that weakens the cohesion of the adhesive. Furthermore, induction-based Debonding on Demand (DoD) concepts have been developed, where functional susceptor additives (e.g., metallic wire meshes [9], carbon fibre non-wovens [10], or particulate fillers [10]) are embedded in the adhesive layer. In the presence of a high-frequency induction field, these susceptors rapidly generate heat, thereby inducing localised thermal shocks. These shocks

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have been shown to compromise the integrity of the adhesive–substrate interface [10].

In the domain of thermoset polymers, the utilisation of specific additives within the adhesive composition is of significance. These additives, when exposed to elevated temperatures, induce a separation process within the polymeric network. The initial example is a primer that has been modified with foaming agents, as illustrated in the patent FR 2 852 965 [11]. Following the application of heat, the primer is degraded, facilitating the separation of the parts. Moreover, the implementation of reactive fillers is a viable option, with the capacity to initiate a reaction that results in either the attack or destruction of the filler following its activation [12].

Another approach is to modify the adhesive's chemical structure so that, when activated (most often by heat), the crosslinked network can be cleaved in a reversible manner. One example of this is the incorporation of reversible chemistries, such as the Diels–Alder reaction [13]. Adhesives based on this mechanism typically have low glass transition temperatures (around 40 °C), with the retro-Diels–Alder reaction initiating at relatively low temperatures (above 90 °C).

The Fraunhofer IFAM has developed an adhesive that can be detached from another adhesive by applying a temperature of 65 °C and an electric potential of 48 V simultaneously. For the adhesive ElectRelease, an electrical voltage between 10 V and 50 V at the interface is sufficient to separate the adhesive, which then fails adhesively [14]. The efficacy of these methodologies is contingent upon the presence of adequate electrical properties in one or both constituent components. Adhesive bonds, especially thermoplastic adhesives, can be dissolved by using organic solvents. In addition to organic solvents, acids and bases are also used to dissolve the adhesive matrix. However, the conventional use of solvents has the disadvantage that, especially in the case of larger, structural bonds, the adhesive surface is difficult to access from the outside. Often, only the outer edges of an adhesive bond can be reached and dissolved. In the case of large-area adhesive bonds, chemical depolymerisation or swelling can be achieved by introducing a filler capillary system made of natural or synthetic fibres, whereby the solvent diffuses into the adhesive bond via the capillary system [15]. Another method of chemical separation involves the introduction of reactive groups that remain in the adhesive matrix even after curing. These can be thermally activated, thereby breaking the chemical bonds of the adhesive [16]. A further common disadvantage of thermal and chemical separation is the potential for damage to the polymer matrix [17].

The presented work in this study proposes a solution to the issue by placing emphasis on the design of FMLs with the objective of facilitating their DoD as illustrated in Fig. 1. The central focus of this research is the development of temperature-activatable adhesive layers. These interlayers are engineered to undergo selective weakening at the end of the FML lifecycle. This deliberate weakening of the material under the influence of temperature facilitates the effective separation of the metal and fibre-reinforced composites, and should enable the recycling or reuse of the individual materials. The ideal outcome of the tests is the formation of a microcrack network or gas-induced interfacial stresses that promote delamination upon thermal activation, while maintaining high interlaminar shear strength in the unactivated condition. The activation temperature of a suitable blowing agent must be higher than the maximum service life and manufacturing temperature to prevent premature damage, yet low enough to avoid degradation of the adherents and matrix constituents during activation.

The study is divided into three main parts. First, the blowing agents are characterised using Differential Scanning Calorimetry (DSC) and Thermogravimetric Analysis (TGA) to assess their thermal stability and identify transition points. In the second part, the interaction between the blowing agents and the epoxy matrix is investigated. For this purpose, various specimens are prepared and analysed using microscopic, gravimetric, and volumetric methods to gain deeper insight into their degradation behaviour. Finally, FMLs are manufactured, and their shear strength is tested both before and after activation to evaluate their suitability for DoD applications.

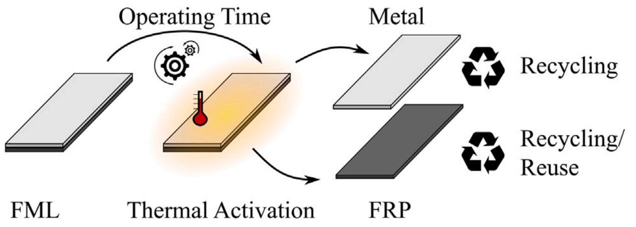


Fig. 1. Systematic debonding for recycling of FML after operating time via thermally activatable blowing agents in the adhesive layer.

Table 1
Blowing agents with chemical base and activation temperatures according to manufacturer.

Blowing agent	Chemical base	Temperature in °C	
		T_{Onset}	T_{Peak}
Genitron OB	OBSH	140–160	
Porofor ADC/M-C1	ADC	214	
Expancel 980 DU100	VHC	169–189	209–229
Kureha S2640D	TMP	190–215	250–265

2. Material and methods

2.1. Materials

Four blowing agents and oxidising agents were considered for the DoD process. Two chemical blowing agents and two physical blowing agents respectively thermally expandable microspheres (TEM) were examined. The first is an 4,4'-oxy-bis(benzenesulphohohydrazide) (OBSH) based blowing agent, marketed under the designation Genitron OB by the company Lanxess. The second chemical blowing agent is marketed under the product name Porofor ADC/M-C1 by Lanxess and its chemical composition is azodicarbonamide (ADC). The physical blowing agents employed in this study are Kureha S2640D with the blowing agent 2,2,4-trimethylpentane (TMP), manufactured by the Japanese company Kureha, and Expancel 980 DU 100, microspheres that are filled with volatile hydrocarbons (VHC), produced by the Dutch company Nouryon. The temperatures necessary for activation according to the manufacturers are shown in Table 1.

The adhesive used is UHU Plus Endfest from the company UHU GmbH. A two-component epoxy resin based on bisphenol-A-(epichlorhydrin) and bisphenol-F-(epichlorhydrin) combined with an aliphatic amine hardener.

The FML laminates consist of aluminium sheets and a carbon-fibre-reinforced epoxy composite with [0°/90°/90°/0°] configuration and an activatable thermoset interlayer incorporated between the metallic and composite layers. While the CFRP stacking sequence affects the in-plane mechanical response, it does not significantly influence the interfacial shear behaviour investigated in this study. The composite constituent was supplied in the form of a prepreg (SIGRAPREG® C U230-0/NF-E320, SGL Technologies GmbH), consisting of a unidirectional HT carbon fibre/epoxy system with a resin mass content of 39%. Each ply had a nominal thickness of 0.22 mm. Aluminium sheets (EN AW-5754 H111, Kaysser GmbH & Co. KG) with a thickness of 1 mm were used. To minimise out-of-plane distortion caused by thermal expansion mismatch, a symmetric lay-up was selected, as shown in Fig. 3.

2.2. Thermal analysis of neat blowing agents

For the DSC measurements of the blowing agents the Netzsch DSC300 was used, the tests are conducted with a temperature range from 20 °C to 300 °C and a heating rate of 10 K/min. In the context of DoD, the blowing agents are embedded within the epoxy matrix.

Therefore, the DSC and TGA are conducted within a protective nitrogen atmosphere.

The decomposition curves of the blowing agents were measured in a nitrogen atmosphere by using the vacuum-tight Netzsch TG 209 F1 Iris and were conducted in a nitrogen atmosphere. A temperature range of 20 °C to 600 °C and a heating rate of 10 K/min are used to conduct the tests. For all TGA and DSC curves, two measurements of each are shown.

2.3. Effect of blowing agent activation on cured epoxy

For the evaluation of the influence of the blowing agents to the neat epoxy, two kinds of specimens are produced. The specimens were produced utilising all four blowing agents, with mass contents of 2%, 5% and 10%, respectively. The bulk specimens are cube-shaped with dimensions of 25 mm × 25 mm × 10 mm. One specimen was prepared for each mass content and additive by combining the epoxy resin and hardener according to the manufacturer's specifications, degassing and mixing the components for approximately 5 min in a vacuum stirrer, and casting it into a negative mould coated with polytetrafluoroethylene (PTFE) spray. For a different specimen type, two glass slides were bonded with epoxy resin. The thickness of the adhesive layer was determined by utilising a 0.2 mm metal spacer. The glued area exhibited a diameter of approximately 20 mm. A weight was used to apply pressure to the glass slides during the curing process to achieve the desired thickness of the metal spacers. Five specimens per configuration were produced for this specimen type. This configuration facilitates an optical evaluation of the temperature-induced effects on the epoxy resin in conjunction with the additives.

The mass and volume of the bulk specimens were measured by using the Mettler-Toledo Excellence XPR504SDR scale. The objective of the experiment was to ascertain the change in mass and volume of the samples over several time intervals at the temperature of 220 °C. To achieve this, the samples were alternately activated in the preheated convection oven for a designated time interval. Thereafter, they were removed, cooled to room temperature, and measured in distilled water according to Archimedes. Subsequently, the volume samples were placed in the oven for the subsequent time interval, and the same process was repeated. The times in chronological order were 9 min, 18 min, 27 min, 54 min and 81 min from the start. The samples between the glass slides were subjected to the same temperature and time intervals.

2.4. Evaluation of fibre-metal-laminate separation

The laminates, with dimensions of 280 mm × 195 mm, were consolidated using a hot-pressing system (LaboPress P200S, VOGT Labormaschinen), resulting in a total laminate thickness of approximately 4.5 mm. The carbon fibre-reinforced polymer (CFRP) prepreg was cured in an initial hot-pressing step and subsequently bonded to the aluminium sheet in a second hot-pressing operation using the activatable thermoset interlayer as an adhesive layer. Through this two-stage fabrication process, the possibility of reusing the CFRP to manufacture a FML can be tested. To improve interfacial bonding between the aluminium and polymer phases, the aluminium sheets were surface textured by using a fibre laser system (gieMARK 30+, Gie-Tec GmbH, wavelength 1064 nm). The resulting sheet surface topography is shown in Fig. 2.

Based on the processing recommendations of SGL Technologies, the consolidation temperature was set to 140 °C. To achieve full curing of the prepreg, the temperature was maintained for 20 min after reaching the setpoint. A process pressure of 5 bar was applied, which differed from the manufacturer's recommended 8 bar, as preliminary trials at higher pressure yielded suboptimal laminate quality, since the polymeric interlayer was squeezed out non-uniformly under pressure, resulting in variable interlayer thickness. The applied pressure was maintained during cooling to minimise residual stresses and avoid warpage. Before processing, the press tooling was lined with PTFE foil and treated with a release agent to ensure reliable demolding of the consolidated laminates. To ensure direct comparability of the mechanical and structural properties, all laminate configurations were manufactured with identical processing parameters. The specimens were cut to the required geometry of 10 mm × 20 mm using a QATM QCUT precision cutting machine.

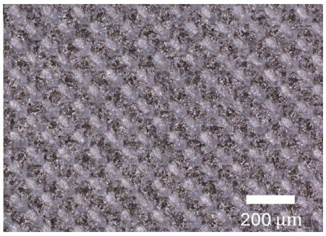


Fig. 2. Laser-generated surface structure on the aluminium sheets.

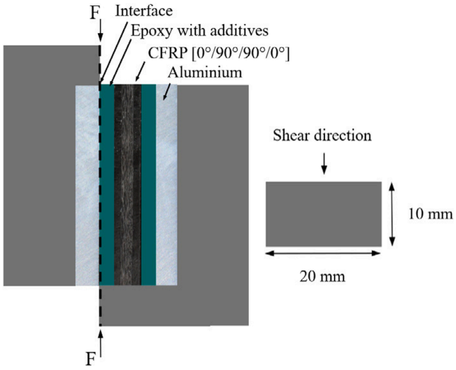


Fig. 3. Schematic representation of the edge shear test fixture used for interfacial characterisation of the FML specimens.

Table 2
Test parameters and specimen geometry for quasi-static edge shear tests.

	Value	Unit
Crosshead velocity	1	mm/min
Load cell	50	kN
Specimen height	10	mm
Specimen depth	20	mm
Number of specimens	min. 3 specimens per FML variant	–

The shear strength of the investigated FML configurations was determined by using the edge shear test according to Weidenmann et al. [18]. A schematic of the test setup is presented in Fig. 3. Through the adjustable positioning of the shear plane, the load can be applied directly to the interface, ensuring controlled interfacial shear failure. The interlaminar shear strength was calculated as the ratio of the maximum applied force to the sheared area with the relevant test parameters summarised in Table 2. As demonstrated by numerous studies, the edge shear test is a valuable tool for the analysis of the interlaminar shear strength and interfacial properties of FMLs with integrated polymer interlayers [19,20].

Based on previous thermal analyses, the suitability of the four investigated blowing agents could be assessed before mechanical testing. Given that OBSH activated below the FML processing range and TMP

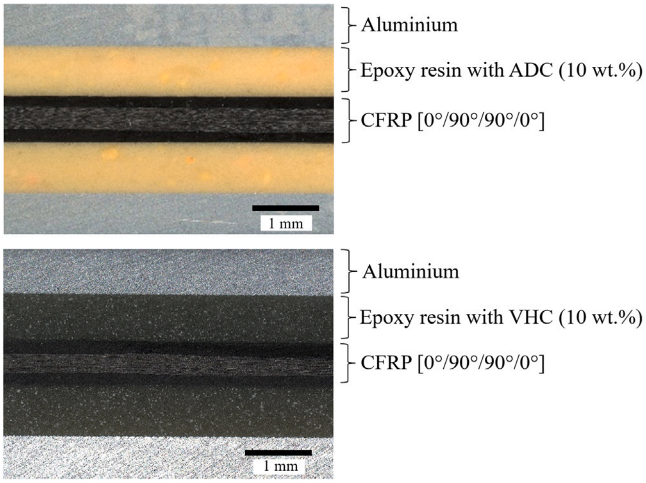


Fig. 4. Cross-sectional micrographs of ADC- and VHC-based specimens with a symmetric laminate lay-up, containing 10 wt.% blowing agent.

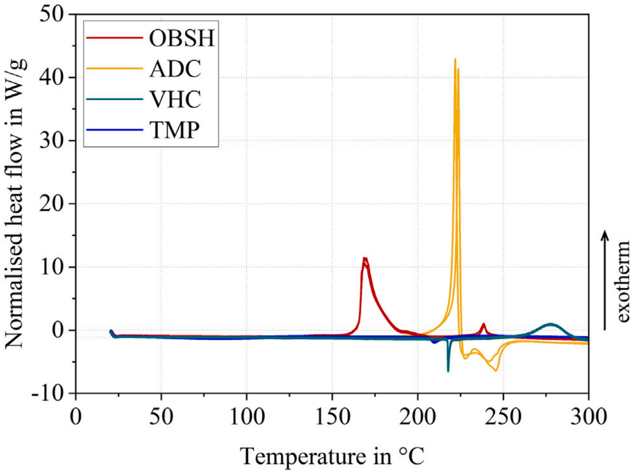


Fig. 5. DSC curves of various blowing agents.

produced neither cracks nor visible chemical degradation in bulk specimens or in the epoxy between glass slides, both blowing agents were excluded from further investigation. The blowing agents ADC and VHC were considered for the subsequent mechanical characterisation of the laminates. Three different laminate variants were mechanically tested, with each variant evaluated in both the nonactivated and activated state, allowing for a quantitative assessment of the dehybridisation strategy. For determining the mean shear strength, at least three specimens per laminate variant were included in the evaluation. Only tests exhibiting interfacial failure were considered valid; cases with purely cohesive failure of the integrated activatable epoxy layer were excluded from the assessment of interlaminar shear strength, as they are not indicative of interfacial performance.

Micrographs of the ADC and VHC laminate variants are shown in Fig. 4. The configuration comprises aluminium, the activatable interlayer – epoxy + ADC or VHC – and CFRP. In addition to the modified FMLs with an activatable epoxy interlayer, reference laminates were manufactured: the reference laminate bonds the metal and CFRP layers solely with the neat epoxy resin. The thermoset interlayer had a nominal thickness of 0.80 mm, with manufacturing-induced variations of ±0.15 mm across the laminate. Owing to the small specimen size, specimens were taken from regions exhibiting uniform layer thickness.

Table 3
Onset and peak temperatures for chemical and physical blowing agents as determined by TGA.

Chemical base	Temperature (°C)	
	T_{onset}	T_{peak}
OBSH	130	165
ADC	190	220
VHC	215	220
TMP	190	210

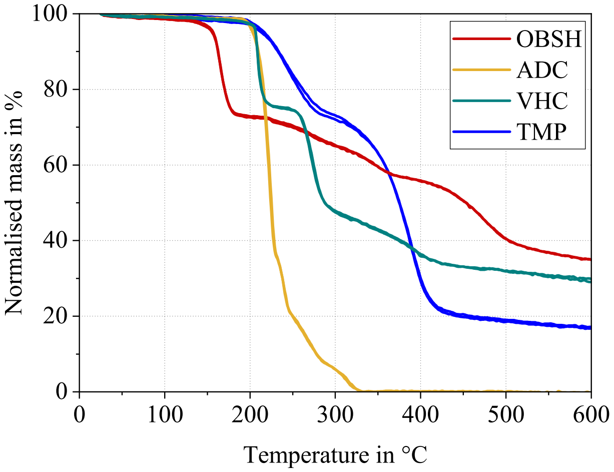


Fig. 6. TGA curves of various blowing agents.

3. Results

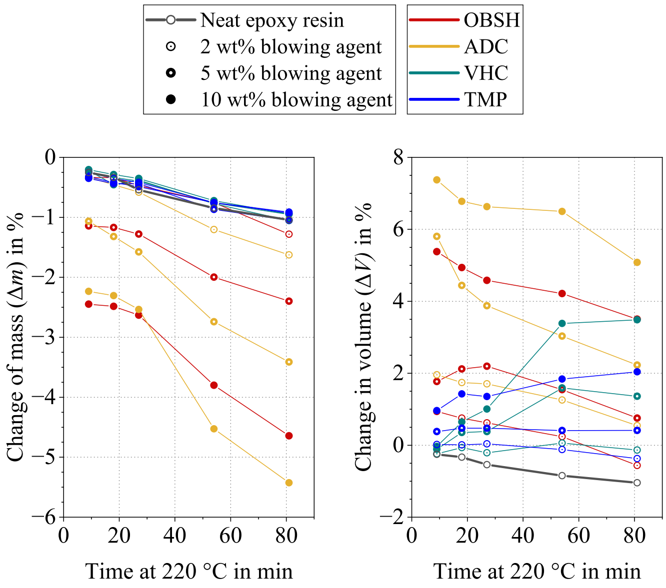
3.1. Thermal analysis of neat blowing agents

In the DSC measurements are shown in Fig. 5, both chemical blowing agents exhibit characteristic exothermic decomposition peaks corresponding to the release of the active gas phase. The OBSH based blowing agent shows the lowest onset of thermal activity among the agents that were examined. It exhibits an exothermic peak at 165 °C, yet its decomposition commences at a relatively low temperature of 130 °C. The ADC based blowing agent exhibits its decomposition with its exothermic maximum at 220 °C with the start at a temperature of 190 °C. In contrast, the two physical blowing agents, VHC and TMP, present endothermic signals associated with the softening of the polymer shell and the expansion of the encapsulated gas. VHC displays a pronounced reaction at 220 °C, with TMP exhibiting a slightly lower temperature of approximately 210 °C. The onset and peak temperatures are summarised in Table 3.

The curves measured with TGA are shown in Fig. 6. Similar onset temperatures are observed for the individual blowing agents in the TGA and DSC measurements. ADC shows with a mass of zero at 330 °C the strongest mass reduction. The mass of OBSH is reduced by 25% at a temperature of 180 °C and decomposes more gradually afterwards with a residual mass of 35% at 600 °C. VHC shows an initial sharp mass reduction of 23% around 210 °C. And a second of 26% between 260 °C and 280 °C. With a remaining mass of 30%. The TMP based blowing agent has its first mass reduction step with 25% between 200 °C and 280 °C. Another significant change in mass occurs between 300 °C and 430 °C, where more than 50% of the original mass is degraded. This is followed by a continuous slow decrease to 16% residual mass at 600 °C.

3.2. Effect of blowing agent activation on cured epoxy

Fig. 7 shows the time-resolved changes in mass and volume for the bulk epoxy resin specimens containing different blowing agents,



(a) (b)

Fig. 7. Time-dependent changes in mass and volume of bulk epoxy resin specimens containing different mass fractions of blowing agents during thermal activation at 220 °C.

subjected to thermal activation at 220 °C. Fig. 7(a) and Fig. 7(b) present the mass loss and volumetric expansion, respectively, for all evaluated blowing agent types over the intervals of 9 min, 18 min, 27 min, 54 min and 81 min.

The physical blowing agents (VHC and TMP) demonstrate a reduction in mass of 1% after 81 min, which is equivalent to the reduction observed in the neat epoxy. The specimens containing chemical blowing agents (OBSH and ADC) demonstrate an increasing reduction in mass with increasing blowing agent content. The ADC containing specimens exhibited the strongest reduction, with a 5.4% decrease observed after 81 min. In comparison, the OBSH specimens demonstrated a slightly less pronounced reduction, with a 4.6% decrease recorded. Both show the strongest reduction in mass after the first 9 min of activation and a stronger slope at the end of the test compared to the difference between the measurements at 9 min and 18 min.

The change in volume of the neat epoxy follows a similar trend and magnitude as the change in mass. With increasing amounts of blowing agents, the volume expansion becomes more pronounced, depending on the exposure time at 220 °C. Differences in the evolution can be observed between chemical and physical blowing agents. Thereby, the physical blowing agents (VHC and TMP) show an increasing volume over time. With the VHC containing specimens showing the maximum of 3.5% after 81 min. The chemical blowing agents demonstrate an initial surge in volume, with ADC exhibiting the most pronounced increase of 7.5% at 9 min, followed by a downward trend of chemical blowing agents containing specimens.

The bulk specimens with various mass fractions of the four blowing agents, after an activation temperature of 220 °C and a total time of 81 min, can be seen in Fig. 8. The specimens with chemical blowing agents (OBSH and ADC) show an increasing amount of cracks in conjunction with an increasing mass fraction of the blowing agents. OBSH containing specimens has been demonstrated to exhibit a more pronounced crack network in comparison to ADC. The physical blowing agents (TMP and VHC) on the other hand show no macroscopically visible cracks.

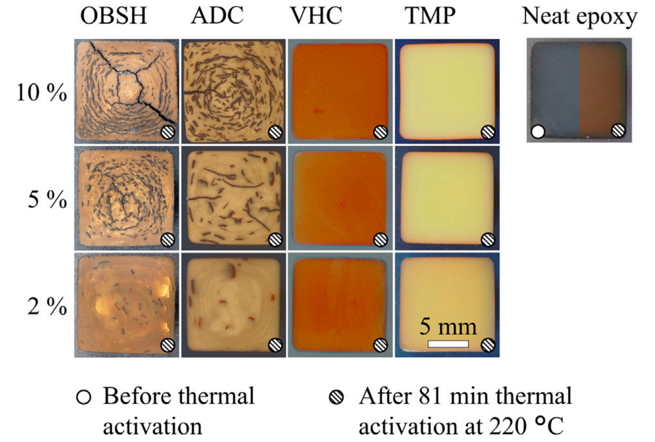


Fig. 8. Bulk epoxy specimens with different mass contents of blowing agents after and before thermal activation.

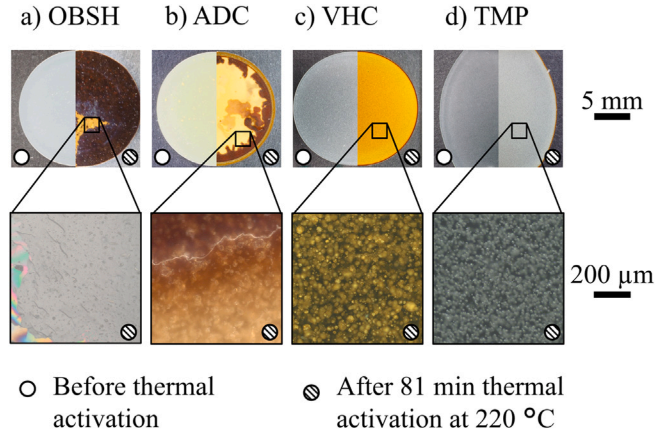
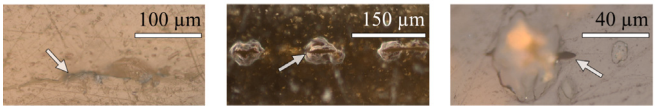


Fig. 9. Epoxy specimens between glass slides with 10% blowing agents before and after thermal activation for 81 min at 220 °C.

The influence of the temperature on the epoxy with 10% blowing agent combination between the glass slides is shown in Fig. 9, with a macroscopic comparison of the specimens before and bottom. Specimens containing chemical blowing agents in Fig. 9 (a) and (b) exhibit delamination between the epoxy layer and the glass slides, resulting in darker areas where the epoxy layer is in contact with surrounding air. This is also visible for the ADC containing specimen in the microscopic image. The material that has not come into contact with the surrounding air is yellow, while the incoming delamination from the right is turning the material a darker shade of brown. Microscopic images of the OBSH containing specimen show cracks up to 200 μm in length. The VHC containing specimens in Fig. 9 (c) exhibit a pronounced alteration in colour, transitioning from white/opaque to yellow. As demonstrated by the microscopic images, the observed change in colour is located at the site of the blowing agent itself. For both kind of specimens containing physical blowing agents, no macroscopic visible cracks were formed in the bulk specimens.

In Fig. 10 representative images of mechanical degradation in the specimens between the glass slides are shown. TMP is excluded, as no mechanical degradation was determined. Specimens containing OBSH as a blowing agent exhibit the most pronounced crack formation. A length of approximately 200 μm has been observed and formed, primarily parallel to the glass slides. The specimens containing the blowing agent ADC regularly exhibit cracks with a length between 40 μm and 120 μm, which are located in the middle of the adhesive layer. The VHC containing specimens exhibit cracks around the TEMs with a length of up to 20 μm.



(a) OBSH (b) ADC (c) VHC

Fig. 10. Cross-sectional microscopic images of specimens containing 5% blowing agent, showing specimens with mechanical degradation after 27 min at 220 °C between two glass slides.

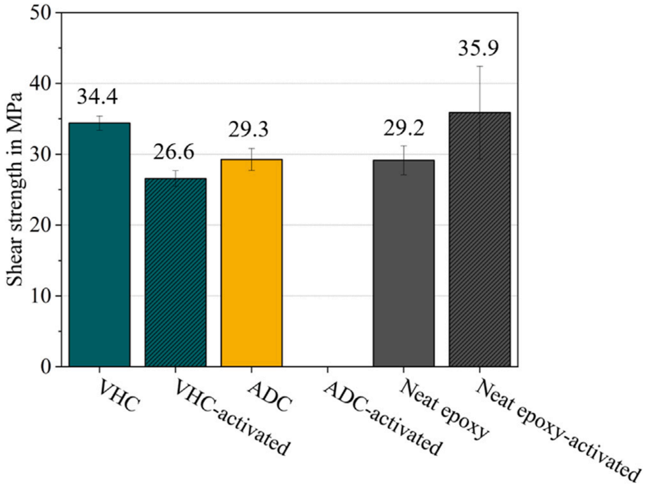


Fig. 11. Mean interfacial shear strength of VHC-, ADC-based and neat epoxy resin laminates in both the activated and non-activated conditions.

3.3. *Evaluation of fibre-metal-laminate separation*

As OBSH exhibited an onset temperature below the manufacturing temperature of the FML of 140 °C, and TMP demonstrated an absence of cracks or visible chemical degradation in the bulk specimens and in the epoxy between the glass slides, these two blowing agents are to be excluded from further investigation.

The effect of thermal activation on the interlaminar shear strength is presented in Fig. 11 for a comparative assessment of the VHC, ADC and reference laminates. In the non-activated state, VHC and ADC variants exhibited comparable strength levels of 34.4 MPa (VHC) and 29.3 MPa (ADC). The reference laminate showed a comparably high interlaminar shear strength of 29.2 MPa, indicating that the incorporation of blowing agents does not adversely affect the shear strength in the non-activated state. Upon thermal activation, the interlaminar shear strength of the VHC laminates decreased by 29%, with adhesive failure occurring at the VHC–CFRP interface. In contrast, no interlaminar shear strength could be quantified for the ADC laminates, as the specimens were already nearly fully delaminated upon removal from the oven. To verify whether the delamination observed in the ADC laminates and the reduction in shear strength of the VHC laminates can be attributed to the activation of the blowing agents, we tested a reference laminate without blowing agents under identical conditions, both before and after defined thermal conditioning. The results show that the interfacial strength of the reference specimens was not reduced by thermal conditioning. On average, the shear strength of the reference laminates was slightly increased after thermal conditioning, although this was associated with a high standard deviation.

In Fig. 12 microscopic examinations of representative laminates conducted after thermal activation are shown. In particular in the ADC laminates, the expansion of the blowing agent induced pronounced microcrack formation, ultimately resulting in almost complete delamination. In contrast, the VHC laminates exhibited predominantly intact interfaces, with activation-induced pores discernible within the interphase. The thermally conditioned neat epoxy resin laminates showed no indication of damage or delamination.

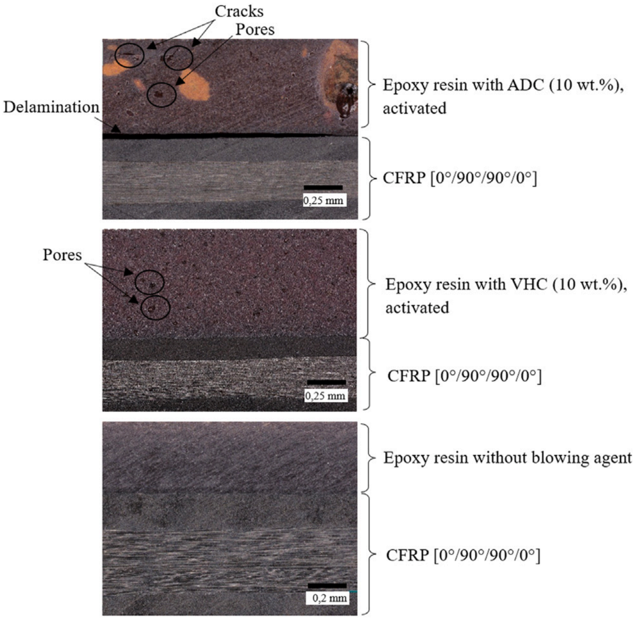


Fig. 12. Cross-sectional micrographs of the activated ADC (10%), VHC (10%) and neat epoxy resin laminates after thermal conditioning at 220 °C for 81 min, showing the thermoset–CFRP interface.

4. Discussion

The behaviour observed in DSC is well reflected by the corresponding TGA measurements. Both chemical blowing agents show significant mass losses in the temperature region of their exothermic decomposition peaks, confirming the evolution of gaseous products during breakdown of the hydrazide and azodicarbonamide structures [21,22]. The TGA curves obtained for OBSH and ADC show similar characteristics to those published in a paper by Emami et al. particularly with regard to the peak decomposition temperature and the kinetics of mass loss [23]. The reaction of OBSH commenced at a temperature of 130 °C, which is to be noted as being lower than the maximal temperature of 140 °C that is employed during the manufacturing process of the FML. For this reason, OBSH was excluded from further investigation. Notwithstanding the encouraging outcomes observed in the experimental tests conducted with the bulk epoxy specimens and the glass slides. In instances where lower temperatures are required for the execution of a given application, the utilisation of a blowing agent based on OBSH such as Genitron OB, will be a suitable DoD solution.

By contrast, the physical blowing agents (VHC and TMP) exhibit endothermic first peaks associated with shell softening and internal gas expansion. A reduction in mass is measured by TGA at the temperatures of the first DSC peaks. The sharp initial drop in the VHC curve indicates that venting occurs as the polymer's encapsulated gas expands [24]. For TMP the mass reduction is stretched towards higher temperatures, indicating a slower venting process. The absence of an increase in mass loss when compared with the pure epoxy in the bulk specimens indicates that no sufficient cracks have been formed in which the gas could be released, or that the gas remains encapsulated within the TEM. This may be owing to the surrounding epoxy withstanding the inner pressure, thus preventing any venting.

Endothermic reactions of the physical blowing agents, are advantageous for short phases of temperature overshoots. This thermal moderating effect helps to prevent runaway heating and unwanted damage to adjacent matrix material. In contrast, exothermic reactions of the chemical blowing agents release, thereby effectively triggering the debonding mechanism by increasing local temperature, promoting adhesive interface failure and crack initiation. In both cases, the adhesive layer may experience transient elevated temperatures during operation, which could unintentionally initiate thermally driven debonding and compromise structural integrity prior to decommissioning. Therefore, FMLs in service cannot be fully isolated from the risks associated with thermally activated interfaces, and this limitation must be acknowledged when evaluating the applicability of the proposed approach. The encapsulating epoxy matrix plays a critical role in the activation dynamics of blowing agents, as it restricts the mobility and diffusion of reactants and gaseous products within the composite structure. This containment effect results in a delayed onset of blowing-agent decomposition, meaning that the precursors and reaction products form at later times or higher temperatures compared to agents dispersed in more permeable matrices [24].

Macroscopic and microscopic evaluation of the bulk and glass slide specimens indicates that TEMs based on TMP, such as Kureha S2640D, are not suitable for a DoD application, as no cracks formed. The other blowing agents exhibited a change in colour after thermal activation, with the colour change in the VHC containing specimens being attributable to the colour change of the blowing agent itself. For the Genitron and ADC containing specimens, the observed change in colour may be indicative of a chemical change in the epoxy. Furthermore, the adhesive failure observed at the glass slide specimens and the subsequent colour change should not be overemphasised, as thermo-oxidative effects are more pronounced in this geometry compared to FMLs [25]. In an FML configuration, the reduced exposure to oxygen is expected to mitigate such surface-driven degradation [26]. Nevertheless, a more intense colour change can still be interpreted as an indication of stronger decomposition. For the neat epoxy specimens, the measured mass loss can be attributed predominantly to the release of volatile compounds, most notably water. The corresponding volume loss, however, originates not only from evaporation of water but also from shrinkage owing to post-curing of the epoxy. In contrast, the chemical blowing agent-containing specimens exhibit an additional mechanism: the decomposition products may either diffuse through the epoxy or escape along cracks, which effectively increase the accessible surface area. By comparison, the TEMs remain encapsulated within their polymer shells. As no or only negligible crack formation was observed for TEM-filled specimens, the release of mass is strongly hindered, which explains their comparatively low weight loss during thermal activation.

Edge shear testing served as quantitative basis for assessing the suitability of the dehybridisation concept. Thermal conditioning of the neat epoxy resin alone did not reduce interfacial strength; if anything, the mean shear strength was slightly higher, albeit with a large standard deviation. VHC laminates exhibited a significant but insufficient weakening of the interface. Their interlaminar shear strength decreased by 29%, and microscopy revealed activation-induced porosity within the interphase; however, the interfaces remained largely intact, i.e., the imposed damage was insufficient to trigger widespread delamination or crack propagation. The increased shear strength compared to the neat epoxy might be owing to absorb energy when the matrix fractures, and crack expansion needs to bypass or shear the PS particles, thus inhibiting the crack expansion and achieving toughening [27]. Goodenough [28] investigated the same blowing agent in an epoxy system and reported that contents exceeding 20 wt% led to a reduction in strength, whereas no such decrease was observed for contents below 15 wt%. However, at 15 wt% no sufficient DoD effect was achieved as well. Banea et al. [7] achieved a more efficient result with polyurethane and a similar blowing agent, Expancel 031 DU 40. Their results showed that the activation time and temperature required to reduce the residual shear strength of laboratory shear specimens below 0.2 MPa depend directly on the concentration of blowing agent in the adhesive formulation. Increasing the blowing agent content to 25% reduced the required conditions from 200 s at 230 °C to 60 at 140 °C, while the initial strength decreased by approximately 50% The ADC variant emerged as the more effective route: after activation, ADC laminates could not be assigned an interlaminar shear strength because the specimens were nearly fully delaminated upon removal from the oven. Micrographs in Fig. 12 corroborate this outcome, showing microcrack formation within the interlayer and agglomerates of the expanded blowing agent; gas release during decomposition likely drove debonding at the CFRP–epoxy interface, culminating in complete separation.

This behaviour supports the conclusion that the observed debonding in ADC and the strength loss in VHC originate from blowing-agent activation rather than from the thermal cycle itself. The high scatter in the conditioned neat epoxy resin set may stem from ageing of the two-component adhesive at elevated temperatures (cf. datasheet), which could introduce variability in interfacial properties. A practical trade-off emerges. Strong interfacial adhesion is desirable in service, but it hinders end-of-life separation. In this study, the laser-structured aluminium interface retained its structural integrity and interfacial bonding capability even after activation, whereas the CFRP–thermoset boundary acted as the preferential release plane (particularly for ADC). Designing for dehybridisation will therefore require balancing in-service adhesion with on-demand separability—e.g., by localising the triggerable weakness to a designated interphase, tailoring blowing-agent type/loading and activation conditions to control gas evolution and microcrack nucleation, and preserving robust adhesion where joints are intended to remain permanent.

Although aluminium–glass fibre laminates are commonly employed to mitigate galvanic corrosion, the aluminium–CFRP configuration was deliberately selected in this study. The presence of a continuous polymer interlayer fully electrically isolates the aluminium from the carbon fibres, thereby effectively suppressing galvanic corrosion. Beyond this, aluminium–CFRP fibre–metal laminates represent a technologically relevant and mechanically demanding hybrid system, particularly for automotive and aerospace lightweight applications. Demonstrating controlled dehybridisation in this system provides confidence that the approach is transferable to other material combinations, such as aluminium–glass fibre laminates.

Srinivasan and Idapalapati summarised four attributes future DoD technologies should focus on [29]. The first two points are “cost of salvageable components” and the “ease of debonding”. It can thereby be posited that the proposed methodology will demonstrate excellent performance, given that the blowing agents are products of low cost and high availability, and the application of heat is a very simple process. The third point to be considered is the time taken for disassembly. The current proposed method incorporated heating cycles lasting in the range of one hour; nevertheless, the change of mass and volume exhibited the most significant alterations during the initial three minutes. Consequently, the optimisation of activation parameters will result in a substantial reduction in the necessary time and total energy. The final point pertains to the process of “reconditioning and surface reactivation”. Owing to the predominantly cohesive nature of the crack propagation within the adhesive layer, machining of the residual surfaces will be necessary to ensure optimal surface reactivation.

5. Conclusion

The findings in this study demonstrate the potential of thermally activatable adhesive interlayers as a viable strategy for debonding on demand in fibre–metal laminates. By systematically comparing chemical and physical blowing agents embedded in epoxy matrices, clear distinctions in their activation behaviour and effectiveness were identified. Chemical blowing agents, particularly ADC, induced pronounced microcracking and enabled nearly complete delamination under thermal activation, thereby validating their suitability for targeted weakening of the adhesive interface. The maximum temperatures to which the blowing agents can be exposed during service life or manufacturing are listed in Table 3 as T_{onset}. For ADC, this temperature is 190 °C. Should the temperature exceed this threshold, the integrity of the material may be compromised. In contrast, the tested physical blowing agents primarily induced volumetric expansion and colour changes without sufficient crack formation to trigger macroscopic separation.

Overall, the findings highlight the feasibility of chemical blowing agents into thermoset adhesive layers to enable the recycling and reuse of FML constituents. Future work should focus on optimising the concentration and distribution of blowing agents, exploring alternative chemistries with tunable activation thresholds, and evaluating long-term durability and scalability of the concept in industrially relevant laminate structures.

CRediT authorship contribution statement

Daniel Esse: Writing – review & editing, Visualization, Validation, Software, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **Sarah Enzler:** Writing – original draft, Visualization, Validation, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **Anselm Heuer:** Methodology, Investigation, Formal analysis, Conceptualization. **Kay A. Weidemann:** Writing – review & editing, Supervision, Resources, Methodology, Conceptualization. **Anna Trauth:** Writing – review & editing, Supervision, Resources, Methodology, Funding acquisition, Conceptualization. **Wilfried V. Liebig:** Writing – review & editing, Supervision, Resources, Methodology, Funding acquisition, Conceptualization.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the authors used deepL and ChatGPT in order to rephrase sentences and text sections. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

References

[1] A. Vlot, J.W. Gunnink, Fibre Metal Laminates: An Introduction, springer science & Business Media, 2011.

[2] R. Marissen, ARALL (Aramidfaserverstärkter Aluminiumschicht-Verbundwerkstoff)-Ein neuer Hybrid-Verbundwerkstoff mit besonderen Schwingfestigkeitseigenschaften, Mater. Werkst. 14 (8) (1983) 278–283.

[3] T. Sinmazçelik, E. Avcu, M. Bora, O. Çoban, A review: Fibre metal laminates, background, bonding types and applied test methods, Mater. Des. 32 (7) (2011) 3671–3685.

[4] G. Roebroeks, Fibre-metal laminates: Recent developments and applications, Int. J. Fatigue 16 (1) (1994) 33–42.

[5] D. Dachev, M. Kazilas, G. Alfano, S. Omairey, Towards reliable adhesive bonding: A comprehensive review of mechanisms, defects, and design considerations, Materials 18 (12) (2025) 2724.

[6] J.L. Thoma, R. Elsener, I. Burgert, M. Schubert, Chemical and physical debonding-on-demand of poly (urethane urea) thermoset adhesives to facilitate the recycling of engineered wooden products, ACS Appl. Polym. Mater. 6 (10) (2024) 5778–5787.

[7] M.D. Banea, L. Da Silva, R. Carbas, Debonding on command of adhesive joints for the automotive industry, Int. J. Adhes. Adhes. 59 (2015) 14–20.

[8] J. Alcorta, E. Papon, J.J. Villenave, Destructuring agent for an adhesive composition, and glue and primer forming said composition, WIPO (PCT) WO2005028583A1, Dtschl. 31 (2005).

[9] B.K. Fink, S. Yarlagadda, J.W. Gillespie Jr., Design of a Resistive Susceptor for Uniform Heating During Induction Bonding of Composites, Technical report, Defense Technical Information Center, 2000.

[10] M. Sauer, Entwicklung einer Debonding-on-Demand-Konzeptlösung unter Verwendung funktioneller Suszeptor-Additivmaterialien zur Erzeugung von Thermoschocks (Ph.D. thesis), Fraunhofer-Institut für Chemische Technologie ICT, 2016.

[11] A. J., P. E., Patent FR 2 852 965: Procede de demontage des revetements et assemblages colles programme par utilisation de primaires d'adhesion modifies, 2005, FR 2 852 965.

[12] A. Concord, Debonding technologies for adhesive bonded structures (Ph.D. thesis), BTU Cottbus-Senftenberg, 2012.

[13] J.H. Aubert, Note: Thermally removable epoxy adhesives incorporating thermally reversible diels-alder adducts, J. Adhes. 79 (6) (2003) 609–616.

[14] D. Haydon, ElectRelease–electrically disbonding epoxy adhesive, Assem. Autom. 22 (4) (2002) 326–329.

[15] P. Schleevoigt, J. Leuthäusser, Neues Konzept für ein leichteres Lösen von Klebverbindungen, Adhäsion KLEBEN DICHTEN 50 (7) (2006) 24–28.

[16] A.M. Schenzel, Advanced debonding on demand systems for dental adhesives (Ph.D. thesis), Dissertation, Karlsruhe, Karlsruher Institut für Technologie (KIT), 2017, 2017.

[17] A. Concord, G. Wachinger, H. Luinge, E. Langkabel, M. Bauer, Klebstoff-Zusammensetzung fuer loesbare Klebeverbindungen und Modifikation der Verkapselungsmaterialien fuer gezielte Energieeinbringung, 2009.

[18] C. Zinn, M. Schaper, J.S. Gonzalez, D. Meiners, Z. Wang, T. Troester, F. Pottmeyer, K.A. Weidenmann, Shear edge tests: a benchmark in investigating the influence of different surface pretreatment methods on the shear stress of intrinsically manufactured metal-CFRP hybrids, Int. J. Automot. Compos. 2 (3–4) (2016) 244–271.

[19] M.M. Stoll, V. Sessner, M. Kramar, J. Technau, K.A. Weidenmann, The effect of an elastomer interlayer thickness variation on the mechanical properties of Fiber-Metal-Laminates, Compos. Struct. 219 (2019) 90–96.

[20] V. Sessner, Charakterisierung und Modellierung des Dämpfungsverhaltens von hybriden Faser-Metall-Elastomer-Laminaten, 2021.

[21] B.A. Hunter, D.L. Schoene, Sulfonyl hydrazide blowing agents for rubber and plastics, Ind. Eng. Chem. 44 (1) (1952) 119–122.

[22] S. Quinn, Chemical blowing agents: providing production, economic and physical improvements to a wide range of polymers, Plast. Addit. Compd. 3 (5) (2001) 16–21.

[23] M. Emami, E. Takacs, J. Vlachopoulos, Rotational foam molding of metallocene catalyzed polyethylene: CBA screening and process characteristics, J. Cell. Plast. 46 (4) (2010) 333–351.

[24] F. Li, Q. Zhang, S. Jiao, Z. Sun, J. Wen, X. Du, R. Liu, L. Li, Preparation, characterization and foaming performance of thermally expandable Microspheres, Mater. Res. Express 7 (11) (2020) 115302.

[25] M.C. Celina, A.R. Dayile, A. Quintana, A perspective on the inherent oxidation sensitivity of epoxy materials, Polymer 54 (13) (2013) 3290–3296.

[26] M. Najafi, R. Ansari, A. Darvizeh, Environmental effects on mechanical properties of glass/epoxy and fiber metal laminates, Part II: Isothermal aging, Mech. Adv. Compos. Struct. 4 (3) (2017) 197–205.

[27] C. Li, Y. Liu, Z. Chen, Study of mechanical properties of micron polystyrene-toughened epoxy resin, Appl. Sci. 13 (6) (2023) 3981.

[28] J.E. Goodenough, Development and testing of a debond-on-demand polyurethane adhesive through induction heating (Ph.D. thesis), University of Bristol, 2024.

[29] D.V. Srinivasan, S. Idapalapati, Review of debonding techniques in adhesively bonded composite structures for sustainability, Sustain. Mater. Technol. 30 (2021) e00345.