THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

in

Applied Mechanics

Cohesive modelling of the temperature dependence of epoxy based adhesives in Mode I and Mode II loading

by

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Gothenburg, Sweden 2013

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THESIS FOR LICENTIATE OF ENGINEERING no 2013:08
ISSN 1652-8565

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Chalmers Reproservice
Gothenburg, Sweden 2013

Abstract

In this work, the influence of the temperature on the cohesive laws for two epoxy adhesives is studied at temperatures below the glass transition temperature for both Mode I and Mode II loading. Cohesive laws are measured experimentally under quasi-static loading conditions in the temperature range $-30 \leq T \leq 80°\mathrm{C}$. Three parameters of the cohesive laws are studied in detail: the elastic stiffness, the peak stress and the fracture energy. Methods for determining the elastic stiffness in Mode I and Mode II are derived and evaluated. With these methods, the results in this work show that it is possible to measure all three parameters for each pure mode loading case by the use of only the DCB- and the ENF-test specimens. Even though the measures tend to spread in values, this can significantly reduce the cost for performing experiments.

It is shown that most of the cohesive parameters are decreasing with an increasing temperature in both loading modes and for both adhesives. An exception is the Mode I fracture energy for one of the adhesives. This is shown to be independent of the temperature in the studied temperature range. For the same adhesive, the Mode II fracture energy is shown to be continuously decreasing with an increasing temperature.

The experimental results are verified by finite element analyses. The simulations only consider uncoupled cohesive behaviours. By use of the experimental results, simplified bi-linear cohesive laws to be used at any temperature within the studied temperature range are derived for one adhesive in both loading modes. This is desired in order to simulate adhesively bonded structures that suffer a wide range in temperature.

Keywords: Cohesive laws, Epoxy adhesive, Fracture energy, Peak stress, Temperature, Regression analyses, Shear modulus, Young's modulus.

Acknowledgements

This work has been carried out during the years 2009 and 2013 at the department of Mechanics of materials at the University of Skövde, Sweden.

I would like to thank my supervisor Professor Ulf Stigh for sharing his expertise in fruitful discussions. I also would like to thank my assistant supervisor, Dr. Anders Biel. Mostly for gladly sharing his great knowledge in test machines and related electronics but also for sharing his experience in experimental research of adhesives.

Further, I would like to thank Dr. Svante Alfredsson and Dr. Stephan Marzi for their help and engagement in my work. I also would like to send my appreciation to my former and present colleagues at the University of Skövde and also at the Fraunhofer institute in Bremen, Germany.

Finally, my deepest gratitude to my family and friends for their support and to Kristina, Vilja, and Maja for making me long to go home when I am at work.

Tomas Walander

Skövde, March 2013

List of appended papers

This thesis consists of two appended papers.

Paper A Walander, T., Biel, A. and Stigh, U., “Temperature dependence of cohesive laws for an epoxy adhesive in Mode I and Mode II loading”. Submitted

Paper B Walander, T., Biel, A. and Stigh, U., “An evaluation of the temperature dependence of cohesive properties for two structural epoxy adhesives”. Proceeding of the 19th European Conference on Fracture, Kazan, Russia, Aug. 26-31 2012.

Contribution to Co-authored papers

Both appended papers in this thesis are published with my supervisors as co-authors. My contribution to these papers is listed below.

Paper A Planned and wrote the paper.
Responsible for the theoretical developments.
Planned and performed the experiments together with one of the co-authors.
Evaluated the experiments.
Responsible for the simulations.
Responsible for the verifications.

Paper B Planned and wrote the paper.
Planned and performed the experiments together with one of the authors.
Evaluated the experiments.
Presented the work at the conference

Contents

Introduction

To test a structure experimentally by use of destructive test methods is often an ineffective method that is associated with high costs. One of the main reason is the fact that it is time-consuming to prepare and perform the experiments. Another reason for the high costs is that several, nominally identical experiments need to be performed in order to get reliable results. This is since experimental results spread. As a substitution, or complement, of full scale experimental tests, it is more cost efficient to analyse a structure using the finite element method. This becomes obvious when several modifications and variants need to be tested and/or where the manufacturing cost is high for each tested component.

With the use of adhesives, the performance of a structure can be optimized by enabling joining of lightweight and high-strength materials. Engineering structures commonly suffer a wide range in temperature. Since the mechanical behaviour of adhesives is known to be strongly temperature dependent, cf. e.g. Kinloch (1987), a numerical model of adhesives needs to take temperature into account as a parameter. No such model has been found in the open literature.

The strength of adhesively bonded, multi-material build-up structures can be adequately predicted using cohesive elements in a finite element analysis, cf. e.g. Carlberger and Stigh (2010a). The constitutive relations for these elements are represented with cohesive laws. A cohesive law is a constitutive relation on a structural length scale between the traction exerted on the interfaces of the adhesive to the adherends and the separation of the interfaces. The deformation of an adhesive layer is dominated by two loading modes, cf. e.g. Klarbring (1991) and Schmidt (2008). These are Mode I and Mode II. Mode I is characterized by peel deformation w and peel stress σ and Mode II by shear deformation v and shear stress τ. An illustration of this is given in Fig. 1.

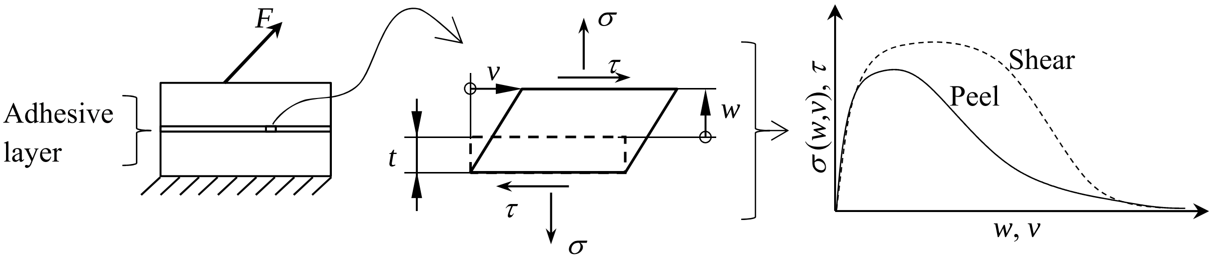

Fig. 1 An adhesive layer with initial thickness t, cohesive stresses (σ, τ) with work-conjugated separations (w, v) and illustrated cohesive laws for pure peel (Mode I) or pure shear (Mode II) deformation.

Cohesive laws for adhesives can be measured experimentally. For this purpose, two commonly used test specimens for each loading mode are the double cantilever beam (DCB) and the end notch flexure (ENF) specimens. By use of Rice's (1968) and Cherepanov's (1967), path-independent J-integral, methods for measuring cohesive laws for these specimens are presented in e.g. Andersson and Stigh (2004) and Stigh et al. (2009). From these methods, the cohesive laws for an adhesive layer can be measured in terms of external forces and rotations of the loading points. Experimental results for adhesive layers using these specimens and methods are presented in e.g. Stigh and Andersson (2000) and Walander (2009). The methods have the advantage that no material data for the adherends need to be known when evaluating the experiments. This is as long as the adhesive is assumed as much more compliant than the adherends. Also, these methods allow for plastic deformation in the adherends as long as unloading of the specimen is avoided. To allow for plastic deformation can significantly reduce the size of the specimens in comparison to methods, e.g. Alfredsson (2004) and Tamuzs et al. (2003), which requires elastic adherends. Small specimens are preferable when performing temperature experiments since climate chambers often are limited in space.

When evaluating experimentally measured cohesive laws, three parameters are of special interest. These are the elastic stiffness, the peak stress and the fracture energy which are derived from the shape of a cohesive law. The elastic stiffness is defined as the initial slope, the peak stress is defined as the maximum value in stress and the fracture energy is defined as the area beneath the entire curve. For the strength of an adhesively bonded structure, the influence of the elastic stiffness of the adhesive is small. This is since the thickness of an adhesive layer normally is small in comparison to other dimensions. Thus, the deformation of a purely elastic structure is normally dominated by the elastic properties of the bonding material. The strength of an adhesively bonded structure is shown to rather be governed by the parameters peak stress and the fracture energy. Industrially, the elastic stiffness and the peak stress are measured using bulk tensile and thick adherend shear tests. For industrially purposes, several standard methods exist for estimating the fracture energy of adhesive layers by use of linear elastic fracture mechanics (LEFM). For a DCB specimen, some of these methods are evaluated in Biel and Stigh (2008) by use of FE-analysis with a known cohesive law. It is shown that most of the standards, by far, fail to predict the fracture energy. In addition, none of the standardized test methods capture the shape of the cohesive relation. However, methods based on the J-integral both capture the shape of the cohesive law and gives the correct fracture energy.

The influence of temperature on cohesive laws in Mode I is studied by Carlberger et al. (2009). In this study, cohesive laws for the epoxy based DOW Betamate XW 1044-3 (DB1044) adhesive are measured using a DCB specimen. From the study, it is shown that both the parameters peak stress and, to a small extent, the fracture energy are decreasing with an increase in the temperature. In Mode II, no previous study of the temperature dependence for cohesive laws has been reported. However, some studies using LEFM have been performed. Chai (2004) studies the Mode II fracture energy of the American Cyanamid toughened thermosetting (BP-907) epoxy adhesive by use of a Napkin ring specimen. From this study it is shown that the Mode II fracture energy monotonically decreases with an increase in the temperature within the region $0.7 < T/T_\mathrm{g} < 1.0$. Banea et al. (2012) study the epoxy based, high temperature, adhesive ChemteX XN1244 using an ENF specimen. The results show a maximum in fracture energy at the temperature $T = 0.88\, T_\mathrm{g}$. In this study, the peak stress at each temperature is not measured. Instead, by use of FE-analysis, the peak stresses are obtained by curve-fitting the numerical results of the force-deformation relation to the experimental results. The procedure is questionable since the influence of the peak stress on the structural behaviour of the ENF-specimen often is small. By performing ENF experiments at several temperatures using the method in Stigh et al. (2009), the influence of temperature for entire cohesive laws and thereby the peak stress and the fracture energy can be measured in Mode II.

Summary of appended papers

Mode I and Mode II experiments are performed on an epoxy based adhesive at different temperatures. The work is performed in a project involving the Swedish automotive industry. This work is the first to report the influence of temperature on cohesive laws in Mode II loading. The main goal is to study the influence of temperature on cohesive laws for an adhesive and to use the results to create a numerical model of the adhesive layer. The adhesive is SikaPower498 (SP498) which is a crash resistant epoxy adhesive that is used today by the automotive industry. The test is performed with a nominal layer thickness of 0.3 mm. Carlberger and Stigh (2010b) show that the thickness of an adhesive layer influences the measured fracture energy in both Mode I and Mode II whilst the peak stress is reported to slightly decrease with an increasing layer thickness in both modes. The used thickness in the present work is decided by the involved industrial project partners. This is with the motivation that it is a common thickness in automotive structures.

The experiments are performed using DCB and the ENF specimens and the experiments are evaluated using the methods in Andersson and Stigh (2004) and Stigh et al. (2009). Experiments are performed at five temperatures for each mode in the span $-40 \leq T \leq 80$°C. This is below the glass transition temperature of the SP498 and the DB1044 adhesive. At least five successful experiments are presented at each of these temperatures and loading modes. Representative cohesive laws for the evaluated temperatures are presented in **Paper A**, cf. Fig 2. Due to limitations of the climate chambers, the Mode I experiments has a lowest temperature at -40°C and the Mode II experiments -30°C.

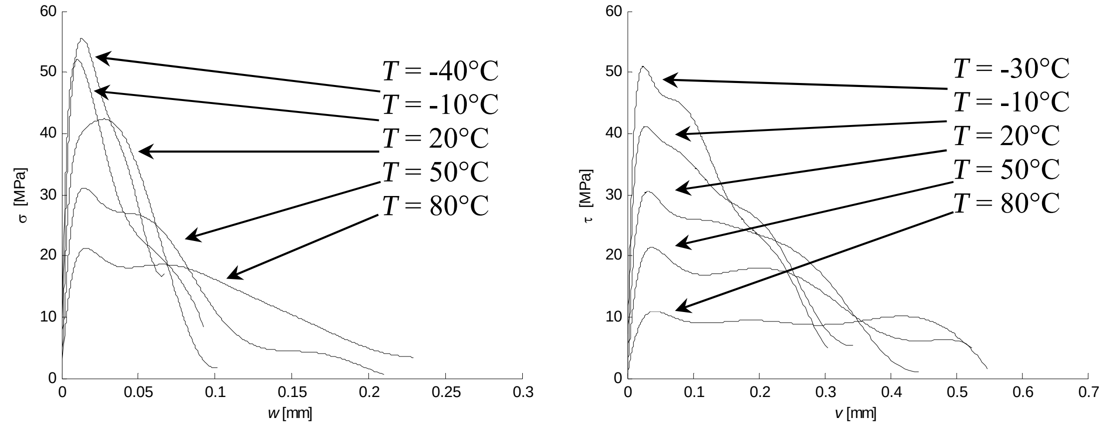

Fig. 2 Representative cohesive laws for each temperature group for SP498.
Left: Mode I. *Right*: Mode II

In **Paper B**, the results in Carlberger et al. (2009) are re-evaluated together with the results of the peak stress and fracture energy for the SP498 adhesive. Regression analysis is used in order to model the influence of the temperature for each cohesive parameter separately. This is done for both adhesives and loading modes. From the evaluation in Paper B, it is shown that the Mode I fracture energy of SP498 can be assumed to be independent of the temperature whilst the other parameters are noticed to monotonically decrease with an increase in the temperature. This is observed for both adhesives and for both loading modes. Another reason to re-evaluate the results in Carlberger et al. (2009) in **Paper B** is to show that the Mode I fracture energy is not generally independent of the temperature within the evaluated temperature span.

Paper A is a continuation of the work presented in **Paper B**. In **Paper A**, two novel methods for determining the elastic stiffness in Mode I using the DCB and in Mode II using the ENF specimen are presented. These methods imply that all three parameters of interest of a cohesive law can be measured using only two types of specimens. This is without any additional measurements in the experiments. Since the methods are sensitive to small variations of the measured forces and deformations, the experimental results are noticed to spread quite large. However, the methods can still be used for estimating the elastic stiffness. Since the elastic stiffness of an adhesive layer has a small influence on the strength of an adhesively bonded structure, estimations of the elastic stiffness can be sufficient. The presented methods can thus significantly reduce the cost for measuring adhesive properties since no additional tests to determine the elastic stiffnesses needs to be performed.

In **Paper A,** only the results of the SP498 adhesive is presented for both loading modes. With the results of the peak stress and fracture energy in **Paper B**, the results of the elastic stiffnesses are also included. Second order regression analyses are performed on all the three cohesive parameters in order to model the influence of the temperature. The elastic stiffnesses are noticed to constantly decrease with an increase in the temperature in Mode I. In Mode II, a minimum of the elastic stiffness is predicted at a temperature of 66°C.

Verification using finite element analysis

Two models for adhesive layers in a FE-code are used in **Paper A**. These are a shape-mimicking and a bi-linear cohesive law. A shape-mimicking cohesive law has the same shape as a measured cohesive law. It is constructed by defining a damage parameter as a function of the displacement using tabulated data from the representative cohesive laws presented in Fig. 2. The advantage by use of a shape-mimicking cohesive law is that the actual behaviour of an adhesive layer is simulated. That is, no simplifications in shape are made. A disadvantage is that this model only can be used at the temperatures where the cohesive laws have been measured. A bi-linear model, cf. Fig. 3, is a simplified cohesive model. For each mode, it is constructed by the three parameters: the elastic stiffness, the peak stress and the fracture energy. Thus by determining these parameters from experiments, an adhesive layer can be simulated. A great benefit with this model is that, by performing regression analyses of these parameters, any temperature within the evaluated temperature span can be simulated.

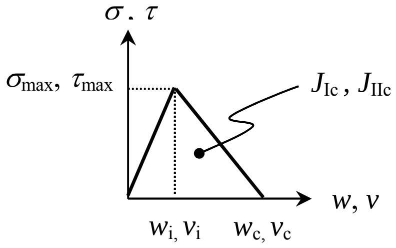

Fig. 3 Bi-linear model with notation for peel and shear

For all evaluated temperatures, simulations of the DCB and the ENF specimens using the bi-linear and the shape mimicking cohesive laws are performed and reported in **Paper A**. The simulated force vs. load point deformation relations are compared to the experimentally measured relations. A good fit is obtained for nearly all simulations. This gives confidence in the simulated cohesive law. Since the simulated cohesive laws are determined from experiments this also give confidence in the experimental results in **Paper A** and **Paper B**. By comparing the outcome of simulations from the analyses using the bi-linear to the shape-mimicking cohesive law, it is shown that the shape of the cohesive law has a minor influence on the structural behaviour for the DCB and the ENF specimens. An exception is at 80°C in Mode II where the bi-linear model is shown to be too much simplified for representing a plateau shaped cohesive law, like the representative cohesive law in Fig. 2.

Intended future work

The results in this work do only consider pure mode loadings. By this, only un-coupled simulations of adhesive layers at different temperatures can be performed. In a FE context, an uncoupled behaviour means that the cohesive laws in one mode are independent the cohesive laws in another. In e.g. a car crash test, a mixed-mode loading of an adhesive layer is common. In order to perform mixed-mode simulations, the mixed mode behaviour needs to be determined experimentally for an adhesive layer. Today there exist only a handful of studies of the mixed mode behaviour of adhesives. By this reason, mixed mode experiments with a prescribed constant mode mix is to be performed by the author.

References

Andersson T, Stigh U (2004) The stress-elongation relation for an adhesive layer loaded in modus I using equilibrium of energetic forces. Int J Sol Str 41:413-434

Alfredsson KS (2004) On the instantaneous energy release rate of the end-notch flexure adhesive joint specimen. Int J Sol Str 41:4787–4807

Banea MD, da Silva LFM, Campilho RDSG (2012) Mode II fracture toughness of adhesively bonded joints a function of temperature: Experimental and numerical study. J Adh 88:534-551

Biel A, Stigh U (2008) Effects of constitutive parameters on the accuracy of measured fracture energy using the DCB-specimen. Eng Frac Mech 75:2968–2983.

Carlberger T, Stigh U (2010a) Dynamic testing and simulation of hybrid joined bi-material beam. Thin-Wall Str. 48:609-619

Carlberger T, Stigh U (2010b) Influence of layer thickness on cohesive properties of an epoxy-based adhesive – an experimental study. J Adh 86:814–833

Carlberger T, Biel A, Stigh U (2009) Influence of temperature and strain rate on cohesive properties of a structural epoxy adhesive. Int J Fract155:155-166

Chai H (2004) The effects of bond thickness, rate and temperature on the deformation and fracture of structural adhesives under shear loading. Int J Fract 130:497-515

Cherepanov GP (1967) The propagation of cracks in a continuous medium, J Appl Math Mech 31:503-512

Kinloch AJ (1987) Adhesion and Adhesives – Science and Technology. Chapman and Hall, London

Klarbring A (1991) Derivation of a model of adhesively bonded joints by the asymptotic expansion method. Int J Engin Sci 29:493-512

Rice JR (1968) A Path Independent integral and the Approximate Analysis of Strain Concentrations by Notches and Cracks. ASME J Appl Mech 33:379-385

Schmidt P (2008) Modelling of adhesively bonded joints by an asymptotic method. Int J Eng Sci 46:1291–1324

Stigh U, Andersson T (2000) An experimental method to determine the complete stress-elongatioin relation for a structural adhesive layer loaded in peel. Fracture of Polymers, Composites and Adhesives (Eds. Williams J.G. and Pavan A.), ESIS publication 27, pp. 297-306

Stigh U, Alfredsson KS, Biel A (2009) Measurement of cohesive laws and related problems. In: Proc ASME, IMECE2009, Lake Buena Vista, Florida

Tamuzs V, Tarasovs S, Vilks U (2003) Compos Sci Technol 63:1423-1431

Walander T (2009) System for measurement of cohesive laws. Dissertation. University of Skövde