International Journal of Adhesion & Adhesives 31 (2011) 530–540

Contents lists available at ScienceDirect

International Journal of Adhesion & Adhesives

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

Evolution of crack path and fracture surface with degradation in rubber-toughened epoxy adhesive joints: Application to open-faced specimens

A. Ameli [a], M. Papini [b], J.K. Spelt [a,*]

[a] Department of Mechanical and Industrial Engineering, University of Toronto, 5 King's College Road, Toronto, Ontario, Canada M5S 3G8
[b] Department of Mechanical and Industrial Engineering, Ryerson University, 350 Victoria Street, Toronto, Ontario, Canada M5B 2K3

ARTICLE INFO

Article history:
Accepted 28 April 2011
Available online 23 May 2011

Keywords:
Crack path
Fracture surface
Degradation
Rubber-toughened
Adhesive
Open-faced
Mixed-mode

ABSTRACT

The crack path and fracture surface in the mixed-mode fracture of two different rubber-toughened epoxy adhesives were evaluated using double-layered open-faced double cantilever beam (ODCB) specimens in which the primary adhesive layer had been environmentally aged. The crack path in the mixed-mode fracture of unaged ODCB specimens was unexpectedly in the secondary adhesive layer, and several hypotheses were examined to explain this. It was concluded that a reduced residual stress in the secondary adhesive layer produced stable crack growth in the secondary layer instead of the expected path in the primary layer. The average crack path depth, fracture surface roughness and maximum elevation in the fracture surface profiles were then measured using optical profilometry as a function of the degree of aging. The results showed a strong relationship between all these parameters and the critical strain energy release rate, G_{cs}, irrespective of the type of adhesive. In the case of adhesive A where significant irreversible degradation was observed, all these parameters varied approximately linearly with G_{cs}. In the case of adhesive B, aging did not result in permanent degradation (G_{cs} was unchanged) and so all these fracture surface parameters also remained unchanged after aging. The results indicate that quantifying fracture surface parameters as a post-failure analysis can be of use in the estimation of the fracture toughness at which a practical joint fails.

1. Introduction

It is well known that epoxy adhesives are susceptible to moisture ingress and their mechanical properties alter upon environmental exposure. Traditional closed joints are not well suited for the study of degradation because the diffusion path of water into the adhesive layer is long and the degradation experiments are hence time-consuming. Moreover, the resulting degradation is not uniform after reasonable exposure times. Since the results from testing such joints do not represent a discrete state of moisture degradation, it is impossible to directly apply the measured properties to other joint configurations or ageing conditions [1,2]. Open-faced specimens can instead be used to achieve a spatially uniform state of degradation and to accelerate the aging process by shortening the diffusion path of water into the adhesive layer [1–6]. The open-faced specimen used in this work was a double cantilever beam specimen in which a layer of adhesive was applied on one adherend and cured using a backing plate, then exposed to humidity and dried. After aging, the open-faced specimen was bonded to another adherend using second layer of adhesive and the completed specimen was cured and prepared for mechanical testing.

Many researchers have studied the crack path in the fracture of adhesively bonded joints. Overall, four types of crack path have been reported: (a) cohesive failure, where the crack propagates through the adhesive layer; (b) interfacial failure, where the failure occurs at the interface between the adhesive and one of the adherends; (c) oscillatory failure, where the trajectory of the crack oscillates about the mid-plane of the bond but remains within the adhesive layer and (d) alternating failure, where the crack alternates between the two interfaces [7–13].

As summarized in Ref. [7], different criteria have been proposed for crack path selection in brittle materials under mixed-mode loading conditions. The mode-I criterion assumes that the crack follows a pure mode-I path, where $K_{II}=0$ [14–16]. Ergodan and Sih [17] suggested that the crack path is perpendicular to the direction of maximum opening stress at the crack tip. Some strain energy based criteria have also been proposed [18,19]. All these criteria yield similar results and no distinguishable differences in predicted crack path have been observed [11,16,20,21].

The directional instability of cracks in adhesively bonded joints was first experimentally addressed by Chai [8–10] who reported that the crack trajectory in the mode-I fracture of certain

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* Corresponding author.
E-mail address: spelt@mie.utoronto.ca (J.K. Spelt).

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doi:10.1016/j.ijadhadh.2011.04.007

aluminum-epoxy joints and graphite reinforced epoxy laminates alternated periodically between the two interfaces. He related this phenomenon to the unavoidable imperfections in the adhesive layer. Cotterell and Rice [22] concluded that the *T*-stress (i.e. the longitudinal stress in the crack plane normal to the crack front) plays an important role in the directional stability of crack propagation, with cracks being directionally stable if the *T*-stress is negative. Chen and Dillard [11] experimentally demonstrated the *T*-stress dependence of crack path selection in adhesively bonded joints using DCB specimens and a mechanical stretching procedure that could impose different *T*-stress levels. They observed that at relatively high positive *T*-stress levels, the crack periodically alternated between the two interfaces.

The thermal residual stress developed in the adhesive layer after the curing cycle plays an important role in the crack path selection since it affects the *T*-stress [16,23]. Yu et al. [24] measured the residual stresses in an epoxy-steel system using curvature in bi-material beams. They concluded that the residual stress due to cure shrinkage was negligible, but considerable stresses developed during cooling due to the differential thermal contraction of the two materials. They also observed that the residual stress increased after a repeat of the thermal cycles and decreased upon moisture exposure. Daghyani et al. [23] studied the crack path in a rubber-modified epoxy adhesive bonding of both aluminum and carbon fiber/epoxy composite adherends. They calculated the thermal residual stress in the joints using a finite element analysis and showed that the type of adherend material influenced the level of the thermal residual stress in the adhesive layer, which consequently resulted in different crack paths in the joints.

There is an extensive body of work on the characterization of fractured surfaces and their correlation with the fracture properties in metals, rock joints and concrete [25–29]. Fracture surfaces are thought to convey inherent fracture properties in response to the failure processes. However, only very limited attention has been paid to fracture surface characterization of adhesively bonded joints. The fractured surface can be related to the strength, fracture toughness and durability of the adhesively bonded joint [30]. Yee and Pearson [31] showed that the fractured surface for rubber-toughened epoxy adhesives is more complex than that for unmodified epoxy adhesives. Naito and Fujii [30,32] studied the fracture surfaces of unmodified and rubber-modified epoxy adhesive joints fractured under static and fatigue mode-I loadings. They observed a larger strain energy release rate for more geometrically complex fracture surfaces (having higher roughness) and derived empirical equations showing the relation between fracture toughness and fractal dimension. They were also able to predict the fatigue crack growth rate by measuring the fractal dimension of the fracture surfaces.

In the present work, the mixed-mode crack paths and fracture surfaces of unaged and aged open-faced double cantilever beam (ODCB) specimens were measured and characterized using optical non-contact profilometry. Two rubber-modified epoxy adhesives were compared. The paper is structured in two parts—the first dealing with unaged specimens and the second with aged specimens. In the mixed-mode fracture of unaged ODCB specimens, the crack unexpectedly propagated in the secondary adhesive layer, instead of growing in the more highly strained primary layer. Several hypotheses were examined to explain this by investigating the effects of curing profile, number of cure cycles, bondline thickness, bondline residual stress and hot-wet exposure on the selection of the crack path. Variations in the adhesive residual stress in the unaged joints appeared to be responsible for the unexpected crack path in these ODCB specimens. The fracture surface profiles of hot-wet aged ODCB joints and their relationships with the fracture toughness were then assessed by measuring the fracture surface roughness and the average crack path depth. A finite element model was used to explain the observed crack paths.

2. Experimental procedures

As indicated below, some of the results used in the present paper have been previously reported in Refs. [2,3,7], and the methods and relevant data are only summarized briefly in Sections 2.1–2.4. The residual stress measurements in the adhesive layers have not been previously reported and are described in Section 2.5.

2.1. Materials

Two different commercial DGEBA-based heat-cured rubber-toughened structural epoxy adhesives were studied (Table 1). Unless otherwise stated, the recommended curing profile of 30 min at 180 °C was used and monitored using a thermocouple embedded in the adhesive layer.

2.2. ODCB specimen fabrication

The DCB adherends were AA6061-T6 aluminum alloy pretreated using the P2 sulfuric acid etch method [3]. The primary adhesive layer was 385 μm thick and was made of either adhesive A or adhesive B. It was cured onto the primary adherend using a backing plate coated with tetrafluoroethylene (TFE) dry lubricant (MS-122N/CO_2 by Miller-Stephenson Co., Connecticut, USA) that had been baked for approximately four hours at 285 °C [33]. After curing, the backing plate was removed and the open-faced specimens were exposed to a 60 °C–95% relative humidity (RH) condition for varying times. The exposed specimens were then dried in a vacuum oven containing anhydrous calcium sulphate at 40 °C for approximately 7 days to remove any reversible effect of water ingress such as plasticization. The primary adhesive layer was then very lightly sanded, wiped with acetone and dried before a layer of adhesive B (termed the secondary adhesive layer) was used to bond the specimen to a second adherend to make a complete ODCB joint. Adhesive B was used as the secondary adhesive in all cases. If the primary layer was adhesive A, the system was called “AB” and if both primary and secondary layers were made of adhesive B, the system was called “BB”. In the case of fresh ODCBs, the secondary bonding was done immediately after the primary bonding without the exposure and drying processes. The configuration and dimensions of a typical ODCB is depicted in Fig. 1. More details about the specimen fabrication can be found in Refs. [2,3]. Single-layered DCB specimens were made following the procedure established in Ref. [34].

2.3. Fracture test procedure

The mixed-mode fracture testing procedure of Ref. [34] was employed using a servo-electric load frame and the load jig of Ref. [35]. All tests were conducted at a loading phase angle $\psi = 27°$,

Table 1
Mechanical and physical properties of toughened epoxy adhesives A and B as provided by the manufacturers [34].

Adhesive	Elastic modulus, E, (GPa)	Poisson's ratio, ν	Tensile strength, σ_y, (MPa)	Glass transition temp, T_g, (°C)	Cured density (g/cm^3)
Adhesive A	1.96	0.45	44.8	125	1.50
Adhesive B	1.73	0.39	N/A	122	1.14