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Identification of interface failure mechanisms of metallized glass fibre reinforced composites using acoustic emission analysis

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ABSTRACT

In this work, interface failure mechanisms of metallized glass fibre reinforced epoxy composites were investigated using acoustic emission analysis. Sandblasting with Al_2O_3 was used to pre-treat the composite surface. The sandblasting time was varied from 2 s to 6 s. A two-step metallization process consisting of electroless and subsequent electroplating was used for depositing the copper coating on the pre-treated composite surface. A significant increase in adhesion strength was obtained due to the sandblasting pre-treatment. SEM and light microscopic investigations confirmed the results of the surface roughness and peel strength. The acoustic emission (AE) from the coating-substrate system was recorded during peel testing to characterize interfacial failure. AE-Signals were analyzed using pattern recognition and frequency analysis techniques. A correlation between the cumulative absolute AE-energy and the surface roughness/peel strength was successfully observed. It was shown that the absolute AE-energy is sensitive to changes in the surface topography and therefore peel strength, and the method is thus suitable for evaluating the peel strength of copper coated glass fibre reinforced composites. Furthermore, two different failure mechanisms could be correlated with the results from AE signal analysis, namely adhesive and cohesive failure. Differences in peak frequency, frequency distribution and the use of pattern recognition techniques allowed classifying the recorded signals. The classified failure mechanisms were confirmed by light microscopic images.

1. Introduction

Cryogenic fluids such as liquid helium (LHe, $T = 4.2$ K), liquid oxygen (LO_x, $T = 90$ 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 FRPs, 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-metallization (e.g. PVD or CVD), indirect metallization (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 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 ($\sim 10^{-8}$ S/m)

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compared to metals ($\sim 10^6$ 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].

The correct characterization of the adhesive strength is of major importance for the work focusing on the optimization of the interface between the substrate and the coating, for which several methods are commonly used. The adhesive strength between electroplated coatings and substrate is usually determined via peel or pull-off tests [11]. In case of peel test many of the criteria of the ideal adhesion test are met. The rate of delamination and the locus of failure can be controlled fairly precisely. This stems from the fact that a very high stress concentration exists at the point where the coating just lifts off the substrate. This tends to narrowly focus the failure region very close to the geometric interface between coating and substrate, which is the region of most interest in any adhesion test. Furthermore, the peel test readily lends itself to use under conditions of controlled temperature and environment [12].

However, the peel test only measures the external force, which is needed to separate the coating from the substrate. No information of failure mechanisms is present to determine weather adhesive or cohesive failure dominates. Furthermore the peel test does not take into account residual stress present after coating application, which can dramatically decrease the load limit of the coating-substrate system [13]. Typical methods for the characterization of failure mechanisms at the interface are light microscopic investigations, scanning electron microscopic investigations and X-ray based measurement techniques (e.g. EDX/WDX). In case of microscopic investigations the fracture surface of tested specimen can be used to visualize adhesive and cohesive failure whereas EDX (Energy-dispersive X-ray spectroscopy)/WDX (Wavelength-dispersive X-ray spectroscopy) analysis helps to qualitatively evaluate the failure mechanisms.

One method that may be used to characterize quantitatively and identify 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 [14].

In the past AE analysis has been applied for health monitoring of pressurized carbon fibre reinforced polymer (CFRP) vessels focusing on CFRP failure [15,16]. In addition several authors investigated damage accumulation in plasma sprayed coatings on metallic substrates [17–19]. First successful investigations on damage behaviour of metallized FRP under mechanical loads (four point bending) using AE analysis were performed by Sause et al. [11,20,21]. In these investigations Sause focused exclusively on AE-signals arising from the failure within the metallic coating.

To our best knowledge studies on the characterization and identification of interface failure mechanisms between the composite substrate and metallic coating during peel testing have not been reported in the literature so far. This study focuses on the identification of interface failure mechanisms of copper electroless-/electroplated fibre-reinforced epoxy composites under peel testing using acoustic emission analysis. Our objective is to closely look on AE arising from the interface between the composite substrate and copper coating. Here we describe the detection of failure initiation by acoustic emission analysis. Stress waves released from microscopic structural changes, which typically originate from crack progress, were detected as a function of the peel strength and surface roughness and attributed to different failure mechanisms. This study presents a correlation between the surface properties of the substrates and accumulated emission energy, as well as correlation between adhesion and identified failure mechanisms.

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 application of release agent Loctite Frekote 770-NC was done thoroughly on the mould surfaces as mould preparation 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 metallization 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 interface failure mechanisms via acoustic emission different surface topographies are produced by setting the blasting time (2 s, 4 s and 6 s). In a previous study [9] the influence of the composite surface structure on the peel strength was 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 stabilized 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 but they create clusters of molecular size. Nevertheless they are packed enough in order to provide a homogeneous Cu layer. After this activating process, a 1 μm thick copper coating was deposited electrolessly on the surface and finally electrolytically plated with the same coating material. The final coating thickness was at least 50 μm. A rigorous surface preparation procedure was employed in this study. The electroless/electroplating process