



Peeling behavior of a viscoelastic thin-film on a rigid substrate



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ABSTRACT

In order to study the adhesion mechanism of a viscoelastic thin-film on a substrate, peeling experiment of a viscoelastic polyvinylchloride (PVC) thin-film on a rigid substrate (glass) is carried out. The effects of peeling rate, peeling angle, film thickness, surface roughness and the interfacial adhesive on the peel-off force are considered. It is found that both the viscoelastic properties of the film and the interfacial adhesive contribute to the rate-dependent peel-off force. For a fixed peeling rate, the peel-off force decreases with the increasing peeling angle. Increasing film thickness or substrate roughness leads to an increase of the peel-off force. Viscoelastic energy release rate in the present experiment can be further predicted by adopting a recently published theoretical model. It is shown that the energy release rate increases with the increase of peeling rates or peeling angles. The results in the present paper should be helpful for understanding the adhesion mechanism of a viscoelastic thin-film.

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1. Introduction

In recent years, investigations on the physical mechanism of interface have been attracting considerable attentions because of the great significance not only for the widely applications of thin-films and coatings in engineering (Kim et al., 1989; Thouless and Jensen, 1992; Wei and Hutchinson, 1998) but also for deeper understanding of the extraordinary adhesion ability of biology, such as gecko (Peng and Chen, 2011; Peng et al., 2010; Pesika et al., 2007; Tian et al., 2006). The adhesion strength and adhesion energy are important properties for materials protecting, connecting and strengthening as well as designing of high-quality interfaces (Wei and Hutchinson, 1998). Peel-test, as a classical technique, is one of the efficient method for assessing the interface mechanical properties (Spies, 1953).

With regard to the problem of an elastic thin-film on a rigid substrate, the classical Kendall's model shows that the peel-off force not only depends on the adhesion energy but also on the elastic deformation of the film as well as the peeling angle (Kendall, 1975). As a pioneering work, it provides a direct method to find the interfacial properties, for example, adhesion energy, by measuring the peel-off force. While for a ductile thin-film, such as a metal film, the measured energy release rate is often much larger than the interfacial adhesion energy due to the plastic dissipation. Bending models were widely adopted to analyze the plastic dissipation (Kinloch et al., 1994; Thouless et al., 1997) following

the work of Kim and his coworkers (Kim et al., 1989; Kim and Aravas, 1988; Kim and Kim, 1988).

What is about a viscoelastic thin-film? It is well known that each material has a viscoelastic feature, which is weak in some materials, e.g. metals, but strong in some other ones, e.g. polymers and biomaterials. Adhesion mechanism of a viscoelastic material on a substrate has been widely investigated theoretically, numerically and experimentally (Andrews and Kinloch, 1973b; de Gennes, 1996; Derail et al., 1997, 1998; Gent and Schultz, 1972; Hui et al., 1992; Kaelble, 1964; Marin and Derail, 2006; Rahul Kumar et al., 2000; Xu et al., 1992), the corresponding force is often a function of the loading rate and temperature. Gent and Petrich (1969) studied the effects of peeling rate and temperature on adhesion of a viscoelastic thin layer on a rigid substrate by T-peeling experiment over a wide range of temperature and peeling rate, a single master relation was yielded in terms of the peeling rate when the temperature was reduced to a reference one by means of the Williams, Landel and Ferry's (WLF) rate-temperature equivalence. Derail et al. (1997, 1998) and Renvoise et al. (2007) experimentally studied the failure criterion of pressure sensitive adhesives at 90° peeling angle and obtained a transition rate from cohesive to interfacial failure. Chivers (2001) studied easy removal techniques of a medical pressure-sensitive adhesive tape for skin application, in which both physical and chemical approaches were introduced to achieve reversible adhesion of the medical adhesive tape. The effect of a flexible substrate on the peeling behavior of a medical pressure-sensitive adhesive tape was studied by Steven-Fountain et al. (2002), which was found to be different from the case of a rigid substrate. The viscoelastic effect of a polymeric film was analyzed

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by Loukis and Aravas (1991), in which the thin-film was modeled as a cantilever beam subjected to a purely bending moment. Based on the work (Loukis and Aravas, 1991), Chen et al. (2013) further investigated the viscoelastic peeling problem theoretically considering both the tensile and bending effects. Poulard et al. (2011) investigated the role of micro-patterning in adhesion properties of a soft deformable PDMS/acrylic adhesive interface, in which it was found that the adhesion energy could be successfully tuned by varying the pattern size.

Though many studies related to the peeling behavior of viscoelastic material exist, the peeling force with a 90° peeling angle was mainly focused on. The peeling mechanism of a viscoelastic thin-film from a substrate at an arbitrary peeling angle is unclear.

Peeling experiment of a viscoelastic thin-film on a rigid substrate is carried out firstly in the present paper. The effects of peeling angle, peeling rate, film thickness, surface roughness as well as the interfacial adhesive on the peeling force or the interfacial adhesion strength are mainly considered. Then, the variation of the viscoelastic energy release rate in our experiment is further predicted with the help of a recently published steady-state peeling model for viscoelastic thin-films on rigid substrates.

2. Peeling experiment of a viscoelastic thin-film on a rigid substrate

2.1. Materials in experiment

3 M Vinyl Electrical Tape (3 M #1500) made of polyvinylchloride (PVC) is used as thin-film in the present experiment. The width w and thickness h of PVC film are 18 mm and 0.13 mm, respectively, with a length of 50 mm. In all the experiments, the interfacial adhesive is polyacrylic acid with a thickness h_1 that is much smaller than the thickness of the thin-film h . Glass slides with a smooth or a rough surface are used as substrates. All the substrates are cleaned two times with ethyl alcohol followed by two times cleaning of acetone, then rinsed with distilled water.

2.2. Peeling experiment

In order to achieve a nearly perfect adhesion interface between the thin-film and glass substrate and avoid air bubbles entrapped at

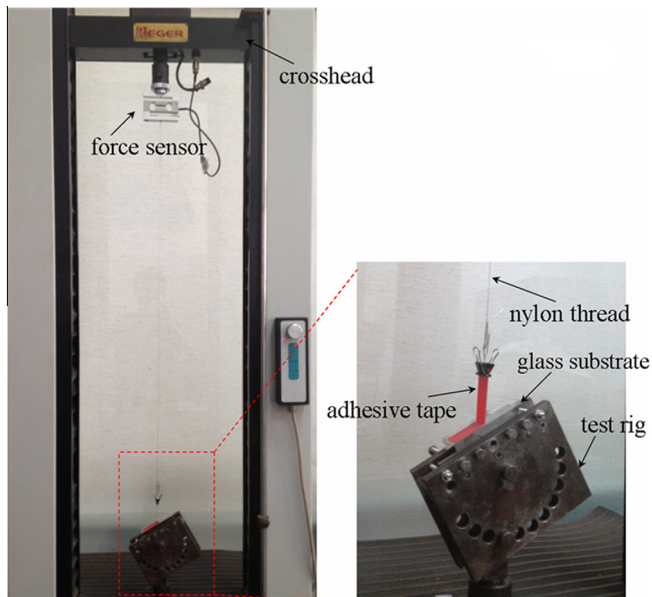


Fig. 1. A standard tensile machine and a rig made specially for tuning the peeling angles from 15° to 165° with an interval 15° .

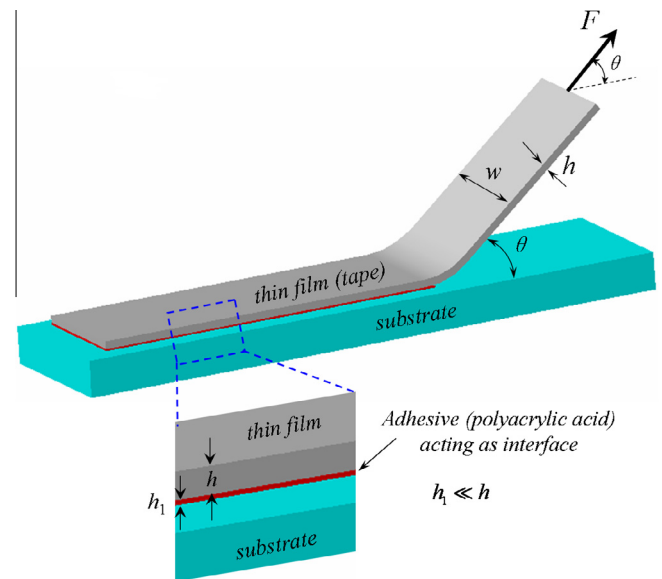


Fig. 2. Schematic of the peel-test with dimension illustrations of the adhesive tape (thin-film), interface (polyacrylic acid) and the substrate.

the interface, a hand roller is used to roll the thin-film on the glass surface five times in both directions. Then, the specimens are placed at a room temperature (about 25°C) for 4 h. All the experiments are conducted with a standard tensile machine as shown in Fig. 1, where a special peel-rig is made in order to vary the peeling angle conveniently. A three-dimensional schematic of the film/substrate system under a peeling load is shown in Fig. 2. The glass substrate is fixed to the rig, with the help of which the peeling angle can be tuned from 15° to 165° (the interval is 15°). To decrease the deviation of a determined peeling angle during peeling process, a thin nylon thread with one meter in length connects one end of the film to the force sensor installed on the crosshead of the tensile machine. Since the length of the thread is much longer than that of the thin-film (50 mm), the peeling angle will approximately keep a constant and the deviation of the peeling angle is about $\pm 1.4^\circ$ during peeling. The relationship between the peeling rate and the speed of the crosshead can be described as $v_{\text{ch}} = (1 - \cos \theta)v$ during steady-state peeling process, where θ is the peeling angle, v_{ch} is the velocity of the machine's crosshead and v is the peeling rate. A long focus Questar microscope is used to observe the images of the interface cohesive zone when the peeling behavior becomes steady-state. All the experiments are carried out at a room temperature 25°C with a relative humidity about 44%.

3. Experimental results and discussion

3.1. The effect of peeling rate on the peel-off force

Due to the viscoelastic properties of the PVC material, the peeling rate should show a significant effect on the peeling behavior. Typical curves of the peeling force vs. peeling distance are measured and exhibited in Fig. 3, where different peeling rates and different peeling angles are considered. Each curve in Fig. 3 shows three distinct regions: (i) the peeling force increases initially up to the onset of interface propagation; (ii) once the interface starts propagating, a slight drop is found in the peeling force; (iii) then it follows a steady-state peeling process and the peeling force approximately remains a constant, which is recorded conveniently and defined as the peel-off force in our experiment. Comparing the peel-off force in the steady-state process, it is reasonable to find

that it increases with the increase of peeling rates. Details of the relation between the peel-off force and the peeling rate can be found in Fig. 4 with three peeling angles 45° , 60° and 90° . Previous studies have shown that the peel-off force as a function of the peeling rate can be described by $F/w = C_0(1 + kv^n)$, where w is the film width, C_0 and k are functions of the peeling angle and related to the thickness of the viscoelastic thin-film, n is a constant related to the intrinsic property of the thin film (Benyahia et al., 1997; Du et al.,

2004; Marin and Derail, 2006; Zhou et al., 2011). In Fig. 4, one can see that our experimental results are well consistent with the above defined relationship. One should note that Xu et al. (1992) modeled a viscoelastic plate as a cantilever beam under purely bending and found that the interfacial toughness increases first and then decreases with the increase of crack propagating velocity. It means there may exist a maximum viscoelastic energy dissipation at an intermediate crack velocity (Xu et al., 1992). From Xu et al. (1992), we note that the change of the intermediate loading rate varies about six orders. The scope of loading rate is too large to verify the special phenomenon experimentally. Numerical study could simulate the theoretical results (Rahulkumar et al., 2000). In our experiment, the maximum loading rate of the tensile machine is 8.33 mm/s and the corresponding results are given in Fig. 5. In the region of peeling rate, the peel-off force increases monotonically with the increase of peeling rate. In fact, at a very large peeling rate, dynamic effects cannot be avoided and it is difficult to achieve a quasi-static loading condition. One can also find from Fig. 4 that, with a fixed loading rate, the peel-off force decreases with the increase of peeling angle, which is very similar to the case of an elastic film detaching from a rigid substrate (Kendall, 1975; Peng et al., 2010).

The images of the interfacial cohesive zone may give some insights on the effect of peeling rate on the peeling behavior, which can be observed and measured with the help of a long focus Que-star microscope. In-situ images of the cohesive zone are shown in Fig. 6, where three kinds of loading rates with peeling angles 60° and 90° are measured. It is found that the adhesive filamentation in the cohesive zone are stretched and the cohesive zone will not propagate until the active filamentation at the right end is broken. Furthermore, the length of the cohesive zone is found to increase with the increasing peeling rate. Actually, the interfacial filamentation is a typical phenomenon for pressure-sensitive interface adhesive (e.g. polyacrylic acid) using in peel-test (Aubrey and Ginosatis, 1981; Kaelble and Reylek, 1969; Niesiolowski and Aubrey, 1981). Niesiolowski and Aubrey (1981) had studied the influence of adhesive filamentation on the normal interfacial stress distribution by analyzing the curvature of the thin-film in the peel-zone. It was found that the stress distribution, and hence the peel-off force, could be affected by the filamentation. The region of stress (or interfacial filamentation) distribution increased with the increasing peeling rate, leading to an increase of the peel-off force, which agrees well with our experimental observation.

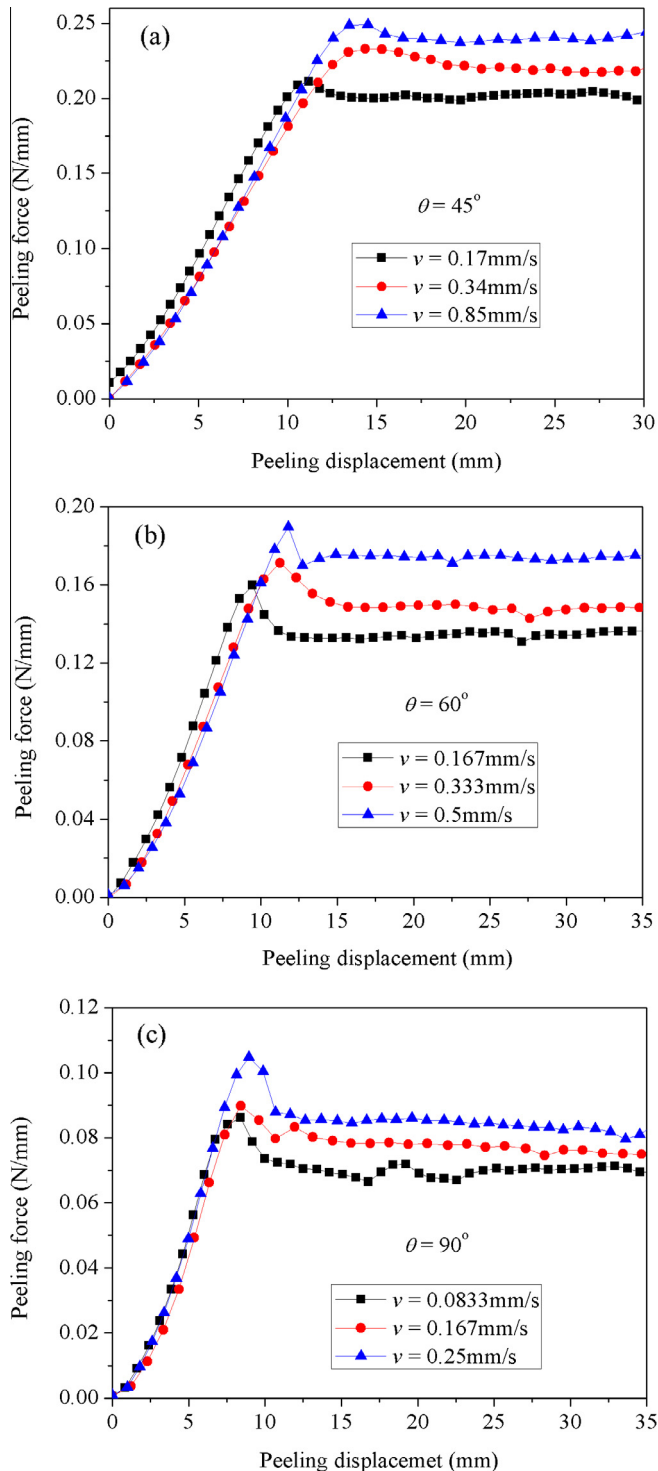


Fig. 3. The peeling force as a function of the peeling displacement with different peeling rates and different peeling angles in the experiment. (a) for the case of $\theta = 45^\circ$; (b) for the case of $\theta = 60^\circ$; (c) for the case of $\theta = 90^\circ$.

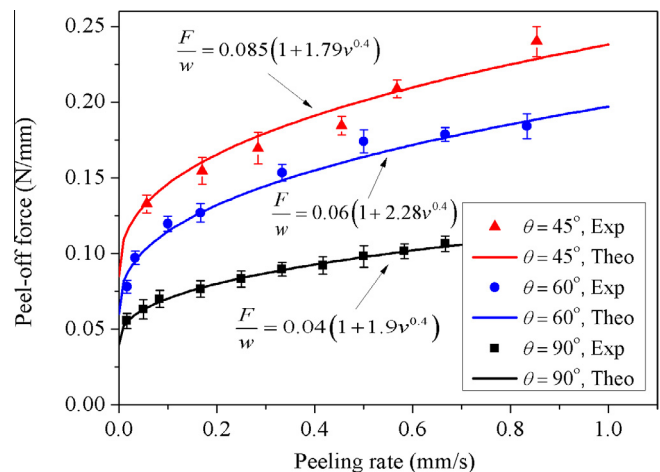


Fig. 4. The peel-off force as a function of the peeling rate with three different peeling angles 45° , 60° and 90° , respectively, in the experiment of a PVC film peeling from a glass substrate. Theoretical predictions are shown for comparison.

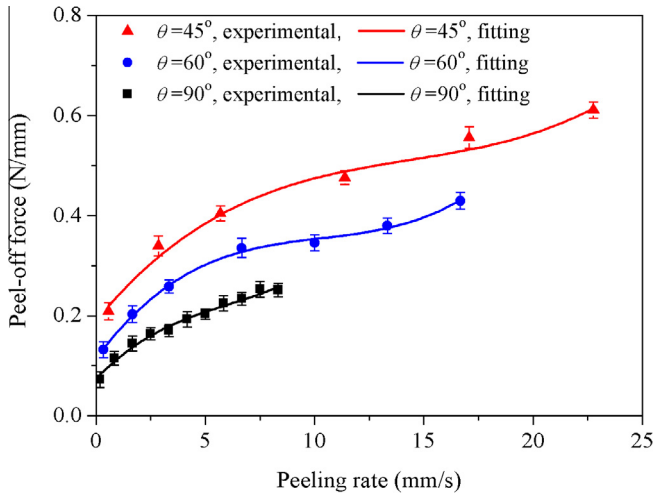


Fig. 5. The peel-off force increases monotonically with different peeling angles in the whole peeling rate region of our present peeling experiment.

3.2. The effect of peeling angle on the peel-off force

The classical Kendall's model gives the relation among peel-off force, peeling angle, elastic deformation and interfacial adhesion energy for an elastic thin-film peeling from a rigid substrate (Kendall, 1975), $P = Eh \left[\sqrt{(1 - \cos \theta)^2 + 2\Delta\gamma/Eh} - (1 - \cos \theta) \right]$, where P is the peel-off force per-unit width of the thin-film, $\Delta\gamma$ is the interfacial adhesion energy, θ is the peeling angle, E and h the Young's modulus and thickness of the film, respectively. Unlike the case of elastic thin-films, theoretical prediction of the peel-off force for a viscoelastic film detached from a rigid substrate is much more complicate due to the unavoidable energy dissipation in both the film and the interface near the crack tip. Experimental investigations might show some insights on the theoretical model. The

effect of peeling angle on the peel-off force of a PVC film adhering on a glass substrate is shown in Fig. 7, where the peeling rates are taken as 0.167 mm/s and 0.333 mm/s, respectively. The peel-off force decreases with the increase of peeling angles at a fixed peeling rate, which is similar to the case of an elastic thin-film. With a determined peeling angle, the peel-off force increases with the increase of peeling rates due to the effect of viscous dissipation.

3.3. The effect of film thickness on the peel-off force

The peel-off force of PVC tapes with different thickness detaching from the glass substrate is also measured. The influence of film thickness on the peel-off force is shown in Fig. 8 with a fixed loading rate 0.0833 mm/s and two different peeling angles 60° and 90°, respectively. It is found that the peel-off force increases with the increasing film thickness since the bending and axial stiffness of the thin-film increases with the increase of film thickness, and a great part of the applied force can be transferred to the interface away from the loading end (De Lorenzis and Zavarise, 2008; Peng et al., 2010; Yuan et al., 2007). Variations of the cohesive zone length with different film thickness are shown in Fig. 9. One can see that the length of the cohesive zone increases with the increase of film thickness, which is very similar to the phenomenon of an elastic thin-film (Peng et al., 2010).

3.4. The effect of surface roughness on the peel-off force

Natural surfaces, even highly polished ones, possess roughness in many different length scales, and surface roughness exhibit significant influences on the adhesion between a thin-film and a substrate (Peng et al., 2010; Persson and Gorb, 2003). In order to investigate the effect of surface roughness on the peeling behavior of a viscoelastic thin-film experimentally, we measure the peel-off force of the PVC film adhering on two different glass substrates. One is a relatively smooth surface with the surface RMS roughness about 1.4–1.6 nm measured by AFM and the other is a rough one with the surface RMS roughness about 3.5–6.2 μm measured by

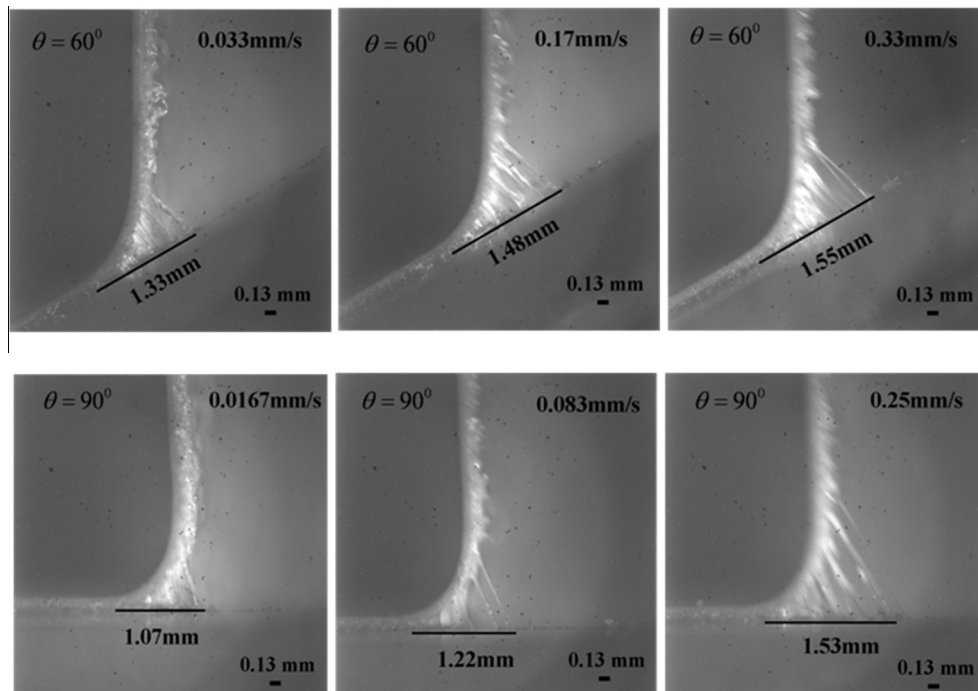


Fig. 6. Images of the cohesive zone with different peeling rates and two different peeling angles 60° and 90°, respectively.

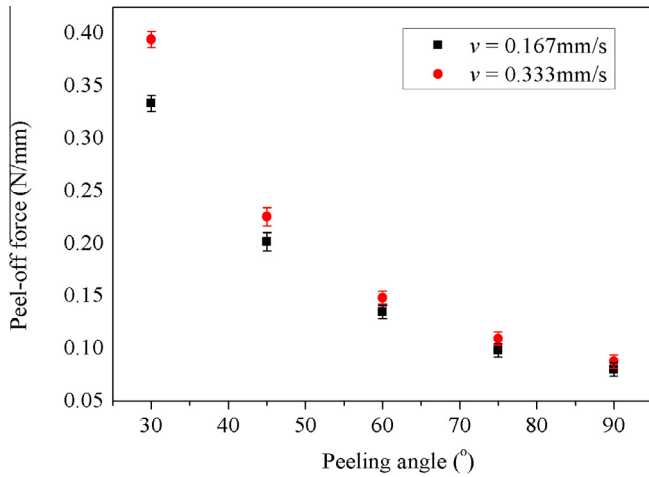


Fig. 7. The peel-off force varying as a function of the peeling angle with two different peeling rates 0.167 mm/s and 0.333 mm/s, respectively.

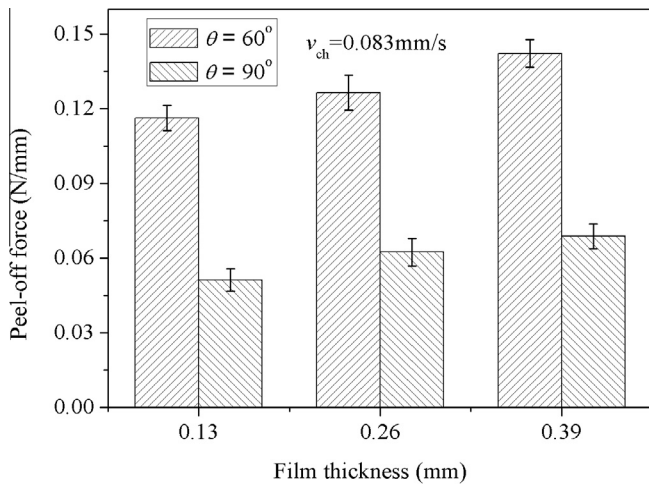


Fig. 8. The influence of film thickness on the peel-off force with a determined peeling rate and two different peeling angles 60° and 90°, respectively.

the contact and non-contact profilometer. The effects of surface roughness on the peel-off force with different peeling rates and peeling angles are indicated in Fig. 10. It is found that the peel-off force increases with the increase of peeling rate on both substrates, but the peel-off force of the film on the rough substrate is much larger than that on the relatively smooth one at the same peeling rate and peeling angle. The effect of surface roughness on the peel-off force can be explained by an effective interfacial adhesion energy $\Delta\gamma_{\text{eff}}$, where $\Delta\gamma_{\text{eff}}A_0 = \Delta\gamma A - U_{\text{el}}$, A_0 is defined as a

nominal contact area, A is the true atomic contact area, U_{el} is the elastic bending energy and $\Delta\gamma$ the adhesion energy of a smooth surface. If the increased adhesion energy $\Delta\gamma(A - A_0)$ induced by substrate roughness is much larger than the stored bending elastic energy U_{el} , we get $\Delta\gamma_{\text{eff}} > \Delta\gamma$, leading to an improved peel-off force on a rough surface in contrast to the case of a flat one (Kendall, 1975; Peng and Chen, 2011; Persson and Tosatti, 2001). Such a mechanism is also proved by the present experiments. However, the peel-off force sometimes will increase first and then decrease after achieving a maximum, which is actually owing to the competition between the stored bending elastic energy and the increased adhesion energy due to the substrate roughness (Martina et al., 2012; Palasantzas and De Hosson, 2003; Persson and Gorb, 2003; Persson and Tosatti, 2001). The problem of a soft viscoelastic adhesive on a periodic rough substrate has been successfully studied by Martina et al. (2012), in which the adhesion force is found to be a function of the surface topology and the viscoelastic properties of the material. Persson and Gorb (2003), Persson and Tosatti (2001) and Palasantzas and De Hosson (2003) also analyzed a thin-film adhering on a randomly rough surface, where the roughness was described by a self-affine fractal function.

4. Viscoelastic energy release rate

In the model of an elastic thin-film peeled from a substrate, the adhesion energy (or energy release rate) was shown to depend significantly on the mode-mixity angle (Evans et al., 1990; Hutchinson and Suo, 1992), which leads to a peeling angle dependent adhesion energy. While for a viscoelastic thin-film, our experiment shows that the deformation of the interfacial adhesives depends obviously not only on the peeling angle but also on the peeling rate (Fig. 6) during the steady-state peeling process. Therefore, besides the effect of peeling angles on the energy release rate, both the viscoelastic energy dissipation in the film and that at the interface near the crack tip should be considered. de Gennes (1996) developed a “viscoelastic trumpet” model in order to analyze the viscous energy dissipation in bulk viscoelastic materials. Xu et al. (1992) analyzed the energy release rate of a viscoelastic double-cantilever-beams model, in which the bending moment per unit beam width M is related to the curvature κ of the viscoelastic beam, i.e., $P(M) = IQ(\kappa)$, where P and Q are differential operators with respect to time t , I is the second moment of inertia of the beam’s cross-sectional area. Deflection of the viscoelastic beam can be successfully obtained considering a given stress distribution in the cohesive zone. The energy release rate in such a viscoelastic beam model equals the work done by the load subtracting the stored elastic bending energy.

Recently, Chen et al. (2013) proposed a theoretical model for a viscoelastic thin-film peeled from a rigid substrate from the energy

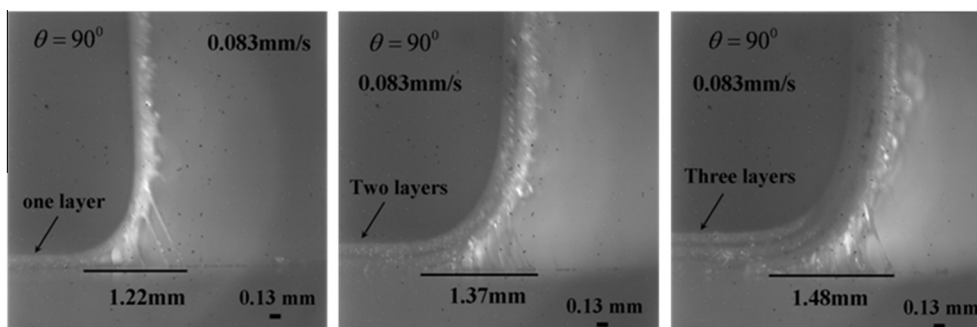


Fig. 9. Images of the cohesive zone in the peeling experiment of PVC films with different thickness, respectively, where the peeling angle is 90°.

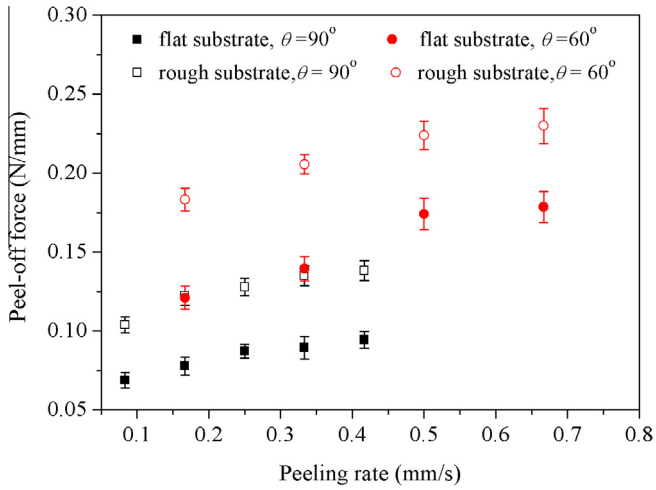


Fig. 10. The effect of surface roughness on the peel-off force with different peeling rates and peeling angles.

conservation point of view, which is more consistent with our experiment. The viscoelastic energy release rate during the steady-state peeling process was well achieved in a closed-form solution, which is very helpful for us to analyze the details of the energy release rate in our experiment.

The theoretical model in [Chen et al. \(2013\)](#) is shown in [Fig. 11](#), in which a virtually infinitesimal peeling length dl is assumed to be peeled off steadily. The work done by the peel-off force F per unit width is

$$dW_F = \frac{F}{w} (1 + \varepsilon_{peel} - \cos \theta) dl \quad (1)$$

where $\varepsilon_{peel} = F/(E_\infty wh)$ is the film strain at the loading end, E_∞ is the long-time relaxation modulus, θ the peeling angle, w the width of the film and h the thickness of the film, respectively.

Considering the whole released energy Gdl of the system and the increase of the strain energy dW_E in the film yields

$$dW_F = Gdl + dU_E \quad (2)$$

where G is the energy release rate.

For a steady-state peeling process, $dU_E = \frac{dl}{w} \int_{peel-arm} (Mdk + Td\varepsilon_m)$, where M , T are the bending moment and the axial force in any cross-section of the film, k , ε_m the curvature and tensile strain in the film, respectively. Detailed calculations of these parameters can be found in [Chen et al. \(2013\)](#).

Then, the energy release rate of a viscoelastic thin-film peeled from a rigid substrate is

$$G = \frac{F}{w} (1 + \varepsilon_{peel} - \cos \theta) - \frac{1}{w} \int_0^\infty \left(M \frac{dk}{ds} + T \frac{d\varepsilon_m}{ds} \right) ds \quad (3)$$

which is further written as

$$G = \frac{F}{w} (1 - \cos \theta) + \frac{1}{w} \int_0^\infty k \frac{dM}{ds} ds + \frac{1}{w} \int_0^\infty \varepsilon_m \frac{dT}{ds} ds \quad (4)$$

For a relatively small peeling strain, i.e., $F/E_0wh \ll 1$, the viscoelastic energy release rate can be approximately written as ([Chen et al., 2013](#))

$$G \approx \frac{F}{w} (1 - \cos \theta) \quad (5)$$

It is interesting to find that [Eq. \(5\)](#) has a similar form to the elastic-film case. However, one should note that the peel-off force F in [Eq. \(5\)](#) depends significantly on the peeling rate in a viscoelastic model.

According to the present experiment ([Fig. 4](#)) and that in the existing literatures ([Benyahia et al., 1997](#); [Du et al., 2004](#); [Marin](#)

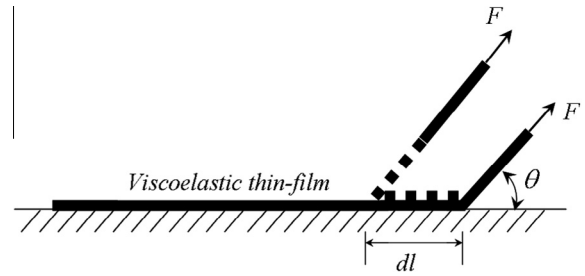


Fig. 11. Schematic of a viscoelastic thin-film peeled from a rigid substrate, where F is the peel-off force, θ is the peeling angle and dl is a virtually infinitesimal peel-off length.

and [Derail, 2006](#); [Zhou et al., 2011](#)), the peel-off force of a viscoelastic thin-film detached from a rigid substrate is a function of the peeling rate, which can be approximately written as

$$\frac{F}{w} = C_0(1 + kv^n) \quad (6)$$

Substituting [Eq. \(6\)](#) into [Eq. \(5\)](#) yields

$$G = C_0(1 - \cos \theta) \left[1 + \left(\frac{v}{k_0} \right)^n \right] \quad (7)$$

where $k_0 = 1/\sqrt[3]{k}$, C_0 and k are parameters related to the peeling angle and the thickness of the viscoelastic thin-film. n is a constant related to the intrinsic property of the thin film. Comparing to the usually adopted solution of the viscoelastic energy release rate, $G = G_0[1 + \phi(a_T v)]$, where G_0 is an intrinsic adhesion energy, ϕ denotes the energy dissipation in the system, v is the peeling rate, and a_T is the WLF (Williams, Landel, Ferry) shift factor ([Andrews and Kinloch, 1973a](#); [Feng et al., 2007](#); [Gent and Schultz, 1972](#); [Kinloch et al., 1994](#); [Rahulkumar et al., 2000](#)), [Eq. \(7\)](#) is a detailed expression with an assumption of a relatively small peeling strain.

[Fig. 12](#) shows the prediction of the viscoelastic energy release rate as a function of the peeling rate with different peeling angles, where $n = 0.4$ is derived from the present experiment. It clearly shows that the viscoelastic energy release rate depends significantly on the peeling rate, and increases with the increase of peeling rates at a given peeling angle. The results agree well with the existing experiments ([Andrews and Kinloch, 1973a](#); [Gent and Schultz, 1972](#); [Kinloch et al., 1994](#)). With a determined peeling rate, the viscoelastic energy release rate increases with

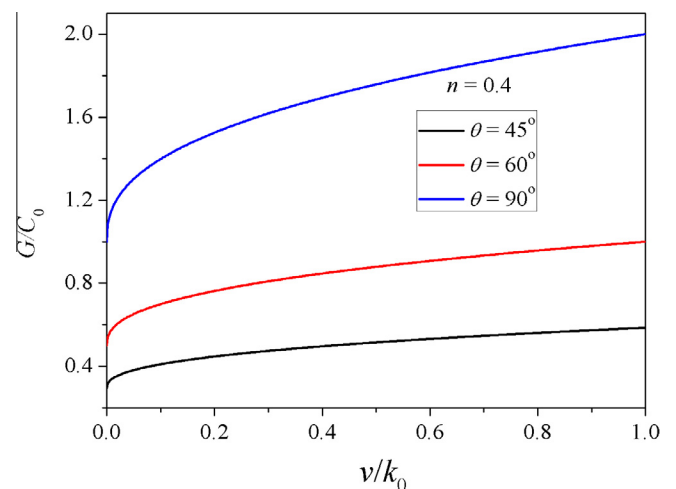


Fig. 12. The non-dimensional viscoelastic energy release rate as a function of the non-dimensional peeling rate for different peeling angles.

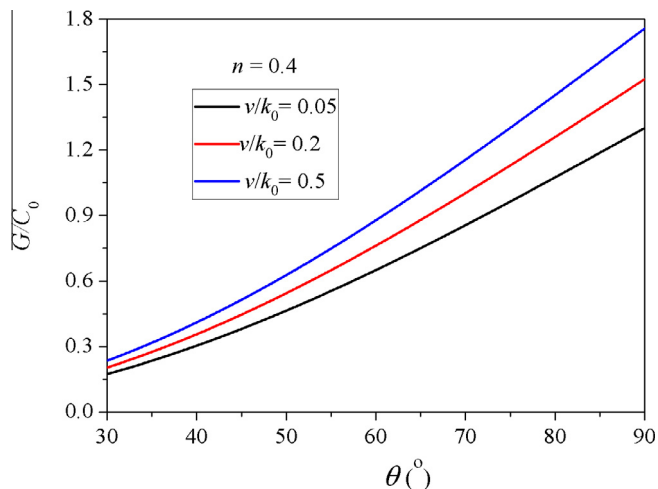


Fig. 13. The non-dimensional viscoelastic energy release rate varying with the peeling angle for different peeling rates.

the increase of peeling angles. The detailed relationship between the viscoelastic energy release rate and the peeling angle is shown in Fig. 13 for different peeling velocities. Here, one should note that the energy release rate in Figs. 12 and 13 includes not only the energy dissipation in the viscoelastic film but also that at the interface near the crack tip. How to distinguish one from another needs further parallel experiments.

5. Conclusions

Peeling behavior of a viscoelastic thin-film adhering on a rigid substrate with a viscoelastic interlayer are experimentally measured firstly in the present paper. It is found that the peeling rate, peeling angle, film thickness and surface roughness exhibit significant influences on the interfacial adhesion. For a fixed peeling rate, the peel-off force decreases with the increasing peeling angle. Increasing film thickness or substrate roughness leads to an increase of the peel-off force. Images of the interfacial filamentation and the length of the cohesive zone are observed and measured, which may partially give some explanations directly on the variation of peel-off forces. With the help of a theoretical analysis in Chen et al. (2013), the viscoelastic energy release rate is further achieved theoretically. It is shown that the energy release rate of the film/substrate system in our experiment increases nonlinearly with the increase of peeling rates and peeling angles. The results in the present paper should be helpful for further study on the peeling behavior of a viscoelastic thin-film with the aim to establish a more generally theoretical model in the future.

Acknowledgments

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