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Influence of the mutable kinetic parameters on the adhesion and debonding of thin viscoelastic films

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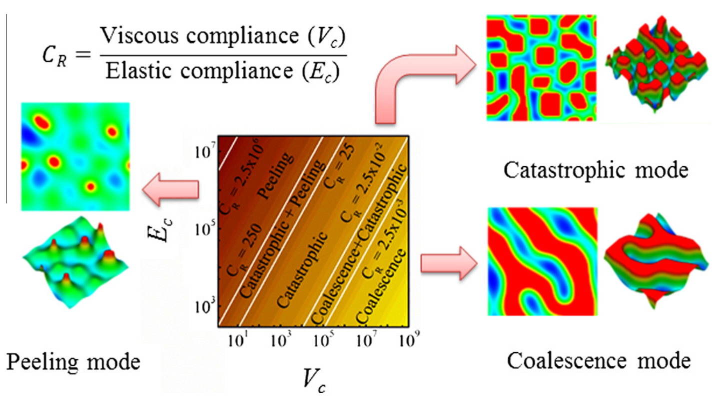
HIGHLIGHTS

- Kinetic factors play crucial role in the adhesion and debonding of viscoelastic adhesives.
- Position, velocity of contactor, and elastic/viscous compliances influence adhesion-debonding.
- The kinetic parameters overshadow the thermodynamically predicted adhesive properties.
- Methodologies for the nonlinear simulations are developed where the contactor is movable.
- Contact instabilities of viscoelastic films develop structures of higher aspect ratio than elastic films.

GRAPHICAL ABSTRACT



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ABSTRACT

Detachment of a surface from a viscoelastic layer, such as a film of glue, engenders bridges between the surfaces until separation. Such surface instabilities arising during contact and detachment of viscoelastic films with rigid contactors have been theoretically explored by linear stability analysis and nonlinear simulations. The contact instabilities of viscoelastic materials are found to manifest in either a 'critical' or a 'dominant' mode in which the former is preferred when the contactor is slowly brought near the film while the latter manifests when the film is 'hard-pressed' against it. The nonlinear analysis considers the movement of contactor during adhesion-debonding cycle, which uncovers that the kinetic parameters can overshadow the thermodynamically predicted area of contact, average force for pull-off, energy of contactor-film separation, and pathways of debonding. Three distinct pathways of debonding – peeling, catastrophic column collapse, and column coalescence, are found to manifest with the variation in the ratio of the elastic to viscous compliances of the viscoelastic film. The study also reveals that in the dominant mode of instability, a smaller length scale with a larger area contact between the contactor and film can develop patterns having aspect ratio ∼10 times larger than the same obtained from elastic film.

1. Introduction

Mesoscale patterns generated on the surface of an ultrathin soft polymeric film during the adhesion with an approaching contactor have been intensively studied in recent times. Periodic and

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miniaturized patterns have influence in improving the efficiency of many applications which include smart adhesives [1–13], lab-on-a-chip devices [14], membrane separation [4,15], MEMS or NEMS devices [16,17], solar or fuel cells [18,19], and micro or nanoelectronic devices [20]. A soft elastic or elastomeric film undergoing contact instability is also a model prototype to study friction, adhesion-debonding [3,21], crack propagation and arrest [2], confinement [1–3,22,23], fabricating structures with higher surface to volume ratio, and heterogeneous nucleation [24]. Thus, in the recent times, a flurry of research activities have been observed to uncover the necessary details of the contact instabilities of thin viscous, viscoelastic, and elastic films stimulated by the intermolecular forces [25–32], external electric field [33], and other adhesive forces [34].

Fundamentally, the surface patterns instigated by the contact instabilities play a crucial role in tuning the extent of adhesion and debonding of an adhesive material. For example, most of the artificial viscoelastic or gel adhesives are found to undergo 'wet' adhesion while attaching a pair of surfaces. Alternatively the natural elastic adhesives such as the pads of common gecko, water striders, or spiders have the capacity of 'dry' adhesion upon pressing followed by facile and 'clean' separation while releasing the pressure [12,35]. It is now well established that the bonding of surfaces with the help of viscoelastic adhesives is initiated by contact instabilities on the surface. This is followed by the capillary bridging of the micro-architectures before the dissipation of the excess energy at the late stage. In such a scenario, when the adhered surfaces are pulled apart, often a cohesive failure of the viscoelastic adhesive material leads to unclean separation of the adhering surfaces [13]. Recent studies indicate that, mimicking the natural adhesives, the 'dry' adhesion can be prepared. The reusability of the artificial adhesives can be improved by tuning the, (a) ratio of viscous to elastic compliance of the adhesive materials [36], (b) laying down sub-surface microfluidic networks [37], (c) decorating hierarchical mesoscale patterns on the surface of the adhesive films [12,13,38,39], or (d) employing multilayer of films [40,41]. The peel or tack test of the artificial adhesives also suggests that for the viscoelastic materials indeed the viscous or elastic [42] compliances, surface morphologies [6,43], and the kinetics of withdrawal and approach [5,7,8] play crucial roles on the pathways to adhesion, debonding, crack propagation and their seizure. Thus, at this stage, for the development of reusable adhesives it is important to systematically explore basic roles of, (i) kinetics of withdrawal and approach of a rigid body (e.g. contactor) towards the adhesive (e.g. viscoelastic film), and (ii) the elastic and viscous compliances of the adhesive materials on the adhesion and debonding of the viscoelastic materials with an approaching contactor.

In this context, previous studies have thoroughly explored the diverse thermodynamic aspects of the contact instabilities of thin elastic films. For example, it is now well established that the contact instabilities on the surface of elastic films can be engendered by the inter-surface adhesive interactions between the film and contactor [38,39,41,44–47]. More precisely, the contact instabilities have finite-wavenumber characteristics because the surface patterns originate only beyond a threshold value of destabilizing van der Waals force when it overcomes the restoring elastic stiffness of the film [27,29]. When the elastic film thickness (h) is more than a few micrometer, the length scale (λ) becomes independent of the inter-surface adhesive interaction and varies only with the film thickness, $\lambda \sim h$ [27,29]. In comparison, when the films are rather thin and elastically less compliant, the short-wave nature of the contact instability elastic films is found to shift towards the longer wavelength regime ($\lambda \sim \mathrm{n}h$, where $\mathrm{n} > 2.96$). This happens because of the increasing influence of the surface tension force for the thinner films [48]. The roles of patterned substrates or contactors [38,39,49], discontinuities on the elastic film [32,50], and multiple layers [40,41,47], on the length scales of the contact instabilities and ordering of these randomly distributed patters have also been explored in detail. In this direction, a very recent work on the contact instability of an adhesively stressed hydrogel film has shown patterns with a relatively longer wavelength ($\sim 7h$) because of the prominence of the elastocapillary effects [51].

Apart from the thermodynamic details, the kinetics of the spatiotemporal evolution of the contact instabilities on the surface of purely elastic films have also been studied with the help of simulations [52,53] and experiments [48]. These studies reveal that, during the adhesion cycle when a flat contactor approaches the free surface of a soft elastic film, the columnar patterns appear on the surface of the film beyond a critical separation distance (s_c). In this situation, the destabilizing van der Waals force marginally overcomes the restoring elastic stiffness of the film to engender the contact instability. If the contactor moves beyond s_c towards the film surface, the contact area between the contactor and the film surface increases because of the lateral bridging of the columns to form a labyrinthine patterns. On further approach, the bridging between the components of the labyrinths leads to an array of randomly placed cavities on the surface of the film. The transition from columnar to cavity patterns is also associated with a large increase in area of contact between the contactor and film surface. In this condition, if the contactor is pulled away from the contact proximity to initiate the debonding cycle, the cavities gradually transform into columnar structures before snapping off from the contactor. Importantly, although the adhesion and debonding can be performed repeatedly in a cyclic manner between a rigid contactor and an elastic film, they are not exactly similar and possess significant hysteresis in a cyclic loop [48,52,53]. In particular, during the debonding cycle the adhesive contact between the columns on the film surface and contactor extends the separation distance much beyond s_c before the columns snap off from the contactor surface. Previous studies have explored the roles of elastic compliance, step size of withdrawal, and external noise on the adhesion-debonding cycle and their hysteresis [53].

Importantly, most of the aforementioned studies employ purely elastic films to study the kinetics of the adhesion and debonding of the contact instabilities. However, most of the commercially available adhesives are viscoelastic materials and there is hardly any theoretical study, which describes the salient features of the contact instabilities of the thin viscoelastic films undergoing adhesion and debonding. In this article, we uncover the theoretical details of the adhesion and debonding of a thin viscoelastic film with the help of linear stability analysis and long-wave nonlinear simulations. We assume that the viscoelastic adhesive to be a zero-frequency linear viscoelastic material [54–57] with permanent storage and loss moduli. The constitutive relation used in the present study is simple and suitable to analyze the situations with small deformations to qualitatively uncover the onset of the different instability modes and their development towards cavities and fibrils while debonding. A brief linear stability analysis (LSA) is performed to identify the length scales associated with the instability. Following this, the contact instabilities of thin viscoelastic film are studied with the help of two-dimensional (2-D) and three-dimensional (3-D) long-wave nonlinear simulations. The framework for the nonlinear simulations considers the movement of the contactor, which helps in exploring the influence of the various kinetic parameters on the contact instabilities of thin viscoelastic films. In particular, we identify the roles of the kinetics of withdrawal or approach of the contactor and the elastic or viscous compliances of the film on the pathways of adhesion, debonding, and adhesion failure of the viscoelastic films. The results obtained for the viscoelastic film are also compared and contrasted with the ones available for the purely elastic film.