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# Ageing of corrosion resistant steel/rubber/composite hybrid structures

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ABSTRACT

One of the major challenges when preparing reliable hybrid structures is the adhesion between different components. Besides enduring the specific stress state, hybrid structures should maintain the required properties in the service environment without degradation. In this study, the environmental resistance of stainless steel/rubber/GFRP (glass fibre reinforced plastic) hybrid structures were tested by exposure to hot, moist and hot/moist environments and after the ageing by peel testing. Two different stainless steel surface finishes and two different rubber grades were investigated. The results were compared with the properties of a mild steel/rubber/GFRP structure. Both mild steel/rubber and composite/rubber structures are used in industrial applications, such as in vibration damping devices and in automotive components.

The peel tests showed that with right rubber compounds, stainless steel/rubber and GFRP/rubber interfaces can maintain their properties even in harsh hot/moist environments to such an extent that the interfacial strength of the joint is higher than the cohesive strength of the rubber. This enables the use of rubber's cohesive fracture properties instead of the substrate/rubber interfacial properties when estimating the strength of the steel/rubber/GFRP hybrid structure. In addition, based on the current study, time-consuming stainless steel pre-treatments are not needed but the stainless steel can be in the as-received stage. According to the chemical analysis even before and after the harsh hot/moist exposure used, none of the studied rubber grades had degraded. Thus, we conclude that it is possible to manufacture environmental resistant stainless steel/GFRP hybrid structures with the aid of EPDM rubbers.



## 1. Introduction

Last decade has shown an increasing interest on polymer/metal hybrid materials and structures in different fields of industry [1]. The high specific properties, i.e. properties divided by the material density, of hybrids offer mainly benefits through weight savings [1]. However, other advantages, such as more beneficial manufacturing methods or improved damping properties, are achievable by these structures as well [2].

Commonly in adhesive bonded polymer/metal structures, different chemical or mechanical pre-treatments are required for the metal surface before joining the polymer by the adhesive [3,4]. The pre-treatment steps are time-consuming and may require the use of hazardous chemicals. Thus the manufacturing method would be highly improved, if an adhesive enabling the use of the metal surface in an as-received stage would be available.

It would be tempting to integrate rubber in a hybrid structure for its good characteristics: rubbers can be compounded to be easily adhered to inorganic and organic materials [5,6] and their damping properties can be tailored. However, the typical mechanical properties of rubbers, namely low modulus and high extensibility [7], do not favour their use as the main component in structural applications. Instead, rubbers can be used as adhesives in polymer/metal hybrid structures simultaneously improving, e.g. the vibration attenuation properties of the structure. The previous studies of the authors [8,9] show, that a thin ethylene propylene diene (EPDM) based rubber layer between stainless steel and glass fibre reinforced epoxy (GFRP) composite enables the use of a simple manufacturing method without substrate pre-treatments and leads to a good contact and adhesion between the rubber and the substrates, as well as to improved damping properties. Thus, the studied stainless steel/rubber/GFRP hybrid structure has a simple manufacturing method and properties which could be utilized in several applications, such as in impact loaded stressed-skin constructions.

The durability of adhesive bonds is more dependent on the environmental resistance than on the fatigue resistance of the joint and in general the fatigue resistance of adhesive joints is superior when compared with mechanically fastened joints [10]. Thus the ageing performance of an adhesive joint is an important topic to be studied before the implementation of the structure in applications. Often when heat and humidity are present in the

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service atmosphere, rubber/metal interfaces tend to fail at the rubber/bonding agent interface, although they have shown cohesive failure in laboratory tests [11]. Within rubber/metal interfaces, the tyre cord/rubber adhesion and its resistance to different environments is widely studied (e.g. [12–14]) and interfaces are shown to degrade due to heat and moisture. Similarly, other polymer/metal interfaces are prone to moist environments and exhibit a significant decrease in interfacial strength after the ageing [15]. Thus, the suitability of the studied stainless steel/rubber/GFRP structures for real life applications has to be verified by environmental testing even though our adhesion studies [8] showed a good adhesion level for non-exposed samples.

Any established general practices for testing the environmental resistance of polymer/metal interfaces do not exist. Instead, it has been studied in various conditions depending on the material combination and application in question. Carbon fibre reinforced epoxy composite/EPDM rubber structure's strength has been investigated after thermal ageing at 100 °C and after hot/moist ageing at 70 °C and 100%RH [16]. Natural rubber/tyre cord strength has been studied after exposure to heat (70 °C), heat and moisture (70 °C, 96%RH) and water immersion at the temperature of 70 °C [12]. Epoxy/copper strength has been tested after heat (85 °C) and four different levels of moisture: ambient atmospheric conditions, 50%RH, 65%RH and 85%RH [17]. The combination of the 85 °C temperature and the 85%RH humidity is also used in the steady state temperature humidity bias life tests for packaged devices [18]. Also the ASTM standard for the hydrolytic stability of rubbers [19] instructs the temperature of 85 °C above a water container, which is close to the humidity of 85%RH. Thus, the 85 °C/85%RH condition has been chosen for this study as well.

In the present study, the environmental resistance of stainless steel/EPDM rubber/GFRP structures is tested after the exposure of the samples for hot, moist and hot/moist environments. In practice, this is done by testing the two interfaces of the structure separately. Two different stainless steel surface finishes, an industrial surface finish and a sand blasted one, as well as two different EPDM based rubbers are used in the studies for the stainless steel and GFRP substrates. The results are compared with the results of a mild steel/EPDM rubber/GFRP system since such structures are used in industrial applications, e.g. in vibration damping devices [20] and in automotive components [2]. The effects of the ageing environments on the interfaces of the hybrids are investigated by peel tests, microscopy, Fourier transform infrared analysis and thermogravimetric analysis.

## 2. Experimental

In the present study, the environmental resistance of steel/rubber and GFRP/rubber interfaces were investigated. Two steel grades, stainless steel AISI 304 (Outokumpu Stainless Oy, Finland) and cold rolled mild steel EN 10130 DC01 (Rautaruukki Oyj, Finland) were studied. The mild steel was passivation treated as is customary for grades used as industrially coated. The aim of the passivation treatment is to enhance the adhesion properties of the steel but the procedure is not public. For the stainless steel grade, two different surface finishes were used, namely the as-received cold rolled, heat treated and pickled (2D) industrial surface finish and the same surface with an additional sand blasting step (SB). The surface finish 2D is defined in the standard EN 10088-2. The 2D surface was chosen for the as-received surface finish, because it showed the best adhesion strength among the different as-received surface finishes in preliminary tests [8]. The thickness of the steel sheets was 0.5 mm, but a thicker metal stiffener was glued on the back side of the metal component to prevent its bending during peel testing. The material combinations used in this study are summarized in Table 1. The sand blasting media was aluminium oxide (grit 36, average particle size 483 μm). A more detailed study of these steel surfaces can be found in [8].

**Table 1**
The used substrates, their average profile roughness parameters ($R_a$) measured with laser profilometer [8] and the studied material combinations.

| *Substrate* | | *$R_a$ [μm]* [8] | *Rubber* |
|---|---|---|---|
| *Code* | *Surface treatment* | | |
| *2D* | *Cold rolled, heat treated, pickled AISI 304* | 0.38 | *A* |
| *SB* | *Sand blasted 2D surface of AISI 304* | 2.46 | *A* |
| *GFRP* | *HexForce® T470 peel ply* | 23.51 | *A* |
| *2D* | *Cold rolled, heat treated, pickled AISI 304* | 0.38 | *B* |
| *SB* | *Sand blasted 2D surface of AISI 304* | 2.46 | *B* |
| *GFRP* | *HexForce® T470 peel ply* | 23.51 | *B* |
| *CR* | *Cold rolled, passivation treated EN 10130 DC01* | 0.43 | *C* |
| *GFRP* | *HexForce® T470 peel ply* | 23.51 | *C* |

The glass fibre reinforced plastic (GFRP) composite was manufactured in-house by vacuum infusion from stitched 0/90 E-glass fibre fabrics (682 g/m$^2$, Ahlstrom Oyj, Finland) and Sicomin SR 1660/SD 7820 epoxy. The thickness of the GFRP sheets was 3.5 mm and its fibre content was about 45 vol%. A metal stiffener was glued on the back side of the GFRP sheets to prevent its bending during peel testing. The heat resistant epoxy was chosen to provide the resistance of the GFRP sheet to the vulcanising temperature of the rubber (varying between 130 and 160 °C for the different rubber grades). From the adhered GFRP surface, a HexForce® T470 (Hexcel Co., USA) peel ply was removed prior rubber attachment.

The EPDM based rubbers adhered to the steel and composite surfaces were manufactured by Teknikum Oy, Finland (grade A) and by Kraiburg GmbH, Germany (grades B and C). The grade A has a trade name Teknikum TRA10 and its ingredients are EPDM rubber, ZnO, stearic acid, polyethylene wax, carbon black, paraffin oil, internal adhesion promoter and peroxide. The grade B is also designed for stainless steel whereas the grade C is designed for mild steels. The main components of the rubbers B and C are EPDM rubber, silica (rubber B) or carbon black (rubber C), paraffin oil, internal adhesion promoters, silane, curing promoters, and peroxide.

The steel/rubber and composite/rubber hybrids were manufactured by vulcanising the rubber to the substrates. The steel surfaces were rinsed with ethanol and acetone and the peel-ply was removed from the composite surfaces just before the rubber bonding but no other pre-treatments for the composite surface were done. A uniform rubber thickness of 2 mm was ensured during the manufacturing of the laminates. A more detailed description of the manufacturing steps of the hybrids can be found in [8]. The peel test samples (size 100 × 12 mm) were cut from larger steel/rubber and GFRP/rubber laminates by water jet cutting.

The environmental resistance of the structures was tested by exposure to isohume (25 °C, 85%RH), isothermal (85 °C, ambient atmospheric conditions) and hygrothermal (85 °C, 85%RH) environments and after the ageing by peel testing. The running time of the exposure tests was 500 h. The EPDM rubber should endure the aforementioned environments without degradation [21]. Between ageing and testing, the samples were stabilised for 72 h in 23 °C and 50%RH.

The adhesion of the steel/rubber and GFRP/rubber interfaces were studied by a floating roller peel test configuration (Fig. 1). The floating roller peel test geometry introduces a constant peel angle of 45° which is shown to be the most convenient to study the adhesion between steel and rubber [22]. The peel tests were