Electronic Supplementary Material (ESI) for Soft Matter. This journal is © The Royal Society of Chemistry 2018



Figure S1 – The stretching unit combined with Probe tack tester used in measurements of films adhesion under uniaxial stretching.

2D Block Model.

Here we represent a short description of the 2D block model developed in [18] and used in the present work with minor changes.

The bulk adhesive layer (grey-rectangle) divided into blocks and motion equations for each block are written.



Figure S2 - Schematic presentation of the actual debonding process (left) and the corresponding 2D block model representation (rights) in 2D block model. (a) Initial state, (b) uniform deformation, (c) cavity expansion, (d) fibrillation. Reproduced with permissions from [18].



Figure S3 – Force balance acting on the *i*-th block. P_i is the pressure in the cavity between (*i*-1)th and i-th blocks; σ_{si} is the shear stress; μX_{si} is interfacial friction; X_i is the center of mass of *i*-th block; X_{si} is the center of block contact line with substrate; ζ is internal variable. Reproduced with permissions from [18].

Model equations:

1. The balance equation for the forces, acting on i-th block:

$$(P_{i+1} - P_i)H_0\lambda = -\frac{2\sigma_{si}W_0}{\lambda}, (S1)$$

2. Slip of the block along the substrate for simplicity is considered to be subject to the law of viscous friction:

$$\mu X_{si} = \sigma_{si, (S2)}$$

3. The real shape of the cavity is ignored, but its volume (in the two-dimensional case - area) corresponds to the volume of the spherical cavity of radius R_i :

$$\pi R_{i}^{2} = H_{0} \lambda \left(X_{i} + X_{i-1} - \frac{W_{0}}{\lambda} \right),$$
(S3)

4. A significant simplification of cavity evolution can be achieved by considering its growth in a viscous Newtonian fluid, which is described by the Rayleigh- Plesset equation:

X-experiment:

$$R_{i} = \frac{R_{i}}{2\eta} \left(P_{c,i} + \sigma_{e} - P_{i} - \frac{\gamma_{a}}{R_{i}} \right),$$
(S4)
Y-experiment:

$$R_{i} = \frac{R_{i}}{2\eta} \left(P_{c,i} - P_{i} - \frac{\gamma_{a}}{R_{i}} \right)$$

5. The process of expansion of a cavity is considered as isothermal:

$$P_{c,i}(t) = P_0 \left(\frac{R_i(0)}{R_i(t)}\right)^2,$$
(S5)

6. The elastic part to debonding stress:

$$\dot{\sigma}_{zz} = -\left(\frac{1}{\tau} - \frac{2\lambda}{\lambda}\right)\sigma_{zz} + \frac{2G\lambda}{\lambda}, \text{ (S6)}$$

7. Total debonding force is calculated as a sum over all blocks:

$$F_{z} - P_{0}S = \frac{S}{N} \sum_{i=1}^{N} \left(\sigma_{zz} - \frac{P_{i} + P_{i+1}}{2} \right) = S(\sigma_{zz} - \overline{P}), \quad (S7)$$

8. Debonding stress is the debonding force divided by contact area (length of the rectangle):

$$\sigma_{tot,z} = \frac{F_z}{S} = \sigma_{zz} + P_0 - P$$
 (S8)



Figure S4 – Dependence of practical work of adhesion W_a (J/m²) on the engineering stress σ_e (MPa) at different initial loads (1, 2, 3, and 5 kg) for PVP-PEG adhesive with 50 wt. % PEG-400



Figure S5 – Dependence of practical work of adhesion W_a (J/m²) on the engineering stress σ_e (MPa) at different initial loads (1, 2, 3, and 5 kg) for PVP-PEG adhesive with 40 wt. % PEG-400.



Figure S6 – Dependence of practical work of adhesion W_a (J/m²) on the engineering stress σ_e (MPa) at different initial loads (1, 2, 3, and 5 kg) for PVP-PEG adhesive with 35 wt. % PEG-400.



Figure S7 – Dependence of peak stress σ_{max} (MPa) on the engineering stress σ_e (MPa) at different initial loads (1, 2, 3, and 5 kg) for PVP-PEG adhesive with 50 wt. % PEG-400.



Figure S8 – Dependence of peak stress σ_{max} (MPa) on the engineering stress σ_e (MPa) at different initial loads (1, 2, 3, and 5 kg) for PVP-PEG adhesive with 40 wt. % PEG-400.



Figure S9 – Dependence of peak stress σ_{max} (MPa) on the engineering stress σ_e (MPa) at different initial loads (1, 2, 3, and 5 kg) for PVP-PEG adhesive with 35 wt. % PEG-400.



Figure S10 – The practical work of adhesion W_a (J/m²) as a function of elongation λ for CommandTM adhesive by 3M. External load 2 kg.



(a)

(b)



Figure S11 – Snapshots of the cavitation process of the stretched PVP-PEG-400 (60/40 wt. %) at elongation $\lambda = 2.76$ (approx. stress 0.81 MPa). (a), initial contact; (b), start of cavities growth; (c), cavitation at maximum of Probe tack curve; (d), surface of the adhesive at the end of experiment. Initial load 2 kg.



Figure S12 – The dimensions L_X, L_Y of the cavities in case of X-experiment.