PEEL ADHESION AS A FUNCTION OF PEEL ANGLE, PEEL RATE, AND PEEL TEMPERATURE

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Introduction

Since its introduction, pressure sensitive adhesive (PSA) tapes and labels have been readily developed for all fields of applications. Prior to its introduction, two foreign objects must be bonded together by mechanical means; PSA surpassed such limitation and allowed surface adhesion of objects without the need of mechanical interlock. Today, PSA products serve essential purposes to our daily life in all fields of applications. In order to produce suitable PSA products for their respective application, adhesive industry have developed standard characterization methods to evaluate the product's performance. Currently, there are several standard testing procedures that evaluate a PSA's peel force. Table 1 lists the conventional standard testing methods.

Table 1. Standard Peel Adhesion Testing Methods

Organization	Method	Peel Angle (˚)	Test Speed	Resident Time
ASTM	D903	180	12 in/min	Open
ASTM	D1000	180	12 in/min	20 min
ASTM	D2860	90	Static	3 min
ASTM	D3330	180	12 in/min	<1 min
TLMI	L1A1	180	12 in/min	Open
PSTC	1	180	12 in/min	<1 min
PSTC	2	90	12 in/min	<1 min
PSTC	3	180	12 in/min	<1 min
PSTC	14	90	Static	3 min
FINAT	FTM1	180	300 mm/min	20 min ~ 24 hour
FINAT	FTM2	90	300 mm/min	20 min ~24hour
AFERA	4001	180	300 mm/min	10 min

ASTM: American Society for Testing and Materials
TLMI: Tag & Label Manufacturers Institute
PSTC: Pressure Sensitive Adhesive Council
FINAT: Féderation Internationale des fabricants et transformateurs d'Adhésifs et Thermocollants
AFERA: The European Association for the Self Adhesive Tape Industry

These standard testing procedures evaluate the peel force of the adhesive under constant temperature and humidity (23 ± 1˚C, 50 ± 5% RH), and under fixed peel rate and angle. The standardized conditions are beneficial in establishing a standard and comparing the relative performance of PSA; however, such performance indicated by the standardized tests may not effectively simulate the conditions of real world application. When a PSA product is used by the end-users, its working environment is never under a controlled environment with constant temperature or humidity. When a PSA product is peeled from its adhered objects, it is never peeled under a fixed peel rate or angle (90˚ or 180˚). Consequently, while the current standard testing methods are great at evaluating PSA products' relative performance at the prescribed conditions, they seldom simulate the real-world applications; in some cases, the evaluation may be simply irreverent. If one does not take the product's application conditions into account and simply develops a PSA product using the current standard testing methods, then the product may fall short of its adaptability and performance in the real-world applications.

It has been long established that peel conditions such as temperature[1,2], rate[3], and angle[4,5] all affect the peel force of the adhesive. In his study[6], Satas suggested that in order to achieve a better judgment of the adhesive, one should observe entire peel force vs. peel rate or peel force vs. peel temperature curve. In order to accurately evaluate a PSA product’s adaptability in the real world conditions, the formulator needs to see the performance curve in all different ranges of experimental conditions. Nonetheless, such studies have only been available in the academia research and have rarely been done in the industry due to the limited resource and machinery capability. Recent advance in the laboratory machinery surpassed such bottleneck and conveniently allowed the precise evaluation over all different ranges of experimental conditions.

In this report, a series of hot melt pressure sensitive adhesive (HMPSA) tapes and their peel adhesion force are evaluated under different peel angles, rates, and temperatures to demonstrate their effect on the adhesion performance. Hopefully through this study, we can find a method to better evaluate a PSA product based on the real world conditions, so it may better assist the research and development efforts in the PSA industry.

Experimental

Materials and sample preparation. A series of HMPSAs are made with the same formulating components in different ratios of solid/liquid tackifying resin (Table 2). Their formulations consist ExxonMobil Vector 4113 (a Styrene-Isoprene-Styrene Block Co-polymer), Yangzi Eastman C-100W (a solid C-5 hydrogenated hydrocarbon resin), Yangzi Eastman C-8010 (a liquid C-5 hydrogenated hydrocarbon resin), and Karamay KN-4010 (a naphthenic oil). The series share the fixed formulation of 30% Vector 4113 and 10% KN-4010. HMPSA-1 has 60% solid tackifier; HMPSA-2 has 45% solid and 15% liquid tackifier; HMPSA-3 has 30% solid tackifier and 30% liquid tackifier. The HMPSAs are coated on a Glassine release paper and transferred onto a 50 μ PET film with a uniform thickness of 20-21μ by a hot melt lab coater.

Table 2. Test Formulations

	Vector 4113 (%)	KN-4010 (%)	C-100W (%)	C-8010 (%)	Coating Thickness (μ)
HMPSA-1	30	10	60	0	20
HMPSA-2	30	10	45	15	21
HMPSA-3	30	10	30	30	21

Basic Characterization Tests. The viscosity of the three HMPSA samples is evaluated at 160, 170, and 180°C by a Brookfield Viscometer accompanied with a Thermosel (ASTM 3236) respectively. Pressure sensitive adhesion performance, such as loop tack (PSTC-16), holding power (PSTC-107), and SAFT (ASTM D-4498-95) are examined based on their specific standard test methods. Rheometrics RDA2 (A strain controlled rheometer) was used to determine the rheological properties of tested samples at varied temperatures. In this study, the test condition used for rheological evaluation is as follow: 1) temperature scan: -20 to +100˚C; 2) tool or geometry: 8 mm parallel plate, 3) oscillatory frequency: 10 rad/sec., 4) sample thickness: 2.0-2.5 mm.

Testing Equipment for Peel Adhesion. A multi-functional peel tester is used to evaluate the peel force in different peel angles, rates, and temperatures (Figure 1). The peel tester is capable of evaluating independent parameters of peel rate from 15-300 cm/min, angle from 0° to 180°, and temperature (by a hot-cold plate) from -20-80°C without using an heating oven and liquid air.

Figure 1. The multifunctional peel tester used for this study that is capable of evaluating the peel force under varying angle, rate, and temperature.

Experimental Mechanism. Figure 2 demonstrates the mechanism and terminology used in this report, where Va is the peel rate, Vb is the sled rate, and angle C is the peel angle. H1 and H2 are horizontal planes in respect to angles A and B. Peel rate represents the velocity of the test sample being peeled from the bonded object. Sled rate represents the horizontal velocity of the test sled being pulled by the equipment's motor. The peeling angle must satisfy the following criteria:

$$C = 180° - (A + B), where\ A = B \qquad (1)$$

The equation can be rearranged to help determine the angle A and angle B:

$$A = B = \frac{(180° - C)}{2} \qquad (2)$$

For example, if a peeling angle C is set at 180˚, then A = B = 0˚. If a peeling angle C is set at 90˚, then A = B = 45˚. If a peeling angle C is set at 30˚, then A = B = 75˚. An experiment with 60˚ peeling angle is shown in Figure 3, where A = B = C = 60˚.

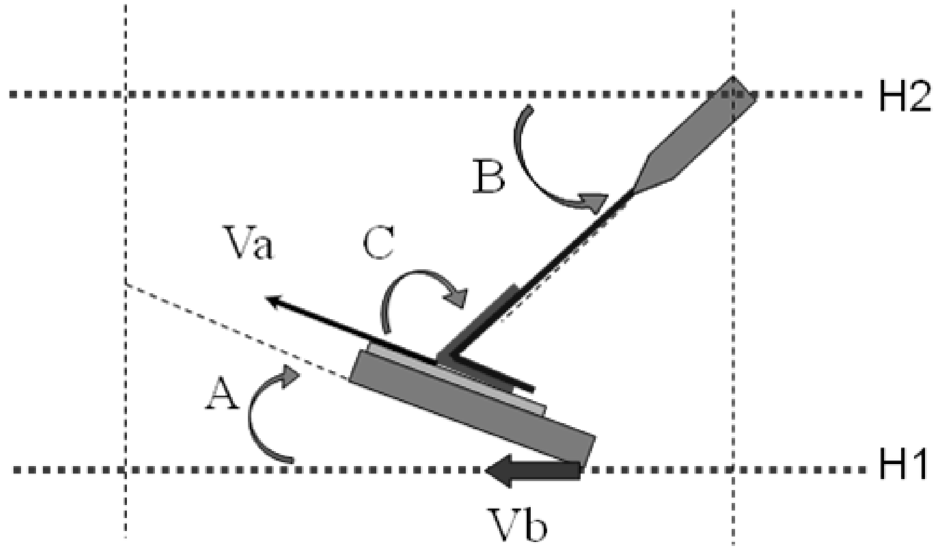


Figure 2. The cross-section of the experimental equipment, which demonstrates mechanism and terminology of this report, where Va represents the peel rate, Vb represents the sled rate, C represents the peel angle, H1 and H2 are horizontal planes in respect to angles A and B.

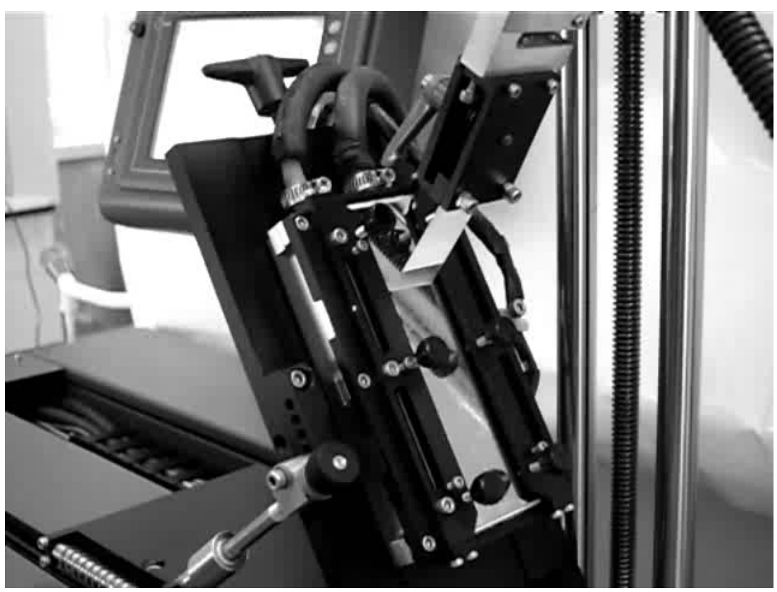


Figure 3. An experiment where the peeling angle is 60˚, using Equation (2), A = B = C = 60˚.

Sled Rate vs. Peel Rate. The sled rate is the horizontal velocity of the test sled being pulled by the driven jaw, and the peel rate is the realistic rate of the testing specimen being peeled away from . Observing Figure 2, an angular relationship between sled rate and peel rate can be described by Equation (3), where Va is the peel rate, Vb is the sled rate, and angle C is the peel angle:

$$Va = \frac{Vb}{cos(A)} = \frac{Vb}{cos\left(\frac{180-C}{2}\right)} \quad (3)$$

Current standard evaluation methods call for a testing parameter of 12 in/min or 300 mm/min (Table 1). The established test speed is in fact the sled rate and not the realistic rate at which the testing sample is peeled. At the 180° peel angle, as abided by most testing methods, the peel rate is identical with the sled rate; however, at any other angle, the realistic peel rate is different from the sled rate. Using Equation (3), the relationship between peel rate and peel angle under a constant sled rate is plotted in Figure 4.

In this study, the angular effect on a PSA tape's performance is examined. The evaluation is conducted in a fixed peel rate throughout all angular conditions, as the peel rate can more realistically relate to the product performance than the sled rate. The peel rate is the actual rate of the tape being removed from its bonded surface, whereas the sled rate is only an instrumental parameter that does not directly simulate any real world application of the tape product.

The peel testers used in this study has the programming capacity to allow a direct control on the peel rate based on the relationship described in Equation (3). For ones that do not have the equipment that allows such capacity, the study into the peel angle effect is still available if the sled rate can be precisely controlled. Using Equation (3), the relationship between sled rate and peel angle under a constant sled rate is plotted in Figure 5. The plot demonstrates the precise sled rate that should be used at each testing angle to ensure the peel rate remains constant in all angular testing parameters.

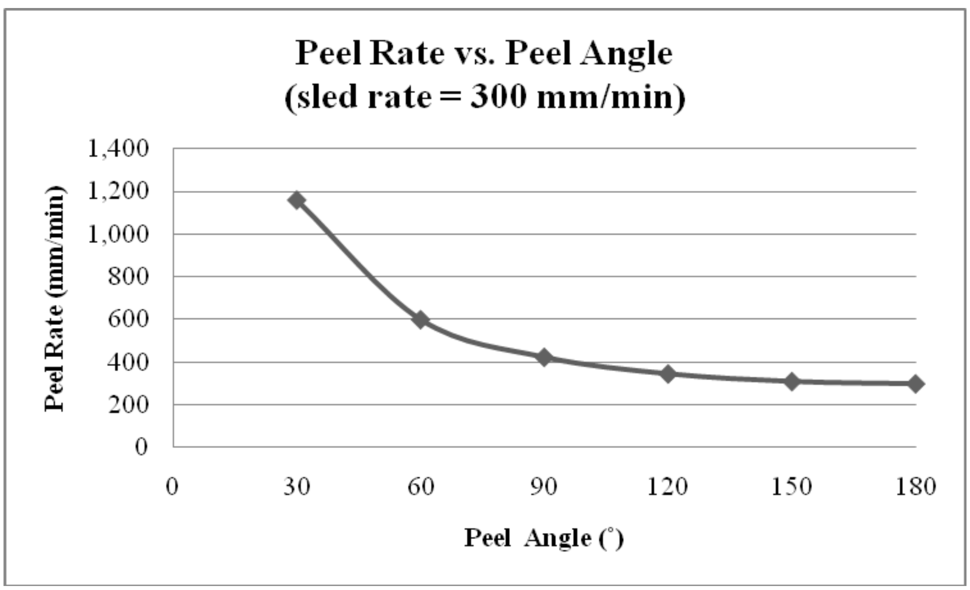


Figure 4. Relationship between realistic peel rate and peel angle under a constant 300 mm/min sled rate. Note the peel rate changes with angle despite the sled rate being constant.

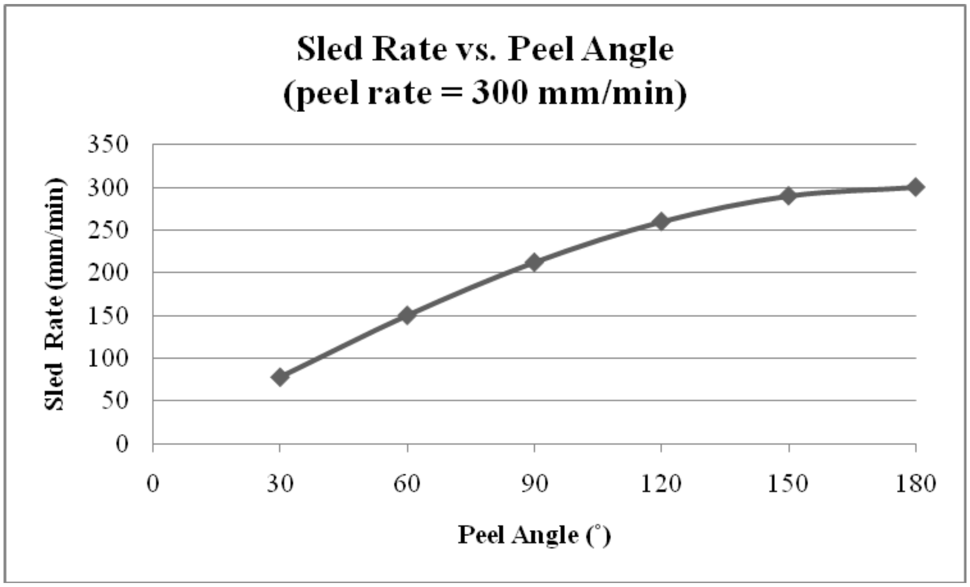


Figure 5. The sled rate used at each testing parameter to ensure the peel rate remains at constant 300 mm/min rate.

Results & Discussion

Basic Characterization Tests

The result of the HMPSAs' basic characterizations tests is listed in Table 3. The viscosity, loop tack, holding power, and SAFT all correspond with the tackifier content. Higher solid tackifier content results in a higher viscosity at all three testing temperatures. The total average molecular weight increases when a higher content of high softening point tackifier is incorporated. Loop tack, holding power, and SAFT also exhibit higher values when higher content of solid tackifier is used. The cause of this performance variation is related to the rheological property of test formulation which has been illustrated in many previous studies[7-9]. The rheological curves and some key rheological properties for tested HMPSAs are shown on Figure 6 and listed on Table 4.

Table 3. Basic Properties, Pressure Sensitive Adhesion Performance

	Viscosity (cps)			Loop Tack (N/25mm²)	Holding Power (4 psi, hr)	SAFT (˚C)
	160˚C	**170˚C**	**180˚C**			
HMPSA-1	21,000	15,000	11,000	38	>300	82
HMPSA-2	17,000	13,000	9,400	31	34	80
HMPSA-3	12,000	9,300	7,000	20	7	78

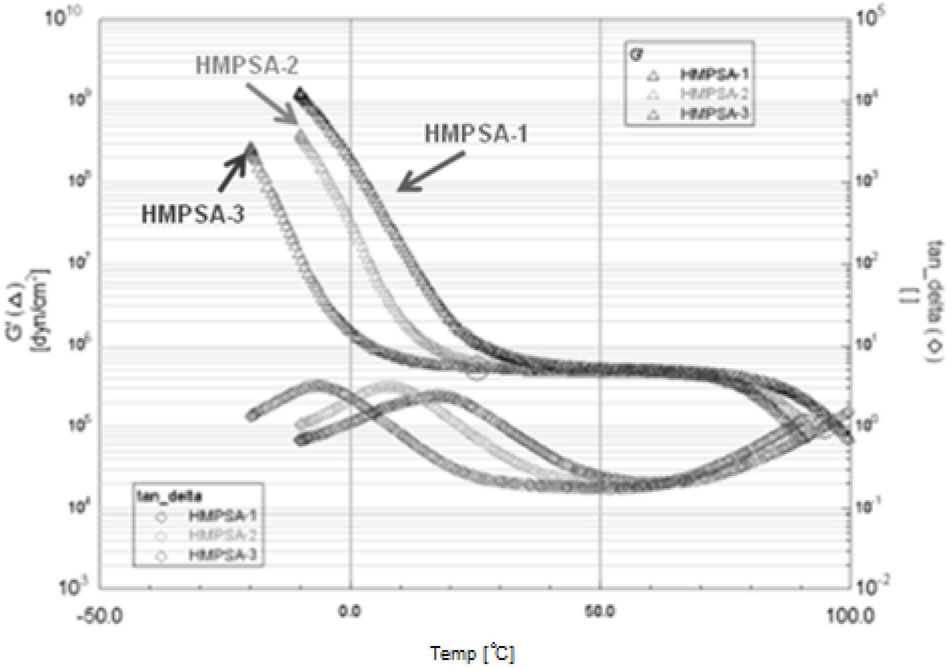


Figure 6. The overlay rheological curves of the three HMPSA samples used in this study

Table 4. Key Rheological Properties

Rheological Properties						
	Tg (˚C)	**G' (25˚C) (dyne/cm^2)**	**Gn˚ (dyne/cm^2)**	**Tan δ min**	**Tan δ min (˚C)**	**Flow Point (˚C)**
HMPSA-1	17.4	1,130,000	497,000	0.20	60	95
HMPSA-2	7.5	605,000	445,000	0.19	58	91
HMPSA-3	-7.1	527,000	471,000	0.17	52	88

It is worthwhile pointing out that the Tg resulted from HMPSA-1, 2, 3 having varied ratio of solid/liquid tackifiers are 17.4, 7.5 and -7.1 ˚C respectively. A blend containing more ratio of high Tg tackifier will result in a higher Tg of the formulation. For blends having the same chemical composition but different Tg, in general, the series PSA performance maximizes when the Tg increases to a value close to room temperature. Further increase of Tg at the vicinity of room temperature will cause an undesired stick-slip facture behavior. This stick-slip phenomenon is also observed at low test or service temperatures for a room temperature oriented formulation.

Angular Effect of Peel

The peel force of the HMPSA tapes are evaluated at 25°C and a fixed peel rate (300 mm/min) over varying angles. The result is plotted in Figure 7.

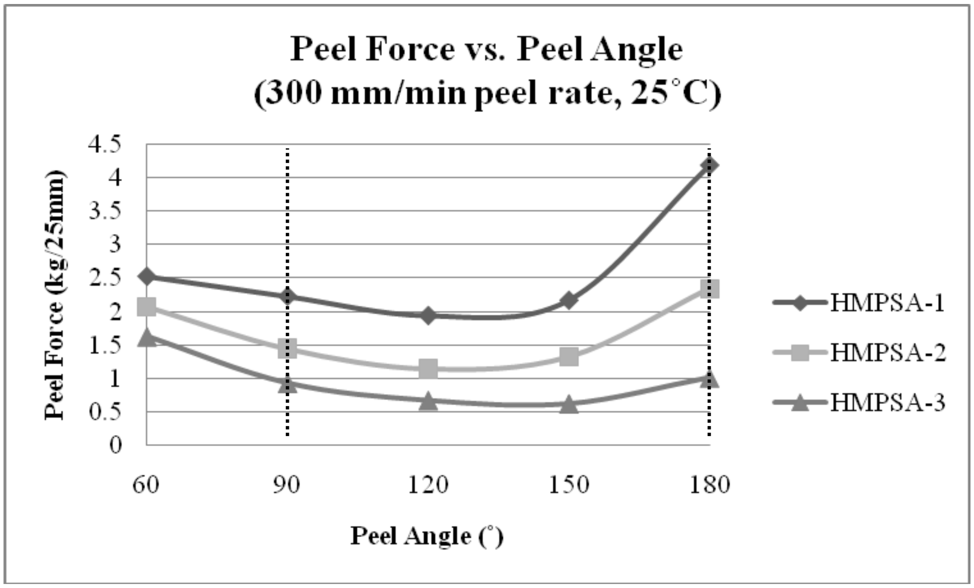


Figure 1. Peel force of the HMPSA series vs. angle at 25°C and 300 mm/min peel rate. The dash lines at 90° and 180° represent the points of evaluation by the current standard testing methods.

In this specific formulation and testing conditions, a higher solid tackifier content results in a higher peel force throughout all angle range of the curves. All curves demonstrate a similar curvature and trend, where the peaks of peel force occur at the two end points (60° and 180° peel angle). This is intuitive, since the two peaks are located at the angle where more peeling force is borne by the tape backing instead of the adhesive. The vortex of the three curves locate around 140-150° peel angle. The vortex represents the angle range where an ordinary PSA tape is the weakest against a peeling force or condition. Note that there is a slight vortex shift toward a higher angle as the liquid tackifier content increases, which suggests a shift in the weakest angular condition for this formulation and testing conditions.

Current evaluation methods focus on experimental conditions of 90° and 180° as marked in Figure 7. They are perfectly adequate for some applications that require such peeling angle; however, for applications that require angled application other than 90° and 180°, one should evaluate the tape products under their respective application condition.

It is so far inconclusive if a mathematical model can be adopted to predict a PSA's peel performance under different angular conditions. From the experimental result gathered from this study, we can only conclude that they display a similar curvature trend and the vortex shifts upon the change in formulation. The shift in vortexes however shows that a linear model cannot be adopted for this formulation system. More research effort into the angular effect is needed in order to develop such a mathematical model that is able to precisely predict the peel performance of PSA product.

Rate Effect of Peel

The peel force of the series of HMPSAs are evaluated at 25°C and 180° peel angle over varying peel rate. The result is plotted in Figure 8.

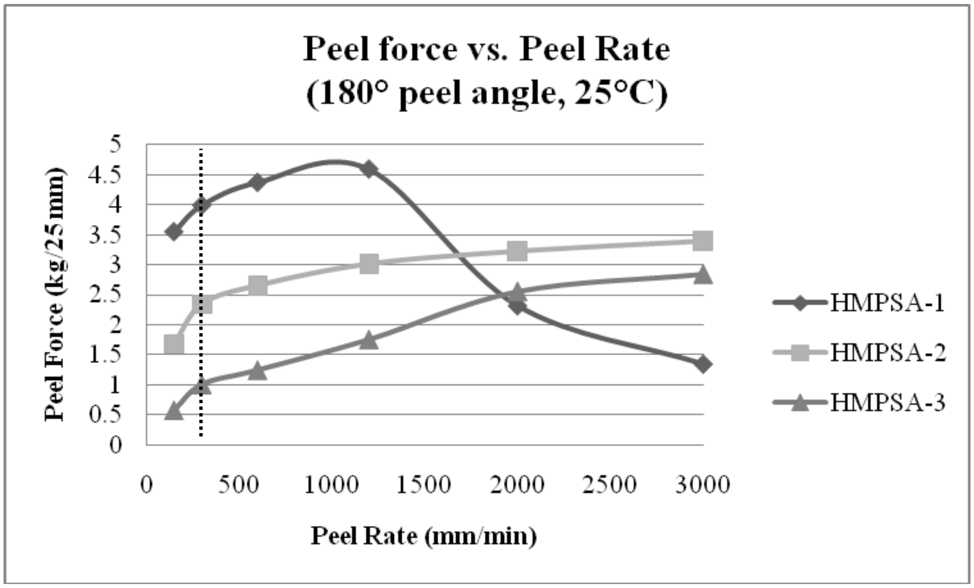


Figure 8. Peel force of the HMPSA series vs. peel rate at 25°C and 180° peel angle. The dash line at 300 mm/min represents the point of evaluation by the established testing methods.

All three curves behave similarly for peel rate up to 1200 mm/min. The higher solid tackifier content once again results in a higher peel force. All three curves increases exponentially at the beginning of the curve, then they hit a plateau zone where the peel force increases slowly with the increasing peel rate. HMPSA-1, however, drops significantly after 1200 mm/min peel rate and exhibits a stick-slip fracture mode.

According to time- temperature superposition principle, the higher peel rate resembles a lower temperature (Figure 9). The skip-slip fracture mode and the significant drop in HMPSA-1's peel force further reflect the adhesive's rheological behavior is similar to having a very high Tg formulation under standard testing parameters. The increasing peel rate, as a superposition of decreasing temperature, may push HMPSA-1's effective condition past and below the Tg. Such scenario is highly possible as the Tg for HMPSA-1 is 17.4°C (Table 4), only 7.6 °C from the testing condition. The other two samples, on the other hand, are still at superposition temperature range higher than their respective Tg’s.

Current evaluation methods follow a standard sled rate of 300 mm/min or 12 in/min as marked in Figure 8. The current standard sled rate does not seem to hold any significant meaning over the other peel rate testing range; however, the standard sled rate does provide a standard testing condition for the relative performance of PSA products. Nevertheless, one should bear in mind of the peel rate's time-temperature superposition principles.

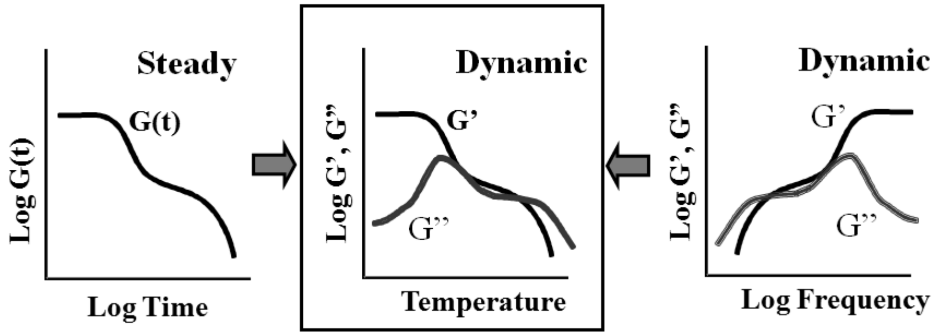


Figure 9. Time-temperature superposition principle

Temperature Effect of Peel

The peel force of the HMPSA tapes are evaluated under a fixed peel rate (300 mm/min) and 180° peel angle over varying temperature. The results are plotted in Figure 10.

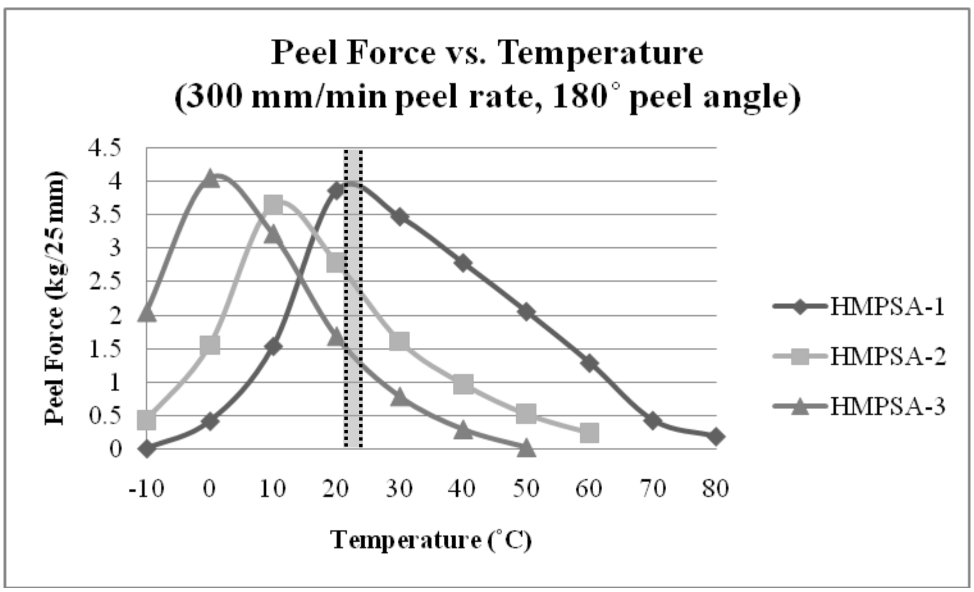


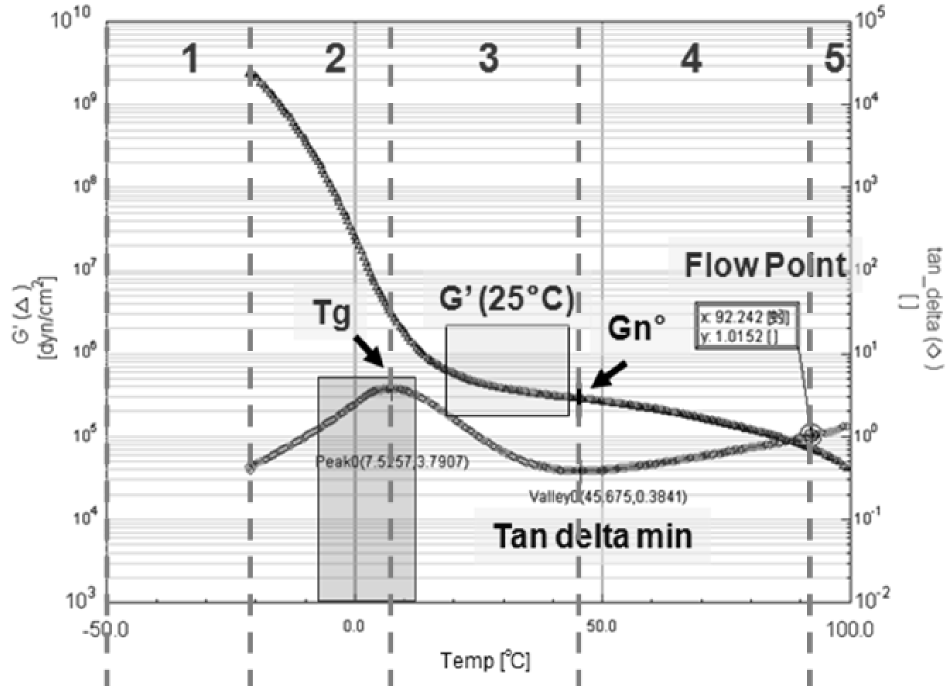
Figure 10. Peel force of the HMPSA series vs. temperature at 300 mm/min peel rate and 180° peel angle. The darken region at 23±1°C range represents the room temperature, also the range of evaluation by the established standard testing methods.

The plots illustrate an almost identical trend of the HMPSA in this experimental series. The curves can almost overlap and fit on top of one another with the shift in temperature range and peak height. The peaks shift toward the lower temperature range as the liquid tackifier content increases. The horizontal shift indicates that an increase of liquid tackifier content in this specific formulation model results in an increase of adhesive performance in a lower temperature range.

Current evaluation methods evaluate the peel force under room temperature, which is defined as 23±1°C. Such precision is hard to enforce such in the laboratory unless a strict temperature control is available. The plot demonstrates a substantial change in the peel force by even one or two degree of difference. If a standard condition is used to compare a product's relative performance, then the standard condition must be strictly enforced. Moreover, the room temperature only focuses on a specific range on the peel performance curve as marked on the plot. From there, it is impossible to predict the product's performance in real world condition, in different temperature range. It is only when we see the full picture of the product performance in all temperature range, that we are able to predict its adequacy in real world applications.

Many PSA researchers have revealed that the Tg or Tan delta value can be used to correlate PSA performance, particularly for peel and tack performance. Figure 11 is a typical rheological curve for a general purposed HMPSA. According to S.G. Chu[8], a general purpose PSA should have the G' (25˚C) within $2E^5$ and $2E^6$ dyne/cm^2 and the Tg between -10 to 10˚C. A rheological curve for a typical HMPSA can be divided into five zones. From low to high temperature, they are glassy zone (1), glass transition zone (2), entangled zone (3), disentangled zone (4), and flow zone (5). The rheological and physical features of each zone are illustrated on the bottom table of Figure 11. It has been reported that the suitable working range of a HMPSA is ranging from Tan delta max and Tan delta min, i.e. the entangled zone.[10]

Figure 12 is a compiled plot of Tan delta curve and peel force for HMPSA-1, 2, 3 evaluated at temperatures ranging from -10 to 80°C. It is notable that peel force and tan delta value show a parallel relation in the polymer entangled zone- between Tan delta max and Tan delta min. Peel force decreases as the Tan delta value is lowered at elevated temperatures for each single formulation above the Tan delta max. At a fixed test temperature, e.g. room temperature, the formulation having a higher value of Tan delta exhibits a higher peel force as well. The level of Tan delta value or Tg is apparently affected by the ratio of used solid/liquid tackifier in this study. This finding is true for all HMPSA systems which have identical chemical composition and simply different in formulation ratio[9].



Zone	Glassy (1)	Glass Transition (2)	Entanglement (3)	Disentanglement (4)	Flow (5)
G'	H	H-M	M	M	L
Tan δ	L	L-H	H-L	L-H	H
Feature	Brittle	Tough	High Cohesion	Low Cohesion	Melt

Figure 11. Features of rheological zones

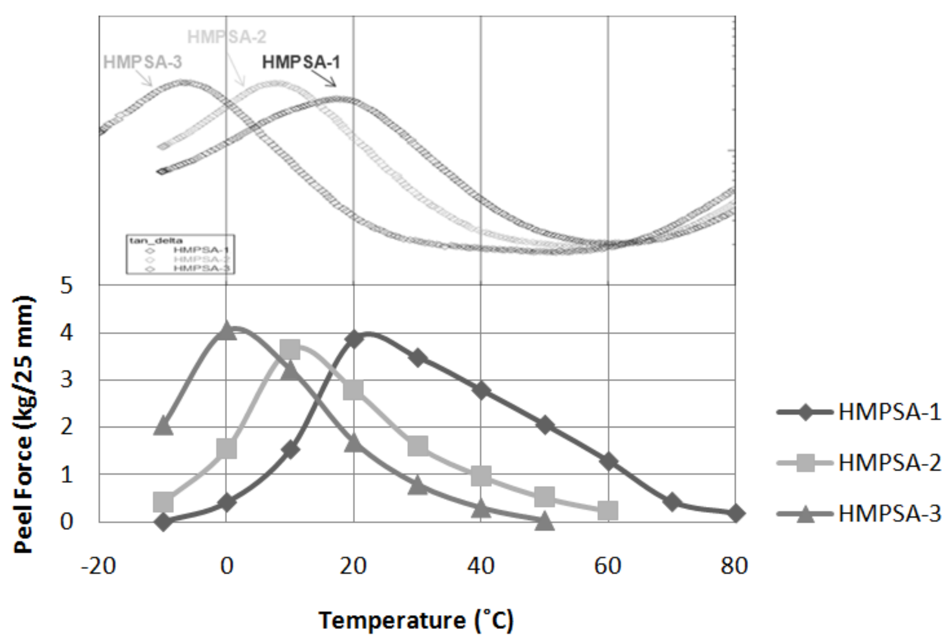


Figure 2. The relationship between Tan delta and temperature curves.

Conclusion

Peel force is affected by peel angle, peel rate, and peel temperature. Peel force determined by current standard peel test methods can't apply to the real world performance. To estimate the real world peel force, one should perform a combination of varied peel angle, rate, and temperature. Tan delta curve correlates very well with peel force generated at entangled zone- temperature range between Tan delta max and Tan delta min.

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Supporting Information Available: All raw basic characterization data, rheological curves, peel force data (recorded every 40 or 100 ms), and laboratory equipment & manuals.