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Identification of failure mechanisms of metallised glass fibre reinforced composites under tensile loading using acoustic emission analysis

E. Njuhovic [a], M. Bräu [a], F. Wolff-Fabris [b], K. Starzynski [c], V. Altstädt [a, *]

[a] University of Bayreuth, Department of Polymer Engineering, Universitaetsstrasse 30, 95447 Bayreuth, Germany
[b] SKZ - KFE gGmbH, Weissenbacher Strasse 86, 95100 Selb, Germany
[c] Lüberg Elektronik GmbH & Co. Rothfischer KG, Hans-Striegl-Strasse 3, 92637 Weiden, Germany

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ABSTRACT

In this work, failure mechanisms of metallised glass fibre reinforced epoxy composites under tensile loading were investigated using acoustic emission analysis. Sandblasting with Al_2O_3 was used to pre-treat the composite surface prior to metallisation, and therefore to improve adhesion. The sand-blasting time was varied from 2 s to 6 s. A two-step metallisation process consisting of electroless and subsequent electroplating was used for depositing the copper coating on the pre-treated composite surface. The mechanical pre-treatment had no significant negative effect on the mechanical properties of the composite laminate. The acoustic emission (AE) from the metallised composite was recorded during tensile testing in order to investigate the failure mechanisms. AE-Signals were analysed using pattern recognition and frequency analysis techniques. A correlation between the cumulative absolute AE-energy and the mechanical behaviour of uncoated and coated specimens during tensile testing was successfully observed. It was shown that a stronger adhesion between substrate and coating leads to a lower release of mechanical elastic energy, which could be recorded by means of AE analysis. Furthermore, differences in peak frequency, frequency distribution and the use of pattern recognition techniques allowed classifying the signal into three failure mechanisms for the uncoated samples and four failure mechanisms for the coated samples, namely matrix cracking, fibre-matrix interface failure, fibre breakage and substrate-coating interface failure. Waveform and frequency analysis of the classified signals supported the identification of the failure mechanisms. Furthermore, optical investigation and SEM images of the tested samples and fracture surfaces confirmed the identified mechanisms evaluated by acoustic emission analysis.

1. Introduction

Cryogenic fluids such as liquid helium (LHe, $T = 4.2$ K), liquid oxygen (LOX $T = 90$ K) or liquid hydrogen (LH_2, $T = 20$ K) are of special interest as energy carrier for aerospace applications due to the much higher gravimetric energy density compared to gaseous and solid stored hydrogen or helium as well as other conventional fuel systems [1]. These cryogenic fluids are typically transported and stored in steel, titanium or aluminium tanks [2]. The use of fibre-reinforced polymers (FRP) instead of metals for the construction of such cryogenic tanks could lead to major mass savings. Studies show that a weight reduction of approximately 60% could be achieved [2].

However, due to the permeable nature of FRP, an extra impermeable barrier is needed to prevent the loss of the respective cryogenic liquids. A metal coating on the surface of the FRP is therefore required as permeation barrier in order to fulfil the strict requirements [1,2].

Suitable coating processes of FRP are vacuum-metallisation (e.g. PVD or CVD), indirect metallisation (e.g. hot foil stamping) and plating processes (e.g. electroless/electrolytic plating) [2–4]. Hot foil stamping is a suitable and economically viable method for relatively simple 2D geometries [3]. However, it cannot be

* Corresponding author. Tel.: +49 921 557471; fax: +49 921 55 7473.
E-mail addresses: edin.njuhovic@uni-bayreuth.de (E. Njuhovic), f.wolff-fabris@skz.de (F. Wolff-Fabris), krzysztof.starzynski@lueberg.com (K. Starzynski), altstaedt@uni-bayreuth.de (V. Altstädt).

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employed for the manufacture of cryogenic storage systems. In the case of these complex shaped 3D parts, plating process is the most suitable coating process mainly due to faster deposition rates, higher ductility of the coatings and lower process temperatures compared to PVD or CVD processes [5].

Independently of which process is selected to coat the FRP with the metallic barrier, it is generally very difficult to achieve a consistently high adhesive strength between the composite and the coating materials [6]. Especially in case of the plating process this challenging issue is due to the much lower polarity of the polymer surface in comparison to the coating material as well as due to the low electrical conductivity of the composite (~10^{-8} S/m) compared to metals (~10^{6} S/m). A high electrical conductivity is a requirement for a successful plating process [7,8]. As consequence of a weak adhesion, the coating can detach from the FRP surface leading to a significant increase of permeability [9].

To increase the adhesion of the coating layer to the polymer substrate, surfaces are often treated to increase the surface roughness (mechanical adhesion) or to modify the surface energy to increase wettability and adsorption [3,10]. In both cases the surface is modified by pre-treatment processes, which can be generally classified as mechanical, chemical or electrical pre-treatments. A detailed description of pre-treatment processes and their mechanisms and effects on their surface structures are presented elsewhere in the literature [9].

Considering the final application, the metallised composite is exposed to extensive mechanical loads during operation due to the high temperature difference and elevated pressure inside of the tank. These loadings, which induce mainly tensile and compressive stresses, could lead to delamination of the metallic coating in case of insufficient adhesion. Tensile stresses are considered to be more critical than compressive stresses [1]. The correct characterisation of the material's mechanical behaviour, especially regarding the failure mechanisms within the composite and at the interface between the substrate and the coating, is therefore of major importance. Uniaxial tensile tests are one possibility for investigating the tensile stresses occurring in the metallised composite. However, the tensile test only measures the external force until final failure providing very limited information about damage evolution and propagation. For instance, it is not possible to clearly identify which components are being damaged and how crack propagation develops. In particular, it is not possible to determine the load level at which first microcracking occurs within the material [11]. Typical methods for the qualitative characterisation of failure mechanisms in the composite and at the interface after tensile testing are light microscopy and scanning electron microscopy. The fracture surface and cross section of tested specimen can be used to examine fibre–matrix adhesion as well as crack propagation. One method that may be used to identify and quantitatively characterise failure mechanisms of coated FRP under mechanical loads is the acoustic emission (AE) analysis. Acoustic emission analysis is a powerful method to investigate materials deforming under stress. Next to real-time capability, it also enables volume monitoring and it has a high sensitivity to any process or mechanism, which generates sound waves, such as mechanical failure [12].

AE analysis has been successfully applied for health monitoring of pressurised carbon fibre reinforced polymer (CFRP) vessels focussing on CFRP failure [13,14]. In addition, several authors investigated damage accumulation in plasma sprayed coatings on metallic substrates [15–17]. First successful results on damage investigation of metallised FRP under mechanical loading (four point bending) using AE analysis were presented by Sause et al. [18–20]. In these investigations Sause focused exclusively on AE-signals arising from the failure within the metallic coating. In a previous work of our group, we showed that different interface failure mechanisms of a metallised composite under peel testing could also be successfully identified by AE analysis [9].

To our best knowledge studies on the characterisation and identification of failure mechanisms of metallised composites during tensile testing have not been reported in the literature so far. The present study focuses on the identification of failure mechanisms of copper electroless/electroplated fibre-reinforced epoxy composites under tensile testing using acoustic emission analysis. Our objective is to closely look on AE arising from the copper coated composite investigating the effect of increased peel strength and the adhesion of the coating respectively. This study presents a correlation between the surface properties of the substrates and accumulated emission energy, as well as correlation between adhesion and damage evolution until final failure.

2. Experimental

2.1. Substrate material

In this study a glass fibre reinforced polymer (GFRP) consisting of E-glass fibres (unidirectional non-crimp fabric from Saertex) with an areal weight of 701 g/m^2 and a toughened epoxy resin as matrix (XU3508/XB3486 from Huntsman) were used. The GFRP laminates with 4 layers of $(+45/-45)_s$ were manufactured by VARTM-process in a 1-part machine setup with a two-sided hard mould. The release agent Loctite Frekote 770-NC was thoroughly applied on the mould surfaces before injection. The laminate thickness of 2 mm corresponds to a fibre volume content of approximately 54%. The laminates were cured at 100 °C for 5 h according to the resin manufacturer's datasheet.

2.2. Surface pre-treatment

The GFRP surfaces were pre-treated prior to metallisation of the material. The method used in this study was sandblasting with aluminium oxide (200–300 μm grit size and a mohs hardness of 10). To identify failure mechanisms in the metallised composite under mechanical loading via acoustic emission, different surface topographies are produced by varying the blasting time (2 s, 4 s and 6 s). In previous studies [9,21] the influence of the composite surface structure on the peel strength and its influence on the adhesion mechanisms under peel using acoustic emission analysis were thoroughly investigated. Furthermore it was also shown that the sandblasting process does not lead to a significant decrease of the mechanical properties of the composite plate. The sandblasting machine ST 1200 ID-Z-SB with a die diameter of 10 mm is used to perform the tests. Constant parameters are blasting distance of 500 mm, blasting pressure of 3 bars and a blasting angle of 90°. All plates including the reference laminate (without surface pre-treatment) were cleaned using an ultrasonic bath with equal parts of ethanol and water for 30 min at 25 °C prior to the coating process.

2.3. Coating process

The GFRP substrates were coated by the electroless/electrolytical plating process. Direct electrolytical plating of GFRP is impossible due to the electrical insulation of the polymer matrix. On account of this, a thin adherent conductive layer was chemically deposited on the GFRP surface. The substrate was dipped into an aqueous solution consisting of a stabilised Pd–Sn colloid. Palladium needs to be protected in order to prevent agglomeration and drop out [22]. In an accelerator bath the enclosed Pd ions are broken free to leave palladium on the surface. At molecular level, the single palladium atoms are not homogeneously dispersed on the surface