ARTICLE IN PRESS

Theoretical and Applied Fracture Mechanics xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Theoretical and Applied Fracture Mechanics

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

The influence of water on the fracture envelope of an adhesive joint

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A R T I C L E I N F O

Article history:
Received 3 October 2016
Revised 7 December 2016
Accepted 8 January 2017
Available online xxxx

Keywords:
Moisture degradation
Structural adhesive
Fracture toughness
Mixed-mode loading
Open specimen
Accelerated ageing

A B S T R A C T

This research aims at determining the fracture envelope of an adhesive as a function of the water content. The fracture toughness of an adhesive joint was determined under pure mode I, II and mixed mode I + II loadings, in three different environments: dry, aged in salt water and aged in distilled water. The fracture toughness under mode I and II were determined using Double Cantilever Beam (DCB) and End-Notched Flexure (ENF) tests, respectively. The characterization of the fracture toughness under mixed-mode was done using an apparatus capable of applying a wide range of loadings that go from pure mode I to almost pure mode II. To accelerate the diffusion process and obtain a uniform water concentration in the adhesive joint, a modified DCB specimen (ODCB specimen) was adopted. Finite Element (FE) analysis was used to determine the gradient of water concentration in both specimens and to validate the use of the modified DCB specimens, comparing the fracture toughness obtained using DCB and ODCB specimens. It was found that the toughness of the adhesive changed as a function of the ageing environment. For the salt water environment, the mechanical properties increased, while for the distilled water environment, degradation of the mechanical properties was observed.

1. Introduction

As an alternative to mechanical joints, the use of adhesive joints has been increasing since they provide several advantages over conventional methods. This can be seen in aerospace, automotive and maritime industries as adhesive joints allow for a uniform stress distribution along the width of the bonded area, enhancing the stiffness, load transmission and fatigue resistance of the structure while reducing the weight and thus the cost [1,2].

This type of joint may be exposed to aggressive environments such as high humidity, extreme temperature or radiation. While fracture mechanics characterization tests for adhesive joints may provide relevant properties to guide the design process, the information available to predict the behaviour of the adhesive after being exposed to aggressive environments is scarce. Therefore, the influence of environmental agents on the mechanical properties of the adhesive should be studied [3].

Water may enter the adhesive joint by Comyn [4]: diffusion in bulk adhesive, transport along the interface, capillary action through cracks and crazes or diffusion through the adherend if permeable. Usually this process can be described with Fick's law of diffusion, where the uptake is a function of time, concentration and thickness [5–9]. However, other models have been developed to describe the diffusion process such as: dual fickian diffusion [8], delayed dual fickian [10] and the Langmuir model [11]. External factors also influence the rate at which water is absorbed and the maximum water uptake, such as temperature [8] and the stress state of the adhesive [12]. Water can act as a plasticizer, reducing the interaction forces between molecules and allowing them to rearrange themselves more easily. As a result, this water uptake can lead to changes on the properties of the adhesive due to the plasticization of the adhesive and adherend, which leads to a change of thermal and mechanical properties, involving: lower rigidity at room temperature, decrease of the glass transition temperature (T_g) [5,9,13] and increase of the strain failure at room temperature [9,14]. However, these changes caused by plasticization can be partially or fully reversed with desorption [9,15]. It has also been reported that the presence of water on the adhesive leads to a reduction of its fracture toughness [3,16,17]. Nonetheless, in some cases, an initial increase due to plasticization effects is observed, followed by a decrease due to degradation [9,16].

Fracture mechanics tests such as DCB, ENF and mixed-mode loadings can be used to assess the influence of water on the

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http://dx.doi.org/10.1016/j.tafmec.2017.01.001
0167-8442/

Please cite this article in press as: P. Fernandes et al., The influence of water on the fracture envelope of an adhesive joint, Theor. Appl. Fract. Mech. (2017), http://dx.doi.org/10.1016/j.tafmec.2017.01.001

adhesive [18]. However, using the standard specimens used in these tests, a long time is required to reach an appreciable level of water concentration. Furthermore, this concentration can also vary in time and space [8,9,16]. As an alternative, some authors used smaller specimens [19] to accelerate the diffusion process, while others modified the specimen by opening it and using a secondary bond [16,18,20]. Based on the second alternative mentioned, a new modified specimen was used in this research, which shortens the diffusion path and avoids the asymmetry of the specimen proposed by Wylde and Spelt [16]. Regardless of the method adopted to accelerate the diffusion process, all the mentioned authors found a reduction in the mechanical properties of the adhesive when it was exposed to aggressive environments for long periods of time. In the particular case of Wylde and Spelt [16], an initial increase of mechanical properties was observed.

In this research, the fracture envelope of a commercial epoxy adhesive used in the automotive industry is characterized as a function of the water content in the adhesive. The fracture characterization of the adhesive joints was performed when the specimens were submitted to pure modes (shear and opening) and mixed mode loadings. Three different environments were tested: dry, a saturated solution of NaCl (salt water) and distilled water. For the dry environment, standard DCB specimens were used, while for the salt water and distilled water environments a modified DCB specimen was adopted.

Furthermore, a FE analysis was performed to determine the gradient of water concentration in the standard DCB specimen, as well as to validate the use of the modified DCB specimens, used to accelerate the ageing process, and predicting the behaviour of the adhesive joint.

2. Experimental details

In order to determine the fracture envelope as a function of water content, DCB specimens standardized by ASTM were used [21]. However, due to the geometry of this specimen, the saturation process would take several years. Thus, in order to accelerate the diffusion process, open-DCB specimens (ODCB) were used, as they are able to replicate the diffusion process that occurs in an adhesive plate.

To be able to compare the influence of water on the fracture envelope using two different specimens, it was necessary to determine the influence of their geometry in the value of the fracture toughness measured experimentally. This analysis was done only for mode I and assumed to be constant for the other modes.

The characterization of the fracture envelopes was done using three loading modes: pure mode I, mixed-mode 55° and mixed-mode 87°. Exceptionally, ENF tests were performed using DCB specimens in a dry environment to determine the G_{IIC} of the adhesive and to be able to input this property in the numerical models.

2.1. Adhesive

The adhesive chosen for this study was SikaPower®-4720 (Supplied by Sika, Vila Nova de Gaia, Portugal) and was used for both DCB and ODCB specimens. It is a two-component high-strength epoxy adhesive specifically designed for metal, particularly aluminium, and composite panel bonding but not intended to be used for body structural parts [22].

The stress-strain curve, as well as the mechanical properties of this adhesive, have been determined previously with tensile tests using bulk specimens [23] (Table 1). On the other hand, the toughness of the adhesive was determined in this research.

Table 1
Mechanical properties of SikaPower®-4720 [23,24].

Property	SikaPower®-4720
Young's modulus, E [MPa]	2170
Tensile strength, σ_{max} [MPa]	25.8
Strain to failure, ε_f [%]	2.7
Shear modulus, G [MPa]	800[a]
Shear strength, τ_{max} [MPa]	14.9[a]
Critical energy release rate in mode I, G_{Ic} [N/mm]	1.15
Critical energy release rate in mode II, G_{IIc} [N/mm]	4.5

[a] Deduced from tensile properties using Von Mises Yield Criterion.

2.2. Ageing environment

The ageing of the adhesive was done by immersing the specimens in a container with either distilled water or salt water (saturated solution of NaCl, which is equivalent to exposure in a 75% RH environment) at 32.5 °C [25,26]. The standard DCB specimens were used to determine the fracture envelope in a dry environment, while the ODCB specimens were used to characterize the fracture envelope in the salt water and distilled water environments.

2.3. DCB specimens

To characterize the fracture envelope, a standard DCB specimen was used, in accordance to ASTM 3433-99 [21]. The adherend's material used for the DCB specimens was aluminium Al7075-T6 supplied by Lanema (Ovar, Portugal). Aluminium was chosen over steel due to the ageing environment, as when exposed to distilled or salt water, the steel adherend would be corroded. This situation can be completely avoided by using phosphoric acid anodized aluminium instead. The choice of this particular aluminium alloy (Table 2) was based on its yield strength, which is high enough to avoid any plastic deformation during the tests.

2.4. ODCB specimens

Open-faced specimens have been used in the past to accelerate the diffusion process [16,18], but this leads to an asymmetric adhesive joint. In this research, a new configuration is proposed. The ODCB specimens are a modification of the standardized DCB specimens. They differ on the fact that, instead of one adhesive layer, the ODCB specimens are constituted by three adhesive layers: one primary bond and two secondary bonds (Fig. 1).

The primary bond is a plate made of the adhesive that is meant to be degraded, in this case SikaPower®-4720. The plate is produced in a mould coated with a release agent and, after 24 h in a hydraulic press, it can be removed from the mould and placed in a dry environment for 5 days, allowing the plate to be completely cured and dried. Afterwards, the adhesive plate is abraded with sandpaper and exposed to the ageing environment. Since the plate is not bonded to any adherend, the area exposed to the environment is much larger than the area of adhesive on a standard DCB specimen, which accelerates the saturation process. After these steps, the aged adhesive plate is abraded with sandpaper again and cleaned with acetone to allow a better adhesion to the secondary bond.

The secondary bond was made with a secondary adhesive, with higher mechanical properties, which is meant to bond the

Table 2
Mechanical properties of aluminium Al7075-T6 supplied by Lanema (Ovar, Portugal).

Maximum strength (Rm)	Yield strength (Rp 0.2)	Hardness (Brinell)
525 MPa	455 MPa	130–150

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