International Journal of Adhesion & Adhesives 49 (2014) 51–57

Contents lists available at ScienceDirect

International Journal of Adhesion & Adhesives

journal homepage: www.elsevier.com/locate/ijadhadh

Adhesion properties of novel corrosion resistant hybrid structures

E. Sarlin [a,*], E. Heinonen [b], J. Vuorinen [a], M. Vippola [a], T. Lepistö [a]

[a] Department of Materials Science, Tampere University of Technology, P.O. Box 589, 33101 Tampere, Finland
[b] Center of Microscopy and Nanotechnology, University of Oulu, P.O. Box 7150, 90014 Oulu, Finland

ARTICLE INFO

Article history:
Accepted 4 November 2013
Available online 10 December 2013

Keywords:
Hybrid structure
Adhesion
Floating roller peel test
FIB
TEM

ABSTRACT

Hybrid structures are widely used since they enable unique combinations of properties for the final structure. Within polymer/metal hybrids, stainless steel instead of more regularly used mild steel or aluminium is a tempting choice but it is difficult to join adhesively. Thus improved solutions for the stainless steel joining with polymeric materials are needed. This study concentrates on the idea of utilisation of thin rubber layer as an adhesive between stainless steel and glass fibre reinforced epoxy composite (GFRP). Both mild steel and stainless steel with different surface finishes together with GFRP laminates were used as substrates for the ethylene propylene diene (EPDM) based rubber. The adhesion and microstructure of the interfaces were characterised. Transmission electron microscopical (TEM) studies indicated that a close contact between the components can be achieved and thus high quality interfaces are created by vulcanising the rubber to the steel or GFRP surface without pre-treatments. Stainless steel/rubber/GFRP hybrid structures where the strength of the steel/GFRP joint is defined by the cohesive strength of the rubber can be manufactured, as seen from the results of the peel tests. Since the fracture is located inside the rubber and not at the interface, the prediction of the structure's behaviour is also easier.

1. Introduction

Hybrid materials are increasingly employed in several fields of industry. In a hybrid structure, the advantages of different materials can be combined and properties unattainable by any existing single material can be achieved. In addition to light weight, attractive solutions can be achieved through improved performance and more economical manufacturing processes. Thus, hybrid structures can solve a wide range of design challenges faced today.

Typically durable interfaces within hybrid structures are challenging to manufacture due to different physiochemical properties of the components from different material groups, such as metals, polymers or ceramics. Several different bonding methods are used within hybrids [1] and the selection of the constituent materials and the structure geometry, for example, are factors affecting the choice of the bonding method. For laminar structures of two rigid components, adhesive bonding is the most obvious choice. An example of the advantages of adhesive bonding over mechanical fastening or welding is the possibility of reducing internal stress concentrations between materials with very different moduli [2].

In adhesive bonding, special surface treatments and adhesives are applied. Both chemical and mechanical surface pre-treatments are used to prepare the surfaces to be adhered responsive to the adhesive. Chemical surface treatments, such as etching or anodising, used for metal surfaces before polymer joining [3,4], may include the use of hazardous chemicals and solvents, whereas mechanical surface treatments are typically time consuming. The selection of the applied adhesive is done according to the adhered materials and the mechanical and environmental requirements of the application. In addition to structural adhesives, coupling agents, like silanes, or coatings can also be used [1,3,4]. Thus, adhesive bonding includes numerous stages. Another drawback of the adhesive method is that it may lead to poor predictability of the joint durability and failure modes compared with mechanical fastening or welding [2].

To simplify the manufacturing of adhesive bonds and to make it more cost-effective, the number of the operation stages should be kept low. In addition, adequate strength of the joint and especially its predictability have to be ensured. To overcome these challenges, new methods and material combinations for adhesive bonding have to be studied.

Stainless steel is a tempting choice for the metal component in corrosion resistant metal/polymer hybrid structures for a load bearing application due to its good corrosion resistance and mechanical properties. However, joining polymeric materials with stainless steel sheets is difficult and requires the application of

* Corresponding author. Tel.: +358 408490146.
E-mail address: essi.sarlin@tut.fi (E. Sarlin).

0143-7496/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ijadhadh.2013.12.009

pre-treatments and adhesives [3], which complicates the manufacturing. As an attempt to simplify the manufacturing process without a loss of the adhesion properties or an increase in the manufacturing costs in relation to the gained benefits, we framed the question if it is possible to replace the conventional adhesives with a new solution.

Rubbers can be modified with additives to be adaptive for both metallic and polymeric materials [5,6]. They are used in composite and hybrid structures together with mild steel, aluminium, plastics, fabric, and cords to increase strength, minimise internal stress concentrations, and simplify mounting of the structure [7]. In addition, added value for the structure, such as improved energy absorption properties, can be achieved by the inclusion of rubber. However, stainless steel/rubber combinations are rather uncommon. In this study, we suggest that a stainless steel/composite hybrid structure, which would otherwise be difficult to manufacture, could be formed by vulcanising a thin rubber layer between the steel and composite sheets. The application of rubber would not only simplify the manufacturing process but also introduce improved dynamic properties to the hybrid structure.

In the present study, the structure and the adhesion properties of steel/rubber/glass fibre reinforced epoxy composite (GFRP) hybrid structures are characterised. Both mild and stainless steel sheets are used in the hybrid together with compatible ethylene propylene diene (EPDM) based rubber and GFRP sheets. In addition, five different surface treatments for the stainless steel were used to study the effect of the steel surface topography on the adhesion strength. Scanning and transmission electron microscopies are used to characterise the components and the structure. The adhesion properties between the rubbers and the steel sheets as well as GFRP sheets are investigated by floating roller peel tests.

2. Materials and methods

In the present study, the adhesion properties of steel/rubber and composite/rubber interfaces were studied. Two steel grades, cold rolled mild steel EN 10130 DC01 (Ruukki Metals Oy, Finland) and stainless steel AISI 304 (Outokumpu Stainless Oy, Finland), were used. The mild steel was passivation treated as it 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. Five different surface treatments for the stainless steel (Table 1) were used to investigate the effect of the steel surface topography on the adhesion properties. The as-received surfaces 2B, 2D and 2J are also defined in the standard EN 10088-2. The industrially polished (IP) surface mentioned in Table 1 is a stainless steel sheet with 2D surface finish which is electrolytically polished by SpecialSteelStudio (Finland). The sand blasting medium used for the SB samples was aluminium oxide (grit 36, average particle size 483 μm). 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 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 resist the vulcanising temperature of the rubber. From the adhered GFRP surface, a HexForce® T470 (Hexcel Co., USA) peel ply was removed prior to 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). 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. Grade B is also designed for stainless steel whereas grade C is designed for mild steels. The main components of 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.

Prior to the rubber bonding, the substrates were studied with a Laser profilometer (UBM-Microfocus Compact, NanoFocus AG, Germany). A length of at least 50 μm was used to measure the average surface roughness parameter R_a of the surfaces in two perpendicular directions, longitudinal and transverse to the rolling direction of the steel. In addition, the steel surfaces were investigated with a Field Emission Gun Scanning Electron Microscope, FEG-SEM (Zeiss ULTRAplus, Germany).

The steel/rubber and composite/rubber samples were manufactured by vulcanising the rubber to the substrate under heat and pressure. The steel surfaces were rinsed in ethanol and acetone prior rubber bonding; otherwise they were in the as-received stage. Any pre-treatments for the composite surface were not done after the removal of the peel ply. The vulcanising conditions are listed in Table 2. The vulcanising of the rubbers was done according to the manufacturers' guidelines, except for the GFRP/rubber A samples, for which the lower vulcanising temperature was compensated with longer vulcanisation time. To ensure that a high enough degree of vulcanisation is reached during GFRP/rubber A sample vulcanisation, differential scanning calorimetry scans (DSC 204 F1, Netzsch, Germany) were done for rubber A.

Methods to study the adhesion of rubber to rigid substrates are standardised [8]. The ASTM D429 standard introduces six different test methods, one of which (method B) is intended for determining the adhesive strength of rubber/metal bonding. In this test method, the rubber is peeled from the substrate at an angle of 90°. However, Cook et al. [9] have found that a peel angle of 45° is optimum for testing rubber/steel adhesion since it leads the fracture into the interface and the cohesive fracture of rubber is minimised. Thus a floating roller peel test configuration (Fig. 1) which provides a constant peel angle of 45° was used. The peel tests were performed with a universal mechanical testing machine

Table 1
The studied substrates and their average profile roughness parameters (R_a) measured with the laser profilometer.

Code	Surface treatment	R_a (μm)
CR	Cold rolled, passivation treated EN 10130	0.43
2B	Cold rolled, heat treated, pickled, skin passed AISI 304	0.35
2D	Cold rolled, heat treated, pickled AISI 304	0.38
2J	Dry brushed AISI 304	0.31
SB	Sand blasted AISI 304	2.46
IP	Industrially polished AISI 304	0.39
GFRP	HexForce® T470 peel ply	23.51

Table 2
The vulcanisation conditions of the different sample types.

Sample type	Temperature (°C)	Time (min)	Pressure (MPa)
Stainless steel and rubber A	160	15	1.2
GFRP and rubber A	130	18	1.2
Stainless steel and rubber B	130	20	1.2
GFRP and rubber B	130	20	1.2
Cold rolled steel and rubber C	130	30	1.2
GFRP and rubber C	130	30	1.2