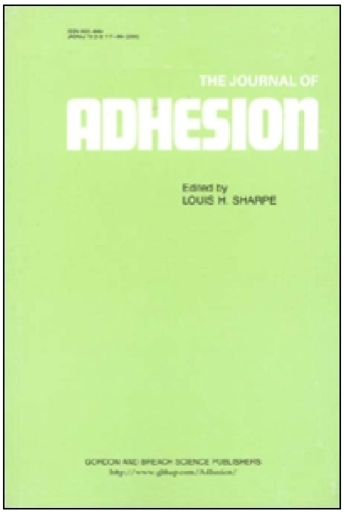

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PREDICTING SHEAR ADHESION OF WATER-BASED PRESSURE-SENSITIVE ADHESIVES VIA TRANSIENT RHEOLOGICAL MEASUREMENTS

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The relation between shear holding power and pressure-sensitive adhesive (PSA) rheological properties is investigated. Two constitutive models of PSA rheology, i.e., Newtonian and Power Law, are considered and experimentally tested for their ability to predict shear holding power. A Power Law model is found to be suitable for this purpose and is used to predict shear holding power with dynamic viscosity and steady shear viscosity measured by means of a transient creep experiment. Dynamic viscosity is shown to fail in predicting shear adhesion of high-viscosity PSA, whereas steady shear produces good agreement. It is demonstrated that the Cox-Merz rule does not hold for high-viscosity PSAs, explaining the need for steady shear viscosity data in modeling shear holding power.

Keywords: Shear holding power test; Adhesion; Rheology; Pressure-sensitive adhesives

1. INTRODUCTION

Shear holding power [1] is an empirical estimation of pressure-sensitive adhesive (PSA) performance under tangential stresses and represents a key parameter to which PSAs are formulated. The test has large uncertainty and is time consuming, thus having spurred

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numerous efforts [2–6] directed at predicting this parameter from mechanical properties of PSAs.

Several methods have been used. For example, an inversion of the stress relaxation modulus of PSAs [2] was used in predicting shear holding power. There have been attempts to predict shear holding power by formulating generalized viscoelastic models for PSAs [3].

The most elegant method from a practical standpoint is to predict shear holding power from PSA viscosity [4, 5]. Dahlquist [4] presented two mathematical models for predicting shear holding power considering two constitutive PSA models, Newtonian (viscosity independent of shear rate), and Power Law ($\eta = a\dot{\gamma}^b$, where η is non-Newtonian shear dependent viscosity, $\dot{\gamma}$ is shear rate, and a and b are constants). Both models assume that PSA bulk viscosity governs PSA deformation in thin films. His results for the shear holding time are

$$\textit{Newtonian:}\ t = \frac{\mu L_1^2 L_2}{2Mg\delta}, \tag{1}$$

$$\textit{Power Law:}\ t = \frac{L_1^{\frac{b+2}{b+1}}}{\frac{b+2}{b+1}\delta\left(\frac{Mg}{aL_2}\right)^{\frac{1}{b+1}}}, \tag{2}$$

where t is the value of shear holding time, L_1 and L_2 are correspondingly the initial overlap length and width, δ is film thickness, μ is Newtonian viscosity, a and b are parameters in the Power Law model, M is the applied mass, and g is the acceleration due to gravity.

An approach similar to Dahlquist's has been used by several authors [5, 6]. Zosel [5] used dynamic viscosity in predicting shear holding values assuming the Cox-Merz [8] rule, which defines an empirical relation between shear and dynamic viscosity, *i.e.*, $\eta(\dot{\gamma}) = \eta^*(\bar{\omega})$, where $\dot{\gamma} = \bar{\omega}$. Zosel found this approach to be reasonably accurate in predicting shear holding power of low viscosity PSAs, *i.e.*, zero shear viscosity 2.5×10^5 Pa.s, 8×10^5 Pa.s at 35 and 23°C, respectively [5]. The method failed, however, in the case of higher viscosity PSAs, *i.e.*, zero shear viscosity of 2.5×10^8 Pa.s at 23°C [5]. This observation agrees with our own experience. We often find that shear holding power can be successfully predicted from the PSA's dynamic viscosity (assuming the Cox-Merz rule) when the adhesive has a viscosity lower than 1×10^6 Pa.s (at the conditions of shear rate of 0.01 s^{-1} and 25°C). On the other hand, modeling shear holding power of higher viscosity PSAs (viscosity larger than 4×10^6 Pa.s at 0.01 s^{-1} and 25°C) using dynamic viscosity results in erroneous predictions.

This inability to predict shear holding power accurately for higher viscosity PSAs motivates this work.

In this work we experimentally tested the validity of Equations (1) and (2) in describing the shear holding test for a water-based PSA. We determined that the Power Law model was accurate in describing the rheology of the water-based PSA under the conditions of the shear holding test. Shear holding power test results were predicted using dynamic viscosity and steady shear viscosity determined by transient measurements. We confirmed that dynamic viscosity was ineffective in predicting shear holding power for a high-viscosity PSA. We established that the reason behind the failure was that the Cox-Merz rule did not hold in the case of a high-viscosity water-based PSA, and we demonstrated how shear holding power of a viscous PSA could be determined using shear viscosity measured by transient rheological measurements.

2. EXPERIMENTAL

Rheology of water-based PSA was measured on a Rheometrics, SR-5000 stress-controlled rheometer using an 8 mm parallel plate at 25°C. To improve adhesive bonding to the surfaces of the parallel plate tool, the samples upon loading were heated to 80°C and cooled down. The steady state acceptance criterion in determining steady shear viscosity in transient rheological testing was set at 3%. The transient experiment consisted of three consecutive stress step functions corresponding to 20667, 31000, and 62000 Pa stresses. Time temperature superposition between 0 and 60°C was used in measuring dynamic viscosity. The data appearing in Figure 1 were shifted to the reference temperature of 25°C. It was found that in order to minimize the possibility of sample failure during the creep experiment, the gap had to be 0.5 mm or less.

3. SAMPLE PREPARATION

Two acrylate PSAs used in producing commercial labelstock were used for shear holding power testing. The samples are referred in this study as samples 1 and 2. The adhesives were coated directly on a Mylar® (PET) substrate (type A, 2 mil [0.025 mm] thick) using a Gardco Automatic Drawdown Machine and Meyer rods # 18 or 20 to obtain dry coverage of 20–24 mg/m^2. The coated films were dried in a convection oven at 75°C for 10 min and then allowed to condition for ~15 hours (overnight) at 23 ± 2°C and 50 ± 5% relative humidity.

Samples for rheological testing were obtained from the PSA films, which were rolled into balls and heat pressed into ~1 mm thick films.

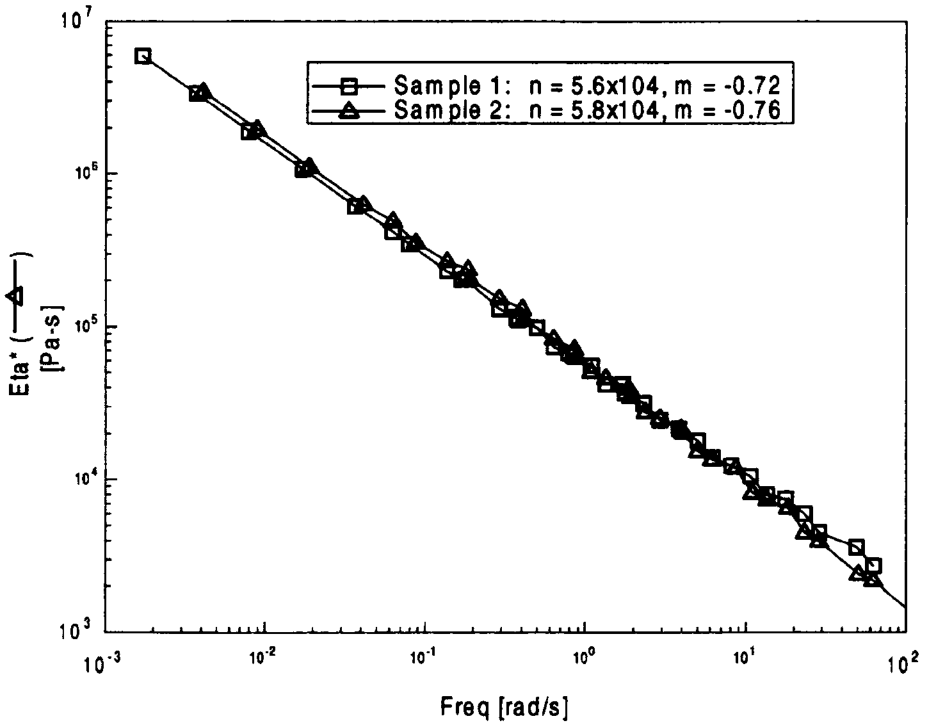

FIGURE 1 Dynamic viscosity of Samples 1 and 2. The figure is obtained by time temperature superposition; reference temperature is 25°C.

The films were allowed to condition overnight at room conditions prior to rheological measurement.

4. MODEL TESTING

There are several approaches by which models (Equations (1) and (2)) can be tested. One possible approach would be to construct a device similar to what has been built by Miyagi et al. [6] and replicated by Zosel [5]. In this device the position of a tape is recorded as a function of time as it slides during the shear holding experiment. We, however, decided to undertake a different approach. Instead of determining displacement as a function of time, we measure holding time as a function of *load*.

Table 1 contains shear holding times measured for sample 1.

SOLVER (Generalized Reduced Gradient (GRG2) nonlinear optimization code protocol in EXCEL) was used to determine which model (Equations (1) or (2)) described the physics of PSA deformation in the shear holding test. This was done by minimizing the average of squares of the difference between model predictions and the experimental data, subject to either Newtonian viscosity (μ, Equation (1)) or a, b Power Law fluid parameters (Equation (2)). Figure 2 shows the result of this exercise and demonstrates that the Power Law, not the Newtonian, constitutive equation describes water-based PSA rheology during the shear holding test.

5. PREDICTION OF SHEAR HOLDING POWER DATA

Viscosity can be measured by a number of methods. In the case of viscous materials, the method of choice is to determine the dynamic viscosity and use the Cox-Merz rule [8]. Figure 1 contains dynamic viscosity data measured with samples 1 and 2 using time-temperature superposition.

TABLE 1 Holding Time as a Function of Load

Sample 1	Load	Holding time (min)
$1'' \times \frac{1}{2}''$ and 0.515 kg	1.03	13300 ± 1000*
$1'' \times \frac{1}{2}''$ and 0.615 kg	1.23	7600 ± 1100
$1'' \times \frac{1}{2}''$ and 0.715 kg	1.43	6500 ± 70*

* The errors are the standard deviation for each condition measured in sets of 3.

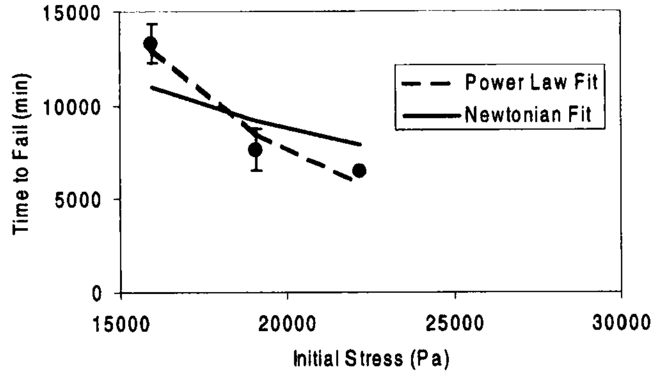

FIGURE 2 Fits of shear holding times by Equations (1) and (2) as a function of initial stress, $\tau_0 = gM/L_1L_2$.

Another means of measuring shear viscosity is by creep, where a constant stress is applied until a specified steady-state criterion is reached. The creep data for samples 1 and 2 are shown in Figure 3. Stress values of 20667, 31000, and 62000 Pa were used, which correspond to the stresses experienced by the PSA at $\frac{3}{4}$-, $\frac{1}{2}$-, and $\frac{1}{4}$-inch (1.9, 1.3, and 0.1 cm) overlap at the standard shear holding test conditions (*i.e.*, initial $1'' \times 1''$ (2.5×2.5 cm) $\times$ 1 kg). Power Law parameters determined from the data are shown in Table 2.

Figure 4 tests the validity of the Cox-Merz rule and demonstrates that the rule does not hold in the case of the high-viscosity PSA materials considered here. This suggests that dynamic viscosity is not suitable for determining the shear holding power of high-viscosity PSAs and provides a plausible reason for the failure of this approach by previous authors.

Shear holding power is predicted using Equation (2) and the Power Law parameters presented in Table 2. Table 3 contains experimental shear holding times along with model prediction. The data demonstrate that steady shear viscosity is a good predictor for shear holding power, and, as expected, dynamic viscosity data produce poor predictions.

6. DISCUSSION

We have demonstrated that the Power Law constitutive equation of the PSAs' rheology successfully describes the physics of the shear holding power test and that experimentally determined Power Law parameters can adequately predict shear holding power. Although the

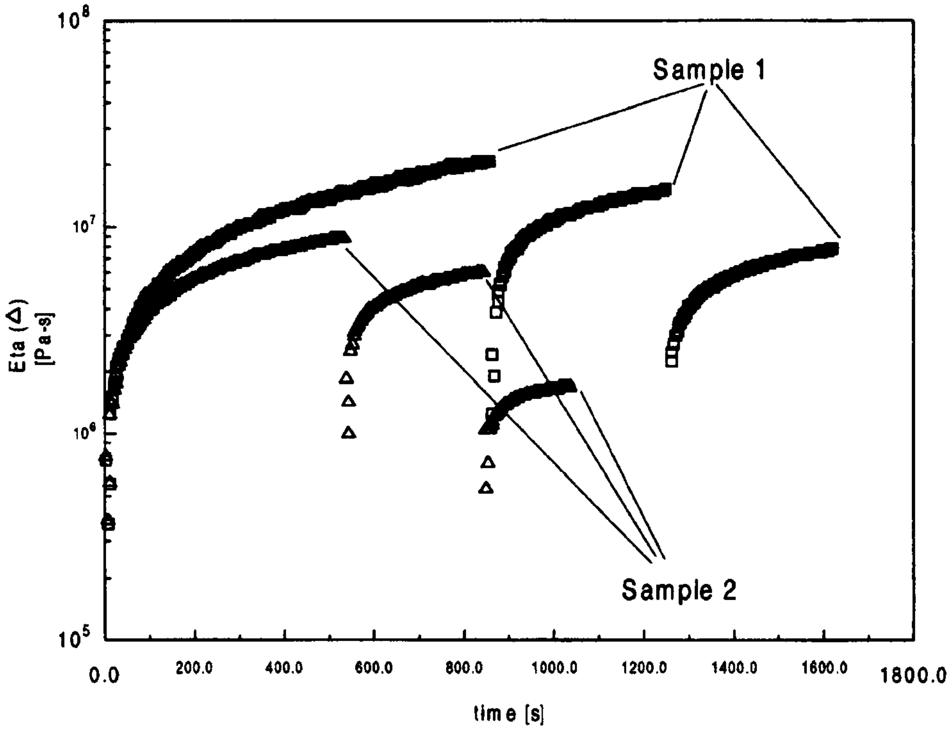

FIGURE 3 Creep experiment data for samples 1 and 2.

TABLE 2 Rheological Parameters

	η (Pa.s) @ 20667 Pa	η (Pa.s) @ 31000 Pa	η (Pa.s) @ 62000 Pa	Parameter *a*	Parameter *b*
Sample 1	2.07×10^7	1.48×10^7	7.85×10^6	8.11×10^5	– 0.470
Sample 2	8.99×10^6	6.05×10^6	1.68×10^6	2.27×10^5	– 0.613

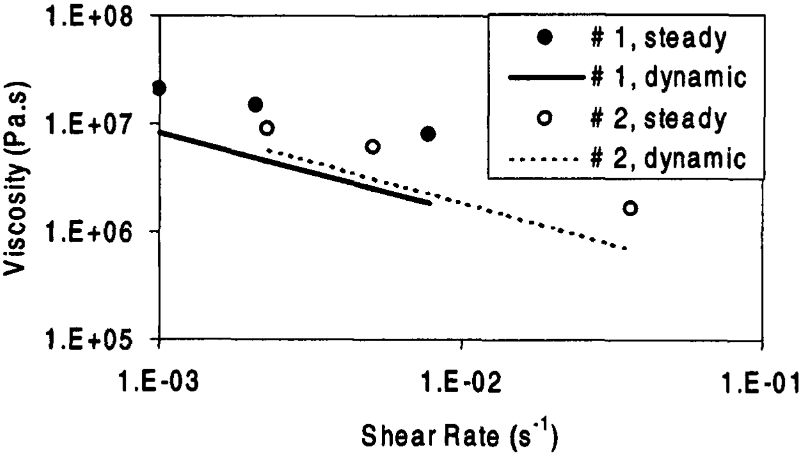

FIGURE 4 Test of Cox-Merz rule, samples 1 and 2.

TABLE 3 Model Prediction

	Shear holding time (min)	Steady shear viscosity data (min)	Dynamic viscosity data (min)
Sample 1	8800 ± 740	10600	350
Sample 2	4900 ± 350	6100	1000

experimental samples were all water-based PSAs studied at 25°C, this approach is general enough to be applicable at other temperatures and for other adhesive materials.

The rheological approach, in addition to eliminating numerous steps of applying PSAs to a substrate, drying, curing, cutting, and hanging PSA-coated tapes, significantly shortens the actual testing time. Sample 1-based data, although extreme, demonstrate this point.

TABLE 4 The Effect of Overlap on Shear Holding Time (Using Sample 2 data)

Standard 1″ × 1″ × 1 kg (min)	Off by 1 mm in vertical direction (min)	Off by 1 mm in tape width (min)
10600	9500 (1 mm less) 11900 (1 mm more)	9800 (1 mm narrower) 11400 (1 mm wider)

While the shear holding test with this sample lasted for 8800 min (~ 6 days) it required only about 30 minutes of rheological testing.

Despite the dramatic improvement of the modeled shear holding power results using steady shear viscosity data, there remains an appreciable difference between the modeled and experimental values. We note that the traditional shear holding test remains the preferred experiment when precise values of shear holding power are desired, whereas the method outlined here offers an attractive option for rapid screening of a large number of tackifiers or when the experimental accuracy is not crucial.

The model (Equation (2)) provides useful insight into both the source and the magnitude of potential errors arising during the execution of the standard shear holding power test. One source of error lies in the variation of the PSA layer thickness and is well documented [4]. Since the shear holding power is inversely proportional to layer thickness (δ, see Equation (2)), the not uncommon PSA coverage variability of 20–26 mg/m^2 will result (assuming a PSA dry density of 1 gm/cm^3) in a more than 10% error in shear holding time.

Another source of error springs from the potential variability of the initial overlap between the PSA-coated tape and the substrate as well as from the variability in tape width. To illustrate this point we calculated the errors in holding time that would result in overlap discrepancies of 1 mm. Table 4 contains the results of this calculation. The estimates are based on Sample 1 rheological parameters and show errors as large as 12%.

7. CONCLUSIONS

It is demonstrated that the Power Law constitutive equation describes the rheology of two high-viscosity water-based PSAs in a shear holding power experiment. The approach chosen in this verification is simple and does not require construction of any additional hardware. Therefore, it can be easily carried out with a host of other adhesive materials and at different temperatures.

It is demonstrated that shear holding power of the high-viscosity PSAs can be successfully predicted using steady shear data. The prediction is impossible using dynamic viscosity. This is explained by demonstrating that Cox-Merz rule does not hold for higher viscosity PSAs.

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