

# MICROMECHANISMS OF ADHESION OF PRESSURE SENSITIVE ADHESIVES

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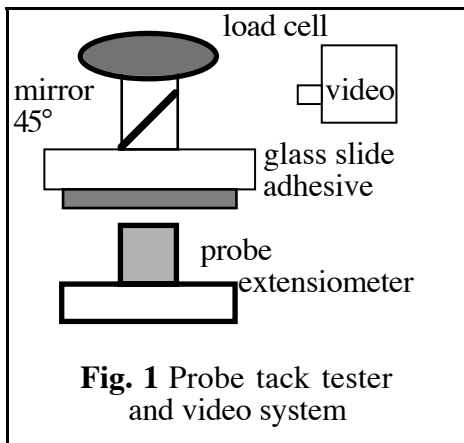
## Introduction

The earliest adhesion theories typically considered only the thermodynamic work of adhesion. However, the energy of adhesion predicted by these models is lower by orders of magnitude than that obtained from a common pressure sensitive adhesive (PSA). In this case, a PSA can be considered a viscoelastic material that sticks under pressure without any solvent, chemical reactions or interfacial diffusion at the interface. Gent and Schultz demonstrated that this discrepancy could be attributed to deformation of the bulk material and could be taken into account by a factor  $\Phi(v,T)$  where  $v$  is the debonding rate and  $T$  is the temperature. The resulting energy or work of adhesion is now given by

$$W_{adh} = W_o \times \Phi(v, T) \quad (1)$$

where  $W_o$  is the thermodynamic work of adhesion [Gent and Schultz, 1972; Andrews and Kinloch, 1973]. While a relationship between the velocity and the temperature during the processes of bonding and debonding is usually observed, a detailed description of the micromechanics of debonding is not yet available. In this paper, we will describe a novel instrumented probe tack tester which allows the simultaneous optical observation of the debonding process. Preliminary results on Poly(2-ethylhexyl acrylate) (PEHA-AA) samples and model Poly(n-butyl acrylate) (PnBA) will be presented.

## Experimental



**Fig. 1** Probe tack tester and video system

A probe tack technique is used in which the flat end of a cylindrical probe (diam = 1cm, roughness  $R_t = 1$  mic) is brought into parallel contact with the adhesive. The tack tester (Fig.1) and PC control have been adapted to a MTS 810 hydraulic testing machine. Test results are acquired in stress and strain with simultaneous video recording of the bonding and debonding process at the face of the probe. Tack tests were performed using three rates of separation ( $v=10,100,1000$ mic/s), four temperatures (-10, 0, 25, 50°C) and two contact times ( $t_c = 0.1, 60$  s). The applied normal pressure was 0.9 MPa for all measurements.

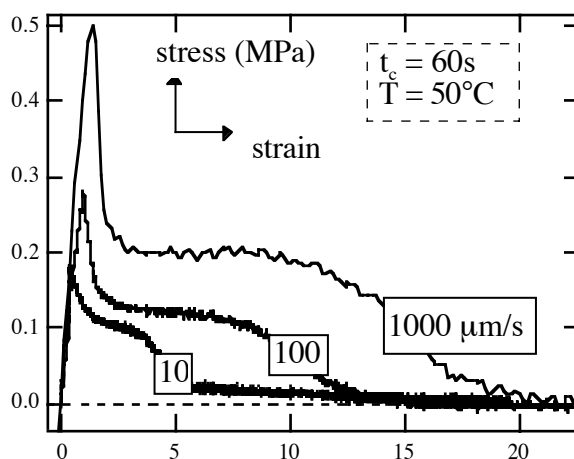
Preliminary studies were done on random copolymers of PEHA-Acrylic acid (97.5:2.5 wt%). The  $T_g$  of PEHA-AA was determined to be about -50°C by DSC. Further studies will include model monodisperse PnBA ranging in

molecular weights (4k - 900k). The samples are spread on clean microscope glass slides by doctor blade and dried at 70°C in vacuum. The thicknesses of films are between 70 and 100 microns.

## Results on PEHA-AA

Fig.2 represents a typical stress versus strain curve during the debonding process of PEHA-AA. The strain is obtained by dividing the crosshead displacement by the thickness of the film and the stress is obtained by dividing the force by the actual contact area as observed with the video. All curves possess three characteristic features - i.e., a maximum peak in stress followed by a plateau region and then a decrease. As expected [Zosel, 1985], temperature and debonding rate have opposite effects. Increasing velocity increases maximum stress and energy of separation. Indeed, for the lowest velocity and the highest temperature, values for the stress and energy were found to be 0.2 MPa and 40 J/m<sup>2</sup>, respectively, values typical for these experimental parameters. In contrast, for the highest velocity and lowest temperature measurements values of 1.3 MPa and 1100 J/m<sup>2</sup> were determined.

These observations can be rationalized if one considers that the ability of the chains in molten polymers to dissipate energy decreases with increasing temperature and decreasing debonding rate. For sufficiently long contact times, meaning quasi-complete relaxation of the polymer during the contact time, time temperature superposition (TTS) of energy or maximum stress is possible. However, for a short time of contact there is incomplete bonding between the probe and the film, bonding and debonding effects are mixed, and TTS is invalid.

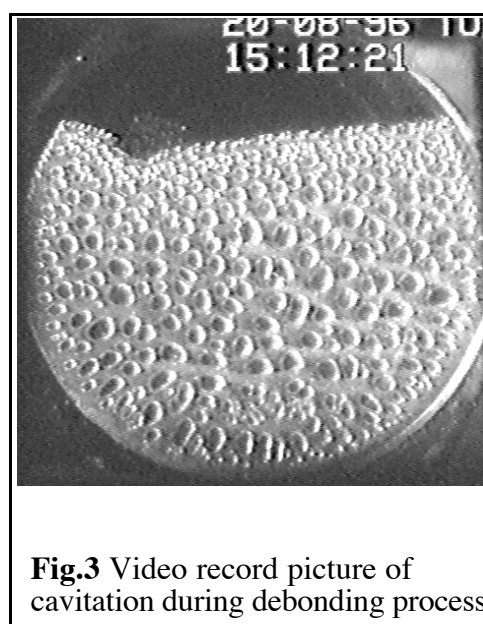


**Fig.2** Influence of the debonding velocity

### **Mechanism of debonding of PEHA-AA: one general description**

Debonding occurs in four steps, regardless of experimental conditions (in the range we have investigated). During the first stage, homogeneous deformation takes place. Stress increases but without any appearance of inhomogeneities at the surface of the probe. Secondly, the onset of cavitation occurs just before achieving the maximum stress. The third step is the extension of the polymer fibrils and appears to occur at approximately constant stress. Finally, fibril rupture (cohesive fracture) or fibril debonding (adhesive fracture) is observed. For the case of PEHA-AA in this study, a transition from cohesive to adhesive rupture occurred around 0°C upon decreasing the temperature.

Fig.3 represents a typical snapshot of the probe (d=1cm) during the cavitation stage (25°C, 0.1s, 10 mic/s). The area of contact appears clear on a dark background



**Fig.3** Video record picture of cavitation during debonding process

d. Video sampling and higher magnification (x4000) show fast growth of the cavities at the steel/polymer interface. During this process the diameter increases (much more in the plane of the probe) by dewetting of the PEHA-AA at the probe surface. Eventually the «walls» between the bubbles (fibrils), become thinner, extend, and, finally air enters the bubbles (which were mostly vacuum up to that stage) from the outer parts of the contact patch by tearing the PEHA-AA boundaries.

### **Conclusion**

It is concluded from this preliminary study on PEHA-AA that time temperature generally holds for long contact times but breaks down for shorter contact times. Moreover, four characteristic stages of the debonding process have been observed - i.e., homogeneous deformation, cavitation, fibrillation, and rupture. Finally, a transition from cohesive to adhesive rupture was observed around 0°C upon decreasing the temperature. Further studies will be presented on model PnBA's to examine effects of molecular weight on adhesion.

### **References**

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