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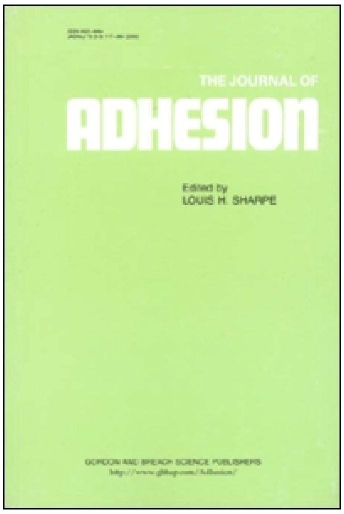

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MODELING THE PEEL PERFORMANCE OF PRESSURE-SENSITIVE ADHESIVES

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MODELING THE PEEL PERFORMANCE OF PRESSURE-SENSITIVE ADHESIVES

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This article presents an approach to analyzing the peel behavior of pressure-sensitive adhesives (PSAs) using the finite element method. The rheological properties and the peel strength of four natural-rubber–based PSAs were experimentally measured to provide input for and comparison with the finite element modeling. A criterion based on stored elastic energy density was used to describe the interfacial debonding. It was shown that the finite element predictions essentially captured the general features of the peel behavior of the PSAs. However, the peel forces predicted were lower than the experimental measurements at intermediate and high peel rates. This might be related to the fact that the nonlinear viscoelastic behavior of the PSAs at large deformation was not considered in this study.

Keywords: Pressure-sensitive adhesive; Peel; Interfacial debonding; Rheology; Viscoelasticity; Finite element modeling

INTRODUCTION

Pressure-sensitive adhesives (PSAs) are materials that can develop significant adhesion to a surface by the application of light pressure, but can be removed from the surface without leaving any residue. The unique properties of PSAs result in their use for applications ranging from medical tapes to adhesive labels. A standard method for characterizing the performance of PSAs is to measure their resistance against being peeled from a given substrate. The measured peel

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forces are frequently used to compare different PSAs and to evaluate the quality of adhesive bonds.

Many efforts have been devoted to understanding the phenomenological aspects of peel behavior [1–4] and to quantifying the peel performance [1, 5]. In general, three failure modes may be observed in a peel process [1, 2]. At high temperatures or low peel rates, the adhesive experiences primarily viscous deformation, allowing the failure strain to be reached. This leads to a cohesive failure in which the adhesive remains both on the substrate and on the backing. At intermediate peel rates or temperatures, the adhesive undergoes a transition from cohesive failure to interfacial debonding. This interfacial failure mode is associated with extensive viscoelastic deformation, where the adhesive stays either on the substrate or on the backing, with the latter being the more common case in practice. At low temperatures or high peel rates, an unstable failure mode may occur in which oscillatory force variations are observed. This type of stick-slip behavior is related to the glassy response of the adhesive. It is interesting to note that whatever the failure mode (cohesive or interfacial), the peel force *versus* peel rate behavior is shown to obey the time-temperature superposition principle, which also describes the bulk properties of the adhesive [3]. This indicates that the peel process is rheological in nature, and hence the peel force depends on the viscoelastic properties of the adhesive. In addition, it is observed that the peel performance in the interfacial failure domain also depends on the physico-chemical properties of the debonding interface, as the peel strength usually scales with the strength of the interfacial interaction [4].

Based on the experimental observations described above, it is generally accepted that the peel adhesion, Γ, in the interfacial failure regime can be evaluated by the following relation [1, 5]:

$$\Gamma = \Gamma_0[1 + \phi(\mathrm{a_T v})], \tag{1}$$

where Γ_0 is the energy required to separate the debonding interface. This quantity is usually assumed to be rate- and temperature-independent and is essentially the thermodynamic work of adhesion when only van der Waals interactions exist at the interface. ϕ characterizes the energy dissipation induced by the viscoelastic deformation of the adhesive. The rate- and temperature-dependence of the peel behavior is captured by this loss function through its dependence on the peel propagation velocity, v, reduced by the Williams-Landel-Ferry (WLF) [6] shift factor, $\mathrm{a_T}$, for the time-temperature superposition. When the energy consumption is confined within the adhesive layer or limited to the vicinity of the peel front, the loss function, ϕ, can usually be calculated based upon knowledge of the failure characteristics of the debonding interface and the viscoelastic properties of the adhesive [7, 8], or using an empirical power-law relation of approximately 0.6 [9]. However, a theoretical prediction for the loss function is usually unobtainable when the backing and/or the substrate undergo irreversible deformation on a large scale. Hence, numerical approaches, such as finite element analysis, are often used to meet the challenge.

Finite element methods are widely used in engineering analysis and product design. In particular, these numerical procedures are extremely well suited to the complex materials and geometries that are usually encountered in the analyses involving PSAs. Furthermore, it is desirable that the techniques for modeling the peel performance of PSAs using finite element approaches be established to assist the design and/or the formulation of PSA-related products and applications. Therefore, a finite element study on the peel behavior of typical PSAs is presented in this article. The rheological properties and the peel performance of the PSAs were experimentally measured to provide input and comparison for the finite element modeling. The analyses were focused on the interfacial debonding regime because this failure mode is practically most important. It was shown that the finite element calculations predicted the general shape of the peel force *versus* peel rate master curves. However, the peel forces predicted were generally lower than the experimental results. This might be associated with the fact that the nonlinear viscoelastic behavior of the PSAs at large deformation was not considered in this work.

EXPERIMENTS

The procedures for preparing the PSAs and for measuring their viscoelastic properties and peel strengths are described in detail in Yarusso [8], and they are briefly summarized here. The PSAs used in this study were prepared by blending two natural rubber bases with a tackifying resin and a small amount of antioxidant. PSAs A and B were formed using a rubber base with relatively low molecular weight, and PSA A has a lower tackifier content than PSA B. PSAs C and D were formed using a rubber base with relatively high molecular weight, and PSA C has a lower tackifier content than PSA D. By modifying the molecular weight and tackifier content, the bulk properties of the PSAs can be systematically varied, while essentially retaining the physico-chemical interaction at the interface on a given substrate.

The rheological properties of the PSAs were measured using a dynamic mechanical analyzer. The tests were performed under a series of frequency sweeps (0.1 to 100 rad/sec) at various temperatures (−40°C to 200°C). Master curves of dynamic moduli as a function of reduced frequency were constructed with 25°C as reference temperature. These curves were then used to extract the relaxation spectra of the PSAs (Figure 1). The effects of the molecular weight of the rubber bases can be seen by comparing PSAs A and B with PSAs C and D. The relaxation modulus decreased with increasing molecular weight. The effects of adding tackifying resin can also be seen by comparing PSA A with PSA B and PSA C with PSA D. The relaxation modulus was depressed and the glass transition was shifted to a lower frequency by adding higher amounts of tackifier.

The peel adhesion of the PSAs to a stainless steel substrate was measured using a universal testing machine. The tests were conducted in a 180° peel mode with crosshead speeds ranging from 1.27 mm/min to 1270 mm/min and testing temperatures ranging from −10°C to 200°C. In particular, the polyethylene terephthalate (PET) backing was specially treated so that debonding would not occur at the interface between the PSAs and the backing. The peel force *versus* peel

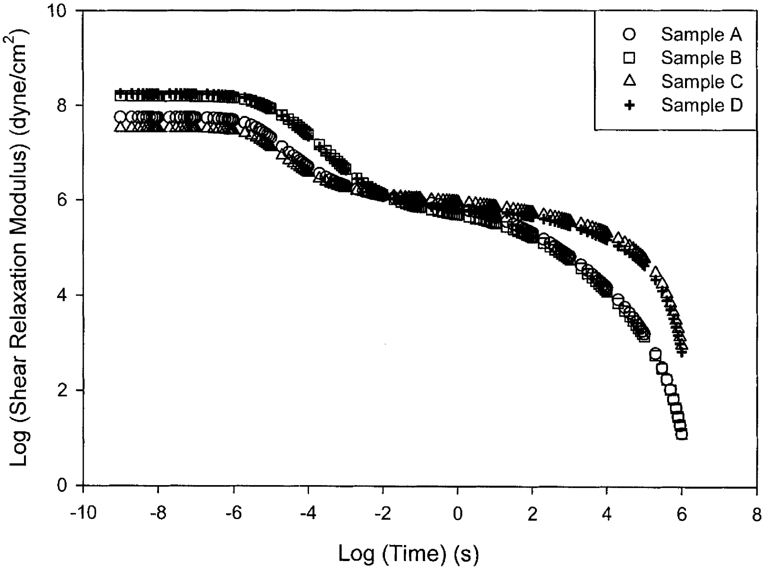


FIGURE 1 Shear relaxation moduli for the PSAs at reference temperature 25°C.

rate master curves were constructed using the WLF shift factors determined from the dynamic mechanical testing. Visual observation indicated that the PSAs underwent a transition from cohesive failure to interfacial debonding at moderate peel rates. Only the results for the interfacial failure region are reported.

FINITE ELEMENT MODELING

A two-dimensional (2D) large-strain finite element model of the 180° peel test was developed using the general-purpose finite element program ABAQUS^{TM} [10]. The instantaneous response of the PSAs was described using neo-Hookean hyperelasticity. The linear viscoelastic response of the PSAs was described in terms of shear relaxation moduli (Figure 1) represented by a generalized Maxwell series. The thickness of the PSAs was assumed to be 25 μm. Both the backing and the substrate were characterized as linear elastic solids. The Young's modulus and the Poisson's ratio of the backing were defined as 2.7×10^{10} dyne/cm^2 and 0.45, respectively. The backing thickness was assumed to be 0.3 mm. The Young's modulus and the Poisson's ratio of the substrate were defined as 1.95×10^{12} dyne/cm^2 and 0.29, respectively.

The finite element mesh was generated using fully-integrated 4-node quadrilaterals. The elements used for the PSAs employed a hybrid formulation to handle near-incompressibility, while the elements used for the backing incorporated incompatible mode shape functions to prevent locking behavior during bending. PSAs often cavitate during peel tests. The energy consumption associated with the cavitational processes of the PSAs is usually much smaller than the energy losses induced by the viscoelastic deformation of the PSAs. Hence, cavitation of PSAs was not included in the model. Displacement boundary conditions were applied on the left and the bottom edges of the substrate to restrain its motion in the horizontal and vertical directions, respectively. A beam element was attached at the left end of the backing as a loading mechanism to obtain the 180° peel angle. The beam element was first rotated by 180° in a sufficiently long period of time (to avoid introducing viscoelastic losses), and was then pulled at a given speed until a steady state was reached.

An energy-based failure criterion was used in the finite element simulations to characterize debonding at the interface between the PSA and the substrate. In this debonding criterion, interfacial failure is assumed to occur when the stored elastic energy density in the PSA reaches a critical value that is expected to depend on interfacial strength. Reasonably good agreement between the predictions based on this failure criterion and the experimental results has been observed [8, 11]. During the peeling process, mechanical energy is provided to stretch the PSAs. This energy is stored as strain energy through elastic deformation, dissipated as heat through viscous loss, or both. Some of the stored elastic energy may be released to drive the interfacial debonding. Therefore, it is expected that the onset of interfacial failure is somehow related to the level of stored elastic strain energy. However, it is noted that the dependence of the interfacial debonding on the stored elastic strain energy *density* is not well understood. The deformation of the PSAs was approximated as uniaxial extension in Yarusso [8]. However, the finite element model incorporated shear and area reduction effects that may result in a more rapid buildup of strain energy. Thus, the critical value of the stored elastic energy density used in this study was taken to be $10\,\mathrm{J/cm^3}$, which is higher than the value of $7\,\mathrm{J/cm^3}$ used in Yarusso [8]. The nodes at the interface between the PSA and the substrate were tied together using multipoint constraints (MPCs). During the analyses, user-defined subroutines were used to evaluate the elastic strain energy density of each PSA element and to compare this quantity with the critical energy density value of $10\,\mathrm{J/cm^3}$ to determine whether the MPCs associated with this PSA element should be released. Using this approach, interfacial debonding was successfully simulated.

RESULTS AND DISCUSSION

Typical finite element predictions of the deformed shape of a 180° peel test are shown in Figure 2. It is seen that the PSAs were significantly stretched in the vicinity of the peel front, which is immediately followed by a compressive region. This is consistent with the experimental observations and the theoretical predictions, such as those based on Beam on Elastic Foundation Theory [12]. In particular, the PSAs may attain a tensile strain of several hundred percent before they debond from the substrate. Such a large extent of viscoelastic deformation may dissipate a substantial amount of energy. This results in peel forces that may be several orders of magnitude higher than the thermodynamic work of adhesion (the value for the PSA/stainless steel interface encountered in this work is approximately $50\,\mathrm{erg/cm^2}$ [8].) Furthermore, it is noted that in contrast to the relatively rapid snap-back of the PSAs after debonding that is usually observed in peel testing, a considerable amount of residual deformation appears to exist in the PSAs even after the steady state is reached. The viscoelastic properties of the PSAs were approximated by a

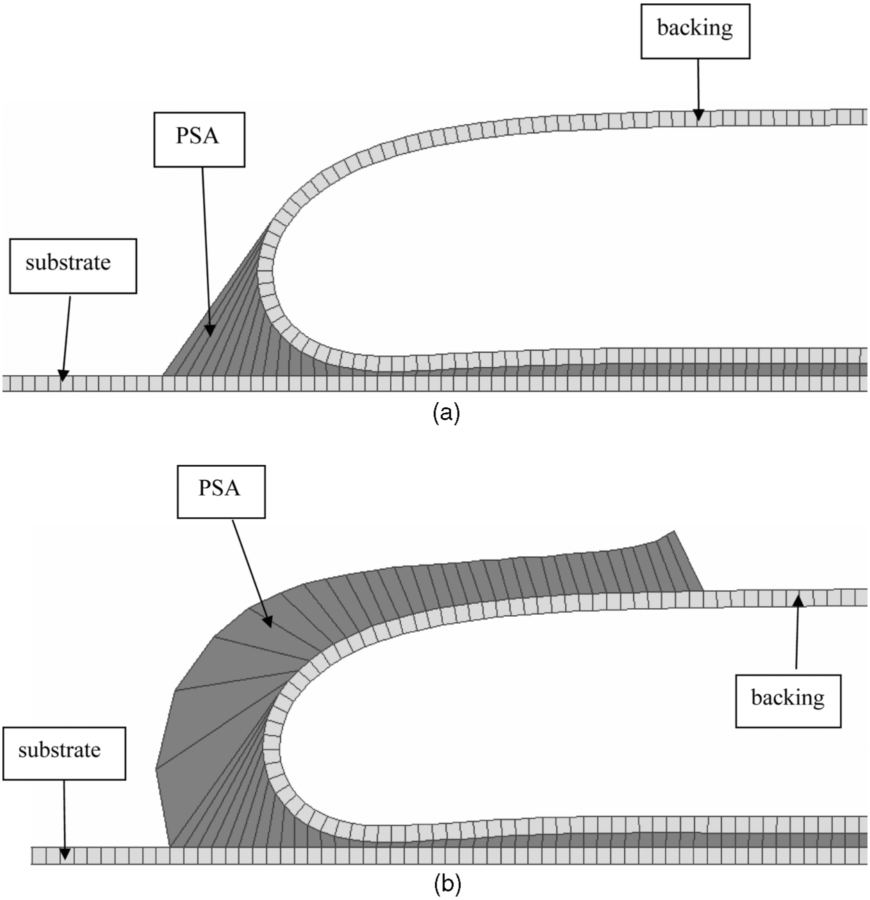


FIGURE 2 A typical finite element prediction of the deformed shape of a 180° peel test (a) before debonding and (b) at steady state.

generalized Maxwell model in this study. This model emphasizes the liquid-like behavior of a viscoelastic material because the parallel representation of the Maxwell elements results in nonrecoverable deformation in the viscous dashpots. Consequently, this may lead to the large residual elongation of the PSAs observed in the finite element calculations.

A typical peel force *versus* peel time curve for a 180° peel test is shown in Figure 3. The peel force increased with the peel time until a peak value was reached at initial debonding. The peel force then rapidly decreased to a relatively constant steady-state value that

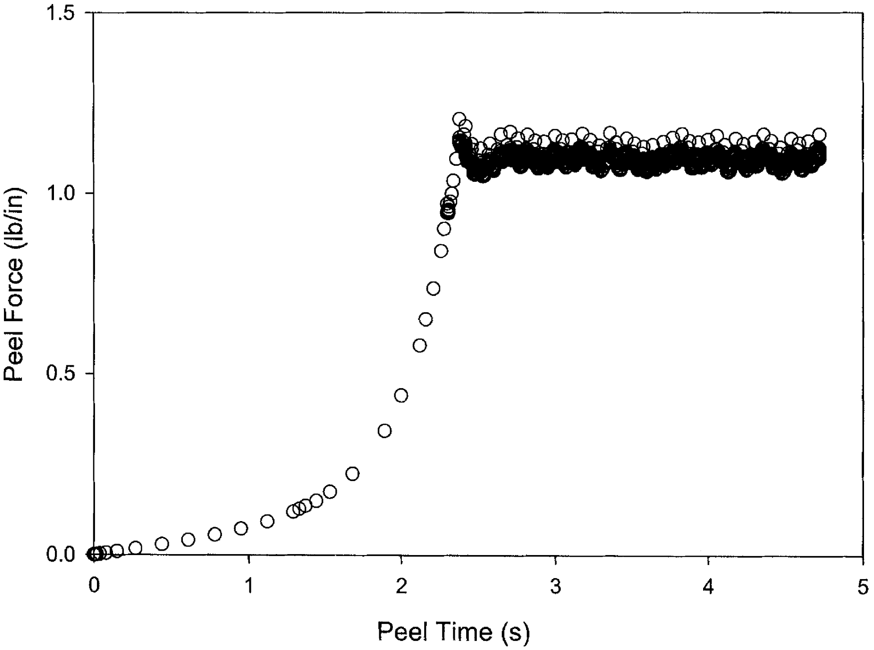


FIGURE 3 A typical finite element prediction of the peel force *versus* peel time curve for a 180° peel test.

was used to characterize each simulation. The variation of the peel force at steady state was due to the loading and unloading of the PSA elements during the progressive release of the MPCs at the PSA/substrate interface. These predictions are also consistent with those observed experimentally.

The peel force *versus* peel rate master curves for the PSAs obtained both from the experiments and from the finite element simulations are shown in Figure 4. It is seen that the finite element modeling predicted the functional form of the peel force *versus* peel rate master curves. The peel force increased with the peel rate until it reached a maximum at some intermediate peel rate, and then dropped as the peel rate further increased. At relatively low or high peel rates, the PSAs were in a relatively relaxed (viscous) or unrelaxed (elastic) state. Little dissipation was induced and, hence, relatively low peel forces were obtained. In contrast, at intermediate peel rates, a large amount of viscous dissipation occurred, and hence relatively high peel forces were observed. However, it is noted that the predicted peel forces were noticeably lower than the experimental

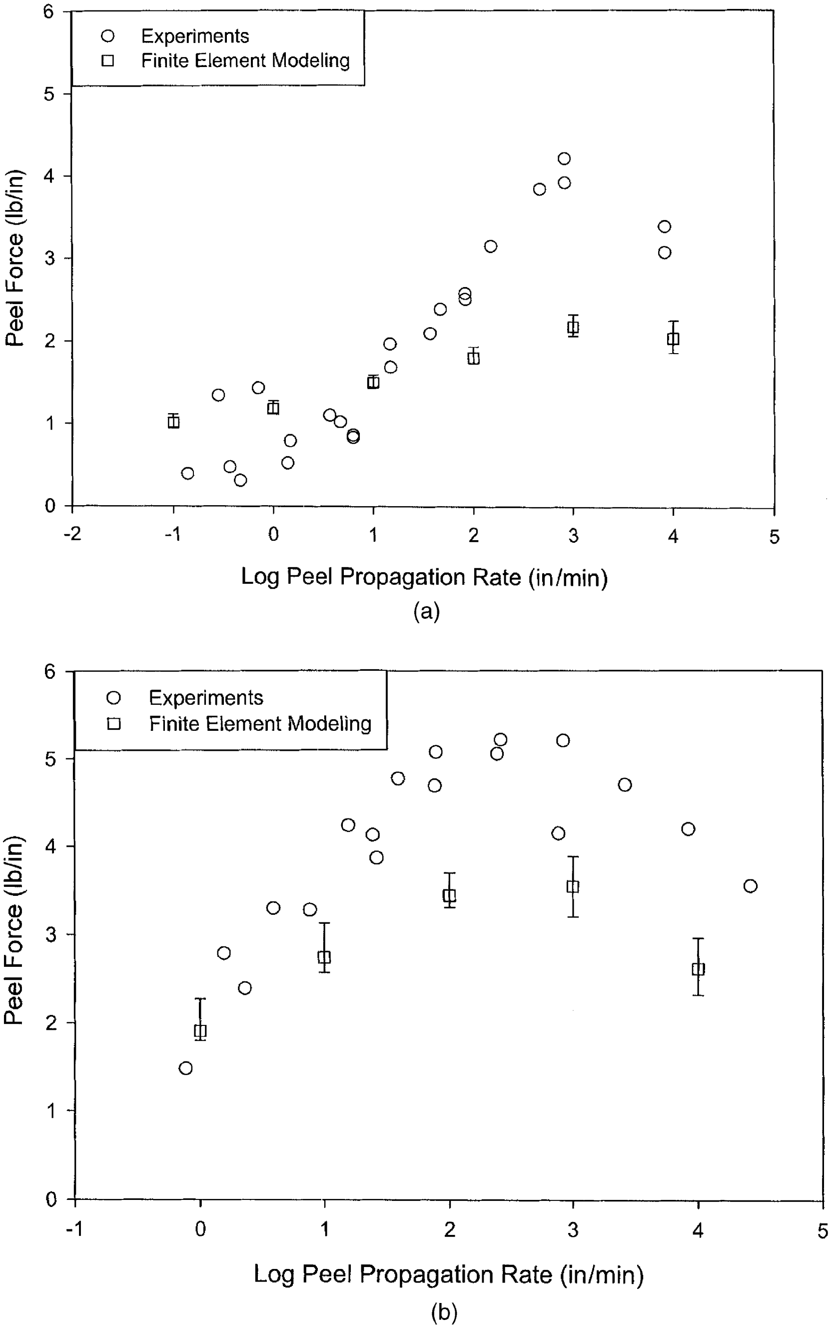
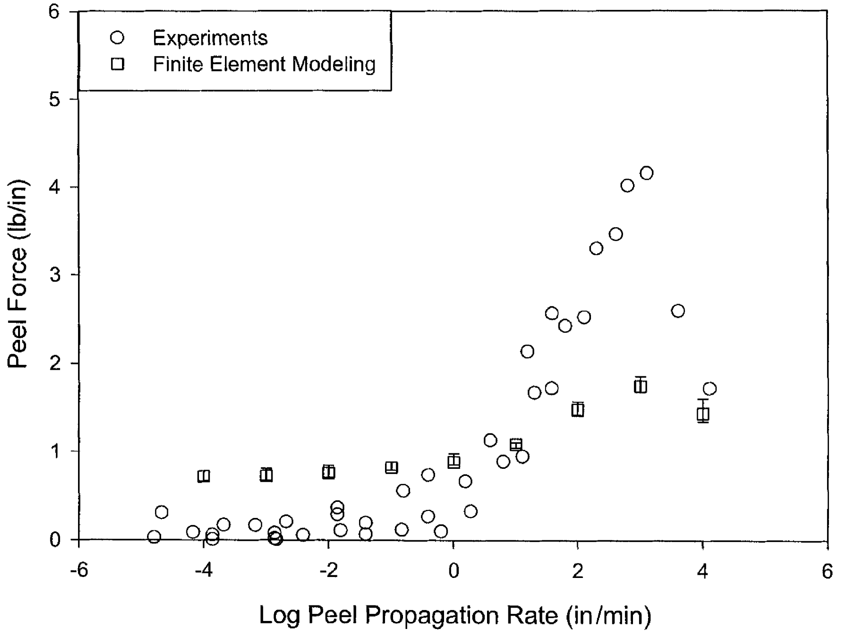
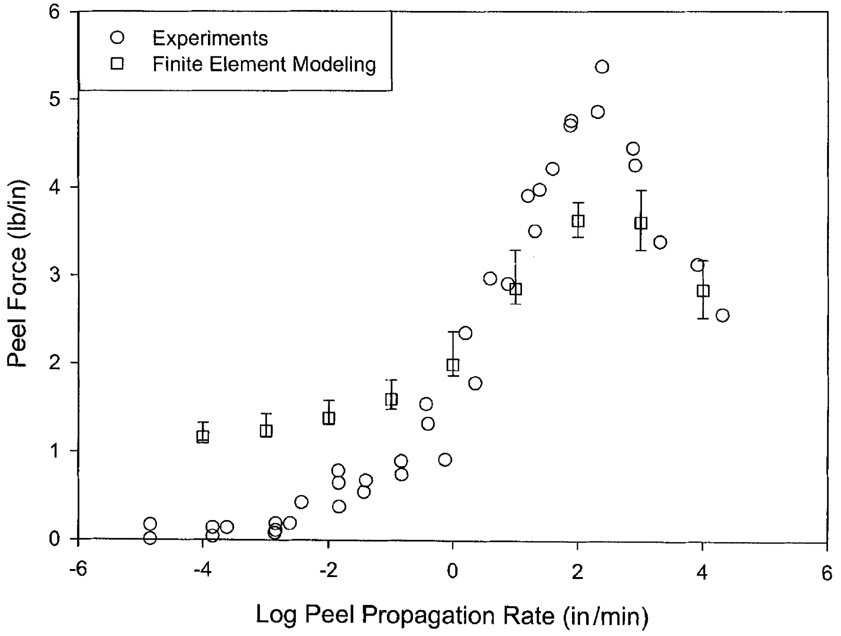


FIGURE 4 Peel force *versus* peel rate master curves at reference temperature 25°C for (a) PSA A, (b) PSA B, (c) PSA C, and (d) PSA D. (*Continued.*)



(c)



(d)

FIGURE 4 (Continued).

results. This discrepancy might be due to several possible reasons, including: the cavitation that is usually observed in peeling of PSAs was not incorporated into the finite element model; the strength of the interfacial interaction may be rate-dependent; and the nonlinear viscoelastic behavior of the PSAs at large deformation was not considered. Compared with the energy losses induced by the viscoelastic deformation of the PSAs, the energy consumption associated with the cavitational processes of the PSAs is expected to be minimal. Several studies have suggested that interfacial fracture may be a rate-dependent process [13, 14], but this concept has not been validated in the area of peeling of PSAs. The shear relaxation moduli of the PSAs, as represented by a generalized Maxwell model, well describes their linear viscoelastic response at strains on the order of several percent. However, as mentioned above, the PSAs may experience strains on the order of several hundred percent prior to debonding. At such strains, the PSAs may significantly harden owing to the alignment of the molecular chains. Such nonlinear behavior may result in substantial energy dissipations. Therefore, it is recommended that nonlinear models, such as those capable of describing the strain-hardening behavior of viscoelastic materials at large deformation [15, 16], should be used to evaluate the observed discrepancy between the finite element predictions and the experimental measurements.

CONCLUDING REMARKS

The interfacial failure regime in the peel behavior of the PSAs was studied using the finite element method. The linear viscoelastic properties and the peel performance of four natural-rubber–based PSAs were experimentally measured to provide input for and comparison with the modeling. Interfacial debonding was simulated using a failure criterion based on the level of the stored elastic strain energy density in the PSAs. It was shown that the finite element calculations predicted the general shape of the peel force *versus* peel rate master curves. However, the finite element modeling underpredicted the peel forces. This might be associated with the fact that the large-strain properties of the PSAs were not considered in this study. Also, it should be emphasized that the finite element modeling captured the general features of the peel behavior of the PSA remarkably well, despite the complexity involved in real peel testing and the simplicity of the approach used to model this process.

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