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A micromechanical damage characterization and the modeling of a mineral filled epoxy adhesive

S. Joannès *

Centre des Matériaux, Mines ParisTech, CNRS UMR 7633, BP 87, 91003 Evry Cedex, France

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ABSTRACT

Current structural adhesives can be used to assemble a variety of similar and dissimilar materials, usually with minimal surface preparation and sometimes even without degreasing. This kind of performance is obtained by using filled structural polymers which, as a consequence, lead to complex multi-phased composite material microstructures. Nevertheless, the use of mineral fillers such as talc also affects the adhesive mechanical behavior which is of major importance for structural applications. In order to study this type of material, a multi-axial mechanical characterization of an epoxy structural adhesive has been undertaken using a modified Arcan setup. An unfilled and not marketed formulation of the epoxy adhesive has also been tested to emphasize the role of the mineral particles on its mechanical behavior. As a first step, this paper reports the mechanical differences that have been observed between the filled and the unfilled versions of the adhesive, and proposes possible explanations related to microstructural processes. Contribution of the mineral fillers leads to a deterministic failure process. This is suggested, most significantly, by microstructural observations which have revealed useful information concerning the damage evolution and its kinetics. Indeed, the applied mechanical load provokes an exfoliation of talc flakes leading to a pattern of micro-cracks. In a subsequent step in the study, based on both macro and micro experimental results of the adhesive, a damage model has been derived from the vectorial description of cracking modes and its performance has been assessed.

1. Introduction

Design safer, lighter and cleaner cars: this is the challenge to manufacturers and OEMs[1] in the twenty-first century. The development of a new vehicle must balance a number of requirements: improve performances in terms of safety and comfort, reduce costs and manufacturing cycle times whilst preserving the environment. In this context, automotive manufacturers and suppliers are increasingly interested in materials previously used for aerospace applications such as magnesium, aluminum and especially organic matrix composites. The overall cost of mono-material solutions, despite some outstanding performances, remains far too high but multi-material designs all have their place for large vehicle series. Prospects for improvement offered by the use of such hybrid structures, however still require the development of specific assembly techniques. Already widely used for windscreen mounting, carrier plastic parts assembly or corrosion protection, high performance adhesives today find more and more applications within the vehicle structure. Adhesive bonding avoids any machining operation and the continuous connection obtained leads to a uniform stress distribution, unlike riveted or bolted joints. In recent years, structural adhesive bonding has therefore become a popular joining method for the transportation industry.

* Tel.: +33 160763015.
E-mail address: sebastien.joannes@mines-paristech.fr

[1] Original Equipment Manufacturer.

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To optimize costs and performances, material suppliers are dealing with multi-phased composite material microstructures by providing filled structural adhesives. The addition of mineral fillers to an adhesive compound can modify the viscosity, opacity and weight of the compound but also reduce shrinkage or speed up the crosslink process of the polymer. Other specific functions can be achieved and, for example, the oleophilic nature of talc, which is often seen as a cost-reducing filler only, makes it popular for minimizing the degreasing operation of metallic surfaces (Greiveldinger et al., 2000). Also hydrophobic, talc can add chemical and water resistance to adhesive products (Mario, 2007). Nevertheless, the use of mineral fillers will also affect the mechanical behavior of the adhesive (Joannès, 2007; Kulkarni et al., 2009) which is of major importance for structural applications. Alongside advanced material developments, there is a need to gain knowledge about the various effects of the mineral fillers and to propose appropriate material models to describe their behavior. With this perspective, the paper focuses on the case of a commercial single component adhesive, comprising a prepolymer DGEBA type epoxy (Bisphenol A diglycidyl ether) and a DDA (dicyandiamide) crosslinking agent. According to the material supplier, the adhesive formulation is completed by additives and mineral fillers (mass fraction around 23%) leading to a composite microstructure. In the work described here, an unfilled version of the adhesive (not marketed) has also been used to emphasize the role of the mineral particles on the mechanical behavior.

The main purpose of this paper is to report the mechanical differences that have been observed between the filled and the unfilled versions of the adhesive, and suggest possible explanations related to microstructural considerations. The second purpose is to build and justify a physically motivated damage model that links the macromechanical behavior to micromechanical mechanisms. In the following section, experimental results will thus highlight some filler effects on the mechanical response of the bonded joints and microstructural observations will provide all the information needed for describing the damage kinetics. The third section is dedicated to the damage model which is based on a vectorial description of cracking modes (Talreja, 1985; Thionnet and Renard, 1993, 1999; Thionnet, 2010) and the invariant theory (Thionnet and Martin, 2006). Experimental and simulation results are compared and the reliability and efficiency of the model is discussed.

2. An in-depth experimental study

The design of an assembly requires a thorough knowledge of the mechanical behavior of materials used, and the mechanical characterization of adhesive systems presents many challenges. Adhesion problems aside, though some authors consider that a bulk characterization of the adhesive polymer might be enough (Lilleheden, 1994), this suggestion is far from universally accepted; mainly due to the hydrostatic pressure effects undergone by the polymer in a confined state between substrates. It is of course possible to take into account the stress triaxiality or the pressure sensitivity effects in a bulk[2] state by using specific notched cylindrical bar specimens (Saï et al., 2011), however, this is made very difficult in the case of an anisotropic behavior which may occur with filled adhesives (Joannès et al., 2010). Then it is often necessary to employ *in situ* characterization devices. Several methods might be considered (Dean et al., 1996), including the thick-adherend shear method, the butt torsion test or the Arcan joint method. Each method has its advantages and drawbacks in terms of ease of use, accuracy, precision, resolution, sensitivity and price.

In this paper, bonded joints were mainly tested thanks to a modified Arcan (named Arcan-Mines) device able to vary the loading direction while maintaining the state of stress as uniform as possible within the adhesive joint. This device, consisting of two half circular discs (see drawings in Fig. 1) connected by an aluminum or steel bonded specimen, is inspired by the original Arcan[3] fixture (Arcan, 1978) and is fully described in Joannès (2007). While maintaining the centering quality of the two discs, the design of the Arcan-Mines device allowed the addition of shims to vary the thickness of adhesive joints from 100 to 1500 μm. In addition to the Arcan fastening system, the accuracy of the tests was however obviously based on the quality of the specimen. A dedicated assembly mount was thus designed with two steel plates maintaining four substrates which provided for each one a bonding surface of $70 \times 10\ mm^2$. Centering pins and steel shims completed the device for the gluing and was then placed in an oven imposing the cure cycle. Crosslinking of the epoxy adhesive was obtained by imposing a 180 °C temperature plateau for 60 min after a temperature rise rate of 10 °C/min; the cooling was then carried out at room temperature. The formulation of the commercial adhesive that we denote in this paper by β consisted of wollastonite (18% by mass) and talc (5% by mass) silica mineral fillers. The wollastonite was in the form of elongated compact aggregates and the talc in the form of a lamellar structure as is shown below. The unfilled and unmarketed version of β is denoted in this paper by β_{NC}.

Dimensions and proper alignment of each specimen were controlled before the mounting on the Arcan-Mines device. Gripped onto the specimen, a high resolution biaxial extensometer was used to record the extension and the shear displacement of the "bonded joint"; an assembly composed of the upper substrate, the adhesive joint itself and the lower substrate on an overall thickness (measurement base) of about ten millimeters. An estimation of strains inside the adhesive joint could then be computed by deducting the deformation of the two substrates. Due to the relative stiffness of the substrate (compared to the adhesive), most of the displacement is induced by the adhesive itself: for the considered thicknesses, more than 90% of the total displacement is induced by the polymer. Applying a loading rate of 70 N/s, it is thus possible to measure the multiaxial rigidity of the assembly.

[2] Considering that the bulk solidification of the polymer is similar to that obtained in a confined area.

[3] The original test applies to an homogeneous and bulk specimen, but with some modifications, the system was used for adhesive joints (Cognard et al., 2005).