

Micromechanics of the debonding process in probe tack experiments

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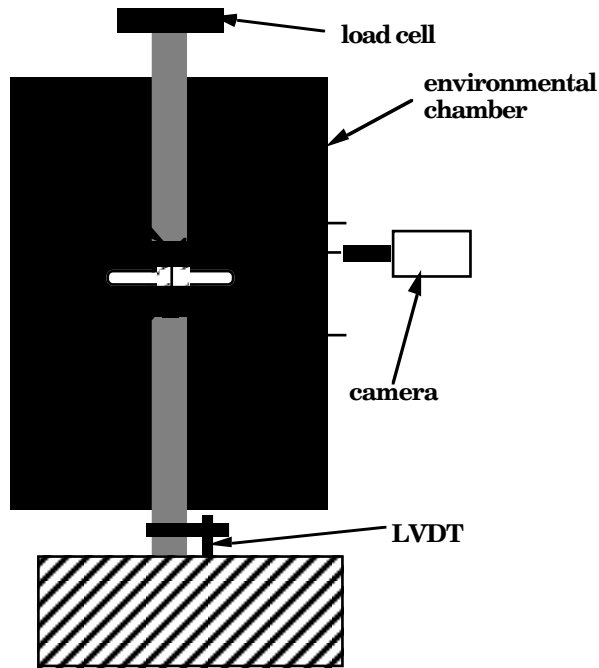
I. Introduction

Soft adhesives have the property to adhere upon contact and light pressure on most substrates. While applications are ubiquitous around us, the detailed mechanisms by which these pressure-sensitive-adhesives work are still not very well known. The reason lies in the interdisciplinary nature of the problem. The study of the coupling occurring between the molecular interactions at the interface between the film and the substrate and the very large deformations of the adhesive layer, which are responsible for the macroscopic mechanical response of the adhesive, require some knowledge of the rheology of polymer melts, of the physical chemistry of interfaces and of wetting dynamics and solid mechanics.

II. Experimental

An important progress has been made recently in our laboratory thanks to the development of a custom made instrument which allows the simultaneous observation of the deformation mechanisms of an adhesive layer while the debonding takes place.

We use the experimental geometry of probe tack where a flat-ended cylindrical probe is brought into contact with a 50-100 μm thick adhesive film (previously deposited on a transparent glass substrate) and kept in contact for a given time (0.1 to 200 s. in our case) and under a controlled pressure (typically 1 Mpa), and subsequently removed at a constant velocity. The force-displacement curve is recorded during the debonding stage and in parallel, a video camera records the debonding of the film through the transparent glass substrate.



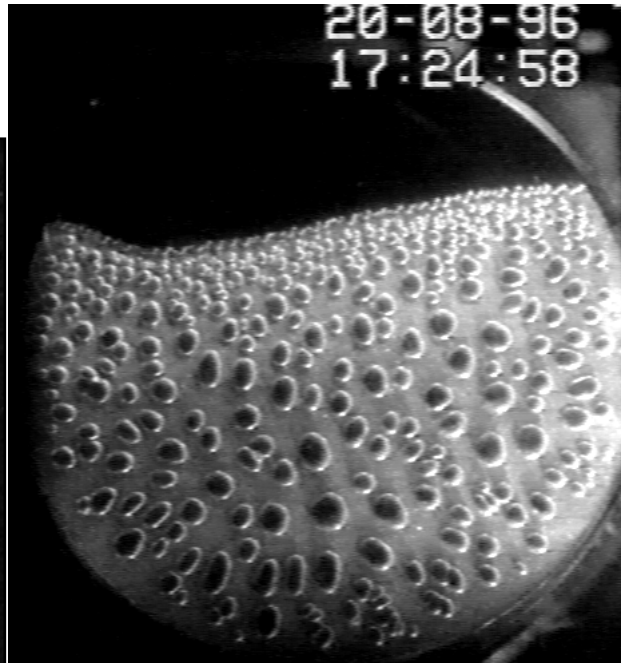
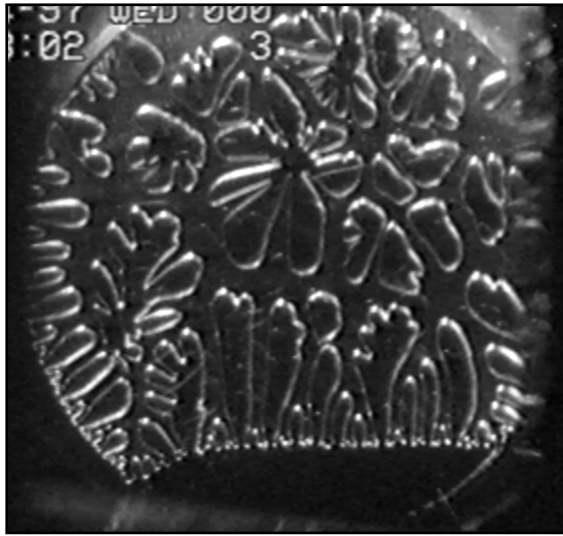
Probe tack geometry for the video observation

The model adhesives are either poly(2-ethylhexyl acrylates) synthesized by emulsion polymerization or anionically synthesized poly(*n*-butyl acrylates).

III. *Results and discussion*

A first analysis of the stress-strain curves shows that the maximum tensile stress attained σ_{\max} , generally follows well the time-temperature superposition and can therefore be represented as a function of an equivalent debonding rate $a_T V$. Interestingly for most cases, σ_{\max} is directly proportional to the shear modulus of the adhesive, measured at an equivalent frequency. On the other hand the adhesion energy W_{adh} (the integral under the stress-strain curve) does not generally follow the time temperature superposition. This apparent discrepancy can be better understood by a description of the debonding mechanism.

When the pressure applied on the film becomes negative (tension), in all experimental conditions, we observe the nucleation and growth of cavities at or near the interface between the film and the probe as shown on figure 1. These cavities initially grow in the plane of the adhesive film in a fashion reminiscent of the propagation of multiple cracks. However in a second stage, the walls between the cavities become thinner and start to deform in the direction perpendicular to the plane of the film. At that stage, the structure resembles a honeycomb structure and eventually the walls will fail either by cohesive fracture within the wall or by adhesive debonding from the surface of the probe. The growth of the interfacial cracks and the subsequent evolution of these cracks into a fibrillar structure is strongly dependent on both the rheological properties of the adhesive and on the nature of the surface of the probe.



The video information provides more insight on the physical meaning of the experimental curve. Clearly the maximum stress is related to the appearance of the cavities. However the maximum stress does not correspond to the appearance of the first cavities but rather to the equilibrium between an increasing local stress and a decreasing load-bearing area. This decrease in load-bearing area can be due to two factors:

- The nucleation of new cavities
- The growth of existing ones

If the leading cause of the decrease in load-bearing area is the nucleation of new cavities, it is understandable then that the maximum stress would be directly proportional to the shear storage modulus of the adhesive at an equivalent frequency. This occurs typically for high energy surfaces such as stainless steel or glass. However when the decrease in load-bearing area is mostly due to the growth of the first cavities, giving rise to a few large cracks rather than many small ones as shown on figure 2, the maximum stress is no longer proportional to the shear modulus but is affected by the relationship between the crack propagation velocity at the interface da/dt and the driving force for such a crack propagation G . G is here a local strain energy release rate in a fracture mechanics sense as it has been used in JKR experiments by Maugis.

The transformation of the cracks into a fibrillar structure requires that the cracks do not coalesce into larger cracks but rather stabilize at a fixed size. This only occurs in a viscoelastic window and requires a distribution of relaxation times. Experiments on monodisperse polymer samples

showed that if the molecular weight of the polymer is too high, the formation of fibrils (but not of cavities) can be completely suppressed.

Once the fibrillar structure is formed, its maximum extension is controlled by the elongational properties of the polymer and cannot be simply predicted by shear experiments. If the polymer exhibits a strain hardening behavior in extension, the final extension of the fibrils was found to be roughly constant independent of the temperature and debonding rate. If on the other hand the polymer does not strain harden much, the extension ratio is controlled by the kinetics of disentanglement and the maximum extension occurs for an intermediate value of the debonding rate.