

Numerical modelling of fibrillation and cavitation in PSA tapes

Krupal Patel

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THÈSE DE DOCTORAT

DE L'UNIVERSITÉ PSL

Préparée à Ecole supérieure de physique et de chimie industrielles de la Ville de Paris

Modélisation numérique de la fibrillation et de la cavitation dans des bandes adhésives

Numerical modelling of fibrillation and cavitation in PSA tapes

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Physique et Chimie des Matériaux

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Introduction

Adhesive tapes are everywhere around us as far as their applications are concerned. They are different from other adhesives as they do not require heat or chemical cross-linking to give a relatively good adhesive strength on a wide range of substrates. The brand name, Scotch tape is almost a synonym for PSA (Pressure-Sensitive adhesives) tapes. The common experimental technique to check the strength of PSA tapes is peeling. In a typical instrumented peeling test, we peel the adhesive tape from the substrate of interest with the help of a tensile testing machine and try to measure the force at different peeling angles with an imposed peeling velocity. However, peeling of PSA tapes seems to be a simple process, but it is not! Generically, the adhesive tape *in operando* consists in a thin soft viscoelastic layer sandwiched between two much more rigid adherends, the tape backing and the substrate. During adhesion rupture, there are many phenomena going on at the micron scale such as cavitation, fibrillation, stringing, etc. in the layer of soft adhesive which affect its adherence properties to different surfaces. As en example, we can see the fibrillation during the peeling of adhesive tape from the substrate with the help of Scanning Electron Microscopy (SEM) in fig. 1.



Figure 1: SEM view of adhesion rupture of a PSA tape, showing fibrillation (X. Morelle, J.Chopin and B. Bresson). After detachment, the stretched fibrils almost completely relax to the original flat surface geometry as seen from the small humps in the front row.

The peeling process of adhesive tapes can occur by different types of failure such as cohesive (bulk) failure or adhesive (interfacial) failure. In this work, we will consider the *interfacial failure* of PSA tapes from the substrate. We can define the fibril as a "failed interfacial failure", where the material remains partly stuck to the opposite surface, resulting in elongated ligaments between the adherends. However, fibrils can also result from the growth of cavities generated in the bulk of the adhesive and undergoing very large strains during peeling. In the case of interfacial failure, these fibrils detach cleanly from the substrate, leaving almost no residuals on the substrate. This work throws light on the numerical modeling of the fibrillation and cavitation processes in the peeling of adhesive tapes from a substrate, with a special focus on the energies expended in the large deformation of these structures and their relative contributions to the total adhesion energy. The LEFM (Linear Elastic Fracture Mechanics) approach can not be expected to properly describe the peeling process out of the small strain limit (very weak adherence). In particular, the key missing ingredient in the modeling of adherence energy of PSA tapes is the criterion for debonding *i.e.* the final detachment of the heavily stretched fibrils from the opposite surface. We need to implement a large strain description of the fibril stretch and of the adhesive behavior since the typical length of these fibrils before debonding is several times larger than the initial thickness of the PSA layer. With such a criterion, we should be able to fully predict the adherence energies for PSAs based on the mechanical properties of the adhesive.

In the initial stages of the debonding process, cavities can nucleate and grow, eventually merging to give rise to fibril arrays. Fibril stretching is supposed to be responsible for dissipation during the later stages of debonding of PSA tapes. The cavities have the function of locally relaxing the confinement provided by the two stiff adherends on the thin incompressible adhesive. One of the main questions of this work is what is the relative contribution from cavitation and fibrillation in the total adherence energy. Can we predict adhesion energies based on only the fibril stretching energy ? is the energy of the cavity expansion negligible for all practical purposes, as it is often considered?

The insight gained by the present numerical modeling of cavitation and fibrillation will help model some recent experiments where PSA tapes are peeled from chemically patterned substrates (the so-called "Gecko adhesion"). More generally, from a technical perspective, uncovering the fundamental science behind the debonding mechanics in PSA will result in better product design strategies, better quality and enhanced product life cycles.

In chapter 1, we give an overview of the work that has been done previously. The history of the invention of the scotch tape at 3M company starts the chapter. Then we discuss the peeling process at length and make it clear that interfacial adhesion is enhanced by the dissipated energy to provide the total adherence energy. As an illustration, previous experiments with custom-made PSA tapes and their properties will be introduced. We then describe a simple model which has been proposed to predict the adherence energy from the large strain mechanical response of the adhesive. In this model the basic assumption is that dissipation mostly occurs in terms of large deformation of the fibrils before snap off. However, both qualitative and quantitative issues arise, and the aim of the subsequent chapters is to contribute to improving this model. In all such discussions, we will also consider the effect of changing the main geometrical factors such as confinement, which have a strong bearing on the actual mechanical response of incompressible and viscoelastic adhesive layers. The chapter will be concluded with a description of patterned substrates and "Gecko adhesion".

In chapter 2, we consider the small strain limit of the fibril extension, *i.e.* the contact of a flat punch on an elastic, homogeneous film, coated over a stiff half-space. We employ the Boundary Element Method to solve this problem. We compare this semi-analytical method to Finite Element Modeling (FEM) of the fibril stretch in the small strain regime. We discuss the implications of this comparison, and especially the breakdown of the BEM method for highly confined, nearly incompressible materials. In order to gain a better understanding, we will also compare the FEM with some scaling models that can describe the limiting conditions of very low and very high confinement of the adhesive patch. The last part of this chapter is devoted to a discussion of the debonding of the adhesive patch in the small strain limits in terms of the LEFM theory for crack propagation and the related stability.

In chapter 3, we present the large strain FEM of the fibril stretch and cavity growth by considering an hyperelastic material behavior. We start the discussion with the selection of the appropriate material models that can describe the large strain behavior of PSA such as Arruda-Boyce and Yeoh and discuss the limitation of their implementations. After that, we discuss the simulation strategy to tackle the large strain behavior associated with the fibril simulations. The effect of geometry on adhesion energy is evidenced by comparing the drawing of stretched fibrils from a soft layer to the large uniaxial strain of cylinders of the same initial height and radius. The results of this comparison will be rationalized through some scaling arguments. Then, an investigation of the effect of the degree of incompressibility on the shape and behavior of the fibril is carried out. We then consider the use of nonlinear hyperelastic fracture mechanics to model the debonding of fibrils after a large stretch. This chapter ends with some modeling of the cavity expansion. First we discuss the effects of strain hardening on the spherically symmetric expansion of the cavities in infinite Arruda-Boyce and Yeoh soft solids. Then we discuss the effect of confinement of cavity growth due to both the two stiff adherends and the interaction with neighboring cavities.

In chapter 4, we discus the implications of these results on fibril drawing and cavity growth for modelling the adherence energy of PSA. We first showcase the peeling experiments done on a homogeneous substrate with standard Scotch 3M600 adhesive tape and the characterization of the debonding process using image analysis. The direct comparison of our fibril simulation with the peeling experiments done on homogeneous substrates is not straightforward since we do not control the fibril dimensions and inter-fibrillar distances. Hence, in this chapter, we compare our fibril simulations with the peel data of PSAs from patterned substrates with well defined geometries. We limit our comparison to the simplest experimental conditions where steady-state peeling is observed and regular fibrillation is obtained. This comparison will be useful both to investigate the fibril debonding criterion and to gain insight into adherence on patterned substrates. At the end of the chapter, we compare the role of fibril stretching and cavity expansion in the peeling process.

The chapter 5 closes this thesis with some general discussion, conclusions and prospects.

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Chapter 1

State-of-the-art on peeling of PSA tapes

RÉSUMÉ

Au chapitre 1, nous donnons un aperçu du travail qui a été fait jusqu'à maintenant. L'histoire de l'invention du scotch par la société 3M commence le chapitre. Ensuite, nous discutons longuement du processus de pelage et précisons que l'adhésion interfaciale est renforcée par la partie dissipée de l'énergie pour donner l'adhésion totale. Cette dissipation se produit principalement par les grandes déformations des fibrilles. Ensuite, nous présentons nos rubans adhésifs faits sur mesure et les différents ingrédients qu'ils contiennent. Un modèle simple a été proposé pour prédire l'énergie d'adhésion à partir de la réponse mécanique en grandes déformations. Cependant, nous verrons que des questions tant qualitatives que quantitatives se posent, et l'objectif des chapitres suivants est de contribuer à améliorer le modèle. Cependant, dans toutes ces discussions, nous introduirons également les concepts de facteurs géométriques tels que le confinement qui ont une forte incidence sur la réponse mécanique réelle des couches adhésives incompressibles et viscoélastiques. Le chapitre se terminera par une description des effets sur l'adhésion de textures apportées au substrat, souvent connus sous le nom d'effet Gecko.

1.1 Introduction about Scotch tapes

Scotch tapes are useful everywhere from gift wrapping to sealing envelops. It was invented by Richard Drew at 3M Company, the USA in 1933, and the first patent was filed by him in 1939. There are many interesting things about the adhesion problem of scotch tape. For example it generates the X-Rays while we peel it under the vacuum at 3 cm/s speed [15] and thin sheets of graphene were first time created by Andre Geim and Konstantin Novoselov [16] with the help of scotch tapes. In this thesis, we will investigate the peeling process, cavitation, fibrillation, and stickiness to high energy surfaces like glass and low energy surfaces like PDMS. Many papers are published on this topic however, it is still relatively mysterious how micron-scale structures called fibrils and cavities affect the adhesive property of the PSA tapes. This work is an attempt at bringing some answers to this question!

As can be seen from fig. 1.1, scotch tape is made up of two layers: one is a stiff backing and the other one is a very soft adhesive layer.



Figure 1.1: Scotch tape and its layers

1.2 Introduction to the peeling

Peeling is the process that is utilized to check the adhesive strength of the interface between the substrate and tapes to be peeled. It looks simple from that point of view but there remains many undiscovered grey areas.

Let us first consider the peeling experiment of PSA tapes wherein we try to peel the commercial tape or custom-made tape from the substrate such as glass or PDMS (Polydimethyl siloxane) or any other surface. Initially, the relatively soft adhesive layer is sandwiched between the stiff backing and the substrate, then gradually it can be peeled from the surface by various means. In this simple experiment the peel force can be mesured by a dead weight as in Ciccotti et al. [1] (fig. 1.2)

They measured velocity as a function of force [1]. The adherence energy Γ can be found by

$$\Gamma = \frac{F}{b} (1 - \cos \theta) \tag{1.1}$$

where F is the dead weight in Newton, b is the width of the tape in meters and θ is the



Figure 1.2: Experimental 90° Peeling setup by Ciccotti et al.[1]

angle of the peel. In the previous experiment, the angle of peel is $\theta = 90^{\circ}$

The very first pioneer work was done by D.H. Kaelble [17] to uncover the theory of peeling by analytical method. His approach stems from observations in instrumented peeling (refer 1.3): people use a tensile testing machine to do such experiments with the camera at the right place to film the process zone (i.e. cohesive zone). Kaelble's model attempts do describe this cohesive zone. It conceives an adhesive layer as a viscoelastic foundation and solves the governing equations for the peeling process. It is different from the singularity-based approach of the Linear Elastic Fracture Mechanics extended to the soft solids by Rivlin et al. [18].



Figure 1.3: Instrumented 90° Peeling setup

1.3 Concepts of fracture Mechanics and Interfacial adhesion

In PSA tapes, fracture occurs at the interface between the soft adhesive layer and the stiff substrate as we are considering only adhesive failure (i.e.interface failures). Then we load the tape mechanically in a controlled manner on the Instron machine to do a peeling experiment. The energy we input by mechanical means is characterized by energy release rate (G). It largely depends on the loading and geometry of the specimen.

According to the energy criteria of Griffith [19], the crack will propagate only if the energy

release rate reaches a critical value G_c which is fracture energy Γ (i.e. Material property).

The intrinsic fracture energy or work of separation is related to surface energy. When two new surfaces are created, it requires to provide energy that is equivalent to $2\gamma A$, where γ is the surface energy of the material and 2A is the new area created. This thermodynamic work of separation $\Gamma_0=2\gamma$ reaches a value in the range of 10^{-2} to 10^{-1} J/m²

However, when we do peeling, the actual energy to peel off the tape from the substrate is in the range of 10^2 to 10^3 J/m² which is at least a thousand times higher than the intrinsic fracture energy Γ_0 . Hence, dissipation plays a vital role. It is seen from the eqn. (1.3). This contribution to the total adhesion energy or fracture energy Γ which is abbreviated as Γ_D depends on the viscoelastic property of the adhesive material used in the PSA tapes.

1.4 Recent models for the peeling energy of PSA

The excellent adhesion of Pressure Sensitive Adhesive (PSA) tapes to the majority of substrates is explained by several factors, including the adhesive's strong viscoelastic behavior and softness, as well as the layer's thin thickness and containment by a stiff backing. Then, depending on the substrate and other peel parameters, there is cavitation and stringing mechanisms during debonding, which are influenced by significant deformation and material response. However, it is still difficult to simulate the intricate connection of all these components at the global level. R. Villey et al. [20] present some significant experiments and modeling claims: They demonstrate a strong relationship between the adhesion energy and the large strain rheology of the adhesives. Any attempt to quantitatively anticipate the adhesion energy must consequently take into account the adhesive's large strain rheology.

During the peeling of a Pressure Sensitive Adhesive (PSA), the adhesive energy Γ (the work which should be provided to peel a unit tape area) is several orders of magnitude above the thermodynamic Dupré's surface energy Γ_0 between the adhesive and the underlying substrate. This demonstrates the dominant role of energy dissipation.

In the slow, steady state regime, Γ is highly depend ant on the temperature T and peeling rate V.

$$\Gamma = \Gamma_0 \left[1 + \Phi(T, V) \right] \tag{1.2}$$

$$= \Gamma_0 + \Gamma_D \tag{1.3}$$

where Γ_0 = is the Intrinsic fracture energy and Φ = is a factor to account for dissipative losses which depends on velocity V and temperature T.

By re-normalizing the velocity axis by the same shift factor a_T that is used for the Time-Temperature Superposition (TTS) of the linear rheological measurements, it has been found that for viscoelastic adhesives, the adherence curves $\Gamma(V; T)$ can be collapsed to a single master curve $\Gamma(a_T V; T_{ref})$ at a reference temperature [21, 22, 23, 24, 25].

Peeling can occur through failure inside of the adhesive layer ("cohesive failure") or through debonding of the adhesive from the substrate ("interfacial" or "adhesive" failure). The latter is the most typical and useful failure mode for PSA since it leaves the substrate clean: we thus focus on interfacial failure in this thesis.

Compositions of the custom-made copolymers used for the adhesives						
Name	EHA	MA	AA	Cross-linker	T_g	
6A	85%	10%	5%	0.2%	-43 ± 5 °C	
6B	85%	10%	5%	0.4%	-43 ± 5 °C	

Table 1.1: EHA stands for 2-ethylhexyl acrylate, MA for methyl acrylate and AA for acrylic acid

By combining real-time microscopic imaging of the debonding region with sound modeling of the mechanical interaction between the bending of the elastic backing and the deformation of the fibrillar network, which will be treated as an effective nonlinear cohesive zone, Villey et al. [2] developed new tools to finely characterize the damage mechanisms at the scale of the debonding region during steady-state peeling of PSA. As can be seen in fig. 1.4, the zone of focus to explain peeling of PSA is the cohesive zone (length L_{dr}) where all the non linear processes occur. Villey et al. [2] performed experiments on custom-made tapes. Out of all, we will be discussing mostly, PSA type 6A and 6B in this thesis for which compositions are given in the table 2.3. Here, PSA type 6B have higher cross-linker than PSA type 6A. Hence, it is having high strain hardening than 6A.

Villey et al. [2] used the lateral camera to film the cohesive zone and fitted the backing profile with the model of elastica theory with the cohesive zone to measure the cohesive stress and crack opening displacement at the given peeling speed.



Figure 1.4: Geometry of a peeling experiment and typical variations of the peeling force F and of the adherence energy G with the peeling velocity V. L_{dr} is the characteristic extension of the debonding region, or cohesive zone, where the adhesive is significantly strained. [2]



Figure 1.5: Adhesion Energy vs. Speed [2]

They made the following conclusions from the fig. 1.5, fig. 1.6 and fig. 1.7:

• Adherence energy increases with speed within the steady-state regime



Figure 1.6: Cohesive stress vs. Speed [2]



Figure 1.7: Crack opening displacement vs. Speed [2]

- Cohesive stress also increases with speed in that regime.
- + Crack opening displacement (i.e., fibrillar height before debonding) is approximately 200 $\mu \rm m$
- There is a slight decrease in the crack opening displacement observed with speed.

In the standard approach, attempts are made to quantitatively model the adhesion energy $\Gamma(V;T)$ using linear viscoelasticity [26, 27, 28]. Subsequently, Chopin et al. [3] used an extensional rheometer to impart the uniaxial extension on the custom-made PSAs 6A and 6B for which he measured nonlinear as well as linear rheology. They used crack-opening displacement data from Villey et al. [20] to get the strain value corresponding to the peeling experiment.

In work done by Chopin et al. [3], they showed that even if the linear rheology is the same for both the PSAs 6A and 6B, it is the nonlinear response or nonlinear rheology (Fig. 1.8a and fig. 1.8c) which plays an important role in determining the adherence energy of the PSA tapes during the peeling process which formulates the complex debonding criteria. It was also deduced that the stress could be divided into a strain rate-dependent term that describes molecular friction and is influenced by the glass transition temperature of the adhesive and a strain-dependent term that describes the strain softening and hardening behavior of the adhesive for more crosslinked Polymer 6B. However, the two elements are no longer easily distinguishable for weakly crosslinked Polymer 6A, and the functional form of strain hardening is dependent on the strain rate, particularly for extremely large strains.

However, the uniaxial extensional rheological data does not fit the peeling data from Villey et al. [2] directly (Fig. 1.8b). In fact the total measured energy must be multiplied by some factor $K_{exp} = 5$ to match the energy of peeling at each velocity in the steady peeling regime as per fig. 1.8d. Hence, they were able to deduce that fibril extension in PSAs during peeling is not simply uniaxial. But the unexplored question remaining in this work is why it is not uniaxial and what is the cause for this $K_{exp} = 5$?



Figure 1.8: Peeling prediction using extensional rheology by Chopin et al. [3]

1.5 Fibrillation and Cavitation



Figure 1.9: Optical image $(3.5\text{mm} \times 5\text{mm})$ showing the cohesive zone in the adhesion rupture of a scotch tape from a substrate (circled green)

1.5.1 Relation between adhesion, cavitation and fibrillation

In fig. 1.9, we can see the cohesive zone or process zone (circled area) in the steady 90° degree peeling of a PSA. In this process zone, one can observe the cavities and fibrils, which have micron-scale dimensions. The thickness of the adhesive layer is 20 μ m and the typical length of such fibrils before interface failure is around 200 μ m. So, it is a very large stretch of the adhesive layer in terms of fibrils.

However, for peeling on a homogeneous substrate such as glass, the initial process in

this interface rupture is cavitation at the interface (in the early stages of rupture in the cohesive zone). After some amount of growth of the cavities due to the incompressible nature of the soft polymer and negative pressure, these cavities merge with each other to give rise to the shape of the fibrils, which cleanly debonds from the surface of the substrate in steady peeling process at the end (at the end of the cohesive zone).

Closely related to the cavitation and fibrillation process in adhesive tape peeling, the probe-tack test is another measure to check the strength and investigate the adhesion mechanisms of Pressure Sensitive Adhesives [29, 30]. We can observe cavitation and fibrillation in this type of test as well. A typical setup for this type of test involves spherical [31] or cylindrical flat ended probes [32, 33, 34, 35]. This probe compresses the adhesive layer till the preset level, and after a preset contact time, it is pulled away at a constant rate. Along with this test, nominal force and displacement are measured. During these experiments, a microscope with camera [33, 36] is placed either at the top or the bottom to visualize the microscopic phenomenons such as cavity growth and fibrillation.

At the macroscopic level, a very high value of tack energy associated with these probetack experiments is due to fracture propagating at the interface along the substrate or very high dissipation associated with the polymer film being split into separate fibrils during the debonding process [33, 37, 38]. As for peeling, this macroscopic description of the process starts with cavitation and growth of the cavities [39, 40] and accompanied by the formation and elongation of the fibrils and finally terminates with their breakage [33, 34, 35, 36, 41, 42].

An initial peak is observed in the stress-strain curve of the tack experiment, which is due to nucleation of the cavity, which is in turn governed by the elastic modulus of the adhesive [30, 33]. This is followed by a stress decrease, which does not go to zero, but approaches a low plateau value or even rises slightly due to cavity growth and multiplication. Finally, the stress approaches zero as the probe separates from the film at a relatively high deformation [35].

However, accurate modeling of fibrillation and cavitation is difficult to perform. People have tried to do some simulations to model the mechanical response of the adhesive in multiple way [43, 44, 45, 46]. The following four parameters play a pivotal role in the peeling or tack experiments which leads to cavitation and fibrillation:

- Confinement between the backing and the substrate
- Incompressible nature of the material
- Non-linear elastic response of the adhesive material such as modulated by the amount of cross-linker in PSAs 6A and 6B
- Visco-elastic nature of the soft adhesive material

1.5.2 Cavitation instability

Understanding how a single cavity expands in an infinite solid is an important topic for the present work. Initial criteria for the spherically symmetric cavity expansion in an infinite soft solid gives a critical internal pressure value $P_c = 5E/6$ where E is Young's modulus. This result is valid for a neo-hookean material but is inadequate when considering the stiffening of the rubber material. A.N. Gent et al. [4] gave the new conditions for propagation of a pressurized crack within a rubber-like solid in terms of the elastic properties of



Figure 1.10: An inflated spherical void and spherical void in a medium under far-field negative pressure [4]

rubber, the fracture energy Γ and the initial radius r_o of the crack. We will use the same approach to derive the relation of pressure in terms of stretch for expanding cavity inside an infinite Arruda-Boyce soft solid and Yeoh soft solid without considering the fracture.

1.6 State-of-the-art on numerical modeling of fibrillation and cavitation

1.6.1 Numerical study of the peel test

Numerical simulations were conducted by Pelfrene et al. [47] to assess the adhesive qualities of a PVB interlayer on glass in the 90peel test. By employing the generalized Maxwell's model and PVB as a visco-elastic material, they are able to solve this issue. An estimate of the cohesive stress ranged from 5 to 15 MPa. At the peel front, a zone of the greatest strain rate was seen.

The zero-degree peel test is an easy approach to determine how adhesive tape reacts to significant shear deformation. The area where the adhesive layer is subjected to high shear can be hundreds of times thicker than the adhesive layer since the backing is very stiff in comparison to the adhesive. A large deformation hyperelastic model is used in this study by Z. Liu et al. [5] to analyze the stress and displacement fields in the adhesive layer under this test. The outcomes of finite element (FE) analysis are then compared to their analytical model. The anticipated stress and deformation accord well with the FE model, with the exception of a tiny area close to the peel front or free edge. They compare their findings to the linear theory and discover that strain hardening and significant adhesive deformation can significantly affect the distribution of shear strain and stress state in the adhesive layer.

They applied the Yeoh model to simulate the significant deformation connected to this problem and obtained the parameters depicted in fig. 1.11. We also used the identical Yeoh parameters to some of those simulations in our numerical computations. We performed data fitting to obtain the proper parameters for PSA types 6A and 6B.

1.6.2 Fibrillation and Cavitation in the confined viscous fluid: Numerical study

To analyse the probe-tack test, the finite-extensibility Giesekus viscoelastic constitutive equation was used by S. Varchanis et al. [6] to model viscoelastic fluidlike adhesives and theoretically examine their behavior during the debonding process from a rigid surface. Together with the constitutive equation that takes into account the non-Newtonian stress contribution, they solve the complete three-dimensional, transient momentum, and mass



Figure 1.11: Nominal stress P (normalizes by the shear modulus μ) versus stretch ratio λ for Yeoh's material with three terms $C_1 = 0.5\mu$, $C_2 = -0.0237\mu$ and $C_3 = 0.00166\mu$ [5]



Figure 1.12: Simulation of tack experiment [6]

conservation equations. According to fig. 1.12, there are initially surface inhomogeneities that result in the small cavities at the adhesive-solid interface in the cylindrical sample of soft pressure-sensitive adhesive sandwiched between two solid disks. As in the experiment, the upper disk begins to move throughout the debonding process, lengthening the sample; meanwhile, the cavities widen laterally, weakening it. The cavities begin to interact with one another and deform primarily in the direction of elongation as the debonding process progresses. This results in the production of thin walls and fibrils, which become thin progressively, and adhesion is lost. Regarding the stress-strain curve and the design of the cavities, their findings qualitatively concur with those of experimental data. Finally, they explore how rheological and geometrical characteristics of the the sample affect the adhesion energy of the material by conducting a parametric analysis.

1.6.3 Fringe instabilities in the fibril

It is readily visible in fig. 1.13 that these fibrils are not axisymmetric. Their edges become warped as a result of what is called a fringe instability. This type of instabilities have been investigated by S. Mora et al. [7] and S. Lin et al. [8]

S. Mora et al. [7] experimentally studied the deformations of heavy elastic cylinders with their axis parallel to the direction of the earth's gravitational field. The specimens, which are constructed of polyacrylamide hydrogels, are fastened to a hard plate at their top







Figure 1.14: Three-dimensional views and horizontal slices at various heights (from the top to the bottom of the samples) of 4 hanging cylinders with different aspect ratios [7]

circular cross-section. The interaction of gravity, which attempts to deform the cylinders downwards under their own weight, and elasticity, which resists these distortions and produces intriguing shapes that can be observed with micro-tomography, leads to an equilibrium configuration. Beyond a critical value of a control parameter that depends on the volume force, height, and elastic modulus, the deformed cylinders for any given initial aspect ratio are no longer axially symmetric: Due to elastic instabilities, self-similar wrinkling hierarchies form and dimples show up at the bottom surface of the shallowest sample. Their findings for four distinct cylinders with various initial aspect ratios are displayed in fig. 1.15.



Figure 1.15: Simulation of fringe instability in the confined elastic layer [8]

S. Lin et al. [8] have shown the coexistence and interaction of the different types of instabilities, such as cavitation, fingering, and fringe instabilities, with the help of a phase diagram by combining numerical, analytical, and experimental methods. This work can help the design of robust adhesives by switching to the desired mode of instabilities while suppressing the other modes.

1.7 State-of-the-art on the experimental study using patterning of the substrate

As seen before, cavitation and fibrillation leads to a self-organised structure in the cohesive zone, which is clearly a key element in understanding adhesion rupture. An interesting strategy to assess its impact is to try and control this structuring through designed heterogeneities at the interface. Indeed, micro-patterning of the substrate leads to a different kind of adhesion when we peel the PSA tapes from it as compared to a homogeneous substrate. It is studied by C. Poulard et al. [9] for soft deformable polydimethylsiloxane (PDMS)/acrylic adhesive interfaces. In this investigation, low aspect ratio cylindrical pillars are used in such patterning as shown in fig. 1.16. In contrast to what has been observed for low aspect ratio rigid patterns, where the adhesion was enhanced due to the increase of the interfacial area due to patterning, it was shown that for soft elastic arrays of cylindrical pillars, the elastic deformation of the patterns could lead to the extra adhesion increase. This study shows that one can tune the adhesion energy by varying the size of the pattern.

Following a different route, E. Chan et al. [10] brought the new concept of wrinkled adhesive layers instead of applying patterning on the substrate. After making the smart adhesive that uses surface wrinkles as patterns to control the adhesion of a poly (n-butyl acrylate) (PnBA) elastomer, they applied it to a glass substrate as shown in fig. 1.17. Following are the advantages of this type of system as compared to substrate patterning:

- Controlled adhesion as compared to substrate patterning
- Fabrication process is simple which does not involve the expensive lithography for patterning
- Applicable to the wide variety of polymer systems.



Figure 1.16: Patterning of the substrate: different morphologies to check the effect on interfacial adhesion [9]

Nature has created surfaces that are reversibly adhesive, and over the past ten years, this stickiness has drawn a lot of research interest. The main takeaway from nature is that fibrillar or "patterned" surfaces, as shown in fig. 1.19 can create stronger adhesion forces to flat and uneven substrates than smooth ones. The principles of fibrillar adhesion are critically examined in this work by M. Kamperman et al. [11] from the viewpoint of contact mechanics. The following are the advantages of splitting the entire contact into fibers (fig. 1.18):







Figure 1.18: Schematic representation of adhesion mechanisms in fibrillar surfaces identified to date ("contact splitting effects"): (a) extrinsic contribution to the work of adhesion;(b) adaptability to rough surfaces; (c) size effect due to surface-to-volume ratio; (d) uniform stress distribution; (e) defect control and adhesion redundancy. The overall effect on adhesion may be a superposition of some or all of these mechanisms [11].

- extrinsic/intrinsic contributions from fibril deformation
- adaptation to rough surfaces by this type of fibril.
- size effects due to surface-to-volume ratio
- uniformity of stress distribution, and defect-controlled adhesion

Therefore, multilevel branching fibers with specific tips make up the intricate structures that enable reversible adhesion. As an example, M. Murphy et al. [12] presented a novel method for creating multilevel structures (fig. 1.19) using polymer materials. They showed how to make arrays of two- and three-level structures with flat mushroom-like points at the

termination of each level. Two-level fiber arrays on a 12-mm-diameter glass hemisphere are the subject of experiments that show increased adhesion compared to samples of onelevel fibers and unstructured control samples. These improvements in adhesion are the outcome of both greater extension and enhanced surface conformation during separation.



Figure 1.19: Scanning electron micrographs of polyurethane hierarchical fibers with flat mushroom tips. The base fibers have approximately 50- μ m-diameter stems with 100- μ m-diameter tips, and the tip fibers have 3- μ m-diameter stems with 5- μ m-diameter tips [12].

In addition to above mentioned articles, there are several others as well [48, 49, 50, 51, 52, 53, 54, 55, 56] which discuss the role of fibrillar structure to improve adhesion property of the given interface.

1.8 Fibril debonding: Experimental study



Figure 1.20: Schematics showing fibril debonding using commercial PSA tapes from PDMS pillar [13].

Fibril debonding is the key missing ingredient in modeling the adherence energy of PSA tapes, according to Chopin et al. [3]. In a recent article by Duigou-Majumdar et al. [13], they study the fibril debonding from the PDMS pillar as shown in fig. 1.20. In this study, they performed experiments showing the detachment of the tape from the top surface of a single micrometric pillar of PDMS elastomer. At a constant displacement rate, the pillar and the adhesive separate resulting in the generation of fibril of adhesive material. They give the power laws for the maximum force and the critical elongation of the fibril at debonding as a function of the diameter of the fibril for the commercial double-sided acrylic adhesive tape using experiments. But, their mathematical model is based on extension of the linear theory to describe the nonlinear fibril debonding phenomenon.

1.9 Problem definition and thesis plan

In this study, we will investigate by numerical modeling the mechanisms of debonding of Pressure Sensitive Adhesives (PSA) and in particular the large stretching of adhesive fibrils and the growth of defects into cavities. We want to model the two phenomena, fibrillation and cavitation, separately and then try to combine their effects.

Recent modeling based on experimental observations has approximated the debonding energy of PSAs during peeling through the work of stretching of the spontaneously formed fibrils (Chopin et al.[3]). The dependency of the debonding energy on the peeling velocity resulted from the combination of the progressive increase of stress in the fibrils with strain rate and the observed progressive decrease of the fibril debonding strain. However, while the description of the fibril stretching by uniaxial large strain tests provided a strong link with the nonlinear viscoelastic rheology of the adhesives, this modeling approach was proven to systematically underestimate the experimental measurements by a constant prefactor of about 5. The main aim of the present work is to provide a mechanical rationale for this prefactor, which we will call "*The Factor 5*"! The secondary, yet very important, objective is to investigate the criterion for fibril debonding to predict the maximum fibril strain instead of measuring it.

Although the strong adherence of PSA is known to stem from the viscoelastic nature of the adhesive, the main hypothesis of this work is that the origin of this *factor 5* must be into the complex mechanics of the large strain drawing a fibril from a flat adhesive film confined between two stiffer adherents, which is expected to require larger stresses than for the simple uniaxial extension of an independent portion of the adhesive material. In order to tackle such a complex phenomenon and to be able to discuss a fibril debonding criterion in a sound and consistent manner, we decided to focus on numerical modeling using a basic hyperelastic material models, with no viscous effect included. We are confident that the most relevant aspects of viscoelasticity can be treated by introducing in a later stage some effective rate dependency based on experimental characterization.

In this representation, the adhesive fibrils derive from the large elastic deformation of an adhesive film of thickness t that is still in perfect contact with the stiff backing, while the contact with the substrate is limited to a circular patch of radius a. In order to represent the interaction with a periodic array of fibrils, it is sound to enforce periodic boundary conditions out of a distance b equal to the half of the interfibrillar distance. In order to reduce the computation time, we also implemented axial symmetry conditions. In this representation, we consider that the interface has previously debonded in the region out of the contact patch of radius a. The perimeter of the contact patch thus corresponds to the front of a crack that can potentially propagate inward, leading to the debonding of the fibril. The additional complications arising from the warping of the fibril surface such as shown in fig. 1.13 and described in S. Lin et al. [8] are not considered in this geometry. Finally, we remark that as there is no interaction between the adhesive and the substrate out of the contact area, this problem is also mechanically analog to the pull-out of a stiff, flat punch of radius a from a soft elastic layer that lays on a stiff substrate.

In chapter 2, we will study the small strain behavior of these fibrils using two numerical methods based on the Boundary Element Method (BEM) and the Finite Element Method (FEM), as well as some simplified analytical arguments such as the Poker-chip test. The flat punch contact to a coated half-space is a good representation of the small strain deformations of this fibril. Hence, we have solved this problem using a semi-analytical approach based on the BEM method. We will also present an algorithm and its implementation

with the Python programming language. The simulation methodology using the FEM is also described in this chapter, and we compare the FEM results with that of the BEM for different aspect ratio of the fibrils and different compressibility levels. Due to discrepancies in the the incompressible case, we have also investigated an approximate analytical model suitable in this case, which we call the Poker chip test. Finally we study the debonding of these fibrils and stability analysis of the crack propagation at the end of the Chapter.

In chapter 3, we will see the large strain behavior of the fibril and cavity. Here, we will discuss the proper material models relevant for large strain, non linear elastomeric response, such as Arruda-Boyce and Yeoh. We will show how we can use these models to fit the extensional rheological data of the custom-made PSA tapes, such as 6A and 6B, thus providing a crude approximation to the viscoelastic response of the fibrils. Then we will use this material model to trace the origin of the "factor of 5" reported in the previous modeling by Chopin et al. [3], meaning that peeling the tape from the given substrate requires five times more energy than predicted from uniaxial extension data, at the different peeling speeds. We will investigate this question by single fibril simulations and we can predict the fibril detachment, which was a missing ingredient in the previous study by Chopin et al. [3]. We will formulate the dependence of fibril debonding on fibril dimensions with the help of J-integral and present the resulting power laws for force and displacement at debonding predicted for PSAs tape 6A and 6B at the given peeling velocity. In addition to that, we will also study cavitation as an initial process in adhesion rupture of the PSA tapes besides fibrillation.

Chapter 4 showcases the peeling experiments done on the homogeneous substrates with a new batch of Scotch 3M600 and the patterned substrates made up of glass and PDMS with custom-made PSA tape 6A and 6B. First, we will compare our simulation results with peel data on the patterned substrate for some cases. Then we will compare the simulations of the confined cavity with mono-fibril simulation. The last chapter discusses the general conclusions and prospects of this work.

Chapter 2

The flat punch contact on a soft confined adhesive: The Small strain limit of the fibril

RÉSUMÉ

Dans le deuxième chapitre, nous commençons avec le problème d'indentation par poinçon plat d'un demi-espace élastique, homogène, ou revêtu d'une couche élastique, en petite déformation. Nous utilisons la méthode des éléments finis (Finite Element Method (FEM)) pour résoudre ce problème. Nous comparons une méthode d'éléments frontière (Boundary Element Method (BEM)) à nos calculs FEM de fibrilles dans un régime de petite déformation. Nous discuterons des implications de cette comparaison, et en particulier de la mise en défaut de la méthode BEM pour les matériaux très confinés et presque incompressibles. Sur la base de ces observations, nous introduisons une méthode d'analyse complète approximative pour traiter ce cas et comparer les résultats avec les méthodes numériques. Nous discutons également de la propagation des fissures dans la limite des petites déformations et examinons en particulier la question connexe de leur stabilité.

2.1 Introduction

Flat punch contact on soft adhesive layer is a good representation of fibrils in the small strain regime. Since semi-analytical and analytical solutions are available for such problem, it will help us to compare the fibril simulations done in the later chapters in the small strain limit here.

Here, the relevant case is of a soft layer deposited on a more rigid substrate. This could be a polymer film on a glass substrate for instance. It is known that confinement of the layer at aspect ratio, (i.e. $a/t \gg 1$) will result in the specific phenomena for nearly incompressible materials [57]. Indeed, for such materials, volumetric deformations are restricted so that shear deformations dominate. In the present case, such deformations are hampered by the confinement. The response is therefore dependent upon the axial compression modulus in the absence of lateral strain–the so-called œdometric modulus of the layer [58]-

$$E_0 = \frac{E(1-\nu_0)}{(1-2\nu_0)(1+\nu_0)} \tag{2.1}$$

In this chapter, first, we will see the previous work done on the indentation problem. In particular, A. Perriot and E. Barthel [59] have developed an algorithm based on the punch (i.e. conical, flat cylindrical, and parabolic) indentation to the coated elastic half-space. We will give a detailed description of the numerical algorithm based on the Boundary Element Method implemented in Python and show a few representative results. Then we will introduce the finite element method for the calculation of flat punch contact on the coated elastic half-space in small strain. Then we will focus on the comparison of the two methods. The discrepancies in the more confined cases will lead us to resort to an approximate analytical model to assess the validity of the numerical methods.

We are going to answer the following questions in this chapter:

- How to use the algorithm based on the Boundary Element Method to solve the problem of flat punch indenting or contacting the coated elastic half-space?
- How do Boundary Element Method (BEM) and finite element method compare?
- Up to what point this algorithm based on BEM can handle the incompressibility associated with the coating layer and its domain of validity?
- How does this semi-analytical method and other analytical approaches such as the Pocker-chip test help us to validate the incompressible case of fibril in the small strain regime?

2.2 Literature review on small strain linear elastic models of indentation problem

Indentation by different shapes of the punch is the most famous experiment to determine the basic mechanical properties locally. Sometimes, it is also known as Vickers' hardness test or Brinell's hardness test. Various types of punch can be utilized in indentation tests such as conical, flat cylindrical, and spherical. In fig. 2.1, conical punch is shown indenting the coated elastic half-space. Now, what is the elastic half-space? An elastic half-space is an elastic material that extends infinitely in all directions including depth with the surface at the top considered as the boundary. In the linear regime, the constants that



Figure 2.1: Conical punch indentation to the coated half space

describe the behavior of an elastic and homogeneous half-space are Young's modulus (E) and Poisson's ratio (ν).

Boussinesq [60] first considered the problem of determining the stress distribution within the elastic half-space when it is deformed by a rigid punch. Boussinesq derived a solution to the problem corresponding to the case of penetration by a solid of revolution whose axis is normal to the original boundary of the half-space, but the form of his solution was not useful for practical computations and partial numerical results based on his solution were derived in the case of conical [61] (fig. 2.1 when t is infinite) and flat [62] (fig. 2.2) indenters by A.E.H. Love.

An important protagonist in developing a simpler solution methodology to Boussinesq's problem (i.e. punch indenting on elastic homogeneous half-space) is I. N. Sneddon [63], who used the Hankel transform and dual integral technique. It makes the calculation simpler. He derived the simple formulas for the depth of penetration of the tip as a function of contact radius of the arbitrary punch profile and for the total load to be applied to the punch to get this penetration.

From these papers, we note some useful results of the flat punch indention on the homogeneous half-space which are the limit cases for flat punch contact on the coated elastic half-space. When we put the thickness of soft layer to the infinity in the coated half-space case we get the homogeneous half-space. Contact stiffness (S_0) and stress intensity factor (K_0) for flat-punch contact on a homogeneous half-space are given by

$$S_0 = 2aE_1^*$$
 (2.2)

$$K_0 = \frac{E_1^* \delta_{fp}}{\sqrt{\pi a}} \tag{2.3}$$



Figure 2.2: Flat punch indentation to the half-space

Where $E_1^* = \frac{E_1}{1 - \nu_1^2}$. E_1 and ν_1 are the moduli and the Poisson's ratio of the half-space respectively, "a" is the radius of the punch, and δ_{fp} is the flat punch displacement.

Li et al. [64] presented the elastic solution of a coated half-space with perfect interfacial bonding under an axisymmetrical compressive loading on the plane surface. It is particularly useful to model the nano-indentation of thin-film coating/substrate systems. This work presents theoretical solutions for the elastic coating/substrate systems. The surface displacement profiles and the stress fields are shown to be sensitive to the thickness of the coating layer and the ratio of the elastic modulus of the coating material to that of the substrate. When the film thickness is comparable to the loading contact radius, the film elastic property cannot be accurately determined by using Sneddon's half-space indentation solution. In addition to that, for the measurement of mechanical property by cone indentation, Sneddon's solution proved to be inadequate as it ignores many important features that are having unavoidable implications. Note that this type of loading, because it only considers the normal displacement, does not model the indenter shape exactly, as has been shown by Hay et al. [65]. The minor corrections taking into account the radial displacement will not be considered here. Hay et al. [65] presented finite elements and pertinent analytical results that show corrections to Sneddon's equations to get the accurate load and contact stiffness.

H.J. Gao et al. [66] studied the contact problem of a rigid cylindrical punch indenting a layered elastic half-space. They used the moduli-perturbation method to derive the first-order accurate analytical solution for the contact compliance of a nonhomogeneous medium with layered or continuously varying moduli in the depthwise direction. In this paper, these solutions are used to estimate the unloading compliance associated with the indentation testing of an Aluminum thin film deposited on a Silicon substrate. Comparison with results obtained from numerical computations indicates that the perturbation estimates are approximately valid for a moderate range of material combinations of practical importance. Also, a finite element analysis is performed to investigate the effects of a penny-shaped debonding crack along the film/substrate interface on the unloading compliance, and to analyze the energy release rate which drives the interface crack.

A.-S. Huguet et al. [67] suggest a synthetic solution to the issue of the adhesive contact of axisymmetric elastic bodies. Thus, a practical and all-encompassing formulation is created, which is proven to immediately give the majority of usable models. Particularly, there is a clear distinction between the functions played by the indenter's shape on the one hand and the characteristics of the attractive interactions on the other. By its very nature, this method can also be applied when the bodies which are interacting but not coming into touch. As a result, long-range interactions and contact features are treated consistently.

Based on the systematic approach by, Huguet et al. [67], E. Barthel et al. [68] proposed an algorithm which can efficiently handle the problem of spherical punch contact to coated elastic half-space which is based on the Boundary Element Method. They gave few numerical examples of the spherical punch indenting the incompressible coating layer over the half-space. In continuation, A. Perriot and E. Barthel [59] developed a numerical algorithm based on the Boundary Element Method for the conical as well as the flat punch in addition to the spherical indentor. We will see this algorithm in detail for the flat punch contact problem in the next section which is not freely available and our aim is to make it available for free and we want to apply it to nearly incompressible film and stiff substrate case.

2.3 Numerical algorithm based on Boundary Element method

A relatively simple numerical algorithm based on an exact integral formulation of the elastic contact of an axisymmetric indenter to a coated substrate is detailed. It provides contact force and penetration as a function of the contact radius.

Consider the system of coated elastic half-space under a load of an axisymmetric frictionless indenter as shown in fig. 2.1. Consider the layer has the thickness t, and the substrate is as usual semi-infinite. The Layer and the substrate have perfect adhesion and are elastic, isotropic, and homogeneous. Now E_0 and ν_0 are elastic moduli and the Poisson ratio of the substrate. Similarly, E_1 and ν_1 are elastic moduli and the Poisson ratio of the layer.

As mentioned in the previous section, Li et al. [64] used Hankel transform to get the relation between the applied normal stress at the layer surface (positive when compressive) and normal displacement (positive when inwards) in their work. However, indentation problem is a mixed boundary problem as one only knows the surface displacement under the contact and the applied stress outside of it.

Hence, the boundary conditions of this problem are the following :

$$\begin{cases} \forall r \le a, \quad u_z(r) = \delta - p(r) \\ \forall r \ge a, \quad \sigma_z(r) = 0 \end{cases}$$
(2.4)

where p(r) the shape of the indenter, a the contact radius and δ is the displacement

Analytically exact form of the relation between normal stress and normal displacement is of little use in their present form given by Li et al.. Hence, in order to solve that problem Barthel and Haiat [69] introduced auxiliary fields to avoid the difficulty. This auxiliary field is defined as cosine Fourier transform of the Hankel transform of a given field (Here, the normal surface stress ($\sigma_z(\mathbf{k})$) and displacement($u_z(\mathbf{r})$). More details on this and related equation one can find in Appendix B. For this problem, equilibrium equation is given by

$$u_z(r) = \int_0^\infty dk \bar{\sigma}_z(k) J_0(kr) \mathcal{C}(kt)$$
(2.5)

where,

$$\mathcal{C}(kt) = \frac{2}{E_1^*} \frac{1 + 4b\,kt\,e^{-2kt} - ab\,e^{-4kt}}{1 - (a + b + 4b(kt)^2)e^{-2kt} + ab\,e^{-4kt}}$$
(2.6)

where,

$$a = \frac{\alpha \gamma_3 - \gamma_1}{1 + \alpha \gamma_3}, \ b = \frac{\alpha - 1}{\alpha + \gamma_1}, \ \alpha = \frac{E_1(1 + \nu_0)}{E_0(1 + \nu_1)}, \ \gamma_1 = 3 - 4\nu_1 \text{ and } \gamma_3 = 3 - 4\nu_0$$

where, $E_1^* = \frac{E_1}{1 - \nu_1^2}$, $J_0(x)$ is the 0th order Bessel function of the first kind and $\bar{\sigma}_z$ the 0th order Hankel transform of σ_z defined as:

$$\bar{\sigma_z}(k) = \int_0^\infty dr r J_0(kr) \sigma_z(k) \tag{2.7}$$

We introduce the auxiliary fields g and θ defined as the cosine transforms of $\overline{\sigma}_z(r)$ and $k \overline{u}_z(r)$ respectively :

$$g(s) = \int_0^\infty dk \,\overline{\sigma}_z(k) \cos(ks) \tag{2.8}$$

$$\theta(s) = \int_0^\infty dk \, k \, \overline{u}_z(k) \cos(ks) \tag{2.9}$$

Expressing eqn. (2.8) and eqn. (2.9) in the real space, we obtain :

$$g(s) = \int_{s}^{\infty} dr \frac{r\sigma_z(r)}{\sqrt{r^2 - s^2}}$$
(2.10)

$$\theta(s) = \frac{d}{ds} \int_0^s dr \frac{r u_z(r)}{\sqrt{s^2 - r^2}}$$
(2.11)

Rewriting eqn. (2.5) with the Hankel transform, we obtain the simple form:

$$k\bar{u}(k) = \mathcal{C}(kt)\bar{\sigma_z}(k) \tag{2.12}$$

Note that g(r) = 0 for r > a. Apply cosine Fourier transform to eqn. (2.12) and we obtain the following equation after some simplification:

$$\theta(s) = \frac{2}{\pi} \int_0^a g(r) \left(\int_0^\infty dk \, \mathcal{C}(kt) \cos(kr) \cos(ks) \right) dr \tag{2.13}$$

Let us now consider eqn. (2.13) for application to an indentation experiment. Let us not make any hypothesis on the shape of the indenter, apart from the fact that it is rigid, convex, axisymmetric, and frictionless. For simplicity, we will consider the contact between the indenter and the coated material to be non-adhesive.

Then, under the contact, our indentation problem turns into an integral equation of the
type $p(r) = \int_0^{\ a} f(s) M(r,s) \mathrm{d} s$ where p and M are known. Here,

$$M(r,s) = \int_0^\infty dk \, \mathcal{C}(kt) \cos(kr) \cos(ks) \tag{2.14}$$

To evaluate the value of the integral equation $p(r) = \int_0^a f(s)M(r,s)ds$, we used trapezoidal rule. The system size (n) is related to the number of integration points of the integral equation of the type $p(r) = \int_0^a f(s)M(r,s)ds$. To evaluate the eqn. (2.14), we used Fast Fourier Transform. The details and the Python code which was originally developed by R.Brossard, are given in Appendix B. For getting the converged value of the stiffness and stress intensity factor in the case of flat punch, we need to tune the system size n and parameter related to Fast Fourier Transform (i.e. V_T and B). Where, $2^{VT} = No$. of points in Fourier space, B = FFT Cut off. The accuracy of the numerical solution depends on the dimension n of the \underline{M} matrix (which is associated to the discretization of [0; a]), the cut-off B for the sampling range of the C(kt) function and the sampling rate $B/2^{V_T}$ for the FFT calculation of the matrix elements. Here, B is the maximum wave vector of the response function C(kt) which defines the resolution in the direct (real) space while the sampling rate defines the maximum characteristic size in the direct space.

In numerical algorithm, we have contact stiffness and stress intensity factor as output variables when shape of the punch is flat. However, the output variables are force as a function of contact radius and displacement when indenters is conical or spherical.

For flat punch contact over the coated half-space problem, we also included the stress intensity factor in the algorithm as there is a crack tip (corner singularity) associated with the problem. The stress intensity factor (K_0) for the case of flat punch contact on homogeneous half-space is given as per follows [70]:

$$K_0 = \frac{E_1^* \delta_{fp}}{\sqrt{\pi a}} \tag{2.15}$$

Derivation of the stress intensity factor (K_I) for the case of flat punch contact on the coated half-space is given below [70] and interestingly, the stress intensity factor is directly related to the auxiliary function g(r) at r=a.

$$K_I = \frac{2g(a)}{\sqrt{\pi a}} \tag{2.16}$$

But in normalized form,

$$G(a) = \frac{2}{\delta E_1^*} g(a) \tag{2.17}$$

$$K_I = \frac{G(a)E_1^*\delta_{fp}}{\sqrt{\pi a}} \tag{2.18}$$

The general treatment explained in Appendix B is applied to the three shapes of indenter as mentioned in the Literature review. However, we are more interested in the flat punch and related outcomes in the subsequent sections as it is useful to compare our fibril calculations in the small strain limit.

For numerical calculations, it is necessary to select adequate value of the parameters n, B, V_T . The convergence has been studied for getting the correct results out of BEM.

Especially the impact of the parameters n, B, V_T for flat punch contact to the coated substrate is studied in fig. 2.3a, fig. 2.3b and fig. 2.3c. As the value of n, B, and V_T increases, the value of contact stiffness converges for modulus contrast 100 and layer Poisson's ratio 0.4. It is found that values of n=700, B=1000 and $V_T=20$ are adequate to find the correct value of the contact stiffness as there is not much change in the value of contact stiffness by further increasing these parameters.



Figure 2.3: Convergence study using parameters (a) B, (b) n, (c) V_T for the case of $a/t=2.2, E_0/E_1=100$ and $\nu_1=0.4$

Numerical algorithm based on Boundary Element Method is shown here in the flowchart 2.4. The notations are useful to understand the Python code given in the section B.3 in Appendix B.



Figure 2.4: Flowchart for implementation of the algorithm based on BEM

2.4 Finite element simulation of the flat punch contact to the coated substrate



Figure 2.5: Flat punch contact to the coated half space

To study the flat punch contact, we have also used FEM. With FEM, we also investigate large strain response in the next chapter. We see in fig. 2.5 that we are pulling on the stiff layer above the soft adhesive layer which is considered as our punch. But, in the linear regime, both pulling and pushing are considered equivalent as we are more interested in comparing the stiffnesses in the case of BEM and FEM calculations.

2.4.1 Simulation strategy

We have decided to keep the domain size of the fibril simulation to be $10 \text{mm} \times 10 \text{mm}$ with 10 strips of size $10 \text{mm} \times 1 \text{mm}$ by using the partition command of ABAQUS. So, progressively we can change the thickness of the adhesive layer as to change the aspect ratio (a/t) easily. Geometry and Boundary conditions are shown in fig. 2.6.

We put the initial crack at the interface between the adhesive layer and the substrate in ABAQUS which means that at the interface, nodes are not connected as explained in fig. 2.6. We need to use appropriate elements type to discretize the domain and choose a meshing strategy for our simulations as well. In order to tackle the singularity at the crack tip, a spiderweb type of mesh was employed as it can be seen in fig. 2.7. With such a mesh the *J*-integral can be calculated over the concentric paths surrounding the node defined as the crack tip. The *J* value is taken on contour number 50. The approximate size of the mesh elements near the crack tip is 1×10^{-6} m which is 0.1% of the punch size "a".

The boundary conditions are represented in fig. 2.6. Axisymmetric and periodic boundary conditions are selected to reduce the domain size and make simulation computationally efficient. Step size selection is also important, so we chose the manual step size available in ABAQUS for small strain calculations. However, for large strain simulation we have



Figure 2.6: Geometry and Boundary conditions for the small strain fibril simulations



Figure 2.7: Mesh at the crack tip

switched to the automatic step size. As we are doing static calculation, we selected static method in step and our aim is to model the small stretch of the fibril so we need not to do anything special here. However, in the subsequent chapters when we need to model the large stretch of the fibril, we selected the NLGEOM option in the step to account for the nonlinear effects which in turn uses Newton-Raphson algorithm with the direct solver by default. The calculations were also done for the different constitutive relations, in anticipation to the next chapter.

2.4.2 Hybrid elements in finite element method for incompressible solids

The following type of elements were used in order to handle Poisson's ratio which is close to 0.5 in the case of incompressible adhesive layer:

- CAX4RH: A 4-node bilinear axisymmetric quadrilateral, hybrid, constant pressure, reduced integration, hourglass control.
- CAX3H: A 3-node linear axisymmetric triangle, hybrid, constant pressure, hourglass control.

Indeed, for the FEM modelling of the deformations of nearly incompressible materials, an efficient strategy is to use hybrid elements [71]. They use a displacement-pressure

formulation for the geometrically and materially nonlinear analysis. The formulation features the a priori replacement of the pressure computed from the displacement field by a separately interpolated pressure; this replacement is performed without reference to any specific material description. Considerations for incremental nonlinear analysis (including contact boundary conditions) are discussed in [71], and various elements are studied. Numerical examples show the performance of the formulation for two- and three-dimensional problems involving isotropic, orthotropic, rubber-like, and elastoplastic materials.

2.5 Results: The Boundary Element Method



Figure 2.8: Flat punch contact to the coated half-space

In earlier work [59], the algorithm based on BEM was tested for modulus contrast between the substrate to soft layer ranging from 0.01 to 100 for different indenter shapes. Computed equivalent moduli showed good agreement with the Gao model [66] for small modulus mismatch ratios ranging from 0.5 to 2 for flat punch calculations. Beyond this range, substantial effects of inhomogeneous strain distribution are evidenced. An empirical function is proposed to fit the equivalent modulus. In this work, we improved the Python implementation of the BEM method for flat punch case and used it for solving the flat punch contact to the compressible soft layer as well as the nearly incompressible soft layer (i.e. Poisson ratio 0.4999) of the coated elastic half-space. We calculated Equivalent Modulus (E_{eq}) and Stress intensity factor (K) as a function of the aspect ratio for the different combinations of the elastic properties for layer and substrate.

To analyse the results, several fit functions have been proposed to find the intrinsic material properties of the thin film from depth-dependent equivalent modulus. Most of them are based on the following eqn. (2.19): [59]

$$E_{eq}^* = E_1^* + (E_0^* - E_1^*)\Phi(x)$$
(2.19)

where E_0^* is the reduced modulus of the substrate and E_1^* is the reduced modulus of the thin film layer. x is the ratio of contact radius a to the layer thickness t and Φ is the 'weight function' which equals 0 when x is zero and 1 when x is infinite.

Representative examples of the equivalent reduced modulus calculated with BEM method for the flat punch contact problem is given in fig. 2.9. Here, we have made a comparison with the Gao model [66] which is based on moduli perturbation and accurate up to the first order. In particular, when the moduli mismatch ratio converges to one, our graph converges to the Gao model. The agreement is good in the range of 0.5-2.



Figure 2.9: Evolution of equivalent reduced modulus with aspect ratio for different modulus contrast and Poisson's ratio: (a) For $\nu_1=0.25$ (b) For $\nu_1=0.40$ (C) For $\nu_1=0.25$ and 0.40 and modulus mismatch ranging from 0.5 to 2 (d) For $\nu_1=0.25$ and 0.40 and modulus mismatch ranging from 0.001 to 1000, where $\frac{E_0}{E_1} = 1$ corresponds to Gao's analytical formula:

From fig. 2.9, it is shown that when the layer thickness is much smaller than the punch radius (i.e. $a/t \gg 1$), we see the equivalent modulus is dominated by the substrate modu-

lus. While, when the layer thickness is much bigger than the punch radius (i.e. $a/t \ll 1$), it is observed that the equivalent modulus is dominated by the layer modulus. This is true for all the modulus contrast or modulus mismatch.

When the mismatch is larger, the shape of the E_{eq} curves are almost unchanged but the transition range, over which the system response changes from one limit behavior to the other, appears to shift from the position given by Gao. In particular, when indenting a soft layer on a stiff substrate, the range over which the behavior of the system is close to that of the film increases with the modulus mismatch, whereas it conversely shrinks when indenting stiff layers.

From fig. 2.9c, it is observed that for the same moduli mismatch ratio and same aspect ratio, the equivalent modulus is more impacted by substrate modulus for a more incompressible layer material. (i.e. for layer having Poisson ratio 0.4). It means incompressibility makes the layer effectively stiffer for the same other parameters. (i.e. same moduli mismatch ratio and aspect ratio). This effect appears in the situation of confinement (i.e. $a/t \gg 1$).

2.6 Comparison of Semi-analytical BEM and FEM methods

In this section we compare the two different methods to calculate the contact stiffness (because it is the linear calculations) and stress intensity factor of underlying problem for the two different values of Poisson's ratio of the soft layer. These two cases of compressible and incompressible layers will help us define the domain of validity of our implementation of the BEM (Boundary element method).

2.6.1 The compressible case



Figure 2.10: von Mises stress distribution: Moduli Mismatch ratio 100:1, $a/t{=}2.2,\,\nu_1{=}0.4,\,\delta{=}0.001~{\rm mm}$

Parameter for simulations						
Name Young's Modulus Poisson Ratio						
rigid substrate	$E_0 = 100 \text{ MPa}$	$\nu_0 = 0.4$				
soft layer	$E_1 = 1$ MPa	$\nu_1 = 0.4$				
rigid Punch	$E_{Punch} = 200 \text{ GPa}$	$\nu_{Punch} = 0.25$				

 Table 2.1: Parameter for simulations: The compressible case



Figure 2.11: Comparison of contact properties calculated with the BEM and the FEM methods: Moduli Mismatch ratio 100:1, Poisson Ratio $\nu_1 = 0.4$, $\delta = 0.001$ mm, a=1.1 mm while t is varying

We first compare the results for a compressible layer for which parameters are given in table 2.1. As our domain of calculations in the case of semi-analytical calculation is semi-infinite (i.e. substrate), in our FEM simulations we have increased box size from 10x10 to 20x20, to check if there is a significant effect of the boundary on the contact stiffness. The results are given in fig. 2.11 which suggest that there is no significant effect on contact stiffness due to increasing the domain size. And there is a good agreement between BEM and FEM for this set of parameters. von Mises stress distribution for aspect ratio 2.2 is given in the fig. 2.10 and is seen to adequately portray the singular field of stress at the crack tip in the small strain limit due to well defined mesh near to the crack tip. The strong modulus contrast induces strong gradient in the iso-stress at the layer-substrate interface.

2.6.2 The near incompressible case



Figure 2.12: von Mises stress distribution: Moduli Mismatch ratio 1000:1, a/t=2.2, $\nu_1=0.4999$, $\delta=0.001$ mm

We now consider the case of a nearly incompressible layer for which parameters are given in the table 2.2 and von Mises stress distribution for aspect ratio 2.2 is given in the fig. 2.12 which shows the stress singularity in the small strain limit at the crack tip for incompressible case. We identify again the diffraction effect of iso-stress lines and find that the stress gradient at the interface is more pronounced which indicate strong effect of larger modulus contrast. In fig. 2.13, one can observe that Normalised contact stiffness (where S_0 corresponds to stiffness value for homogeneous half-space case) increase as the aspect ratio increases. As can be seen from the fig. 2.13a, there is an agreement between finite element results of the fibril small strain calculations with an aspect ratio less than 1 (i.e. $a \ll t$) with that of the Boundary Element Method. But, when $a \gg t$, FEM and BEM are not in good agreement. Hence, we checked the issue of the singular value of the response function (C(kt)) at the larger aspect ratios which we will see in the detail in the subsection related to it. Here, one can notice that all FEM material models such as Neo-Hookean, Arruda-Boyce and Linear theory agrees well in the linear limit as expected.

We can see the stress intensity factor for various aspect ratios of the fibril in the small strain limit using two different methods which are BEM and FEM in fig. 2.13b for parameters given in the table 2.2. As the aspect ratio increases with a constant flat punch radius or fibril radius, (i.e. a=1.1 mm and t is varying) value of the stress intensity factor increases. However, the two methods give a diverging trend as there is a issue with the BEM as pointed out earlier with these sets of parameters.

In fig. 2.14, we plot the normalised contact stiffness for the different aspect ratios by changing the Poisson ratio of soft layer from 0.4999 to 0.45. We can see from the fig. 2.14 that as we decrease the Poisson ratio of the soft layer from nearly incompressible limit to 0.45, there is a better agreement between BEM and FEM for the given modulus mismatch ratio (Here, 1000:1).



Figure 2.13: Moduli Mismatch ratio 1000:1, Poisson ratio $\nu_1 = 0.4999$, a=1.1 mm while t is varying

Parameter for simulations					
Name	Young's Modulus	Poisson Ratio			
rigid substrate	$E_0 = 2950.81 \text{ MPa}$	$\nu_0 = 0.4754$			
soft layer	$E_1 = 3 \text{ MPa}$	$\nu_1 = 0.4999$			
rigid Punch	$E_0 = 2950.81 \text{ MPa}$	$\nu_0 = 0.4754$			

Table 2.2: Parameter for simulations: The near incompressible case



Figure 2.14: Impact of soft layer's Poisson ratio change on the normalised contact stiffness: Modulus mismatch 1000:1, a=1.1 mm while t is varying

2.6.3 Discussion on the probable breakdown of the BEM at high aspect ratios for incompressible soft layer

To better understand the breakdown of the BEM method at high aspect ratios for nearly incompressible soft layer, we investigate the form of kernel of eqn. 2.6. Using the notation

$$\mathcal{C}(kt) = \frac{2}{E_1^*} \frac{N}{D} \tag{2.20}$$

Since, we are interested in the highly confined limit, we will look at the large aspect ratios (i.e. the small k values, where k is the wave vector in the Fourier space). Expanding at small kt, we find that the numerator is given by

$$N = 1 + 4b \, kt \left[1 + \frac{-2kt}{1!} + \frac{4k^2t^2}{2!} + \dots \right] - ab \left[1 + \frac{-4kt}{1!} + \frac{16k^2t^2}{2!} - \frac{64k^3t^3}{3!} + \dots \right]$$

= $(1 - ab) + (kt)(4b + 4ab) + (kt)^2(-8b - 8ab) + (kt)^3(8b + ab\frac{32}{3}) + \dots$ (2.21)

The denominator is given by

$$D = 1 - (a + b + 4b(kt)^2) \left[1 + \frac{-2kt}{1!} + \frac{4k^2t^2}{2!} \dots \right] + ab \left[1 + \frac{-4kt}{1!} + \frac{16k^2t^2}{2!} - \frac{64k^3t^3}{3!} + \dots \right]$$
(2.22)
= $(1 - a - b + ab) + (kt)(2a + 2b - 4ab) + (kt)^2(-2a - 6b + 8ab) + (kt)^3(8b - ab\frac{32}{3}) + \dots$

From the expression of the C(kt) given in eqn. 2.6, it can be seen that as k goes to zero, E_0 goes to infinity and ν_1 goes to 0.5 then

$$\alpha = 0, a = -1, b = -1. \tag{2.23}$$

We see that D is well behaved but first three terms in the expansion of the N go to zero. This is the direct consequence of confined incompressible layer. This is the probable source of problem with this BEM method in this regime.

In fact, at finite thickness t and large radius a, (i.e. taking the limit kt goes to zero), we recover from eqn. (2.6), the uniaxial strain compliance $C = \frac{(1+\nu_1)(1-2\nu_1)}{E_1(1-\nu_1)}$. It duly goes to zero when the material becomes incompressible. This is an effect of confinement. Hence, the apparent reason for the breakdown of BEM method at high aspect ratio and

the incompressible coating layer is investigated.

In numerical analysis, the condition number of a function measures how much the output value of the function can change for a small change in the input argument. This is used to measure how sensitive a function is to changes or errors in the input, and how much error in the output results from an error in the input. A problem with a low condition number is said to be well-conditioned, while a problem with a high condition number is said to be ill-conditioned. For example, the condition number associated with the linear equation AX = b gives a bound on how inaccurate the solution X will be after approximation. For a given system AX = b, condition number is given by $\kappa(A) = \frac{\lambda_{max}(A)}{\lambda_{min}(A)}$. Where, $\lambda_{max}(A)$ and $\lambda_{min}(A)$ are maximum and minimum eigenvalue of the matrix A.

Condition Number of $\mathcal{C}(kt)$ matrix						
Aspect Ratio (a/t)	Poisson's ratio of the	Condition Number				
	soft layer	$\kappa(A)$				
3.14	0.4999	1946				
2.2	0.4999	1459				
1.1	0.4999	962				
3.14	0.4	1470				
2.2	0.4	1220				
1.1	0.4	899				

Table 2.3: Condition Number of $\mathcal{C}(kt)$ matrix: Modulus mismatch 1000:1

In the table 2.3, we gave the calculated value of condition number when $a \gg t$ and for Poisson ratio 0.4999 and 0.4. Contrary to what is expected, the condition number is not high enough to reach to the conclusion that inversion of C(kt) matrix is causing the numerical problem for these sets of parameters (*i.e.*, aspect ratio 3.14, 2.2 and Poisson's ratio 0.4999).

2.7 Simplified analytical arguments for some limiting cases

The Poker-Chip test is a way to test the strength of the elastomer. In this test, the relatively compliant elastomer of thickness t, colored red is as shown in fig. 2.15 is sandwiched between two rigid substrates which are pulled apart. Here the elastomer is incompressible and confined between the two substrates (i.e. $a \gg t$). The corresponding force vs displacement curve is measured. This work has been done with the help of Prof. Herbert Hui, Sibley School of Mechanical and Aerospace Engineering, Cornell University, USA.

The radius of the elastomer sheet is $a \gg t$. Since the modulus of substrate is about 6 orders of magnitude larger than the elastomer, the substrates can be treated as rigid and the elastomer is considered as linear elastic since it is a small strain calculations. Let F denote the force applied by the loading machine. We assume that the elastomer is linear elastic with Young modulus E and Poisson's ratio ν respectively. Here, F= force, δ = displacement, μ =shear modulus, t=thickness of the soft layer, a= radius of the punch.

Using a simple argument, the force required to pull the substrate scales according to the following equation,

$$F = \frac{Ea\delta}{t}\phi(a/t,\nu) \tag{2.24}$$



Figure 2.15: Poker-chip test schematics

The problem is axisymmetric and has non-trivial radial displacement u and vertical displacement w in the vertical direction.

An approximate solution can be obtained by assuming that w depends on z only while the radial displacement u has the form shown in the following eqn.

$$u = u_0(r) \left[1 - \frac{4z^2}{t^2} \right]$$
 (2.25)

Here, $u_0(r)$ is an unknown function to be determined. Substituting these displacements into the stress-strain equations, then we can integrate the stresses through the thickness to obtain a through-thickness average for all the relevant stress components.

$$\bar{\sigma}_{rr}(r) = \frac{1}{t} \int_{-t/2}^{t/2} \sigma_{rr}(r, z) dz$$
(2.26)

With this averaging operation, the equilibrium equation leads to an ordinary differential equation (ODE) which allows us to solve for $u_0(r)$. This equation is as follows:

$$u_{0}^{''} + \frac{u_{0}^{'}}{r} - \left[X^{2} + \frac{1}{r^{2}}\right]u_{0} = 0$$
(2.27)

where

$$X = \frac{2\sqrt{3/2}}{t} \sqrt{\frac{1-2\nu}{1-\nu}}$$
(2.28)

The general solution of the eqn. (2.27) is as follows:

$$u_0(r) = C_1 I_1(Xr) + C_2 J_1(Xr)$$
(2.29)

where, C_1 and C_2 are constants while $I_1(Xr)$ and $J_1(Xr)$ are 1^{st} order Bessel functions of the first and second kind respectively.

Since, $u_0(r) = 0$ at r=0 and $J_1(Xr)$ is ∞ at r=0 which implies C_2 has to be zero. The second boundary condition comes from the volume conservation which gives the value of C_1 .

After applying all the stated boundary conditions, we have the solution of the eqn. (2.27) as follows:

$$u_0 = C_1 I_1(Xr) (2.30)$$

where

$$C_1 = -\frac{(3/2)\nu\varepsilon a}{(1-\nu)\eta I_0(\eta) - (1-2\nu)I_1(\eta)}$$
(2.31)

Here, $\eta = Xa$ and $\varepsilon = \frac{\delta}{t}$, $I_0(x)$ and $I_1(x)$ are the 0^{th} order and 1^{st} order Bessel functions of the first kind.

From this solution, the stresses can be calculated :

$$\sigma_{zz}(r,z) = \lambda(\nabla \cdot \bar{u}) + 2\mu\varepsilon$$
(2.32)

$$= \lambda \left[\frac{1}{r} \frac{\partial(ru_r)}{\partial r} + \varepsilon \right] + 2\mu\varepsilon$$
(2.33)

$$= \lambda X \left[C_1 I_0(Xr) \left(1 - \frac{4z^2}{t^2} \right) \right] + (2\mu + \lambda)\varepsilon$$
 (2.34)

The vertical force F acting on the soft layer is obtained by integrating the normal stress over the area of the soft layer and is found to be

$$\frac{F}{2\pi\mu\varepsilon a^2} = \frac{1}{1-2\nu} \left[\frac{2\nu^2}{(1-\nu)\beta - (1-2\nu)} + (1-\nu) \right]$$
(2.35)

where, $\beta = \frac{\eta I_0(\eta)}{I_1(\eta)}$. When $\nu \to 0.5$ and a/t any finite number, $\eta \to 0$. Hence, eqn. (2.35) reduced to the following expression:

$$F = 3\pi\mu \frac{\delta}{t}a^2 \left(1 + \frac{1}{2}\left(\frac{a}{t}\right)^2\right) \tag{2.36}$$

Integrating eqn. (2.36), we get the strain energy is as per the following eqn.:

$$U_{d} = \frac{3}{2}\pi\mu \frac{\delta^{2}}{t}a^{2} \left(1 + \frac{1}{2}\left(\frac{a}{t}\right)^{2}\right)$$
(2.37)

From strain energy, one can derive the energy release rate:

$$J = \frac{1}{2\pi a} \frac{\partial U_d}{\partial a} \tag{2.38}$$

$$= \frac{3}{2}\mu \frac{\delta^2}{t} \left(1 + \left(\frac{a}{t}\right)^2\right) \tag{2.39}$$

In the regime where BEM and FEM solutions are diverging we also employed the Pokerchip test to validate our FEM simulations. To better understand the results, the FEM solution has been calculated for the exact geometry of the poker chip. To do so, we have changed the boundary conditions for stringent parameters such as modulus contrast 1000:1 and layer Poisson ratio 0.4999 in FEM as shown in fig. 2.16. Here, we put a=b and relaxed the lateral confinement by removing the periodic boundary condition (i.e. $U_1=0$ boundary condition on the surface of the cylinder) in FEM. The results are shown in fig. 2.17 for stiffness and in fig. 2.18 for stress intensity factor. There is a good agreement with the analytical calculations of Pokerchip test. As we can see from fig. 2.17, when $a \ll t$, all flat punch results converge towards S_0 which is the stiffness of the homogeneous half-space case except for the results of the Pokerchip test. The results for the small strain, flat punch simulation have also been reported. We conclude that the poker chip solution gives a smaller stiffness than the full flat punch model. This is expected since in the former the edges are free. However, the edge effects will play a more and more negligible role as the radius increases. Indeed, the evolution of the stiffness of the poker chip parallels the full flat punch result. Without surprise, the same conclusion applies to the stress intensity factor (Fig. 2.18).



Figure 2.16: FEM: Boundary conditions for Pokerchip test



Figure 2.17: Normalised contact stiffness (S/S_0) , $\nu_1=0.4999$, a=1.1mm, t is varying, $S_0 = 2aE_1^* =$ stiffness of the homogeneous half-space indented by the flat cylindrical punch. Where, $E_1^* = \frac{E_1}{1-\nu^2}$.



Figure 2.18: Stress Intensity Factor (K_I): $\nu_1=0.4999$, a=1.1mm, t is varying

2.8 Analytical analysis of debonding criteria in the small strain

Consider that debonding happens when the energy release rate reaches the critical value (i.e. fracture energy) $J=J_c$. As we know, for the half-space, energy release rate is given by,

$$J = \frac{E_1^* \delta^2}{2\pi a} \tag{2.40}$$

Now when $J=J_c$, we can define a critical displacement for contact rupture, which is given by $\delta = \delta_{cr}$. Hence,

$$\delta_{cr} = \sqrt{\frac{2\pi a J_c}{E_1^*}} \tag{2.41}$$

Then using eqn. (2.2) (flat punch stiffness), we get the critical debonding force as follows:

$$F_{cr} = 2\sqrt{2E_1^* J_c} a^{\frac{3}{2}} \tag{2.42}$$

We should note that for the half-space, the critical debonding force is directly proportional to 1.5 power of the patch size or the punch radius in the small strain limit. This is the baseline for more involved fibril debonding in the later chapters.

2.8.1 Stability analysis of the crack propagation at the small strain



Figure 2.19: Stress Intensity Factor with varying punch radius



Figure 2.20: Correction Factor with varying punch radius

The relation between energy release rate (J) and stress intensity factor (K_I) is given below:

$$J = \frac{K_I^2}{2E_1^*} \tag{2.43}$$

We can study the debonding at the given displacement while decreasing the punch radius for the constant thickness of the adhesive layer in the small strain limit with the help of BEM method. We employ eqn. (2.18) in BEM to calculate the stress intensity factor. In fig. 2.19 stress intensity factor is plotted against the punch radius for given other parameters. From fig. 2.19, it can be observed that as we reduce the patch size or the radius of the punch, the stress intensity factor decreases till a becomes comparable to the thickness of the layer t. After that when $a \leq t$, the stress intensity starts to increase with the reduction in radius which is supported from the fact that for the flat punch contact on homogeneous half-space problem, it is also increasing. So, from that point onwards, the crack propagation becomes unstable as we know that $\frac{\Delta J}{\Delta a}$ has to be positive for the stable crack propagation and this variation of the J-integral with the decrease of radius becomes negative when $a \leq t$ as per fig. 2.19¹. Here, correction factor (G(a)) is also decreasing with the patch size or punch radius but becomes stable at unity value when $a \ll t$ as shown in fig. 2.20. This discussion will pave the way for more involved J-integral calculations in the large strain in the later chapters which is useful to describe the fibril debonding phenomenon.

We can also understand stability of the crack propagation at the small strain from the scaling analysis for the limit cases. In the case of flat punch contact on homogeneous half-space, J-integral scales according to the following equation:

$$J \sim \frac{E_1^* \delta^2}{a} \tag{2.44}$$

$$\frac{\partial J}{\partial a} \sim -\frac{E_1^* \delta^2}{a^2} \tag{2.45}$$

From eqn. (2.45), it is observed that variation of the energy release rate with the punch radius is negative, it means when $a \ll t$, crack propagation becomes unstable as it will

¹It can be noted that $\frac{\Delta J}{\Delta c}$ has to be negative for the stable crack propagation. Where "c" is the crack length while in our analysis we use punch radius "a" instead.

reach to the half-space limit.

Similarly, from Poker-chip test analysis we can deduce the following scaling equation for J-integral when $a \gg t$:

$$J \sim \frac{E_1^* \delta^2 a^2}{t^3} \tag{2.46}$$

$$\frac{\partial J}{\partial a} \sim \frac{2E_1^* \delta^2 a}{t^3} \tag{2.47}$$

From eqn. (2.47), we can see that variation of the energy release rate with the punch radius is positive, it means when $a \gg t$, crack propagation is stable in that regime.

2.9 Partial Conclusions

Following are the conclusions or take-home messages from this chapter:

- We described linear models based on the Boundary Element Method, Finite Element Method and analytical Poker-chip test with focus on the compressible as well as the nearly incompressible soft layer to calculate the contact stiffness and stress intensity factor for the flat punch contact to coated elastic half-space problem and more importantly to validate the Finite Element Results with the help of semi-analytical BEM and analytical Poker-chip test.
- The numerical algorithm based on the Boundary Element Method works very well for modulus mismatch (i.e. E_0/E_1) up to 100:1 and till the layer Poisson's ratio is up to 0.4. Moreover, there is a close agreement between contact stiffness and stress intensity factor given by FEM and BEM methods for the different aspect ratios for these parameters. We have also checked the boundary effect in the case of FEM calculations by increasing the domain size from 10×10 to 20×20 , and there is no significant change in the contact stiffness.
- When we change the parameters to modulus contrast (i.e. E_0/E_1) 1000:1 and soft layer Poisson's ratio 0.4999, it is found that the contact stiffness and the stress intensity factor given by BEM match with that of FEM for the cases when $a \ll t$. However, when $a \ge t$, BEM has some issues. The possible reason is the very high condition number of C(kt) matrix (i.e., Green's function) which makes its inversion difficult for these sets of parameters.
- In the regime where BEM has some issues for stringent parameters of modulus mismatch and layer Poisson's ratio, we employed analytical methods such as the Poker chip test, and it seems that the slope of the results given by the Poker-chip test parallels the FEM fibril calculations for the contact stiffness and the stress intensity factor. However, when we changed the boundary conditions in FEM calculations to match that of the Poker chip test, it agreed with the analytical results of the Poker chip test. Hence, it is demonstrated that FEM calculations can handle the incompressibility of the soft layer when it is confined. Still, there is a pending issue with the BEM in this regime as it has to match with the full flat punch FEM simulations.
- In small strain limit, we observe the stable crack propagation as the stress intensity factor decreases with the patch size for the constantly applied displacement and the given thickness "t" of the compressible soft layer (i.e., Poisson's ratio= 0.4) till "a" is greater than "t". However, when "a" becomes smaller than "t/2", it becomes unstable as stress intensity factor starts to increase.

Chapter 3

Fibrillation and Cavitation: Large strain study

RÉSUMÉ

Dans le troisième chapitre, nous commençons la discussion avec les modèles de comportement d'élastomères en grande déformation : Arruda-Boyce, Yeoh et Neo-Hookéen. Les différences entre ces modèles et leurs limitations sont expliquées. Après cela, nous discutons de la stratégie de simulation pour s'attaquer au comportement en grande déformation de fibrilles. L'effet de la géométrie sur l'énergie d'adhérence est mis en évidence en comparant une fibrille étirée et une grande déformation uniaxiale. Il est rationalisé par des arguments d'échelle. Ensuite, une étude de l'effet du degré d'incompressibilité sur la forme du fibrille est réalisée. Après cela, le phénomène de décollement des fibrilles est discuté en détail. Enfin, nous abordons la relation entre cavitation et fibrillation. Nous présentons d'abord des résultats analytiques sur l'expansion sphérique de cavités dans des solides élastiques infinis décrits par des modèles de Arruda-Boyce et de Yeoh. Puis nous abordons par quelques résultats numériques l'expansion de cavités confinées. Les répercussions de ces résultats sur l'adhésion aux PSA sont étudiées dans le chapitre suivant.

3.1 Introduction

In the previous chapter, we focused on the small strain fibril calculations. We presented a numerical algorithm written in Python to implement the Boundary Element Method from Perriot et al.[59] to solve the flat punch contact problem and its comparison with the small strain FEM fibril results. In addition, we presented approximate analytical results to compare with these FEM calculations in the case of nearly incompressible material and large confinement. In the present chapter, we will focus on the large strain behavior of fibrils and cavities.

The first section details the constitutive relations which we will use, going through the Arruda-Boyce and the Yeoh hyper-elastic models. We will also show how they can be used to model the extensional rheological data at different strain rates.

The second section discusses how we will perform the simulation using ABAQUS software and answer the following questions: what are the boundary conditions? How do we come up with these boundary conditions? What is the meshing strategy? How to implement the presence of a crack front and its propagation? More precisely, to perform these fibril simulations, we adopted the following strategy:

- Selection of the proper constitutive model
- Choice of the correct boundary conditions and especially the domain size to account for boundary effects and mimic the interaction between adjacent fibrils
- Modeling of the crack, *i.e.* the debonding at the top of the fibril
- Meshing strategy to tackle the associated singularity
- Proper simulation step size
- Selection of the solver to perform the static fibril simulation.

The next section focuses on the large strain response of fibrils. We first describe the boundary conditions and initial geometry. Comparing fibrils and uniaxial stretching helps us understand how the cohesive zone (Kaelble) works in the peeling process of PSA tapes and how its dissipation can be evaluated based on the uniaxial results [3]? We also assess the results of this large strain fibril simulation by performing a convergence test and interpret the results using scaling arguments. Then we investigate the change in the shape of the stretched fibrils as a function of material compressibility in the large strain regime. A related point is the energy release rate in the large strain simulations, and we will show how it can be used to predict the fibril debonding criterion, which is key to the effective adhesion energy prediction.

Finally, we turn to the the initial stages of peeling, before fibril formation and consider the physics of cavitation. After a review of previous works on expanding cavities in infinite Neo-Hookean solids, we apply the same approach to the more relevant Arruda-Boyce material model, which involves strain hardening and point to significant differences for stability. Then through numerical simulations of *confined* cavities, we try to connect the initial stage of cavitation and the later stage of fibrillation in the peeling of PSAs.

3.2 Constitutive models

We first select some pertinent constitutive models to simulate the large strain behavior of fibrils and cavities. For modeling the nonlinear visco-elastic response of the polymers used in [3], we have considered the three following hyperelastic models:

- 1. Neo-Hookean (Physical model based on polymer chain theory, without strain hardening)
- 2. Arruda-Boyce [72] (Physical model based on polymer chain theory, with strain hardening)
- 3. Yeoh model (Phenomenological Model, with strain hardening)

The Neo-Hookean model has only one parameter, the shear modulus μ , but also contains physically motivated softening at intermediate strains. It has no strain hardening. The other models add hardening resulting from the finite extensibility of the chains. Therefore in the Arruda Boyce model [72] there are two parameters: (1) shear modulus μ (2) locking stretch of the polymer chain λ_L . In the Yeoh model there are three parameters: C_1, C_2, C_3 , which respectively set the shear modulus, the softening regime and the strain hardening regime.



Figure 3.1: Uniaxial deformation using different material models. Arruda-Boyce adds stiffening to the Neo-Hookean model, while the negative C_2 amplifies the native softening of the Neo-Hookean model.

Now, we will give general treatment for the hyperelastic material model with compressibility. Then, we will show how to specialize it for the incompressible elastomer. First, consider W as the strain energy potential for compressible hyperelastic material. Hence, it has two parts: W_1 , which depends on two invariants which are I_1 and I_2 , and is called a deviatoric part, while the other is W_2 which depends on only J and is called a volumetric part.

$$W(I_1, I_2, J) = W_1(I_1, I_2) + W_2(J)$$
(3.1)

Here, I_1 and I_2 are invariants of the right Cauchy-Green tensor. J is the determinant of the deformation gradient tensor **F**. Typically for incompressible materials, J = 1. So, $W_2(1)=0$.

For the implementation of the hyperelastic constitutive model, such as Arruda-Boyce in ABAQUS, we are using the following relations:

$$W(I_1, I_2, J) = C_1 f_1(I_1, I_2) + \frac{1}{D_1} f_2(J)$$
(3.2)

where C_1 is related to the material's shear modulus, D_1 is the compliance of compressibility *i.e.* $D_1 = \frac{2}{K}$ where K is the bulk modulus of the material. For all practical purposes, we put a very high value of the bulk modulus (K) for the incompressible elastomer, *i.e.* D_1 is very small.

More precisely, the approximate Arruda-Boyce strain energy density function is given by the following expression:

$$W = \mu \left[\frac{1}{2} (\bar{I}_1 - 3) + \frac{1}{20\lambda_L^2} (\bar{I}_1^2 - 9) + \frac{11}{1050\lambda_L^4} (\bar{I}_1^3 - 27) + \frac{19}{7000\lambda_L^6} (\bar{I}_1^4 - 81) \right] + \mu \left[\frac{519}{673750\lambda_L^8} (\bar{I}_1^5 - 243) \right] + \frac{1}{D} \left[\frac{J^2 - 1}{2} - \ln(J) \right]$$
(3.3)

On a similar line, a typical Yeoh strain energy density function is as follows:

$$W = C_1(\bar{I}_1 - 3) + C_2(\bar{I}_1 - 3)^2 + C_3(\bar{I}_1 - 3)^3 + \frac{1}{D_1}(J - 1)^2 + \frac{1}{D_2}(J - 1)^4 + \frac{1}{D_3}(J - 1)^6$$
(3.4)

3.2.1 Arruda-Boyce Model for incompressible elastomer

ABAQUS uses the polynomial approximation eqn. (3.3) to the Arruda-Boyce model. But, the exact work of deformation in the case of the Arruda-Boyce model is given by the expression [72]:

$$W = nk\Theta N \left[\frac{r_{chain}}{Nl} \beta + ln \frac{\beta}{sinh\beta} \right] - \Theta c'$$
(3.5)

where, n is the chain density per unit volume, Θ is temperature, r_{chain} is the current chain end to end distance, N is the number of independent chain segments and $\beta = \mathscr{L}^{-1} \left[\frac{r_{chain}}{Nl} \right]$

The locking stretch represents the maximum stretch ratio for an individual chain. Hence at the locking stretch, $r_{chain} = Nl$ and $\lambda_L = \frac{r_{chain}}{r_0} = \frac{Nl}{\sqrt{N}l} = \sqrt{N}$. Do not confuse the chain stretch and the macroscopic stretch, which is larger than λ_L at divergence. This point will be made clearer at the end of this section. Hence, β goes to infinity at the locking stretch, as we know from the property of the inverse Langevin function. As the inverse Langevin function diverges at the locking stretch, Arruda-Boyce diverges at λ_L .



Figure 3.2: Inverse Langevin Function and polynomial approximation.

It can be inferred from fig. 3.2 that the polynomial approximation of the inverse Langevin function does not diverge at 1 and -1. However, the full function diverges at 1 and -1. As ABAQUS uses a polynomial approximation, there is no such divergence at finite strain. We get the finite result at and beyond the locking stretch, but it does not account for the physics.



Figure 3.3: True Stress vs. Stretch for uniaxial extension: Divergence of Arruda Boyce model

To show this point more clearly, we plot the stress vs. stretch diagram for the uniaxial extension for $\lambda_L = 2$ using the Arruda-Boyce model using ABAQUS software in fig. 3.3. It does not diverge at the locking stretch, whereas the same calculations done by solving the full function from eqn. (3.5) using the MATLAB function 'fsolve' can lead to the divergence of the Arruda-Boyce model at the locking stretch.

Here, to establish the range of validity of our later calculations (fig. 3.19 and fig. 3.21), we first calculate the range of validity for uniaxial stretch. As we know that Arruda Boyce

diverges at the chain's locking stretch (λ_L) , we need to compute the corresponding stretch of the uniaxial fibril. Let's call it a bulk locking stretch for the uniaxial case (λ_{BL}) . For that, we should use [72]:

$$\lambda_L = \frac{(\lambda_1^2 + \lambda_2^2 + \lambda_3^2)^{1/2}}{\sqrt{3}}$$
(3.6)

For uniaxial extension, let $\lambda_1 = \lambda_{BL}$ and from the condition of incompressibility (i.e. $\lambda_1 \lambda_2 \lambda_3 = 1$), we have $\lambda_2 = \lambda_3 = \frac{1}{\sqrt{\lambda_{BL}}}$, so that

$$\lambda_{BL}^3 - 3\lambda_L^2 \lambda_{BL} + 2 = 0 \tag{3.7}$$

Solving for each λ_L value to get the corresponding λ_{BL} , we find the evolution shown in fig. 3.4 which can be approximated by $\lambda_{BL} = \sqrt{3}\lambda_L$, the constant being negligible in this stretch range.



Figure 3.4: Uniaxial bulk locking stretch vs. locking stretch of the single chain

$$\mu \left[\frac{19}{7000\lambda_L^6} (8\lambda^8 - \frac{64}{\lambda^4} + 40\lambda^5 + 48\lambda^2 - \frac{52}{\lambda}) + \frac{519}{673750\lambda_L^8} (10\lambda^{10} - \frac{100}{\lambda^5} + 160\lambda^4) \right] + \mu \left[(80\lambda + 70\lambda^7 - \frac{160}{\lambda^2}) \right]$$
(3.8)

We find that the initial shear modulus μ_0 is related to μ by:

$$\mu_0 = \mu \left[1 + \frac{3}{5\lambda_L^2} + \frac{99}{175\lambda_L^4} + \frac{513}{875\lambda_L^6} + \frac{42039}{67375\lambda_L^8} \right]$$
(3.9)

which will turn out useful in the following comparisons between models.

Uniaxial simulation using Arruda-Boyce Model

Uniaxial fibril is defined as homogeneous deformation of a cylinder subjected to uniaxial traction. We will try to understand the boundary condition for such a simulation first. It is represented in fig. 3.5 where one edge is free from normal stress. We use the axisymmetric boundary conditions to make the simulation computationally efficient.



Figure 3.5: Boundary condition for the uniaxial simulation



Figure 3.6: Schematic representation of the uniaxial extension of a cylinder

Then, we want to test the uniaxial simulation using Arruda-Boyce hyperelastic model in ABAQUS software against the analytical expression shown in eqn. (3.8) for the parameters shown in the table 3.1. Here, a = 1.1mm is the initial radius of the cylinder, and t = 1mm is the height of the cylinder. Numerical and analytical results for the force vs. stretch are shown in fig. 3.7 as well as true stress vs. stretch in fig. 3.8. The results are in the good agreement showing that our analytical evaluation and our implementation of the Arruda-Boyce model in ABAQUS is correct. In addition, we have tried to change the locking stretch parameters, which relate to the limiting polymer chain extensibility, as shown in

fig. 3.9. As the locking stretch decreases, the material starts to strain-harden at the low value of the corresponding uniaxial stretch as per fig. 3.9.

Parameter for simulations								
Name Modulus of rigidity Compliance of compress- Locking stretch								
ibility								
soft layer	soft layer $\mu = 1MPa$ $D = 0.0001MPa^{-1}$ $\lambda_L = 2$							

Table 3.1: Parameter for uniaxial simulations: Arruda-Boyce

Figure 3.7: Comparison of analytical and numerical value of force for uniaxial deformation using Arruda-Boyce Model: Large Strain



Figure 3.8: Comparison of analytical and numerical value of Trues stress for uniaxial deformation using the Arruda-Boyce Model: Large Strain



Figure 3.9: Variation of the chain locking stretch (λ_L) for uniaxial deformation using Arruda-Boyce Model: FEM



3.2.2 Arruda-Boyce Model for compressible material

Figure 3.10: True stress vs. stretch for the compressible Arruda-Boyce material: $\mu=1$ MPa, D=1 MPa⁻¹, $\lambda_L=2$

Similarly, we have derived an analytical expression for Nominal stress in the case of a compressible Arruda-Boyce material (eqn. (3.10)). Using this expression, we have plotted the true stress against the numerical simulations in fig. 3.10 showing good agreement up to volume of 2. To get the value of true stress, use the following relation: $\sigma_T = \frac{\sigma_N}{\lambda_2^2}$

$$\sigma_N = \mu \left(\frac{\lambda_1}{(\lambda_1 \lambda_2^2)^{2/3}} - \frac{(\lambda_1^2 + 2\lambda_2^2)\lambda_2^2}{3(\lambda_1 \lambda_2^2)^{5/3}} - \frac{2(\lambda_1^4 + \lambda_1^2 \lambda_2^2 - 2\lambda_2^4)}{(15\lambda_L^2 \lambda_2^2 \lambda_1^2 (\lambda_1 \lambda_2^2)^{1/3})} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^2 + 16\lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} \right) + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^2 + 16\lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^2 + 16\lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^2 + 16\lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^2 + 16\lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^2 + 16\lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^2 + 16\lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^2 + 16\lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^2 + 16\lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^4 \lambda_2^4 \lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^3)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^4 \lambda_2^4 \lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^4 \lambda_2^6 \lambda_1^6)} + \frac{11(-4\lambda_1^6 - 12\lambda_1^4 \lambda_2^4 \lambda_2^6 \lambda_2^6)}{(1050\lambda_L^4 \lambda_2^4 \lambda_1^4 \lambda_2^6 \lambda_1^6 \lambda_2^6 \lambda_2$$

$$\frac{19\mu}{7000\lambda_L^6} \left(-\frac{8\lambda_1^8\lambda_2^2}{3(\lambda_1\lambda_2^2)^{11/3}} - \frac{64\lambda_1^6\lambda_2^4}{3(\lambda_1\lambda_2^2)^{11/3}} + \frac{8\lambda_1^7}{(\lambda_1\lambda_2^2)^{8/3}} - \frac{64\lambda_1^4\lambda_2^6}{(\lambda_1\lambda_2^2)^{11/3}} + \frac{48\lambda_1^3\lambda_2^4}{(\lambda_1\lambda_2^2)^{8/3}} - \frac{256\lambda_1^2\lambda_2^8}{3(\lambda_1\lambda_2^2)^{11/3}} + \frac{96\lambda_1^3\lambda_2^4}{(\lambda_1\lambda_2^2)^{8/3}} \right) = \frac{1000}{1000} + \frac{1000$$

$$-\frac{19\mu}{7000\lambda_L^6} \left(\frac{128\lambda_2^{10}}{3(\lambda_1\lambda_2^2)^{11/3}} - \frac{64\lambda_1\lambda_2^6}{(\lambda_1\lambda_2^2)^{8/3}}\right) - \frac{1384\mu(\lambda_1 - \lambda_2)(\lambda_1 + \lambda_2)(\lambda_1^2 + 2\lambda_2^2)^3}{(336875\lambda_L^8(\lambda_1\lambda_2^2)^{2/3}\lambda_2^4\lambda_1^3))} + \frac{(\lambda_1\lambda_2^4 - 1/\lambda_1)}{D}$$
(3.10)

3.2.3 Yeoh model for incompressible material

We use the Yeoh model as an alternative choice to model the nonlinear elastic response of the incompressible material. The Yeoh strain energy density function is given by

$$W = C_1(\bar{I}_1 - 3) + C_2(\bar{I}_1 - 3)^2 + C_3(\bar{I}_1 - 3)^3$$
(3.11)

where $C_1 = 0.5\mu$. As an example of values assigned to C_2 and C_3 , the uniaxial response of an acrylate PSA tape fitted in [5] is well modelled by $C_2 = -0.0237\mu$ and $C_3 = 0.00166\mu$.

As in section 3.2.1, we tested the implementation of the model in ABAQUS by evaluating the uniaxial extension of a cylindrical fibril with free sliding on the loading boundaries against the analytical calculations. We found excellent agreement between the analytical expression and numerical result given by ABAQUS, as shown in fig. 3.11.



Figure 3.11: Comparison of analytical and numerical results for uniaxial elongation of the cylindrical fibril: Yeoh model.

3.2.4	Characterization	of the	uniaxial	response	of	acrylate	adhesives
	with the Yeoh mo	odel					

Fit parameters					
Name	Strain rate	C_1	C_2	C_3	
	(S^{-1})				
PSA type 6A	2	0.034 MPa	0.0024 MPa	0.0012 MPa	
PSA type 6A	1	0.018 MPa	0.00021 MPa	$2.4 \times 10^{-6} \text{ MPa}$	
PSA type 6A	0.2	0.0082 MPa	0.00012 MPa	$1.2 \times 10^{-6} \text{ MPa}$	
PSA type 6A	0.01	0.0074 MPa	0.00011 MPa	$1.2 \times 10^{-6} \text{ MPa}$	
PSA type 6B	2	0.034 MPa	0.0011 MPa	0.00002 MPa	
PSA type 6B	1	0.019 MPa	-0.00013 MPa	$4.01 \times 10^{-6} \text{ MPa}$	
PSA type 6B	0.2	0.020 MPa	0.00095 MPa	$3.5 \times 10^{-5} \text{ MPa}$	
PSA type 6B	0.01	0.0064 MPa	-3.17×10 ⁻⁵ MPa	$1.44{\times}10^{-6}$ MPa	

Table 3.2: Parameter for simulations for the different strain rate using Yeoh Model

Chopin et al. [3] used two custom-made acrylate tapes made by 3M and labeled 6A and 6B in their work to model peeling adherence based on measurements of the extensional rheology. They measured the extensional data for several different strain rates. In this section, we characterize the same data to get the pertinent constitutive model parameters for our fibril simulations. We started data fitting with the Arruda-Boyce model, but the residuals after the fit procedure were too large. Hence, we eventually used the Yeoh model that provides a better fit for the two PSA tape as illustrated in fig. 3.12 and fig. 3.13. The fitting parameters are reported in table 3.2.

In the following sections, we used the Arruda-Boyce model to simulate the fibril stretch with controlled values of the locking stretch and compare it to the uniaxial stretch (sec. 3.3) in order to investigate the origin of the mismatch found by Chopin [3] when modeling the peeling energy based on extensional rheology. On the other hand, we preferred to use the Yeoh model when the aim was to make quantitative comparisons of the fibril debonding parameters with experimental measurements of peeling on the patterned substrate by Morelle and Bresson (sec. 3.4).



(a) For several strain rates (b) For the reference strain rate of 2 s^{-1} Figure 3.12: Yeoh model fit for PSA tape 6A



(a) For several strain rates (b) For the reference strain rate of 2 s^{-1}

Figure 3.13: Yeoh model fit for PSA tape 6B

3.3 Stretching the fibril

Our aim is to compare fibril stretching to uniaxial stretching. Let us first consider an SEM image of the fibrillation by X. Morelle and B. Bresson at SIMM Lab in fig. 1.13. To approximate this process, we use basically the same model as in chapter 2 and in particular in section 2.4.1. Here we detail the edge boundary conditions: a schematics is shown in fig. 3.14. In our model, we have used axisymmetric boundary condition to simplify the finite-element calculations and reduce our task to simulating a 2D section of a single fibril instead of several fibrils taken in 3D, as seen in the image of adhesion rupture. The interaction with the other fibrils is implemented approximately by imposing periodic boundary conditions ($U_1 = 0$ on the edge). Note also that it can be seen in fig. 1.13 that the real fibrils seen in the PSA peeling process are not axisymmetric but present displacement modulations on the edges: this fringing effect will be discussed later.

The geometrical parameters for the initial configuration of the fibril confined between substrate and backing is described in fig. 3.15, where 'a' is the radius of the fibril, 't' the thickness of the adhesive layer, 'b' the half of the inter-fibrillar distance and the fibril density is $\Phi = \frac{a^2}{b^2}$

The material parameters for the Arruda-Boyce constitutive law for our simulations are shown in table 3.3. The boundary conditions for the fibril simulation can be seen in fig. 3.16.



Figure 3.14: Schematics of fibrils showing boundary conditions

Parameter for simulations						
Name Modulus of rigidity Compliance of compress- Locking stretch						
		ibility				
rigid substrate	$\mu_2 = 1000 MPa$	$D_2 = 0.0001 M P a^{-1}$	$\lambda_{L2} = 3$			
soft layer	$\mu_1 = 1MPa$	$D_1 = 0.0001 M P a^{-1}$	$\lambda_{L1} = 2$			
rigid backing	$\mu_2 = 1000 MPa$	$D_2 = 0.0001 M P a^{-1}$	$\lambda_{L2} = 3$			

Table 3.3: Parameter for simulations: Poisson ratio=0.4999



Figure 3.15: Initial configuration of the fibril

We have first tested the mesh sensitivity of our model. Representative results for a/t = 1.1and $\lambda = 3$ are shown in fig. 3.17a for 219545 elements and in fig. 3.17b for 329682 elements. We find that the distribution remains constant except in the neighborhood of the



Figure 3.16: Boundary conditions for fibril simulations

singularity. Hence, our solutions are said to be grid-independent or robust with respect to the degree of mesh refinement.

To understand the deformation of the fibril, it is interesting to compare the values of shear stress and hydrostatic pressure. Tri-axiality is defined as the ratio of the von Mises stress to the absolute value of pressure. Representative results are shown in fig. 3.18: it can be said that the stress tri-axiality value is three at the center of the stretched fibril, and it decreases at the top and the bottom.

We have calculated fibril stretching for various aspect ratios a/t ranging from 0.275 to 2.2. In particular we have calculated the pulling force, which will be denoted F. As shown in the previous sections, we have also calculated the force F_0 for uniaxial traction, for the same material parameters. In fig. 3.19, we plot F/F_0 versus λ for the different aspect ratios. These results show that the force needed to draw fibrils from a flat soft film is larger than the force to extend cylindrical fibrils by a factor of roughly 4.

This order of magnitude is already set by the small strain results (section 2.6.2). The small strain values of F/F_0 are shown in fig. 3.20 and are seen to first decrease with a/t then increase again. The small a/t regime can be explored easily by hand waving arguments. For a flat-punch indenting a homogeneous half-space, the force in the linear regime is given by following equation: (see chapter 2 for a discussion on homogeneous half-space)

$$F \sim Ea\delta$$
 (3.12)



(a) No. of finite elements = 219545



Figure 3.17: Fibril simulations for a/t=1.1, $\lambda_L=2$, $\lambda=3$: Mesh invariance. von Mises stress distribution



Figure 3.18: Fibril simulations for a/t=2.2, $\lambda_L=2$, $\lambda=3$. Shear and pressure distribution.

and the uniaxial fibril force scales according to:

$$F_0 \sim E \frac{a^2}{t} \delta \tag{3.13}$$

Hence,

$$\frac{F}{F_0} \sim \frac{t}{a} \tag{3.14}$$

where $a \ll t$ and $\delta \ll a$. It can be noticed in fig. 3.20 that when $a \ll t$, the evolution of F/F_0 follows the scaling argument as per eqn. (3.14), at the very small strain, and it is the intercept of F/F_0 with the vertical axis, which represents the ratio of linear contact stiffnesses.

Considering now larger stretches, for smaller a/t (fig. 3.19a), we find that the ratio of F/F_0 increases roughly linearly with stretch as we move from small to moderate stretch. It is clear that as a/t decreases the slope of this linear relation increases.

In contrast, for a/t roughly larger than 1 (fig. 3.19b), the evolution of F/F_0 with λ has a negative initial slope as the stretch increases from 1 to roughly 2. Above 2, there is



Figure 3.19: Normalised force in the fibril simulations for (a) $a \le t$ and (b) $a \ge t$ using the Arruda-Boyce model: $\mu_{Rigid}/\mu_{Soft}=10^3$, $\lambda_L=2$, a=1.1 mm, b=10 mm and t is varying



Figure 3.20: Normalised force of the fibril in the linear regime: $\delta = 0.001$ mm

a change in behavior, and the slope becomes positive again. We also note that as a/t increases, the absolute value of the slope in the first regime, where we have a negative slope, increases. The same reasoning is true for the positive slope regime. (i.e., the stretch of 2 onward)

This behaviour can be explained qualitatively. Initially, when the adhesive layer is confined between the backing and substrate, the shear energy is dominant and the effective modulus is high (cf section 2.7). When the stretch increases, the confinement decreases, and the uniaxial part becomes dominant; hence we have an initial negative slope. However, due to the strain hardening, a positive slope is restored at larger stretches.

3.3.1 Impact of strain hardening

Calculations were carried out with the parameters shown in table 3.4 for the Arruda-Boyce model. We varied the locking stretch values from 2 to 5. From fig. 3.21, we observe that the values of F/F_0 in the small strain for all the different locking stretches remains the same. In a large strain regime, as the value of the locking stretch increases the value of F/F_0 decreases for a given value of the stretch.

In fig. 3.22, we plotted the normalised force against normalised strain. From this graph, we see that all the curves have the same slope but the effect of strain hardening is visible by shift of the curve according to strain hardening value.

We now investigate the impact of the constitutive relation on the previous results. We now turn to the Yeoh potential, which also exhibits hardening. For fig. 3.23, the Yeoh parameters are shown in table 3.5. We observe the same evolution as for the Arruda-Boyce model in fig. 3.19b when $a \gg t$ which suggest that the behaviour shown here is generic for strain hardening elastomers.

Parameter for simulations					
Name	Locking stretch				
Rigid substrate	$\mu_2 = 1000 MPa$	$D_2 = 0.0001 M P a^{-1}$	$\lambda_{L2} = 2, 3, 4, 5$		
Soft layer	$\mu_1 = 0.001 MPa$	$D_1 = 0.0001 M P a^{-1}$	$\lambda_{L1} = 2, 3, 4, 5$		
Rigid backing	$\mu_2 = 1000MPa$	$D_2 = 0.0001 M P a^{-1}$	$\lambda_{L2} = 2, 3, 4, 5$		

Table 3.4:	Parameter	for	simulations:	Arruda-Boyce Model
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Parameter for simulations						
Name C_1 C_2 C_3 $D_1 = D_2 = D_3$						
Soft layer	0.5 kPa	-0.0237 kPa	0.00166 kPa	$40.3 MPa^{-1}$		
Rigid backing	0.5 GPa	-0.0237 GPa	0.00166 GPa	$0.0403 \ GPa^{-1}$		
Rigid substrate	0.5 GPa	-0.0237 GPa	0.00166 GPa	$0.0403 \ GPa^{-1}$		

Table 3.5: Parameter for simulations: Yeoh Model



Figure 3.21: Normalised force in the fibril simulations for the different strain hardening value using the Arruda Boyce model: a/t=1, $\mu_{Rigid}/\mu_{Soft}=10^6$



Figure 3.22: Normalised force vs. Normalised strain for the different strain hardening value using the Arruda Boyce model: a/t=1, $\mu_{Rigid}/\mu_{Soft}=10^6$



Figure 3.23: Normalised force in the fibril simulations using Yeoh model: $\mu_{Rigid}/\mu_{Soft}=10^6$

3.3.2 Explanation for the initial positive slope when $a \ll t$

We now give a more elaborate argument to explain the response at moderate strain and small a seen in fig.3.19a. From fig. 3.24, it is clear that the affected area for this regime is less than the thickness of the adhesive layer in the small strain. Hence, it is near the half-space approximation as the thickness of the adhesive layer increases for the given radius.



Figure 3.24: Kinematics for the cases where $a \ll t$ in the small strain regime
For the flat-punch indenting over a homogeneous half-space, force in the large strain regime $(a < \delta)$ scales according to

$$F \sim E\delta^2 \tag{3.15}$$

instead of eqn. (3.12) because the volume affected by the deformation is now of size δ and not a. Hence

$$\frac{F}{F_0} \sim \frac{\delta^2 t}{a^2 \delta} \sim \frac{t^2}{a^2} (\lambda - 1) \tag{3.16}$$

where $\lambda - 1 = \frac{\delta}{t}$. Eqn. (3.16) qualitatively explains the linear behavior with λ at moderate strains and the increasing slope when $a \ll t$ in fig. 3.19a.



Figure 3.25: Evolution of F/F_0 at small stretch for $a \ll t$ and various a/t values as predicted from eqn. (3.14) and eqn. (3.16).

Combining eqn. (3.14) and eqn. (3.16), we can predict the intercept and slope obtained by the scaling models: this prediction is plotted in fig. 3.25 and is seen to qualitatively match the evolution of F/F_0 versus λ in fig. 3.19a.

3.3.3 Explanation for the initial negative slope when $a \gg t$

In fig. 3.19b, we see the initial negative slope of the F/F_0 versus λ graph. A qualitative argument is provided here for this initial negative slope. It is based on the Neo-Hookean material model. In brief, approximate expressions for the elastic energy for uniaxial stretch is given by

$$W_{Uniaxial} \sim a^2 t \mu \left(\lambda^2 + \frac{2}{\lambda} - 3\right)$$
 (3.17)

For the fibril in the confined case (see Fig. 3.26), we will show below that we have

$$W_{Fibril} \sim a^2 t \mu \left[\left[\lambda_z^2 + \frac{2}{\lambda_z} - 3 \right] + \left[\frac{a^2}{t^2} \left[\frac{1}{\lambda_z} - \frac{1}{\lambda_z^{3/2}} \right]^2 \right] \right]$$
(3.18)

We can see that W_{Fibril} consists of two terms. The first is due to the uniaxial extension, and the second is due to shear. When a/t is large and λ is close to one, the shear part is dominant, but as λ increases, the uniaxial part takes over. Hence, we have a negative slope, as seen in the fig. 3.27.



Figure 3.26: Kinematics for the cases where $a \gg t$



Figure 3.27: Neohooken material: Initial slope for a/t=2.2

To complete the argument, we give the derivation of W_{Fibril} . In the geometry of fig. 3.26

$$\varepsilon_{shear} \sim \frac{\partial u_r}{\partial z}$$
 (3.19)

$$\sim \frac{a(1-\lambda_a)}{\lambda_z t}$$
 (3.20)

$$= \frac{a}{t} \left[\frac{1}{\lambda_z} - \frac{1}{\lambda_z^{3/2}} \right]$$
(3.21)

where, $\lambda_z = \lambda_i \lambda_a = \lambda_{\theta} = \frac{1}{\sqrt{\lambda}}$ because the material is incompressible. From fig. 3.26, one can write the deformation gradient using the cylindrical co-ordinate as per below:

$$\underline{\underline{F}} = \begin{bmatrix} 1 + \frac{\partial u_r}{\partial R} & \frac{1}{R} \frac{\partial u_r}{\partial \theta} - \frac{u_\theta}{R} & \frac{\partial u_r}{\partial z} \\ \frac{\partial u_\theta}{\partial R} & 1 + \frac{1}{R} \frac{\partial u_\theta}{\partial \theta} - \frac{u_r}{R} & \frac{\partial u_\theta}{\partial z} \\ \frac{\partial u_z}{\partial R} & \frac{1}{R} \frac{\partial u_z}{\partial \theta} & 1 + \frac{\partial u_z}{\partial z} \end{bmatrix}$$
(3.22)

$$\underline{\underline{F}} = \begin{bmatrix} \lambda_a & 0 & \frac{a}{t} \left[\frac{1}{\lambda_z} - \frac{1}{\lambda_z^{3/2}} \right] \\ 0 & \lambda_a & 0 \\ 0 & 0 & \lambda_z \end{bmatrix}$$
(3.23)

Here, the Green Lagrange strain tensor is given by $\underline{\underline{E}} = \frac{1}{2} [\underline{\underline{F}}^T \underline{\underline{F}} - \underline{\underline{I}}]$

$$\underline{\underline{E}} = \begin{bmatrix} \lambda_a^2 - 1 & 0 & \lambda_a \frac{a}{t} \left[\frac{1}{\lambda_z} - \frac{1}{\lambda_z^{3/2}} \right] \\ 0 & \lambda_a^2 - 1 & 0 \\ \lambda_a \frac{a}{t} \left[\frac{1}{\lambda_z} - \frac{1}{\lambda_z^{3/2}} \right] & 0 & \lambda_z^2 + \frac{a^2}{t^2} \left[\frac{1}{\lambda_z} - \frac{1}{\lambda_z^{3/2}} \right]^2 - 1 \end{bmatrix}$$
(3.24)
$$W_{Fibril} \sim \mu \ trace(\underline{E}) \ V$$
(3.25)

$$W_{Fibril} \sim a^2 t \mu \left[\lambda_z^2 + \frac{2}{\lambda_z} + \frac{a^2}{t^2} \left[\frac{1}{\lambda_z} - \frac{1}{\lambda_z^{3/2}} \right]^2 - 3 \right]$$
 (3.26)

The derivation of the expression for force is given as follows:

$$F = \frac{\partial W}{\partial \delta} \tag{3.27}$$

$$F = \frac{1}{t} \frac{\partial W}{\partial \lambda} \tag{3.28}$$

$$F_{Fibril} \sim a^2 \mu \left[2\lambda_z - \frac{2}{\lambda_z^2} + \frac{2a^2}{t^2} \left[\frac{1}{\lambda_z} - \frac{1}{\lambda_z^{3/2}} \right] \left[\frac{3}{2\lambda_z^{5/2}} - \frac{1}{\lambda_z^2} \right] \right]$$
(3.29)

$$F_{Uniaxial} \sim a^2 \mu \left[2\lambda_z - \frac{2}{\lambda_z^2} \right]$$
 (3.30)

When we put $\lambda_z = 1 + \frac{\delta}{t}$ in the eqn. (3.29) and consider the small strain limit by neglecting the higher order terms $(\lambda_z^{5/2} \text{ and } \lambda_z^{3/2})$, we would recover the force equation given by the poker chip model (eqn. (2.36) in chapter 2).

3.3.4 Explanation for the positive slope in the strain hardening regime when $a \gg t$



Figure 3.28: Slope of force against stretch: Arruda-Boyce, a/t=2.2, $\lambda_L=2$, b=10 mm

In fig. 3.28, we plotted the slope of the force vs. stretch curve against the stretch using FEM results for fibril and uniaxial simulation. From graph, we can say that slope of the force for fibril is decreasing approximately till stretch of 1.7 and then it starts to increase while for the uniaxial case, it remains almost constant till the stretch of 2.5. Hence we can say that strain-hardening sets in earlier in the case of fibril than its corresponding uniaxial simulation for the high aspect ratio such as 2.2. Hence, we have positive slope in F/F_0 curve starting from stretch of 2 when $a \gg t$.

3.3.5 Impact of changing the inter-fibrillar distance



Figure 3.29: F/F_0 vs. stretch graph for the different b/t ratio for a given a/t: Arruda-Boyce, $\mu_{Rigid}/\mu_{Soft}=10^3$, $\lambda_L=2$

Here, we study the effect of changing the inter-fibrillar distance b on the F/F_0 ratio for the given a/t ratio. In fig. 3.29, we plotted the F/F_0 vs. λ graph for the given a/t ratio and different b sizes. It is observed that there is a moderate increase in the F/F_0 ratio when decreasing b and a moderate increase only appears for b/t ratio smaller than 2, *i.e.* for rather densely packed fibrils. Even in these conditions and despite the upward shift due to the lateral confinement, the evolution of the graph remains similar.

3.3.6 Change in the shape of the fibril due to change in the bulk modulus (i.e. compressibility)



(a) $D_1=1$ MPa⁻¹, $K_1=2$ MPa



(c) $D_1=0.01 \text{ MPa}^{-1}, K_1=200 \text{ MPa}$



(e) D_1 =0.0001 MPa⁻¹, K_1 =20000 MPa

Figure 3.30: The change in the defined interface angle of fibril due to the change in bulk modulus: a/t=1, $\mu_1=1$ MPa, $\lambda_{L1}=2$, $\lambda=3$

Since confinement plays a role in the fibril response, it is interesting to study the impact of compressibility on the shape of the fibril. We have used our standard Arruda-Boyce model with $\mu_1 = 1$ MPa and an aspect ratio a/t = 1 and changed the bulk modulus from 2 to 2×10^4 MPa. We can observe in fig. 3.30 from (a) to (e) that compressibility indeed plays a crucial role in determining the shape of the fibril. As we increase the bulk modulus for the same other parameters, the angle at the interface increases



(b) $D_1{=}0.1 \text{ MPa}^{-1}, K_1{=}20 \text{ MPa}$



(d) $D_1=0.001 \text{ MPa}^{-1}, K_1=2000 \text{ MPa}$

 $(\theta_a=100^\circ, \theta_b=133^\circ, \theta_c=140^\circ, \theta_d=153^\circ, \theta_e=150^\circ)$, which seems directly related to the force required to pull these fibrils. Indeed, from fig. 3.31, one can see that as the bulk modulus increases, the force necessary to stretch the fibril increases for a given value of the stretch. However, the initial increase of the force is visible but after a particular value of the bulk modulus, there is not much increase in the force at the specific stretch. It is clear that the angle θ increases monotonously when decreasing compressibility, as shown in fig. 3.32a. However, for the particular compressibility value, when we vary the applied stretch on the fibril, this angle first increases in the small strain and after sudden increases, remains almost stable in the large strain as shown by fig. 3.32b.

In fig. 3.33, we plotted the fibril force normalized by the force of the corresponding uniaxial simulation of the cylinder of the same patch diameter and thickness of the adhesive layer as that of the fibril. We observe that as the bulk modulus decreases for the given shear modulus value, the force required to pull these fibrils decreases. And it costs less energy to pull the compressible fibril than the relatively incompressible one. The value of the F/F_0 factor also reduces with the reduction in the bulk modulus.



Figure 3.31: Effect of change in Bulk Modulus on the force required to pull the fibril: Arruda-Boyce, a/t=1, b=10 mm, $\mu=1$ MPa, $\lambda_L=2$





(a) Change in angle at the interface with the bulk modulus for the particular stretch: $\lambda=3$

(b) Change in angle at the interface with the stretch for the given bulk modulus: $K_I=20000$ MPa

Figure 3.32: Change in angle at the interface: Arruda-Boyce, a/t=1, b=10 mm, $\mu=1$ MPa, $\lambda_L = 2$



Figure 3.33: Effect of change in Bulk Modulus on F/F_0 ratio: Arruda-Boyce, a/t=1, b=10 mm, $\mu=1$ MPa, $\lambda_L=2$

3.3.7 Release of the local confinement in the fibril simulation

Suppose we remove the no-slip constraint at the interface of the substrate and soft adhesive layer and allow the local sliding of the adhesive on the substrate (without debonding) in the simulation of the fibril as per fig. 3.34. With the simulation parameters given in table 3.5, we observe a reduction of the pull force, as can be seen from fig. 3.35, but we still do not fall back to the uniaxial case. There is still a residual factor by which the force to pull the fibril is larger than the force in the uniaxial case. This observation tells us about the dominant role played by lateral no-slip confinement. When we notice the von Mises stress distribution and pressure distribution in the fig. 3.36a and fig. 3.36b, respectively, we can take the green area roughly in both the figures and the division of the values near that color-code of von Mises stress to Pressure. We arrive at the value of 3. Hence, those parts are similar to the uniaxial extension because the tri-axiality value for uniaxial extension is 3.



Figure 3.34: Release of the local confinement: Sliding allowed at the interface of substrate and adhesive



Figure 3.35: Normalised fibril force : Yeoh Model, PSA tape 6B, $a/t=2.5, t=10 \ \mu m$, $\Phi=5\%$, Sliding allowed



Figure 3.36: Stress distribution: Yeoh Model, PSA tape 6B, a/t=2.5, $t=10\mu$ m, $\Phi=5\%$, Sliding allowed

3.4 Fibril debonding criteria

In this section, we will try to understand the fibril debonding process by numerical analysis. The motivation for this section comes from work by Chopin et al. [3], which points out that the fibril debonding process is the missing ingredient for predicting the adherence energy in the peeling of PSA. In the current version of the model from Chopin et al. [3], the evaluation of the adherence energy requires the measurement of the maximum fibril stretch at debonding, leaving the theory incomplete.

We try to approach the fibril debonding by using the Griffith criterion for the propagation of the triple line at the edge of the contact patch between the adhesive and the substrate. Although the fibril undergoes large strains before debonding, yet it is modeled as hyperelastic, the *J*-integral method [73] is thus a good candidate for evaluating the strain energy release rate G.

First, we will check that the ABAQUS implementation of the J-integral on our stretched fibril provide a result that is in good agreement with an independent estimation based on an energy difference method according to $J = \Delta U_{el}/\Delta A$ at constant applied stretch λ . Here U_{el} is the elastic energy stored in the stretched fibril and A is the contact patch area. Then we investigate the effect produced on the energy release rate from changing the confinement by changing the relative value of the geometrical parameter a and t, and then changing the adhesive material behavior in order to mimic the nonlinear rheology of our custom PSA tapes (6A and 6B) as well as the degree of incompressibility. Imposing the debonding criterion (i.e., constant critical energy release rate) eventually allows deriving a prediction for the maximum values of the force and stretch where the fibril debonding is expected to start.

3.4.1 Energy release rate at large strain (*J*-integral)

To calculate the energy release rate for the debonding of the fibril, we use the crack model in ABAQUS, as in section 2.4.1. The energy release rate G is evaluated by calculating the J-integral [73] on any of the circular contours surrounding the crack.



(a) Arruda-Boyce, $\lambda_L = 2$ (b) Yeoh Model: PSA tape 6A

Figure 3.37: J-integral validation by Energy difference method

In order to validate the ABAQUS implementation of J, we first consider the energy difference between two simulations where everything is constant except the radius of the contact patch a (which corresponds to the crack front position), letting the aspect ratio change from a/t = 1 to 1.1 (for fixed thickness t). More precisely we evaluate the quantity:

$$J = \frac{\Delta U_{el}}{\Delta A} = \frac{U_{el}(a+) - U_{el}(a-)}{2\pi a_{-}(a_{+} - a_{-})}$$
(3.31)

where $U_{el}(a)$ is the strain energy of the fibril simulation having initial radius a and thickness t, and a+ and a- are two close values. The calculations were carried out for both the Arruda-Boyce model (with the parameters of table 3.4 and $\lambda_L = 2$) and the Yeoh model (with the parameters of tape 6A from table 3.6). In figs. 3.37a and 3.37b we show the good agreement between the *J*-integral evaluated by ABAQUS and the one determined by the energy difference method as a function of the applied stretch for the two material models. We recall here that in chapter 2, we had already compared the *J*-integral calculations of our FEM results in the small strain limit with some theoretical solutions such as the method based on the Boundary Element Method when and Poker-chip test solutions.

Once we validated the J-integral evaluation performed by ABAQUS in our large strain simulation, we can explore the dependency of the J-integral on our simulation parameters. In fig. 3.38, we plotted the J-integral curves for different values of the geometrical parameters a and t. We can appreciate that for a given applied displacement the J-integral decreases when increasing the thickness t of the adhesive layer for constant patch diameter a.

In fig. 3.39, we show that the *J*-integral curve as a function of the large applied displacement can be well described by a quadratic dependency $J \propto \delta^2$ for the parameters shown in the caption. We can remark that this is the same dependency as for the linear domain in the case of both the flat punch solution on half space given by eqn. (2.40) and of the poker chip solution for confined patches give by eqn. (2.38). The prefactor for the simulation and the two models is respectively 2.2×10^{10} for the large strain solution, 3.7×10^{10} for the poker chip and 8.54×10^8 for the flat punch. The better agreement with the poker chip is coherent with the elevated confinement of the considered simulation.



Figure 3.38: *J*-integral curves as a function of the applied displacement for different values of the contact radius a and thickness t of the layer, Arruda-Boyce, $\lambda_L=2$



Figure 3.39: Fit of the *J*-integral curve as a function of the applied displacement for $a = 50 \mu \text{m}, t = 20 \mu \text{m}, b = 111 \mu \text{m}$ and Yeoh model (PSA tape 6B).

3.4.2 Prediction of the maximum force and displacement at fibril debonding

We will now consider the consequences of the large strain numerical simulations of the *J*-integral to discuss the consequences of the change of geometrical and material parameters on the debonding of the stretched fibril, in order to compare them with the data of 90° peeling tests in next chapter. In order to be close to the properties of our custom tapes 6A and 6B, we used the Yeoh model fit from table 3.2 for the reference strain rate $\dot{\varepsilon} = 2$ s⁻¹, which corresponds to a peeling speed of 0.1 mm/s. The parameters are reported in tables 3.7 and 3.6.

PSA tape 6A				
Name	C_1	C_2	C_3	$D_1 = D_2 = D_3$
Soft layer	0.034 MPa	0.0024 MPa	0.0012 MPa	$6 \times 10^{-1} MPa^{-1}$
Rigid backing	0.5 GPa	-0.024 GPa	0.0017 GPa	$0.04 \ GPa^{-1}$
Rigid substrate	0.5 GPa	-0.024 GPa	0.0017 GPa	$0.04 \ GPa^{-1}$

Table 3.6: Parameter for simulations: Yeoh Model, PSA tape 6A.

PSA tape 6B				
Name	C_1	C_2	C_3	$D_1 = D_2 = D_3$
Soft layer	0.034 MPa	0.0011 MPa	0.00002 MPa	$6 \times 10^{-1} MPa^{-1}$
Rigid backing	0.5 GPa	-0.024 GPa	0.0017 GPa	$0.04 \ GPa^{-1}$
Rigid substrate	0.5 GPa	-0.024 GPa	$0.0017 \mathrm{~GPa}$	$0.04 \ GPa^{-1}$

Table 3.7: Parameter for simulations: Yeoh Model, PSA tape 6B



(a) PSA tape 6A (b) PSA tape 6B



We reported in fig. 3.40, *J*-integral simulated curves for several patch sizes and a constant tape thickness $t = 20 \ \mu m$ as a function of stretch. By imposing the criterion of debonding based on $J = \Gamma_0$, where Γ_0 represents the interfacial adhesion energy, we can determine the value of the critical stretch for debonding. Then we can evaluate by the force curve, such as fig. 3.41, the corresponding critical force for debonding. By considering a hypothetical constant value of $\Gamma_0 = 30 \text{J/m}^2$ we can thus evaluate the dependency of both the





critical force and stretch as a function of the size a of the contact patch for the two custom PSA tapes (6A and 6B). Both are given in figs. 3.42 and 3.43.



(a) Debonding force

(b) Debonding stretch

Figure 3.42: PSA tape 6A, $t=20 \ \mu m$, $\Phi=5\%$.

We can describe the observed trends by power laws. For both tapes 6A and 6B the debonding force is observed to increase with the patch diameter with power laws of the same exponent 1.61, while the debonding displacement decreases with the patch diameter with the same negative power law exponent of -0.2.



Figure 3.43: PSA tape 6B, $t=20 \ \mu m, \ \Phi=5\%$.

In fig. 3.44, we represent the effect of changing the compressibility parameter D of the adhesive on the *J*-integral curves as a function of stretch. This allows predicting that for the constant interfacial energy Γ_0 , incompressible fibrils will debond earlier than more compressible fibrils.



Figure 3.44: J-integral at different compressibility level: Arruda-Boyce, a/t=1, b=10 mm, $\mu=1$ MPa, $\lambda_L=2$

3.4.3 Effect of sliding contact on debonding

Since the no-sliding condition is one of the main causes of the confinement effects on the fibril stretch and debonding, we decided to perform a new set of simulations where we allowed for the contact region to slide freely on the substrate without debonding. The stretching energy and J-integral curves with and without sliding are reported in fig. 3.45 as a function of applied stretch. As expected, both the energy of stretching and the J-integral are found to be decreased when allowing the sliding, since this reduces the confinement due to the release of lateral constraint. However, when considering the debonding criterion for a constant value of the interface energy Γ_0 , we obtain the counter intuitive prediction

that allowing sliding will make the fibrils debond at a later stage of stretching. This would imply a higher adherence energy in peeling, which is opposite to the observation from Newby et al. [74] that sliding decreases the adherence energy. As an example, if we consider $\Gamma_0 = 20 \text{ J/m}^2$, then the stretch for debonding from fig. 3.45a is 3.2 when sliding is not allowed and 3.7 when sliding is allowed. For corresponding adherence energies would be respectively 3 J/m^2 and 3.54 J/m^2 , based on the stretching energy plots in fig. 3.45b. The reasonable explanation of this counter-intuitive prediction, is that loss of adherence observed in experiments of peeling on sliding substrates is mainly due to a reduction of the effective interface energy Γ_0 when sliding is allowed. This kind of effect can not be captured by our hyperelastic simulation, since it is probably originated by subtle changes of viscoelastic dissipation due to changes in the boundary conditions for sliding as proposed by Newby et al. [74]. This is a major motivation for developing future enhanced simulations accounting for the role of large strain viscoelasticity.



Figure 3.45: Effect of sliding on the stretch energy and J-integral for the PSA tape 6B represented by Yeoh model, with $a = 25 \mu \text{m} t = 10 \mu \text{m}, b = 111 \mu \text{m}, \Phi = 5\%$

3.5 Analytical study of the cavitation

In the previous sections, we focused on the fibrillation process in PSA tapes through numerical simulations. However, cavitation is also an essential ingredient in the initial stages of peeling. So we devote the end of this chapter to some investigations about cavitation that are pertinent for our peeling problem, such as the subtle effect of strain hardening and the role of confinement of the cavity into an adhesive strand.

3.5.1 Literature Survey on Cavitation

Cavitation is the unstable growth of a void or bubble inside of a liquid or solid under a negative hydrostatic pressure [75]. Cavitation is a source of damage in soft materials, including biological tissues, while being primarily investigated in fluids. A broader and more in-depth understanding of cavitation inside soft matter is required to explore the intricate pathways leading to damage and rupture of soft materials, such as PSAs.

Cavitation in solids can occur in the solid itself or liquid phases within a solid, such as water within a tissue or hydrogel that has swelled. Cavitation in solids can harm the substance and surroundings throughout both the expansion and collapse, unlike cavitation in liquids, which results in damage to adjoining solids on collapse. Since at least the 1930s, cavitation in solids has been recorded; nonetheless, this phenomenon and associated pathways to damage have gotten much less attention than cavitation in liquids [76, 77].

Early cavitation-like phenomena were associated with failure in soft solids. Still, the relationship between the critical pressure for cavity expansion and the characteristics of the material was not established until the 1950s. Gent and colleagues [78, 79] connected the radial expansion, λ , and elastic modulus, E, of a neo-Hookean material (such as rubber, hydrogels, liver [80], etc.) to the pressure P within a spherical void [78]. The resistance offered by the interfacial energy of the void was later included [81]. The elastic component and the Laplace pressure are so superimposed to form the pressure equation:

$$P = P_{elastic} + P_{surface} = \frac{E(5 - 4\lambda^{-1} - \lambda^{-4})}{6} + \frac{2\gamma}{r\lambda}$$
(3.32)

where γ is the interfacial energy, r is the undeformed radius of the cavity and E is the modulus of elasticity.

3.5.2 Cavity expansion using Neo-Hookean Model

For a material obeying the simple kinetic theory of rubber elasticity with Neo-Hookean behavior (cf. Section 3.2), the relation between inflating pressure P and expansion ratio λ of the cavity radius is given by

$$\frac{P}{E} = \frac{(5 - 4\lambda^{-1} - \lambda^{-4})}{6} \tag{3.33}$$

The evolution of pressure with stretch is represented in fig. 3.46. From eqn. (3.33), it is evident that the cavity will expand without limit at the critical pressure $P_c = 5E/6$. A remarkable feature of eqn. (3.33) is that cavity expansion does not depend upon the initial cavity radius.

We will go through the details of the derivation of eqn. (3.33) with the help of [82] in the Appendix C. Here we will give some basic equations necessary for continuity. We can first



Figure 3.46: Pressure vs. radial stretch for an expanding cavity in a Neo-hookean solid.



Figure 3.47: Geometry for the expansion of a thick spherical shell of inner radius r_1 and outer radius r_2 .

consider the expansion of a thick spherical shell as shown in fig. 3.47. Let us consider the curvilinear coordinate system θ_i in the strained spherical shell as the spherical coordinate system $(\mathbf{R}, \theta, \phi)$, which has its origin at the center of the shell. We will denote y_i a cartesian reference frame. Hence,

$$\theta_1 = \mathbf{R}, \ \theta_2 = \theta, \ \theta_3 = \phi$$

 $y_1 = \mathbf{R} \sin\theta \, \cos\phi, \ y_2 = \mathbf{R} \sin\theta \, \sin\phi, \ y_3 = \mathbf{R} \, \cos\theta$

We assume that a thick spherical shell has spherical symmetry so that the point (\mathbf{R},θ,ϕ) was originally at (\mathbf{r},θ,ϕ) . Let's consider a thick spherical shell having an initial inner radius r_1 and outer radius r_2 . The values of the inner and outer radius R_1 and R_2 can be deduced directly from the incompressibility condition:

$$r^{3} - R^{3} = r_{1}^{3} - R_{1}^{3} = r_{2}^{3} - R_{2}^{3}$$

$$(3.34)$$

Define the deformation field,

$$Q(R) = \frac{r}{R} = \left[1 + \frac{(r_1^3 - R_1^3)}{R^3}\right]^{1/3}$$
(3.35)

Let's take the x_i -axes to coincide with the y_i -axes. Then, using eqn. (3.35), we can write

$$x_1 = RQ\sin\theta\cos\phi, \quad x_2 = RQ\sin\theta\sin\phi, \quad x_3 = RQ\cos\theta$$
 (3.36)

From eqn. (3.35), we can derive

$$\frac{dQ}{dR} = \frac{1}{R} \left[\frac{1}{Q^2} - Q \right] \tag{3.37}$$

Please find the detail of the derivation of the eqn. (3.38) in Appendix C .Using equilibrium equations and considering normal pressure on the inner surface of the shell as a boundary condition, we have the following equation for stress component:

 $K(R) = 2 \int_{Q_1}^Q \left[(1+Q^3)\Phi \right] dQ$

$$\sigma_{11} = K(R) - P \tag{3.38}$$

(3.39)

where,

$$\begin{array}{c}
 1 \\
 0.8 \\
 0.6 \\
 0.4 \\
 0.2 \\
 0 \\
 10 \\
 12 \\
 14 \\
 16 \\
 18 \\
 20 \\
 R$$

Figure 3.48: Variation of deformation field with deformed radius

Consider the case of the infinite outer radius of the shell. In that case, the body is an infinite solid containing a spherical cavity. At infinity, stresses will vanish. Hence, using eqn. (3.38) and noting that $Q(R) \rightarrow 1$ as $R \rightarrow \infty$, one can deduce the following relation from eqn. (3.38) and eqn. (3.35),

$$P = K(\infty) \tag{3.40}$$

For Neo-Hookean material, $W = \frac{\mu}{2} (I_1 - 3)$ and $\Phi = 2\frac{dW}{dI_1}$ Let's denote $Q_1 = \frac{r_1}{R_1} = \lambda^{-1}$ and we know that $E = 2\mu(1 + \nu)$, where μ is the shear modulus, ν is the Poison's ratio and E is the young modulus. Simplifying, we get the eqn. (3.33). Hence,

$$P = \int_{Q_1}^{Q} \left[4(1+Q^3) \frac{dW}{dI_1} \right] dQ$$
 (3.41)

$$= \int_{Q_1}^{Q} \left[4(1+Q^3)\frac{\mu}{2} \right] dQ \qquad (3.42)$$

$$= 2\mu \left[\left(\frac{Q^4}{4} + Q \right) \right]_{Q_1}^1$$
 (3.43)

$$\frac{P}{E} = \frac{(5 - 4\lambda^{-1} - \lambda^{-4})}{6} \tag{3.44}$$

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3.5.3 Energy required for cavity inflation without fracture (Neo-Hookean)

We can estimate the energy requirement for the void growth in the infinite solid by integrating the work done by pressure from previous expression:

$$W = \int_{V_0}^{V} P dV \tag{3.45}$$

$$= \int_{V_0}^{V} \frac{E(5 - 4\lambda^{-1} - \lambda^{-4})}{6} dV \qquad (3.46)$$

$$= \int_{1}^{\lambda} \frac{E(5 - 4\lambda^{-1} - \lambda^{-4})}{2} \lambda^{2} V_{0} d\lambda \qquad (3.47)$$

$$= \frac{EV_0}{2} \left[\frac{5\lambda^3}{3} - 2\lambda^2 + \frac{1}{\lambda} - \frac{2}{3} \right]$$
(3.48)

As an example, if we consider the growth of a cavity from an initial diameter of 1 μ m, to the final diameter of 20 μ m ($\lambda = 20$), which corresponds to the typical thickness of 20 μ m of PSAs, the energy of inflation is estimated to 0.00022 μ J for μ =68 kPa (E=204 kPa).

However, we considered a Neo-Hookean material model, which does not account for the strain hardening, as it will be considered in the next section.

3.5.4 Cavity expansion using Arruda-Boyce model

$$\frac{P}{\mu} = \frac{1}{1238242005000\lambda_1^{20}\lambda_L^8} [-9538368840 - 11221610400\lambda_1^3 - 3052278028800\lambda_1^{18} - 11221610\lambda_1^3 - 3052278028800\lambda_1^{18} - 112216\lambda_1^3 - 3052278028800\lambda_1^{18} - 112216\lambda_1^3 - 305227802800\lambda_1^{18} - 112216\lambda_1^3 - 30522780\lambda_1^3 - 30520\lambda_1^3 - 30520\lambda_1^3 - 30520\lambda_1^3 - 3052\lambda_1^3 - 3052\lambda_1^3 - 3052\lambda_1^3 - 305\lambda_1^3 - 305\lambda_1^3 - 300\lambda_1^3 - 30\lambda_1^3 - 30\lambda_1$$

$$915683408640\lambda_1^{15} - 109009929600\lambda_1^6 - 138739910400\lambda_1^9 -$$

 $572302130400\lambda_1^{12} - 2494891896120\lambda_1^{20} + 6104556057600\lambda_1^{21} + 763069507200\lambda_1^{24} + 6104556057600\lambda_1^{21} + 6004500\lambda_1^{21} + 600450\lambda_1^{21} + 600450\lambda_1^{21} + 6004\lambda_1^{21} +$

$$+ 436039718400\lambda_1^{20} + 3095605012500\lambda_L^8\lambda_1^{20} - 334325341350\lambda_1^{20}\lambda_L^6 + 591341227785\lambda_1^{20}\lambda_L^2$$

$$+\ 89363073800\lambda_L^4\lambda_1^{20}-8648039400\lambda_L^4\lambda_1^8-4136544720\lambda_L^2\lambda_1^7-61912100250\lambda_L^6\lambda_1^{12}-$$

$$11530719200\lambda_L^4\lambda_1^{11} - 32265048816\lambda_L^2\lambda_1^{10} - 99059360400\lambda_L^6\lambda_1^{15} - 69184315200\lambda_L^4\lambda_1^{14} - 6000000\lambda_L^4\lambda_L^{14} - 60000\lambda_L^4\lambda_L^{14} - 6000\lambda_L^4\lambda_L^{14} - 600\lambda_L^4\lambda_L^{14} - 600\lambda_L^4\lambda_L$$

$$3360942585\lambda_L^2\lambda_1^4 - 619121002500\lambda_L^8\lambda_1^{16} - 2476484010000\lambda_L^8\lambda_1^{19} + 990593604000\lambda_1^{21}\lambda_L^6 + 60000\lambda_1^{21}\lambda_L^{21}\lambda_L^{21} + 60000\lambda_L^{21}\lambda_L^{21}\lambda_L^{21} + 6000\lambda_L^{21}\lambda_L^{21}\lambda_L^{21} + 6000\lambda_L^{21}\lambda_L^{21}\lambda_L^{21} + 6000\lambda_L^{21}\lambda_L^{21}\lambda_L^{21} + 6000\lambda_L^{21}\lambda_L^{21}\lambda_L^{21} + 6000\lambda_L^{21}\lambda_L^{21}\lambda_L^{21} + 6000\lambda_L^{21}\lambda_L^{21} + 600\lambda_L^{21} + 600\lambda_L$$

$$86040130176\lambda_1^{25}\lambda_L^2 + 215100325440\lambda_1^{22}\lambda_L^2 + 138368630400\lambda_1^{23}\lambda_L^4 - 495296802000\lambda_L^6\lambda_1^{18} - 48000\lambda_L^6\lambda_L^{18} - 4800\lambda_L^6\lambda_L^{18} - 4800\lambda_L^6\lambda_L^{18} - 4800\lambda_L^6\lambda_L^{18} - 4800\lambda_L^6\lambda_L^{18} - 4800\lambda_L^{16}\lambda_L^{18} - 4800\lambda_L^{18}\lambda_L^{18} - 4800\lambda_L^{16}\lambda_L^{16} - 4800\lambda_L^{16}\lambda_L^{16} - 4800\lambda_L^{16}\lambda_L^{16} - 4800\lambda_L^{16}\lambda_L^{18} - 4800\lambda_L^{16}\lambda_L^{18} - 4800\lambda_L^{18}\lambda_L^{18} - 4800\lambda_L^{16}\lambda_L^{18} - 4800\lambda_L^{16}\lambda_L^{18} - 4800\lambda_L^{16}\lambda_L^{18} - 4800\lambda_L^{16}\lambda_L^{18} - 4800\lambda_L^{16}\lambda_L^{18} - 4800\lambda_L^{18}\lambda_L^{18} - 480\lambda_L^{18}\lambda_L^{18} - 480\lambda_L^{18}\lambda_L^{18}$$

$$138368630400\lambda_L^4\lambda_1^{17} - 161325244080\lambda_L^2\lambda_1^{16} - 645300976320\lambda_L^2\lambda_1^{19} - 46092926880\lambda_L^2\lambda_1^{13} + 645300976320\lambda_L^2\lambda_1^{19} - 645300020\lambda_L^2\lambda_1^{19} - 645300020\lambda_L^2\lambda_1^{19} - 64530\lambda_L^2\lambda_1^{19} - 64530\lambda_L^2\lambda_1^{19} - 6453\lambda_L^2\lambda_1^{19} - 645\lambda_L^2\lambda_1^{19} - 645\lambda_L^2\lambda_1^{19} - 645\lambda_L^2\lambda_1^{19} - 64\lambda_L^2\lambda_1^{19} - 64\lambda_L^2\lambda_1^{19$$

 $415105891200 ln(\lambda_1) \lambda_1^{20} \lambda_L^4] \quad (3.49)$

To investigate the role of strain hardening, we use the Arruda-Boyce model to study the expansion of the cavity in an infinite incompressible solid. For making computation easier, we will use the polynomial approximation for the Arruda-Boyce model that has been

provided by eqn. (3.3). Since we consider an incompressible material, we can drop the compressibility terms and omit the bar over I_1 .

Hence, following the same procedure as that of Neo-Hookean solid. By inserting the expression for the strain energy density of the Arruda-Boyce model from eqn. (3.3) into eqn. (3.41), we obtain the expression as per eqn. (3.49) for the pressure required to expand the cavity by a factor λ

This expression is plotted in fig. 3.49 for two different values of the locking stretch. When comparitng with the Neo-Hookean solution, the first striking remark is that in the presence of strain hardening cavity inflation is not an unstable process anymore, and any pressure will result in a finite stretch.

Figure 3.49: Equibiaxial expansion of the cavity inside the infinite solid

Using this expression we can evaluate by integration the work of expansion of the cavity in an infinite Arruda-Boyce solid. The results are shown in fig. 3.50. As an example, for the expansion of $\lambda = 20$ (from $r_1 = 0.5 \ \mu \text{m}$ to $r_2 = 10 \ \mu \text{m}$ as in the previous section, this gives an energy of 0.00068 μ J for the material parameters $\mu = 68$ kPa and locking stretch $\lambda_L = 18$.

Figure 3.50: Energy required for equi-biaxial expansion of the cavity inside the infinite solid

We should not forget that the polynomial approximation of the Arruda-Boyce model loses its validity close to the maximum extensibility. We need to express here the maximum extensibility in equi-biaxial extension as a function of the locking stretch (λ_L), which is defined for single chains. For that, we should use eqn. (3.50) [59] which relates the chain locking stretch to the average of principal stretches of the Arruda-Boyce solid cube:

$$\lambda_L = \frac{(\lambda_1^2 + \lambda_2^2 + \lambda_3^2)^{1/2}}{\sqrt{3}} \tag{3.50}$$

Since for equi-biaxial extension $\lambda_1 = \lambda_2 = \lambda$ and from the condition of the incompressibility $\lambda_3 = 1/\lambda^2$ (since $\lambda_1 \lambda_2 \lambda_3 = 1$), we can rewrite eqn. (3.50) as:

$$2\lambda^6 - 3\lambda_L^2 \lambda^4 + 1 = 0 \tag{3.51}$$

The pertinent numerical solution of eqn. (3.51) is reported in fig. 3.51 as a function of the locking stretch λ_L . For a locking stretch $\lambda_L = 18$, the maximum admissible equi-biaxial stretch is thus $\lambda_{BBL} = 22$.

Figure 3.51: Equibiaxial expansion bulk locking stretch

3.5.5 Cavity expansion using Yeoh model

We now apply the same methodology to derive the analytical expression for the pressure for a symmetrically expanding spherical cavity in an infinite and incompressible Yeoh solid:

$$P = \frac{1}{4} \left(4C_1 + 16C_2\lambda^2 + \frac{8C_2}{\lambda^4} - 24C_2 + \frac{48C_3}{\lambda^2} - 144C_3\lambda^2 + \frac{12C_3}{\lambda^8} - \frac{72C_3}{\lambda^4} + 108C_3 \right)$$

$$\frac{1}{4} \left(1 - \frac{1}{\lambda^4} \right) + 4C_1 \left(1 - \frac{1}{\lambda} \right) + 16C_2\lambda^2 \left(1 - \frac{1}{\lambda} \right) + 8C_2 \frac{\left(1 - \frac{1}{\lambda} \right)}{\lambda^4} - 24C_2 \left(1 - \frac{1}{\lambda} \right) + 48C_3\lambda^4 \left(1 - \frac{1}{\lambda} \right) + 48C_3 \frac{\left(1 - \frac{1}{\lambda} \right)}{\lambda^2} - 44C_3\lambda^2 \left(1 - \frac{1}{\lambda} \right) + 12C_3 \frac{\left(1 - \frac{1}{\lambda} \right)}{\lambda^8} - 72C_3 \frac{\left(1 - \frac{1}{\lambda} \right)}{\lambda^4} + 108C_3 \left(1 - \frac{1}{\lambda} \right)$$
(3.52)

This expression is plotted in fig. 3.52 for the two custom PSA tapes (6A and 6B) with the parameters shown in table 3.8 and for an initial cavity radius of 0.5 μ m. Once again the

strain hardening suppresses the inflation instability. As we can observe in the table 3.8 that strain hardening plays an essential role in determining the energy required to expand the cavity.

Figure 3.52: Equibiaxial expansion of the cavity inside the infinite solid: Yeoh Model. The fit provided by Hui et al. [5] is also reported for comparison.

Parameter for analytical calculations						
Name	C_1 (MPa)	C_2 (MPa)	C_3 (MPa)	Strain	Energy	
				Rate		
				(1/s)		
C.Y.Hui et al.	0.5	-0.024	0.0017	NA	$1.57 \times 10^{-4} \mu J \text{ (Till } \lambda = 4)$	
PSA tape 6A	0.034	0.0011	0.00002	2	$1.11 \times 10^{-5} \mu J \text{ (Till } \lambda = 4)$	
PSA tape 6B	0.034	0.0024	0.0012	2	4.15×10 ⁻⁵ μ J (Till λ =4)	

Table 3.8 :	Energy	for	cavity	expansion	using	Yeoh	Model
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3.6 Numerical simulation of cavitation in a confined layer

The fibrils are often generated starting from cavitation *within a thin layer*, *i.e.* within a system of finite thickness. Therefore, in this section, we present an axisymmetric numerical analysis of cavity expansion with a finite-size domain. We use the Arruda-Boyce and the Yeoh models and compare the results with the analytical solution derived in the previous section for an infinite medium.

In fig. 3.53a we present the initial geometry, and boundary conditions applied in ABAQUS for the numerical analysis of cavity expansion within a layer. In our simulations we fixed the following parameters: X=b=5 mm, Y=t=5 mm, $\mu_0 = 1$ MPa ($\mu = 0.845$ MPa), $\lambda_L = 5$ and D=0.001 MPa⁻¹ with the Arruda-Boyce model. We will vary the radius R of the unstrained cavity.

A major difference with respect to the spherical expansion considered in previous sections is that the control parameter is now the vertical stretch λ_V applied to the layer, instead of the radial stretch of the cavity λ . Due to incompressibility of the material, we can connect the applied vertical stretch to the radial stretch of the expanding spherical

(a) Geometry and Boundary conditions

Figure 3.53: Finite element boundary conditions (a) and mesh (b) for the axisymmetric expansion of a cavity in a confined cylinder undergoing vertical expansion with no lateral contraction at the boundaries.

cavity by eqn. (3.53).

$$\lambda = \left[\left[\frac{3}{(4R^3)} (\lambda_V - 1) t^3 \right] + 1 \right]^{1/3}$$
(3.53)

The results for the pressure as a function of the radial stretch λ are shown in fig. 3.54. The pressure was obtained from a finite element adjacent to the surface of the initial cavity and the stretch was taken as the radial stretch of the expanding cavity. We remark that while in the small strain regime, the cavity expansion in the confined cylinder and infinite body are very similar, the pressure required for the expansion in the confined conditions becomes much larger under large strains. Confinement resists cavity expansion, in a manner somewhat similar to strain hardening. We can remark that smaller cavities remain close to the the infinite medium regime up to a larger stretch, since the confinement effect sets in later.

For larger initial cavities or very large applied stretches, the cavity expansion in the confined cylinder is ellipsoidal. This morphology is reminiscent of what is observed during the peeling of scotch tape as shown in fig. 3.55. In order to investigate the stress distribution in the stretched cavity, the von Mises stress (shear) and the pressure distributions are shown in fig. 3.56. We can remark that shear stresses are mostly concentrated in a very thin layer at the surface of the expanded cavity, as well as on the thin residual layers close to the two stiff adherends. On the other hand, pressure is rather localized on the lateral stretched walls (which are mechanically analog to fibrils), where it is rather homogeneous through the thickness of the layer.

In order to evaluate the energy for inflating the confined cavities in a manner that is pertinent for the comparison with the adherence energy, we will express the energy to apply a vertical stretch λ_V to the cylindrical strands and divide it by the base area of the cylinders. If we implement the material parameters given in table 3.6 and table 3.7 (fitted using Yeoh Model) for our custom PSA tape (6A and 6B) and evaluate the energy per unit area we obtain 2.38 J/m² for PSA tape 6A and 4 J/m² for PSA tape 6B when applying a vertical stretch of $\lambda_V = 3.3$ to the cylindrical strand. We remark that these values have the same order of magnitude as the energy to stretch the fibrils starting from a circular contact patch as in the first part of this chapter.

Figure 3.54: Comparison of the FEM simulation of the axisymmetric cavity expansion in a confined cylinder (X = b, Y = t, Z = R) with the analytical expression for the spherically symmetric cavity expansion in an infinite medium (independent of the cavity radius): $\mu_0 = 1$ MPa, D = 0.001 MPa⁻¹

Figure 3.55: Schematics of cavitation in peeling of PSA: ellipsoidal cavities [14]

Figure 3.56: FEM simulation of the axisymmetric cavity expansion in a confined cylinder (X = b = 5 mm, Y = t = 5 mm, R = 1 mm), Arruda-Boyce Model, $\mu_0 = 1$ MPa, D = 0.001 MPa⁻¹

3.7 Partial conclusions

We summarize here the conclusions or take-home messages from this chapter:

- Simulation confirms that the force to draw fibrils from a flat soft adhesive film is larger than the force required to uniaxially extend hypothetical independent strands of the adhesive, as considered in our previous modeling [3], by a factor of several units, between 3 and 7 depending on the aspect ratio of the fibrils (cf. section 3.3). This provides a sound rationale for the 'factor of 5' observed between the adherence measured in peeling experiments and the model based on uniaxial stretching of the PSA tapes.
- The simulation allows to monitor the evolution of the local contact angle formed by the strained adhesive "meniscus" near the perimeter of the contact patch with the substrate. The external contact angle was observed to stabilise to some finite value after an initial stage of increase in the initial small strain limit (i.e. from 90⁰ in the small strain limit) for an incompressible soft layer. Moreover, for a given stretch of the fibril, this angle increases as we increase the elastic modulus of the soft adhesive layer.
- Our hyperelastic simulation allows the definition of a fibril debonding criteria based on critical energy release rate (evaluated by the *J*-integral) that provides a valuable candidate to predict the debonding stretch of fibrils, which is the most important missing ingredient for a predictive modeling of the peeling energy. Based on this model we could predict that the debonding force for the custom PSA tapes (6A and 6B, adjusted by the Yeoh model) should increase with the patch diameter as a power law of exponent 1.6, while the debonding stretch should decrease with the patch diameter as a power law with negative exponent -0.2. Moreover, the model predicts that, for a given value of the interfacial fracture energy, more incompressible fibrils should debond at smaller stretches than relatively compressible fibrils.
- Thanks to our simulation, we could appreciate that if we allow for local sliding of the adhesive on the substrate, then the fibrils would be expected to debond for larger stretches, and thus to induce a larger adherence energy in peeling. However, these estimates are based on an elastic behavior and a constant value of the interface energy Γ_0 , while for real PSA adhesives the sliding condition is expected to reduce the effective interfacial energy and to be strongly affected by viscoelasticity. In other words, if we release confinement, mode II (shear) decreases. This reduces the energy release rate, but it can also reduce the fracture energy (Γ_0). Therefore, only a reduction of the fracture energy (Γ_0) can explain the observed loss of adherence of PSA on PDMS substrates.
- Depending on the defects distribution in the adhesive and interface, the fibrillation process can be represented as either the drawing of fibrils from more sticky patches in the interface, of from the result of the inflation of cavities in the bulk of the adhesive. Our simulations show that in the large strain domain the two representations provide similar values of the energy to stretch an equivalent adhesive strand. Moreover, we could clarify that the strain hardening of the adhesive makes the cavity growth stable and provides an important contribution to the stretching energy.
- Our present simulations of the large strain fibrils are hyperelastic. We fitted the uniaxial extensional rheometer data of PSA tape 6A and 6B (for a given reference strain rate) to obtain the parameters for our simulations. The first step to better account for viscoelasticity will be to introduce a rate dependency in the elastic

modulus, which is supported by the nonlinear rheology measurements by Chopin et al. [3].

Chapter 4

Peeling of PSA tapes: Experiments and Numerical simulation

RÉSUMÉ

Dans le quatrième chapitre, nous présentons les expériences de pelage effectuées sur les substrats homogènes avec une bande adhésive de Scotch 3M600 et la caractérisation de la zone cohésive à l'aide d'une analyse d'image. La comparaison directe entre notre simulation numérique d'etirement d'une fibrille unique avec les expériences de pelage effectuées sur les substrats homogènes est une tache très ardue puisque nous ne contrôlons pas les dimensions des fibrilles et les distances inter-fibrillaires. Par conséquent, dans ce chapitre nous comparons nos simulations de fibrilles avec les données de pelage sur des substrats texturés présentant des motifs reguliers à géométrie variable. Cette comparaison sera aussi utile pour obtenir des éléments de modélisation de l'adhérence sur les substrats texturés. À la fin du chapitre, nous comparons les énergies d'étirement des fibrilles et d'expansion de cavité.

4.1 Introduction

The main aim of this chapter is to link the numerical simulations that we performed for the fibrillation and cavitation processes to the experimental investigations that have been performed on peeling by previous investigations at SIMM Laboratory.

Although the more accomplished experimental investigations of the debonding mechanisms through the instrumented peeling test have been performed on homogeneous substrates (Villey et al. [20, 2], Chopin et al. [3], Pandey et al. [83]), their direct comparison with simulations is very difficult due to the lack of control on the morphology of the spontaneously formed complex arrays of fibrils and cavities in the debonding region as shown in fig. 4.1.

Figure 4.1: Complex geometry of the debonding region during peeling of a PSA from a homogeneous substrate. Imaging from below the glass substrate.

This has motivated the recent development of a new strategy based on the realization of custom patterned substrates where an organized array of sticky patches (fig. 4.2) was found to trigger the morphology of the fibrillar debonding region as shown in fig. 4.3. These patterned substrates are produced by covering a glass slide with a nanometric layer of PDMS and then etching the layer on a square array of circular patches as in fig. 4.2, thus uncovering the glass substrate. While adhesion of acrylate based PSA is known to be very weak on PDMS, their adhesion on glass is very strong. By imaging the debonding region by Scanning Electron Microscopy (SEM) it was evidenced that in the first stages of debonding the interfacial crack front propagates faster on the PDMS regions, leaving behind some fibrils stuck on the glass patches, which are then progressively stretched to very large strains before eventually debonding. It is clear that this latest phase is physically very close to our simulation of fibril drawing from circular contact patches, where we control the patch radius a, the film thickness t and the interfibrillar distance 2b.

We can thus proceed to a quantitative comparison between our simulation and the dataset for peeling of the two custom PSA tapes (6A and 6B) over some patterned substrates with variable patch radius a, and variable interfibrillar distances 2b, which result in an area coverage ratio $\Phi = a^2/b^2$ of the glass phase. Since these datasets correspond to some still unpublished work from Morelle, Chopin, Bresson, Ciccotti and Barthel. We will report here some of these results with their permission.

Figure 4.2: Schematic of the patterned substrate geometry: sticky glass patches of radius a are arranged on a square lattice of step 2b and surrounded by low adhesion PDMS covered regions.

Figure 4.3: SEM view of adhesion rupture of a PSA tape, showing fibrillation on a patterned substrate (X. Morelle, B. Bresson, J. Chopin)

4.2 Peeling experiments on homogeneous substrates

Since checking a theory against experiments in a proper way is a very difficult task for a theoretician, I decided to spend a few months to practice myself on the instrumented peeling experiments and on the technique to invert the cohesive stresses in the debonding region developed by Villey et al. [2]. On the one hand, this allowed me to assess the reproducibility of these experiments. On the other hand, this was useful to gather some additional information about peeling on the target PDMS and glass substrates, while most of the published work regards peeling from the backing of the tapes.

Here we will compare our instrumented peeling experimental results with Villey et al. [2]. In this section, we will describe the general instrumented peeling setup. Peeling experiments are performed at room temperature $(23 \pm 2 \ ^{\circ}C)$ on a commercial PSA (3M Scotch®600). The substrate is made by applying scotch tape on the glass slide. Hence the backing of the scotch tape is the substrate itself as when peeling from a roller. The adhesive tape is peeled by an Instron testing machine (model 3343), which records the peeling force F while imposing a constant pull-out velocity $V \in [3: 3000] \ \mu ms^{-1}$. The substrate forms an angle $\theta = 90^{\circ}$ with respect to the pulling direction of the testing machine and is mounted on a translation stage which moves at the same velocity V, resulting in a steady-state peeling at constant angle.

The shape of the tape backing bending profile close to the debonding region (where the adhesive is deformed into fibrils before debonding) is monitored during the peeling experiments using a 1536 \times 2048 pixels camera equipped with a microscope objective (frame

rate between 5 and 30 fps, resolution 2.26 μ m/ px). For each frame of the movie, the outer profile of the tape backing is measured using a binarization algorithm that detects the interface between the dark background and the illuminated tape. The tape backing profile does not significantly change with time during steady peeling. We can thus average this profile over five frames.

Figure 4.4: 90° peeling setup.

The measured values of the adherence energy $\Gamma = F/b$ data are reported in fig. 4.5 as a function of the peeling velocity V. These data can be fitted by a power law with an exponent m = 0.16, which is close to the value of m = 0.20 reported by Villey et al. [2]. However, the average value of the adherence energy (on the common velocity range) is twice as that of Villey et al.'s data. By applying the algorithm reported in [2], we extracted the bending profile of the backing with the help of image analysis and then we fitted it with a model based on elastica theory for the bending of an inextensible beam, subjected to a constant cohesive stress in the direction of the fibrils within the debonding region. This allows to obtain the value of both the cohesive stress, the maximum fibril extension as shown in fig. 4.6

Figure 4.5: Adherence energy vs. peeling velocity for 3M600 tape peeled from its backing in 90° peel test

Once we checked the application of the technique on the same substrate as the developer, we now perform the same kind of measurements on the two homogeneous substrates that are used to form the patterned substrates, i.e. glass and PDMS. Concerning the peeling

Figure 4.6: Cohesive zone parameters for 3M600 tape peeled from its backing in 90° peel test

of tape 3M600 from PDMS substrates my personal adherence measurements are reported in fig. 4.7a along with some values reported by Amouroux et al. [84].

(a) PDMS substrate (b) Glass substrate

Figure 4.7: Adherence energy vs. peeling velocity for 3M600 tape peeled from different homogeneous substrates in 90° peel test.

From instrumented peeling measurements on the two custom tapes 6A and 6B over homogeneous substrates of glass and PDMS at a peeling velocity V = 0.1 mm/s, Morelle et al. characterized the dependency of the morphology of the debonding region (cf. Fig 4.8) by extracting the following geometrical parameters: 1) the length L_{dr} of the debonding region, 2) the maximum length of fibrils before debonding a_c , 3) the typical distance d_{int}

Figure 4.8: Side and bottom view of the debonding region in the peeling of custom tapes 6A and 6B from homogeneous substrates of PDMS and glass (Data from X. Morelle, B. Bresson, J. Chopin)

between fibrils or fingers that spontaneously form along the debonding front. The results are summarized in the following tables:

Adherence Energy			
PSA Name	PDMS	Glass	
Crystal 600	$7 J/m^2$	$117 \ J/m^2$	
6A	$41 \ J/m^2$	$180 \ J/m^2$	
6B	$21 \ J/m^2$	$105 \ J/m^2$	

Table 4.1: Adherence energy on homogeneous substrate. Data from Morelle et al.

PSA tape 6A				
PSA tape	PDMS	Glass		
6A				
a_c	$101 \ \mu m$	$130 \ \mu { m m}$		
L_{dr}	$85 \ \mu m$	$211~\mu{ m m}$		
d_{int}	$55 \ \mu m$	110 μm		

Table 4.2: Length scales of the debonding region in peeling of PSA tape 6A on the homogeneous substrate. Data from Morelle et al.

PSA tape 6B				
PSA tape	PDMS	Glass		
6B				
a_c	$37 \ \mu m$	$74 \ \mu m$		
L_{dr}	$74 \ \mu m$	$112~\mu{ m m}$		
d_{int}	$45 \ \mu m$	$72 \ \mu \mathrm{m}$		

Table 4.3: Length scales of the debonding region in peeling of PSA tape 6B on the homogeneous substrate. Data from Morelle et al.

4.3 Peeling experiments on patterned substrates and comparison with fibril simulations

As anticipated in the introduction to this chapter, the peeling experiments on patterned substrates represent the optimal choice for comparing our numerical simulations for several values of the sticky patch radius a and square mesh step 2b. Since our numerical simulation only represents axisymmetric fibrils, we need to focus on selected patterns which exhibit only steady-state single fibril extensions. In these conditions, which were found to occur only on small size patches, the adherence energy can be related to the energy to debond a single fibril W_{fibril} by:

$$\Gamma_{peel} = \frac{W_{fibril}}{b^2} \tag{4.1}$$

We will now proceed to the comparison of the adherence energy of the mono-fibril simulations and the peeling data on the patterned substrate. Different phenomena affect the peeling which we will not discuss here: we select the experimental runs that correspond to simple steady-state peeling with pure fibril stretch, which happen at lower patch density and small patch diameters of 100 and 200 μ m.

We report in fig. 4.9 the adherence measurements of the PSA tapes 6A and 6B on a series of patterned substrates where the area density Φ was modified by changing the distance 2b between sticky patches with constant radius $a = 50 \ \mu m$.

(a) PSA tape 6A (b) PSA tape 6B

Figure 4.9: Adherence energy Γ_{peel} : Patch diameter=100 μ m, t=20 μ m, Velocity=0.1 mm/s. Experimental data from Morelle et al.

To evaluate the energy dissipated during peel, we add the stretched fibril energy to the peel energy on the PDMS surface so that we have

$$b^2 \Gamma_{peel} = W_f + (b^2 - \pi a^2) \Gamma_{PDMS}$$

$$\tag{4.2}$$

$$\Gamma_{peel} = \Gamma_{PDMS} + \Phi\left(\frac{W_f}{\pi a^2} - \Gamma_{PDMS}\right) \tag{4.3}$$

or

Therefore, we apply a linear fit to the data Γ_{peel} in fig. 4.9 at small Φ . We find intercept values $\Gamma_{PDMS,6A} = 10.5$ and slope $S_{6A} = 66.1$. For 6B, we have $\Gamma_{PDMS,6B} = 3.80$ and slope $S_{6B} = 47.1$. From

$$W_f = \pi a^2 (S + \Gamma_{PDMS}) \tag{4.4}$$

with $a = 50 \ \mu m$ we obtain $W_{f,6A} = 0.60 \ \mu J$ and $W_{f,6B} = 0.40 \ \mu J$.

We now calculate the stretched fibril energy based on the results of chapter 3 and some experimental parameters. Using the maximum measured fibril length from tables 4.2 and 4.3, we can calculate the fibril debonding stretch $\lambda = (a_c - t)/t$ from the glass substrate (Here, t is the initial thickness of the adhesive layer), which is 6.5 in the case of PSA 6A and 4 in the case of PSA 6B for the peeling speed of 0.1 mm/s. We also need to calculate the correct strain rate from the experimental data. For that we use the length L_{dr} of the debonding region from tables 4.2 and 4.3. Using that data, we calculate the strain rate $\dot{\epsilon} = (V(\lambda - 1))/L_{dr}$ for the glass substrate, which is 2.61 s^{-1} for PSA 6A and 2.41 s^{-1} for PSA 6B. We can then calculate from our simulations the energy for stretching the fibril up to the measured debonding stretch, for the two PSA tapes 6A and 6B. To do the simulation, we use the material parameters given in table 3.6 and table 3.7 for PSA tape 6A and 6B respectively. We got these parameters by fitting the uniaxial extensional rheological data of PSA tape 6A and 6B as shown in fig. 3.12b and fig. 3.13b respectively at peeling speed of 0.1 mm/s and strain rate of 2 s^{-1} . Then, using those material parameters, we calculate the energy of the mono-fibril for both PSA tapes by doing simulations with the help of boundary conditions described for fibrils in chapter 3 of this thesis. We used the following geometrical parameters: $a=50 \ \mu\text{m}, t=20 \ \mu\text{m}$ and $b=222 \ \mu\text{m}, i.e. \ \Phi=5 \ \%$. We have the following values for stretch energy from these simulations: (1) PSA tape 6A: $W_{sf,6A}=1.31 \ \mu J$, (2) PSA tape 6B: $W_{sf,6B}=0.51 \ \mu J$. These values are reasonably consistent with the measured values, given the fact that the stretch at break has not been measured directly on the patterned substrate, but on the homogeneous substrate. Moreover, we obtain the adherence energy from these simulations by employing $\Gamma_{sf} = \frac{W_{sf}}{h^2}$, which is 8.5 J/m^2 for PSA 6A and 3.3 J/m^2 for PSA 6B.

4.4 Interplay between cavitation and fibrillation

In chapter 3 we argued that fibril drawing from contact patches and cavity growth are two different pathways to fibrillation. We will now evaluate each contribution to the debonding energy in peeling by using the results of our simulations.

We select the geometry where a/t = 2.5 and $\Phi = 5\%$ ($a=50 \ \mu m$, $t=20 \ \mu m$ and $b = 222 \ \mu m$) since we have also experimental data for it. We evaluated the energy per unit area for the mono-fibril simulation in this chapter for the two PSA tapes 6A and 6B as shown in fig. 4.9. They are represented by the fitted Yeoh model parameters in table 3.6 and table 3.7 respectively. We also have the confined cavity simulation in section 3.6 of chapter 3 for the same given material parameters. Hence, we compare the stretch energies for these two cases in table 4.4. The results show that for PSA tape 6B, the adherence energy given the mono-fibril and the mono-cavity simulations are similar but for PSA tape 6A, monofibril simulation gives 4 times more adherence energy than the mono-cavity simulations. This result tells us that even if initial configuration is different, we are getting the same adherence energy. Hence, initial configuration has little role to play in determining the total adherence energy.

Figure 4.10: Schematics of fibrils drown as an array of contact patches (top) and by an array of inflating cavities (bottom).

Adherence Energy				
PSA Name	Confined Cavity Simulation	Fibril simulation		
6A	2.4 J/m ² (till $\lambda = 3.3$ of strand)	8.5 J/m ² (till $\lambda = 6.5$)		
6B	4.0 J/m ² (till $\lambda = 3.3$ of strand)	$3.3 \text{ J}/m^2 \text{ (till } \lambda = 4)$		

Table 4.4: Adherence energy comparison: Fibrillation and Cavitation

4.5 Partial Conclusions

Following are the conclusions or take-home messages from this chapter:

- We have performed a few experiments to better understand previous results. For 90^0 peeling experiments using a new batch of scotch 3M600, we fitted our data in fig. 4.4 by the linear polynomial. We got the slope m=0.16 which is close to Villey et al.'s data of m=0.20 [2]. However, our average value of the Adhesion energy is twice that of Villey et al.'s data [2]. This observation highlights the variability inherent to adhesion measurements.
- Ideally, our simulation should answer why fibrils seen in peeling PSA tapes from the homogeneous substrate are not uniaxial [3]. However, on the homogeneous substrate, we do not control the parameters such as inter-fibrillar distance and radius of the fibril. Hence, we consider peeling data from patterned substrates to compare to our simulations, where the morphological parameters are controlled.
- When we compare the stretched fibril energy from experiments $(i.e., W_f)$ and simu-

lation $(i.e., W_{sf})$ for fibril density 5% and patch diameter 100 μ m, they are of same order of magnitude, given the fact that the stretch at break has not been measured directly on the patterned substrate, but on the homogeneous glass substrate.

• For all practical purposes, the initial stages of cavitation can be neglected compared to fibrillation because if we consider the large strain growth of an array of confined cavities up to the formation of a stretched foam, the energy density per unit area is similar to that of fibril expansion.

Chapter 5

General Discussion, Conclusions and Prospects
In this study, we investigated by numerical modeling the mechanisms of debonding of Pressure Sensitive Adhesives (PSA), such as large stretching of adhesive fibrils or the growth of defects into cavities. We represented the fibril as the hyperelastic deformation of a soft layer of thickness t in contact with a flat punch of radius a (Fig. 5.1a). When considering changes of a, this is mechanically analog to a crack propagating inward and leading to fibril debonding after some critical stretch. This new modeling is compared against the previous representation of the fibril as the uniaxial stretch of some independent strands of adhesive in order to provide the mechanical rationale for the prefactor observed in peeling experiments, which we called "*The Factor 5*". Moreover, we could provide a prediction of the maximum fibril stretch at debonding from the substrate, which should otherwise be measured in previous methods. In these concluding remarks, we first discuss the small strain response of the fibril using numerical methods and its future scope. Then we discuss the large strain behavior of the fibril and the cavity leading toward new paths for future studies. Finally, we compare our numerical insights with previous experiments and discuss the prospects of our modeling.

5.1 The small strain fibril calculations



Figure 5.1: Comparison of (a) Fibril FEM simulation and (b) FEM Pokerchip simulation

In chapter 2, we validated the linear limit of our numerical model for fibril stretching against some existing solutions, concerning both the elastic stiffness of the contact and the estimation of the stress intensity factor acting on the perimeter of the adhesive patch. For the limiting case of very small adhesive patches $(a \ll t)$, the FEM simulation was found to be in good agreement with the analytical contact model for a flat punch over a semi-infinite substrate. For large adhesive patches $(a \gg t)$, the FEM simulation was found to behave in a similar way to the pokerchip test (Fig. 5.1b). In order to assess the FEM simulation in the intermediate regime, we provided a numerical implementation of

the Boundary Element Method proposed by Perriot et al. [59]. We observed an overall good agreement, except when the material parameters approach to the incompressibility limit ($\nu = 0.5$), especially for elevated confinement ($a \gg t$), where the conditioning of the BEM method is questionable and should be improved in future developments.

The linear analysis allowed to appreciate that even for small strains, the contact stiffness of realistic models for elastic fibril drawing from a film is larger than the stiffness for homogeneous uniaxial extension by some factor between 3 and 6.

We have also investigated the stability of the crack propagation during fibril debonding. By using the Boundary Element Method for a compressible soft layer case (i.e., $\nu_1 = 0.4$), we could highlight a change from stability to instability when the patch radius decreased below a cross-over value $a_c = t/2$, which is half of the layer thickness. This is consistent with the stable nature of the crack front in the pokerchip configuration and the unstable character in the flat punch configuration on half-space.

5.2 The large strain simulations of fibril and cavity

Chapter 3 focuses on why pulling the fibril from an adhesive layer requires more energy than stretching it uniaxially, as well as fibril debonding criteria and cavitation. In this chapter, we performed the numerical simulation to assess the quantitative model of peeling presented by Chopin et al. [3]. In their work, they model the adherence energy in 90° peeling of custom-made PSA tapes by multiplying the uniaxial extensional rheological response with unified factor of 5. The results of our simulations show that the force for drawing a fibril by pulling a circular contact area of radius a from a soft film of thickness t is systematically larger by a factor ranging from 3 to 7 than the force required of the uniaxial extension of a cylindrical portion of the soft film with the same area. This provides a very robust confirmation that the necessity of drawing the fibrils in a confined condition is responsible for the large observed prefactor. However, numerical simulation allow to appreciate the dependency of such prefactor on geometrical parameters, such as the radius a, the thickness t and the interfibrillar distance 2b, as well as on the specific strain hardening behavior of the adhesive and the degree of compressibility.

Since our present numerical modeling is hyperelastic, we proposed to consistently model the fibril debonding by using the *J*-integral calculations provided by Abaqus (which we tested to be in agreement with energy difference calculations of the strain energy release rate *G* between two fibrils with slightly different area). This allows to derive a prediction of the maximum stretch λ_{deb} that the fibril can withstand before debonding. This requires the assumption that the interfacial fracture energy to let the perimeter of the adhesive patch propagate inward can be described by a constant (unknown) value Γ_0 . We can thus determine the force at debonding, as well as the total energy injected in the fibril before debonding, that is the required ingredient for feeding an improved version of the adherence model for peeling PSA.

Since we aimed at comparing these predictions with peeling data on the two custom PSA tapes (6A and 6B), we modeled their large strain response by Yeoh model that was shown to better adjust the extensional data (Hui et al. [5]). We found that under these assumptions the force at debonding increases with the fibril patch diameter as a power law of exponent 1.6, while the stretch at debonding decreases with the fibril patch diameter as a power law of negative exponent -0.17. We remark that the interfibrillar dis-

tance 2b had only a minor effect on the fibril debonding for the explored values of b > 1.5a.

We also investigated the consequences of allowing free sliding of the fibril on the substrate, in order to assess the adherence energy on the PDMS portions of the patterned substrates, where sliding is known to occur (Newby et al. [74]). Since sliding releases local confinement, we were not surprised to find that the local pressure and stress field are decreased and so does the total energy for stretching the fibril, as well as the strain energy release rate. In the framework of this simplified hyperelastic model, this would lead to the conclusion that the fibril should debond later than on a non-sliding substrate, which is opposite to experimental observations. However, the sliding condition is known to also decrease the interface fracture energy (Newby et al. [74]), so our present model is too crude to account for this phenomenon. Moreover, our simulation also allows to show that increasing the compressibility of the adhesive would also lead to the reduction of Gand thus to the increase of the stretch at debonding.

The main limitation of our simulation is that we could not obtain stretches larger than 4-5, which is still very large, yet not as large as the maximum stretch of 10 observed in experiments. In future development we could enhance the maximum reachable stretch by employing some suitable re-meshing scheme that would prevent strong mesh distortions in the regions close to the crack front. Moreover, improving the meshing strategy could allow us to get closer to the incompressibility limit ($\nu = 0.5$) while in the present simulations, we used the moderate Poisson ratio of 0.49 to avoid numerical difficulties. In future, we can implement Poisson ratio of 0.4999 in the simulation.

In order to study the fibril debonding in large strain, we decided to first perform a hyperelastic analysis. We can in future tentatively model the effect of viscoelasticity by considering the rate dependency of the extensional rheology data from Chopin et al. [3] and evaluating the predictions of the present model on the maximum force and stretch at debonding.

In this first simulation, we assumed that our stretched fibril is axisymmetric. However, from SEM imagining of the peeling experiments (Fig. 1.13), it is clear that these fibrils are not axisymmetric for large stretch, where they develop complex fringe patterns close to the crack front region. One of the prospects of this work is to perform a 3D simulation in order to allow for the description of these fringes and model their effect on the fibril debonding condition and thus on the adherence energy.

Since fibrils can also originate from the large stretch of small initial cavities, we devoted the end of chapter 3 to discuss the energy required for cavity expansion during the peeling process. We first explored the role of strain hardening on the initial growth of a small cavity, which can be modeled as embedded in an infinite medium submitted to a negative pressure. While current models describe the cavity expansion as unstable when the applied pressure overcomes some fraction of the elastic modulus, we could show by analytic calculation with both Arruda-Boyce and Yeoh material models that strain hardening suppresses such an instability. In order to consider later stages of the cavity expansion, we performed a simulation of cavity expansion in the middle of a cylinder of radius b and height t due to the vertical stretch of the cylinder. This allows to model the effect of the confinement by the two stiff adherends and the interaction with other cavities at distance 2b.

5.3 Patterned substrate peeling experiments and corresponding simulations

In chapter 4, we used patterned substrate peeling data of our custom PSAs (6A and 6B) to compare our fibril simulations. As we are controlling the parameters such as the radius a of the adhesive patches and the interfibrillar distance 2b in the patterned substrates, it became relatively easy to connect the total adherence energy of the peel data to that of the mono-fibril simulation.

We found that the adherence energy for stretching the simulated fibril up to the same debonding stretch observed in the experiments on homogeneous substrate is first order comparable to the measured peeling energy corresponding to that patch. For example, in the case of $2a = 100\mu m$ patch diameter on a $t = 20\mu$ m thick film with cell radius $b = 222\mu$ m, the simulated adherence energy is half of that derived from measurements (which is generally 5x larger than uniaxial estimates). However, our present simulations do not account for the initial stages of debonding from the weakly adherent portions of the substrate, that are covered with PDMS and where the adhesive is supposed to slide while debonding. Since the surface ratio of sticky patches for the considered pattern is $\Phi = 5\%$, we expect the debonding from PDMS to provide a contribution of the same order of magnitude, although the adherence energy on PDMS is less than 25% of the one on glass. In addition to that, our simulated stretch fibril energy (*i.e.*, W_{sf}) is of same order of magnitude as that of derived from the patterned substrate experimental data (*i.e.*, W_f). We can remark that the stretch at break has not been measured directly on the patterned substrate, but on the homogeneous glass substrate.

In chapter 3, we determined the energy required for stretching a basic strand of the confined adhesive containing a single cavity in the large strain by using the material parameters which we got from the extensional rheology data of our custom PSA (6A and 6B). In chapter 4, we compared it with the energy required to draw a mono-fibril from a circular contact patch on a soft film with the same material parameters. We concluded that while for all practical purposes, the initial stages of cavitation can be neglected in the total debonding energy, when considering the whole large stretch of the strand, the energy obtained by representing fibrils as the deformation of cavities or adhesive patches leads to comparable results, and the two approaches should be considered as complementary.

Future developments should be devoted to the complex task of considering the role of viscoelasticity in the simulations. This will affect both the global stretching of the adhesive strands and the mechanics of local debonding of the stretched adhesive from the adherend surface.

Appendix A

Derivation related to Arruda-Boyce model This derivation proves that there is a typo in the true stress expression given in the research paper by E.M.Arruda and M.C.Boyce [72].

The work done of the deformation in case of Arruda-Boyce Model is given by the following expression:

$$W = nk\Theta N \left[\frac{r_{chain}}{Nl} \beta + ln \frac{\beta}{sinh\beta} \right] - \Theta c' \tag{A.1}$$

Where, n is the chain density per unit volume, Θ is temperature, r_{chain} is the current chain length, N is the number of monomers in the chain, $\beta = \mathscr{L}^{-1} \left[\frac{r_{chain}}{Nl} \right]$ is inverse Langevin function.

$$\sigma_1 = \lambda_1 \frac{\partial W}{\partial \beta} \frac{\partial \beta}{\partial \lambda_1} \tag{A.2}$$

$$\frac{\partial W}{\partial \beta} = nk\Theta N \left[\frac{\partial \mathscr{L}(\beta)}{\partial \beta} \beta + \mathscr{L}(\beta) - coth\beta + \frac{1}{\beta} \right]$$
(A.3)

$$\mathscr{L}(\beta) = \operatorname{coth}\beta - \frac{1}{\beta} \tag{A.4}$$

$$\frac{\partial W}{\partial \beta} = nk\Theta N \left[\frac{\partial \mathscr{L}(\beta)}{\partial \beta} \beta \right]$$
(A.5)

$$\frac{\partial \mathscr{L}(\beta)}{\partial \beta} = \frac{\partial}{\partial \beta} \left(\frac{\beta}{3} - \frac{\beta^3}{45} + \dots \right)$$
(A.6)

$$\frac{\partial \mathscr{L}(\beta)}{\partial \beta} = \frac{1}{3} \tag{A.7}$$

$$\frac{\partial \beta}{\partial \lambda_1} = \frac{\partial}{\partial \lambda_1} \left(3 \frac{r_{chain}}{Nl} \right) \tag{A.8}$$

$$= \frac{\sqrt{3\lambda_1}}{\sqrt{N}\sqrt{I_1}} \tag{A.9}$$

$$= \frac{\lambda_1}{\sqrt{N}\lambda_{chain}} \tag{A.10}$$

$$\frac{\partial W}{\partial \beta} = nk\Theta N \left[\frac{\beta}{3}\right]$$
(A.11)

$$\sigma_1 = \lambda_1 \frac{\partial W}{\partial \beta} \frac{\partial \beta}{\partial \lambda_1} \tag{A.12}$$

$$\sigma_1 = \frac{nk\Theta}{3}\sqrt{N}\mathscr{L}^{-1}\left[\frac{r_{chain}}{Nl}\right]\frac{\lambda_1^2}{\lambda_{chain}}$$
(A.13)

Appendix B Boundary Element Method The Boundary Element Method for obtaining a stress/displacement relation for indentation of a coated solid by a rigid indenter like a flat punch is explained here. First, we will understand the general treatment for conical punch indenters and then try to develop a specific case of flat punch. For the following discussion, please refer to fig. 2.1.

$$\theta(s) = \frac{2}{\pi} \int_0^\infty g(r) \left(\int_0^\infty dk \, \mathcal{C}(kt) \cos(kr) \cos(ks) \right) dr \tag{B.1}$$

Let us now consider eqn. (B.1) for application to an indentation experiment. Let us not make any hypothesis on the shape of the indenter, apart from the fact that it is rigid, convex, axisymmetric and frictionless. For simplicity, we will consider the contact between the indenter and the coated material to be non-adhesive.

The boundary conditions of this problem are the following :

$$\begin{cases} \forall r \le a, \quad u_z(r) = \delta - p(r) \\ \forall r \ge a, \quad \sigma_z(r) = 0 \end{cases}$$
(B.2)

where p(r) the shape of the indenter and a the contact radius.

Note that this type of loading, because it only considers the normal displacement, does not model the indenter shape exactly, as has been shown by Hay et al. [65]. The minor corrections taking into account the radial displacement will not be considered here.

Introducing auxiliary fields:

However, the cosine transform of the equilibrium equation works marvels. We introduce the auxiliary fields g and θ defined as the cosine transforms of $\overline{\sigma}_z(r)$ and $k \overline{u}_z(r)$ respectively :

$$g(s) = \int_0^\infty dk \,\overline{\sigma}_z(k) \cos(ks) \tag{B.3}$$

$$\theta(s) = \int_0^\infty dk \, k \, \overline{u}_z(k) \cos(ks) \tag{B.4}$$

Expressing eqn. (B.3) and eqn. (B.4) in the real space, we obtain :

$$g(s) = \int_{s}^{\infty} dr \frac{r\sigma_{z}(r)}{\sqrt{r^{2} - s^{2}}}$$
(B.5)

$$\theta(s) = \frac{d}{ds} \int_0^s dr \frac{r u_z(r)}{\sqrt{s^2 - r^2}}$$
(B.6)

Combining eqn. (B.5) and eqn. (B.2), we have :

$$\forall r \ge a, g(r) = 0 \tag{B.7}$$

Eqn. (B.1) then becomes :

$$\theta(s) = \frac{2}{\pi} \int_0^a g(r) \left(\int_0^\infty dk \, \mathcal{C}(kt) \cos(kr) \cos(ks) \right) dr \tag{B.8}$$

while $\theta(r)$ is known on [0; a] through eqn. (B.6) and eqn. (B.2).

Then, under the contact, our indentation problem turns into an integral equation of the type $p(r) = \int_0^a f(s)K(r,s) \, ds$ where p and K are known.

where,

$$\mathcal{C}(kt) = \frac{2}{E_1^*} \frac{1 + 4b\,kt\,e^{-2kt} - ab\,e^{-4kt}}{1 - (a + b + 4b(kt)^2)e^{-2kt} + ab\,e^{-4kt}} \tag{B.9}$$

where,

$$a = \frac{\alpha \gamma_3 - \gamma_1}{1 + \alpha \gamma_3}, \ b = \frac{\alpha - 1}{\alpha + \gamma_1}, \ \alpha = \frac{E_1(1 + \nu_0)}{E_0(1 + \nu_1)}, \ \gamma_1 = 3 - 4\nu_1 \text{ and } \gamma_3 = 3 - 4\nu_0$$

As the method is similar whatever the indenter shape, we will only detail here the calculation in the case of the cone indentation, while the case of the flat punch and of the sphere will be developed.

One thing is noticeable from the expression of the C(kt) that as k goes to zero, E_0 goes to infinity and ν_1 goes to 0.5, C(kt) goes to zero, which is a bad sign for this type of calculations. At finite thickness t and large radius a, ie at the limit of small kt, we recover from eqn. B.9, the uniaxial strain compliance $C_{11} = \frac{(1+\nu_1)(1-2\nu_1)}{E_1(1-\nu_1)}$. It duly goes to zero when the material becomes incompressible. This is an effect of confinement.

Introducing,

$$\mathcal{Z}(x) \equiv \frac{\alpha E_1^*}{2} \mathcal{C}(x) - 1 \tag{B.10}$$

$$\Theta(\varsigma) = G(\varsigma) + \frac{2}{\pi} \int_0^1 G(\rho) \left(\int_0^\infty d\eta \, \mathcal{Z}(\eta\tau) \cos(\eta\rho) \cos(\eta\varsigma) \right) d\rho \tag{B.11}$$

from which Δ and G can be calculated and

$$\Pi = 4 \int_0^1 d\rho \, G(\rho) \tag{B.12}$$

Here the normalization parameter α is $\alpha_{fp} = 1$ for the flat punch, $\alpha_{cone} = 2$ for the cone and $\alpha_{sphere} = 8/3$ for the sphere. With

$$\epsilon = \frac{u(a)}{\frac{P}{S}} \tag{B.13}$$

we have for the cone:

$$\epsilon = 4 \frac{\Delta - \frac{2}{\pi}}{\frac{\Pi}{E_{eq}}} \tag{B.14}$$

and for the sphere:

$$\epsilon = 4 \frac{\Delta - \frac{1}{2}}{\frac{\Pi}{E_{eq}}} \tag{B.15}$$

B.1 Numerical Implementation of Boundary Element Method

Several model has been proposed to find the intrinsic material properties of the thin film from depth dependent equivalent modulus. Most of them are based on following eqn. B.16: [59]

$$E_{eq}^* = E_0^* + (E_1^* - E_0^*)\Phi(x)$$
(B.16)

where, E_0^* is the reduced modulus of substrate and E_1 being the reduced modulus of thin film layer. x is the ratio of contact radius a to the layer thickness t and Φ is the 'weight function' which equals 1 when x is zero and zero when x is infinite.

Normalized quantities in the case of flat punch:

In the normalised form, with

$$\tilde{r} = \frac{r}{t} \tag{B.17}$$

$$S = 2aE_1^* \mathcal{E}_{eq}(\tilde{a}, t, [E]) \tag{B.18}$$

$$g(r;a,t,[E]) = \frac{\delta_{fp} E_1^*}{2} \Gamma(\tilde{r};\tilde{a},t,[E])$$
(B.19)

where the normalized variables can be numerically calculated by the simple algorithm presented in [59]

Following [59] we introduce the following normalized quantities

$$\rho \equiv \frac{r}{a}; \ \tau \equiv \frac{t}{a}; \ \eta \equiv ka \tag{B.20}$$

Variable	cone	sphere	flat punch
Δ	$\frac{2}{\pi} \frac{\tan(\beta)}{a} \delta$	$\frac{R}{a^2}\delta$	$\delta/\delta = 1$
$\Theta(\rho)$	$\Delta - \rho$	$\Delta - \rho^2$	Δ
$G(\rho)$	$\frac{2}{\pi} \frac{\tan(\beta)}{aE_1^*} g(r)$	$\frac{3R}{4a^2 E_1^*}g(r)$	$rac{2}{\delta E_1^*}g(r)$
П	$\frac{2}{\pi} \frac{\tan(\beta)}{a^2 E_1^*} P$	$\frac{3\ddot{R}}{4a^3 E_1^*}P$	$P/\delta = 4\mathcal{E}_{\mathrm{eq}}^*$

Table B.1

B.2 Implementation of the model

Let us consider eqn. B.21 and discretize [0; 1] into N. From now on, we shall use $\varsigma_i = i/N$ and $\rho_j = j/N$. If we approximate the integral on ρ by a discrete sum, we then get :

$$\Delta - \varsigma_i = G(\varsigma_i) + \frac{1}{N\pi} G(0) M(\varsigma_i, 0, \tau) + \frac{1}{N\pi} G(1) M(\varsigma_i, 1, \tau) + \frac{2}{N\pi} \sum_{j=1..N-1} G(\rho_j) M(\varsigma_i, \rho_j, \tau)$$
(B.21)

with $M(\varsigma, \rho, \tau) = \int_0^\infty d\eta \, \mathcal{Z}(\eta \tau) \cos(\eta \rho) \cos(\eta \varsigma)$

From eqn. B.21 we have G(1) = 0. Then, for a given τ — that is to say for a given contact radius — we have to solve the $(N+1) \times (N+1)$ linear system introduced in eqn. B.21 for

the remaining N values of the G field and the normalized penetration Δ . The <u>M</u>-matrix elements can be calculated with a Fast Fourier Transform (FFT) algorithm for numerical efficiency. The accuracy of the numerical solution depends on the dimension N of the <u>M</u> matrix (which is associated to the discretization of [0; 1]), the cut-off B for the sampling range of the \mathcal{Z} function and the sampling rate $B/2^{V_T}$ for the FFT calculation of the matrix elements. Finally, the applied load II and equivalent modulus \mathcal{E}_{eq}^* are obtained through eqn. B.12 and table B.1 respectively.

B.3 Python Code

This code gives value of stiffness and stress intensity factor in the case of flat ended cylindrical punch indenting on the coated elastic half-space for the small strain limit.

```
# -*- coding: utf-8 -*-
.....
Created on 08/10/2020
Cauthor: krupal Patel & Etienne Barthel
.....
from math import pi,exp,floor
import numpy as np
import matplotlib.pyplot as plt
import scipy.linalg as linalg
from numpy import linalg as LA
from decimal import Decimal
#Indentation free parameter
# contact radius
a=1.1
#Layer thickness
t=0.35
#Tip parameters
#Radius for the sphere or the flat punch
R = 10
#tan(w) of the cone
tanw=2.8
#Bilayer real parameters
#Young's & Poisson's ratio of the half-space (Mpa)
E0=2950.81
       #2900
v0=0.4754
        #0.45
#Young's & Poisson's ratio of the layer (MPa)
E1=2.9999#2.999 #0.003
v1=0.4999#0.4999 # 0.5
#Reduced Normalized variables
tau=t/a
#numeric
#System size - typical 1000 - increase at large a/t
n=700
#FFT cut off B/2 and points number 2<sup>vT</sup>
B=1000# this is cVmax in the original routine
```

```
vT=22
#Tables
G=np.array([0,1])
ZF=[]
M=np.eye(n)
V=np.array([0,1])
X=np.array([0,1])
#Fonction Z du papier
def GreenNormal(kt):
   gamma1=3-4*v1
   gamma3=3-4*v0
# difference HERE
   alpha=E1*(1+v0)/(E0*(1+v1))
   A=(alpha*gamma3-gamma1)/(1+alpha*gamma3)
   B=(alpha-1)/(alpha+gamma1)
   denominator=1-(A+B+4*B*(kt**2))*exp(-2*kt)+A*B*exp(-4*kt)
   C=(1+4*B*kt*exp(-2*kt)-A*B*exp(-4*kt))/denominator
   return C-1
#A smooth periodic approx
def makeZforFFT():
   global vMax
   vMax=float(B)/tau
   1=2**vT
   table=[1.2]*1
   dx=float(vMax)/(l-1)
   for i in range(int(1/2)):
       x=i*(dx)
       val=GreenNormal(x*tau)
       table[i]=val
       table[-i-1]=val
    plt.plot([i*dx for i in range(l)], table)
#
   return table
#cosine transform
def makeTFC():
   global ZF
   Tinput=makeZforFFT()
   T_FFT=np.fft.rfft(Tinput)
   1=2**vT
#
   print 1./l*vMax
   ZF=[x/2*vMax/l for x in T_FFT.real]
   #np.savetxt('result.txt',ZF)
#generates the coeficients of linear system
def generateMatrixTerms():
   global M
   makeTFC()
```

bufferM=np.eye(n+1)

```
#
     plt.figure("oh")
#
    plt.plot([2*pi*y/vMax for y in range(len(ZF))],ZF)
    for i in range(n+1):
        for j in range(1,n):
            s=float(i)/n
            r=float(j)/n
            index_r_moins_s=abs(int((r-s)*vMax/(2*pi)))
            index_r_plus_s=abs(int((r+s)*vMax/(2*pi)))
        j=0
        s=float(i)/n
        r=float(j)/n
        index_r_moins_s=abs(int((r-s)*vMax/(2*pi)))
        index_r_plus_s=abs(int((r+s)*vMax/(2*pi)))
        bufferM[i,0]+=(ZF[index_r_moins_s]+ZF[index_r_plus_s])/(2*n*pi)
        #bufferM[i,0]/=1.
        if shape=='FLAT':
            j=n
            s=float(i)/n
            r=float(j)/n
            index_r_moins_s=abs(int((r-s)*vMax/(2*pi)))
            index_r_plus_s=abs(int((r+s)*vMax/(2*pi)))
            bufferM[i,n]+=(ZF[index_r_moins_s]+ZF[index_r_plus_s])/(2*n*pi)
            #bufferM[i,n]/=1
        else:
            bufferM[i,n]=-1.
    M=bufferM
#Right hand vector
def generateRightVector():
    global V
    Ar=[0]*(n+1)
    if shape=='CONIC':
        for k in range(n+1):
            Ar[k] = -float(k)/n
    elif shape=='SPHERE':
        for k in range(n+1):
            Ar[k] = -(float(k)/n) **2
    elif shape=='FLAT':
        for k in range(n+1):
            Ar[k]=1
    V=Ar
#Let's go!
def runFEBM():
    global X,G,DELTA,PI,delta,P,G_fp,S_fp
    print (shape,a,tau*a,R,tanw,E0,E1,v0,v1)
    CON=LA.cond(M)
    print(CON)
```

```
generateMatrixTerms()
    generateRightVector()
   # X=np.linalg.solve(M,V)
    LU=linalg.lu_factor(M)
    X=linalg.lu_solve(LU,V)
    G=np.array([0.0]*(n+1))
    for k in range(n):
        G[k] = X[k]
    PI=4./n*np.sum(G)
    G_rho=X[n]
    if shape == 'CONIC':
        delta=(pi/2)*(a/tanw)*G_rho # penetration
        P=(pi/4)*(a**2*(E1/(1-v1**2))/tanw)*PI # force
        e_eq=PI/(2*G_rho**2)
        E_eq=E1*e_eq/(1-v1**2)
        print (G_rho,PI,delta,P,E_eq)
    elif shape=='SPHERE':
        delta=(a**2/R)*G_rho # penetration
        P=0.5*(a**3*(E1/(1-v1**2))/R)*PI # force
        e_eq=(3./8.)*PI/(G_rho**(1.5))
        E_eq=E1*e_eq/(1-v1**2)
        print (G_rho,PI,delta,P,E_eq)
    elif shape=='FLAT':
        delta=0.001
        #G_fp=(G_rho*2*(1-v1**2))/(E1*delta)
        G_fp=(G_rho*E1*delta)/((1-v1**2)*(np.pi*a)**(1/2))
                                                               # Stress Intensity
        #G_fp=(delta*E1)/((1-v1**2)*(2*(2*np.pi*a)**(1/2)))
        S_fp=PI*E1/(1-v1**2)/2*a# stiffness
        e_eq=PI/4.
        E_eq=E1*e_eq/(1-v1**2)
        print (G_rho,delta,PI/4.0,G_fp,S_fp)
# shape='CONIC'
# runFEBM()
# shape='SPHERE'
# runFEBM()
shape='FLAT'
runFEBM()
```

Appendix C

Cavitation using Neo-Hookean Model

Here, we will derive the equation of pressure as a function of stretch for the spherically symmetric expanding cavity in the infinite Neo-Hooken solid. We will go through the details of the derivation of eqn. (3.33) using [82]. To obtain eqn. (3.33), we can first consider the expansion of a thick spherical shell as shown in fig. 3.47. Let us consider the curvilinear coordinate system θ_i in the strained spherical shell as the spherical coordinate system (\mathbf{R}, θ, ϕ), which has its origin at the center of the shell. We will denote y_i a cartesian reference frame. Hence,

$$\theta_1 = R, \theta_2 = \theta, \theta_3 = \phi \tag{C.1}$$

$$y_1 = Rsin\theta cos\phi, y_2 = Rsin\theta sin\phi, y_3 = Rcos\theta$$
(C.2)

The definition of the Metric tensor in the strained reference frame is as follows:

$$\boldsymbol{G_{ik}} = \frac{\partial y^r}{\partial \theta_i} \frac{\partial y^r}{\partial \theta_k} \tag{C.3}$$

$$\boldsymbol{G}^{\boldsymbol{i}\boldsymbol{k}} = \frac{\partial \theta^{\boldsymbol{i}}}{\partial y^r} \frac{\partial \theta^{\boldsymbol{k}}}{\partial y^r} \tag{C.4}$$

We can deduce the Metric tensor G^{ik} of the strained body with the help of eqn. (C.1),(C.2), (C.3) and (C.4)

$$\boldsymbol{G}^{i\boldsymbol{k}} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \frac{1}{R^2} & 0 \\ 0 & 0 & \frac{1}{R^2 sin^2 \theta} \end{pmatrix}$$
(C.5)

We assume that a thick spherical shell has spherical symmetry so that the point (\mathbf{R},θ,ϕ) was originally at (\mathbf{r},θ,ϕ) . Let's consider a thick spherical shell having an initial inner radius r_1 and outer radius r_2 . The values of the inner and outer radius R_1 and R_2 can be deduced directly from the incompressibility condition:

$$r^{3} - R^{3} = r_{1}^{3} - R_{1}^{3} = r_{2}^{3} - R_{2}^{3}$$
(C.6)

Define the deformation field,

$$Q(R) = \frac{r}{R} = \left[1 + \frac{(r_1^3 - R_1^3)}{R^3}\right]^{1/3}$$
(C.7)

Let's take the x_i -axes to coincide with the y_i -axes. Then, using eqn. (C.7), we can write

$$x_1 = RQ\sin\theta\cos\phi, \quad x_2 = RQ\sin\theta\sin\phi, \quad x_3 = RQ\cos\theta$$
 (C.8)

From eqn. (C.7), we can derive

$$\frac{dQ}{dR} = \frac{1}{R} \left[\frac{1}{Q^2} - Q \right] \tag{C.9}$$

The definition of the Metric tensor in the unstrained reference frame is as follows:

$$\boldsymbol{g_{ik}} = \frac{\partial x^r}{\partial \theta_i} \frac{\partial x^r}{\partial \theta_k} \tag{C.10}$$

$$\boldsymbol{g}^{\boldsymbol{i}\boldsymbol{k}} = \frac{\partial \theta^{\boldsymbol{i}}}{\partial x^r} \frac{\partial \theta^{\boldsymbol{k}}}{\partial x^r} \tag{C.11}$$

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Using eqn. (C.9), (C.8), (C.10) and (C.11), we have the following Metric tensor in the unstrained reference frame:

$$\boldsymbol{g}^{\boldsymbol{i}\boldsymbol{k}} = \begin{pmatrix} Q^4 & 0 & 0 \\ 0 & \frac{1}{R^2 Q^2} & 0 \\ 0 & 0 & \frac{1}{Q^2 R^2 sin^2 \theta} \end{pmatrix}$$
(C.12)

Constitutive relations are shown here in eqn.(C.13), where τ is stress tensor, p is pressure, and $\Phi = 2 \frac{dW}{dI_1}$. Here I_1 is the invariant of the symmetric second-order tensor or Green-Lagrangian strain tensor.

$$\tau^{ij} = \Phi g^{ij} + pG^{ij} \tag{C.13}$$

Using eqn.(C.13), (C.5) and (C.12) altogether we have the following stress components:

$$\tau^{11} = Q^4 \Phi + p \tag{C.14}$$

$$\tau^{22} = \frac{\Phi}{R^2 Q^2} + \frac{p}{R^2} \tag{C.15}$$

$$\tau^{33} = \frac{1}{\sin^2\theta} \left[\frac{\Phi}{R^2 Q^2} + \frac{p}{R^2} \right] \tag{C.16}$$

$$\tau^{23} = \tau^{12} = \tau^{31} = 0 \tag{C.17}$$

The equilibrium equation is as follows:

$$\tau_{,i}^{ik} + \Gamma_{ir}^i \tau^{rk} + \Gamma_{ir}^k \tau^{ir} = 0 \tag{C.18}$$

where the non-zero Christoffel symbols of the second kind are as follows:

$$\Gamma_{22}^{1} = -R, \Gamma_{33}^{1} = -Rsin^{2}\theta, \Gamma_{33}^{2} = -sin\theta cos\theta, \Gamma_{12}^{2} = \Gamma_{13}^{2} = \frac{1}{R}, \Gamma_{23}^{3} = \frac{cos\theta}{sin\theta}$$
(C.19)

The second and the third equations in eqn. (C.18) indicate that p is the only function of r only. Using eqn. (C.7), (C.19), (C.14), (C.15) and (C.17), the first equilibrium equation reduces to

$$\frac{dp}{dQ} + Q^4 \frac{d\Phi}{dQ} + 2(Q^3 - 1)\Phi = 0$$
 (C.20)

Integrating and using the rule of integration by parts we can obtain

$$p = -Q^4 \Phi + 2 \int^Q (Q^3 + 1) \Phi dQ = 0$$
 (C.21)

Using equilibrium equation as shown in eqn. (C.14) and considering normal pressure on the inner surface of the shell as a boundary condition, we have the following equation for the physical component of stress:

$$\sigma_{11} = K(R) - P \tag{C.22}$$

where,

$$K(R) = 2 \int_{Q_1}^{Q} \left[(1+Q^3)\Phi \right] dQ$$
 (C.23)

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Consider the case of the infinite outer radius of the shell. In that case, the body is an infinite solid containing a spherical cavity. At infinity, stresses will vanish. Hence, using eqn. (C.22) and noting that $Q(R) \rightarrow 1$ as $R \rightarrow \infty$, one can deduce the following relation from eqn. (C.22) and eqn. (C.7),

$$P = K(\infty)$$
 (C.24)
For Neo-Hookean material, $W = \frac{\mu}{2} (I_1 - 3)$ and $\Phi = 2\frac{dW}{dI_1}$
Let's denote $Q_1 = \frac{r_1}{R_1} = \lambda^{-1}$ and we know that $E = 2\mu(1 + \nu)$, where μ is the shear modulus μ is the Poiscov's ratio and E is the young modulus. Simplifying we get the

Let's deno modulus, ν is the Poison's ratio and E is the young modulus. Simplifying, we get the eqn. (3.33). Hence,

$$P = \int_{Q_1}^{Q} \left[4(1+Q^3) \frac{dW}{dI_1} \right] dQ$$
 (C.25)

$$= \int_{Q_1}^{Q} \left[4(1+Q^3)\frac{\mu}{2} \right] dQ$$
 (C.26)

$$= 2\mu \left[\left(\frac{Q^4}{4} + Q \right) \right]_{Q_1}^1 \tag{C.27}$$

$$\frac{P}{E} = \frac{(5 - 4\lambda^{-1} - \lambda^{-4})}{6}$$
(C.28)

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RÉSUMÉ

Dans les bandes PSA (Adhésifs sensibles à la pression), la rupture d'adhésion est accompagnée de cavitation et de fibrillation. Au niveau microscopique, les deux phénomènes impliquent une grande déformation. Comme le matériau de cet adhésif sensible à la pression est viscoélastique, cette grande déformation conduit à la dissipation de l'énergie. Cette dissipation d'énergie est de plusieurs ordres de grandeur supérieure au travail thermodynamique d'adhésion entre substrat et adhésif. Ces mécanismes de rupture d'adhérence par cavitation et fibrillation vont au-delà du cadre de la mécanique linéaire standard de rupture élastique. Par conséquent, nous utilisons des modèles de matériaux hyper-élastiques tels que Arruda-Boyce et Yeoh qui peuvent tenir compte de l'écrouissage et aussi de la nature viscoélastique de ces matériaux d'une manière élastique efficace en considérant la dépendance au taux de déformation. Ce type de mise en œuvre de modèles de matériaux dans la modélisation numérique de la cavitation et de la fibrillation conduit à aider à comprendre l'énergie de pelage total.

Dans cette étude, nous modélisons numériquement le mono-fibrille de différents rapports d'aspect en utilisant la méthode des éléments finis (FEM). La faible réponse de déformation des simulations FEM est comparée au contact plat du poinçon avec le demi-espace enduit. Pour implémenter le problème de contact de poinçon plat, nous utilisons la méthode Boundary Element (BEM). En dehors de cela, nous utilisons certaines méthodes analytiques simplifiées telles que le test Poker-chip pour la comparaison. La simulation confirme que la force require pour tirer les fibrilles d'un film adhésif plat est plus grande que la force require pour étendre uniaxialement des brins indépendants hypothétiques de l'adhésif. Nous étudions également le décollement du fibril pour nos bandes PSA sur mesure de la société 3M et donnons la loi de puissance pour la force et le déplacement au décollement du substrat de verre à une vitesse de peeling particulière. De plus, nous étudions l'effet du glissement et de la compressibilité sur le décollement fibrillaire. Nous vérifions également l'effet de l'écrouissage sur la cavitation dans le milieu infini par méthode analytique. Ensuite, nous montrons la simulation axisymétrique en expansion et la cavité confinée et comment elle se compare aux simulations du fibril.

Comme il est difficile de contrôler les paramètres tels que le rapport d'aspect et la distance interfibrillaire des fibrilles dans les expériences de peeling sur le substrat homogène, nous avons utilisé des données expérimentales de pelage de substrat modélisé pour comparer nos résultats de simulation. Cette comparaison est essentielle pour avoir un aperçu de la conception future du motif qui peut améliorer l'adhésion.

MOTS CLÉS

Caviation, Fibrillation, FEM, BEM, Rupture, Matiere Molle, décollement, PSA

ABSTRACT

In PSA (Pressure Sensitive Adhesives) tapes, adhesion rupture is accompanied by cavitation and fibrillation. At the microscopic level, both phenomena involve large deformation. As the material of this pressure-sensitive adhesive is visco-elastic, this large deformation leads to the dissipation of energy. This dissipation of energy is several orders of magnitude higher than the thermodynamic work of adhesion between substrate and adhesive. These mechanisms of adhesion rupture by cavitation and fibrillation go beyond the framework of the standard linear elastic fracture mechanics. Hence, we employ hyper-elastic material models such as Arruda-Boyce and Yeoh which can account for strain hardening and also for the viscoelastic nature of these materials in an effective elastic manner by considering strain rate dependency. This type of implementation of material models in numerical modeling of cavitation and fibrillation leads to help in understanding the total peeling energy.

In this study, we numerically model the mono-fibril of different aspect ratios using the finite-element method (FEM). The small strain response of FEM simulations is compared with the flat punch contact to coated half-space. To implement the flat punch contact problem, we use Boundary Element Method (BEM). Apart from that, we use some simplified analytical methods such as the Poker-chip test for comparison. Simulation confirms that the force to draw fibrils from a flat soft adhesive film is larger than the force to uni-axially extend hypothetical independent strands of the adhesive. We also study the fibril debonding for our custom-made PSA tapes made by 3M company and give the power law for the force and displacement at debonding from the glass substrate at a particular peeling velocity. In addition to that, we also study the effect of sliding and compressibility on fibril debonding. We also check the effect of strain hardening on cavitation in the infinite medium by analytical method. Then we demonstrate the axisymmetric simulation of expanding and confined cavity and how it compares with the fibril simulations.

As it is difficult to control the parameters such as aspect ratio and interfibrillar distance of fibrils in peeling experiments on the homogeneous substrate, we used experimental peeling data of patterned substrate for comparing our simulation results. This comparison is essential to gain insight into the future design of the patterning which can improve the adhesion.

KEYWORDS

Caviation, Fibrillation, FEM, Boundary Element Method, Fracture, Soft Matter, debonding, PSA