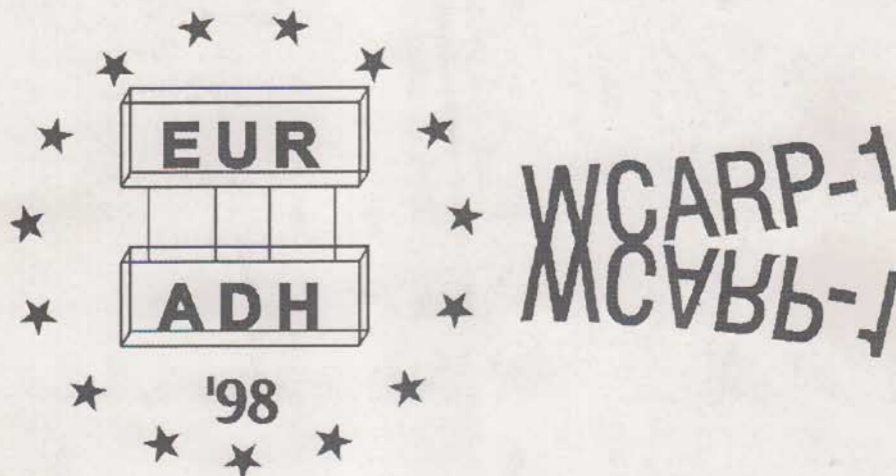




DECHEMA e.V.



EURADH '98  
4th European  
Conference  
on Adhesion

WCARP-1  
1st World Congress  
on Adhesion and  
Related Phenomena

September 6-11, 1998  
Kongreßhaus Garmisch-Partenkirchen/Germany

- 1.1 A. Hartwig, K. Albinsky, U. Diekmann, O.-D. Hennemann, Bremen/D  
Analysis of the surface composition of pretreated polymers by dye reactions
- 1.2 A.D. Crocombe, H.O. Hambly, J. Pan, S. Hadidimoud, Guildford/UK  
Measurement and prediction of the strength of bonded joints exposed to moisture
- 1.3 H.-J. Kim, Stony Brook, NY/USA; S. Hayashi, Shizuoka/J; H. Mizumachi, Tokyo/J  
Miscibility and fracture energy of probe tack for acrylic pressure sensitive adhesives
- 1.4 Z. Li, G. Li, H. He, M. Yu, Shenyang/PRC  
The whiskers of calcium sulfate's strengthening effect on the epoxy resin adhesives
- 1.5 V.V. Shibanov, I.J. Marshalok, Lviv/Ukraine  
The photocuring adhesives
- 1.6 G. Richardson, A.D. Crocombe, S. Hadidimoud, Guildford/UK  
Time dependant constitutive data and enhanced material models for adhesives
- 1.7 H. Kleinert, J. Gehrke, G. Hadlich, Dresden/D  
Properties of bonded joints under typical conditions of use for media in mechanical engineering
- 1.8 M. Pröbster, Heidelberg/D  
New elastic adhesives based on MS-Polymer-Technology
- 1.9 R. Häbler, Dresden/D  
Modulated DSC (MDSC) for adhesives
- 1.10 R. Häbler, K. Mai, Dresden/D  
Interphase characterization of joints by AFM
- 1.11 P.E. Dubois, C. Vittoz, J.C. Joud, Saint-Martin d'Hères/F;  
M. Mantel, Ugine/F;  
Wettability of metallic oxides and passive films. Application to stainless steels
- 1.12 J.C. Joud, P.E. Dubois, C. Vittoz, S. Roger, Saint-Martin d'Hères/F  
Effect of carbon contamination on the metallic oxide wettability
- 1.13 J.P. Jeandrau, Saint-Etienne/F  
Corona-discharge treatment of low surface-energy substrates for adhesive bonding: optimisation of parameters
- 1.14 D. Dengler, Landsberg/D  
Fast curing adhesives answering the demands of modern production technologies

## Measurement and prediction of the strength of bonded joints exposed to moisture

Crocombe AD, Hambly HO, Pan J and Hadidimoud S

Mechanical and Materials Eng, University of Surrey, Guildford, GU2 5XH, UK

### Introduction

Environmental degradation is a major limitation to an increase in the use of adhesives in structural bonding. Whilst a considerable amount of work has been undertaken investigating accelerated testing<sup>1</sup>, much less has been done on modelling the environmental degradation<sup>2</sup>. Such modelling tends to be based on the loss of interfacial integrity or on defining crack growth laws in various environments. In both approaches the change in adhesive material properties by absorbed water is only accounted for indirectly. Preliminary research in a more fundamental modelling approach<sup>3</sup> based on coupled stress-diffusion finite element methods appeared to offer considerable potential. Current research is investigating this approach further. The work reported in this paper is subset of a larger programme of research. Here we consider only a single adhesive type, joint configuration and exposure environment. These being a two part mineral filled epoxy, a butt joint and immersion in deionised water at room temperature respectively. Models for the kinetics of moisture uptake are determined, the effect of moisture on the mechanical properties of the adhesive assessed, residual strengths of adhesive joints are found and all three aspects are combined in coupled FEA-based mechanical-diffusion failure analyses of the degraded joints.

### Moisture uptake model

Varying thickness films of the adhesive were manufactured by compressing and curing the adhesive between two rigid plattens. Strips measuring about 80x30 mm were cut from these films and the kinetics of water uptake were determined using gravimetric methods. The resulting data for two thicknesses of the adhesive can be seen in fig 1. There is a clear thickness effect in these data with what appears to be a two stage uptake process. This is supported by similar tests on another adhesive, not reported here, with a greater range of specimen thicknesses. A clear implication of this is that it is not appropriate to use thin films to generate uptake data to be used in adhesive joints where the characteristic length will be much larger than the thin film thickness.

$$m = m_{1\infty} \left[ 1 - \sum \frac{2L^2 \exp(-\beta_n^2 D_1 t / l^2)}{\beta_n^2 (\beta_n^2 + L^2 + L)} \right] + m_{2\infty} \left[ 1 - \frac{8}{\pi^2} \sum \frac{\exp(-D_2 (2n+1)^2 \pi^2 t / 4l^2)}{(2n+1)^2} \right]$$

where  $L = l\alpha / D_1$  and  $\beta \tan \beta = L$

$a$ ( $10^{-11} \text{ms}^{-1}$ )	$D_1$ ( $10^{-14} \text{m}^2 \text{s}^{-1}$ )	$m_{1\infty}$ (%)	$D_2$ ( $10^{-14} \text{m}^2 \text{s}^{-1}$ )	$m_{2\infty}$ (%)
6.383	15.0	6	10	3.70

Table 1 - Parameters used in the uptake model

Various uptake models have been investigated. A concentration dependent diffusion coefficient model only produces small perturbations from the constant coefficient model and can model neither the two stage uptake nor the thickness effect. A dual absorption model can reflect the two stage uptake but cannot predict the thickness effect. To fully model this data it is necessary to use a dual absorption model for the continuum in conjunction with a boundary absorption model which prevents the boundary reaching equilibrium moisture

uptake immediately. A specific form of this model is shown in Table 1 with the specific values used for the parameters. The fit to the data can be seen in fig 1.

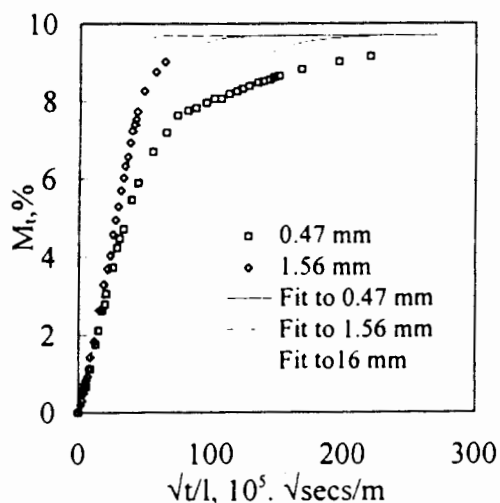


Fig 1 - Experimental and modelled moisture uptake data

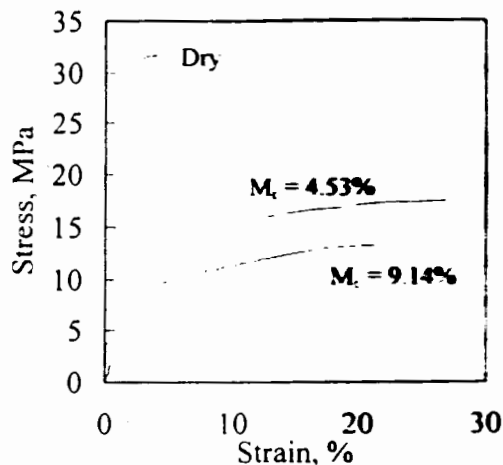


Fig 2 - Variation of the constitutive properties of the adhesive with moisture content

### Moisture dependent material properties

Flat dumbbell specimens of the bulk adhesive, 0.4 mm thick, were manufactured on a CNC machine from blanks cut from thin films. These were then immersed in deionised water for various periods of time and subsequently removed and tested in a servo electro-mechanical Instron 6025 testing machine to provide mechanical constitutive data for the adhesive at varying levels of saturation. The rate of testing was such that the time to failure of the specimens was of the order of a few minutes. The resulting data is shown in Fig 2. Average curves are presented for a given moisture content level. From these data it is clear that the elastic modulus and yield and ultimate stresses decrease with increasing moisture content whilst the opposite is true for the adhesive ductility. These curves form the basis of the definition of moisture dependent material properties that will be used in subsequent mechanical-diffusion stress analysis. The stress strain curves are entered in multilinear format for various moisture levels and properties at intermediate moisture levels are determined by interpolation.

### Residual strength of the exposed butt joints

Butt joints, shown schematically in fig 3, have been manufactured using mild steel substrates of thickness and bond lengths of 2.9 and 10 mm respectively. The substrates were cleaned ultrasonically in acetone both before and after a grit blast with 180/220 white alumina. Finally a silane primer was applied. The adhesive layer thickness was about 0.5 mm. Three periods of immersion in deionised water at room temperature were considered, 0, 19 and 75 days. After immersion the joints were tested to failure and the nominal failure stresses were found to be 50.6, 39.8 and 29.9 MPa. Crosshead rates were used such that failure occurred within a few minutes in each case. As can be seen there is an increasing reduction in joint strength with increasing time of exposure. It should be emphasised that all failure was cohesive within the adhesive and thus the degradation of joint strength should be attributed to the degradation of the adhesive within the joint and not the interface.

### Modelling joint failure

A significant number of preliminary, investigative analyses were carried out. Only 1/8 of the butt joint was modelled because of symmetry and it was found that the substrate could be

replaced by rigidly connecting all the nodes on the adhesive-substrate interface. A total of 1836 20-noded brick adhesive elements were used to create the FE model which had 28038 degrees of freedom. A conventional von Mises non-linear model was found to be inappropriate for the adhesive which exhibits significant pressure sensitivity. Instead a Drucker-Prager model was used. By using both tensile and compressive adhesive data the angle of the yield surface in the deviatoric-pressure stress plane was found to be 25.6° and the dilatation angle was determined from the volumetric and deviatoric strains as 12.7°. Thus the flow rule is non-associated.

Initially the 3-D diffusion analysis was undertaken for the butt joint for time periods of 18.7 and 75 days. The moisture uptake model discussed in a previous section was implemented and this gave the spatial distribution of moisture concentration within the butt joint at the corresponding time of exposure. An automatic time stepping algorithm was used which limited changes of concentration at a node to 5% of the saturation value. The resulting time step size was increased steadily from about 10 secs to a prescribed maximum of 7.5 days resulting in 123 time steps in the worst case. A mechanical stress analysis was then undertaken using the moisture dependent material properties data discussed in a section above. A displacement was applied to the rigid upper surface and a limit state approach was used to determine joint failure. The variation of load with substrate displacement for the three levels of moisture content is shown in Fig 4. By comparing this data with the experimental data it can be seen that a limit state approach enables the residual strength of these joints to be predicted to better than 10% error and this is considered to be a considerable achievement.

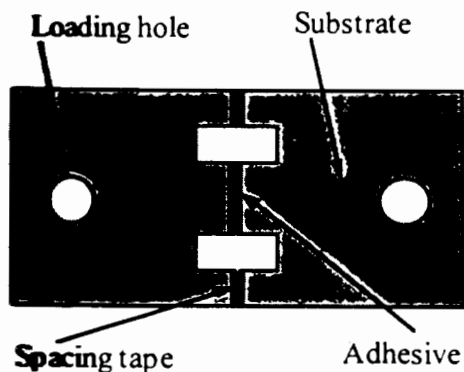


Fig 3 - Schematic of the butt joint being tested

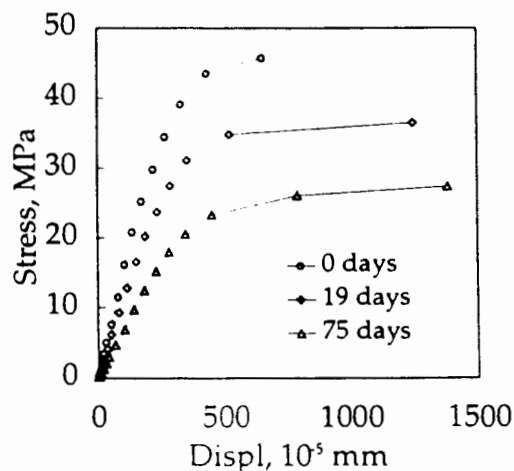


Fig 4 - Load-displacement plots for the three configurations analysed.

## Conclusions

Moisture uptake kinetics and its effect on adhesive constitutive properties have been characterised and used to predict the residual strength of exposed adhesive joints from a sequentially coupled mechanical diffusion finite element analysis.

## References

1. Sang NH, Eng. Matls H'book vol 3 Adhesives and Sealants, ASM Intl, Ohio, 622-627, 1990
2. Kinloch AJ, MTS Adhesives Proj 3, Report 5, DTI London, 1994
3. Crocombe AD, Int J Adhesion and Adhesives, 17, 229-238, 1997