

Performance of Adhesives Joints Programme 1996-1999

Project PAJ1

Failure Criteria and Their Application to
Visco-Elastic/Visco-Plastic Materials

Report No 9

**Study of a Range of Variables on Tack of
Heat Activated Adhesives**

By P Barraclough

SATRA
SATRA House
Rockingham Road
Kettering
Northants, NN16 9JH

February 1999

Performance of Adhesives Joints Programme 1996-1999

Project PAJ1

**Failure Criteria and Their Application to
Visco-Elastic/Visco-Plastic Materials**

Report No 9

Study of a Range of Variables on Tack of Heat Activated Adhesives

By P Barraclough

SATRA
SATRA House
Rockingham Road
Kettering
Northants, NN16 9JH

February 1999

Abstract

Tack is a material property associated with the immediate formation of an adhesive bond between two surfaces. It is a complex property that depends on the properties of the adhesive and surfaces. Measurement and control of tack are important in the shoemaking industry where soles and uppers are bonded during manufacture. In this report, the SATRA/NPL tack tester was used to study the influence of material and process variables on initial tack strengths. The effects of a number of different factors have been identified. These effects point to improved process conditions for the manufacture of footwear.

Contents

1.	Introduction	1
2.	Experimental Method	2
3.	The Influence of Experimental Variables	4
3.1	The Influence of Temperature and Time	4
3.2	The Influence of Compliance of Substrate Materials	7
3.3	The Influence of Test Conditions	12
3.4	The Influence of Preparation	15
4.	Findings on Improving Tack Strength	17
5.	Conclusions	19
6.	Acknowledgements	20

Study of a Range of Variables on Tack of Heat Activated Adhesives

By P Barraclough

1. Introduction

This report covers work carried out by SATRA on the measurement of tack as part of Project 1 of the DTI Materials Metrology Programme - Performance of Adhesive Joints 1996-1999. This programme is managed by NPL and sponsored by the Department of Trade and Industry.

Tack has been defined as 'the property of a material that enables it to form a bond immediately on contact with another surface which can be an adherend or another layer of adhesive'. In footwear sole bonding and similar applications of rubbery adhesives, where coats are applied to both adherends, tack is essential to enable them to form a bond on contact and coalesce to form a single coherent adhesive layer. Shoemaking is particularly demanding as the 'spotting tack' force must be sufficient to secure the portion of the sole 'spotted' onto the upper while the remainder is brought into proper alignment. It must also maintain the integrity of the assembly while it is consolidated in the bonding press.

Most sole bonding adhesives are not tacky at room temperature and are heat activated to provide the necessary level of tack, with rapid cooling and setting during a short pressing time. The tack is therefore transient making the measurement of tack force and influences on it very difficult. Historically tack has been assessed by subjective means. During a previous MTS programme 1993-1996 an instrument was developed by NPL and SATRA for measurement of tack of heat activated adhesives. One coated adherend is heated. The second coated adherend is brought into contact at a controlled speed until a specified contact pressure is achieved. The force to separate the joint is determined as a measure of the tack achieved.

Under the current programme the operation of the tester has been improved and it has been used in a comprehensive work programme with the following aims:

- i) to investigate the factors influencing tack
- ii) to identify effective means of optimising tack strength
- iii) to demonstrate the versatility of the SATRA/NPL tack tester.

2. Experimental Method

In this project, the operation of the SATRA/NPL tack tester is as described in detail in the report “An instrument for the measurement of “Tack” for the footwear industry”, MTS Adhesive Project 5, Measurements For Optimising Adhesives Processing Report 14, written by A Olusanya and L. A. Lay, September 1996 [NPL Report CMMT(B)91]. The major features and improvements to the tack tester since then are described below.



Fig 1. SATRA/NPL Tack Tester

The basic concept is to use rectangular adhesive coated specimens of leather, cotton, rubber or PVC positioned at right angles to each other. The adhesive is heated on the rubber or PVC and the specimens are pressed together using a known force. The ‘tack force’ is then defined as the tensile force required to cause separation.

The equipment consists of a Mecmesin M2500N Motorised Test Stand, mounted on a base plate, and a Mecmesin AFG-2500N Advanced Force Gauge. The original experimental equipment is shown in Fig 1. The force gauge is connected to the test stand so that when the selected downward force is achieved, the crosshead direction is reversed.



Fig 2. Modified Loading Stage

The original tensioning clamps were replaced with adjustable clamps so that the effect of compliance could be studied and any variability introduced by tension in the samples was removed. The lower specimen holder is mounted on a linear stage that allows the sample to be positioned under the adjacent short-wave radiant heater (fitted with thyristor power control and timing device). After heating it is moved under the upper specimen.

The speed range for both upward and downward movement can be set independently. The force gauge records the maximum compression force reached and the maximum separation force. The new sprung connector (Fig 2) was introduced to reduce the amount of overshoot on the equipment and also so that the equipment could be used with

the automatic return facility and repeatable contact pressures. The machine measurement cycle can now be run at full speed without overshooting the required pressure. The time between heat activation and adhesive contact is reduced and, thus, the accuracy and the repeatability of the method are improved.

The adhesives used were solvent based polychloroprene (Neoprene) or polyurethane. Both were supplied by Bostic UK. These were applied with a coating layer weight of 5-10 mg cm⁻² which corresponds to a layer thickness of ca. 50µm to 100µm. The tests were performed using standard unprepared cotton upper (1 mm thick) and either standard resin rubber (95 IRHD) or standard PVC (70 IRHD) at 3 mm thick soling. The resin rubber was prepared by scouring for use with Neoprene adhesive or halogenated for use with polyurethane. The PVC was solvent wiped with ethyl acetate or MEK. The adhesives were all applied by handbrush. All preparation and adhesive application was in accordance with SATRA test method AM2: 1997. Adhesives were dried for 60 minutes after application.

The standard test conditions are laid out in below.

Loading Rate/Separation Rate	10 mm s ⁻¹
Applied Pressure	2 kg cm ⁻²
Dwell Time	ca. 1 s
Activation Temperature	83°C
Drying Time	60 Minutes
Upper Slack	0 mm
Sole Clamp Separation	75 mm

The detailed work programme was set out to examine the following areas of interest:

i TEMPERATURE AND TIME

To assess the influence on tack strength of activation temperature and the heating regime.

- Examination of cooling rates.
- Assessment of thermal effects on tack strength.

ii COMPLIANCE OF SUBSTRATE MATERIALS

To assess the influence of the distortion of substrates on tack strength.

- Assessment of upper compliance.
- Influence of soling compliance.
- Investigation into the interaction of the two compliances.
- Assessment of the influence of substrate compressibility and thickness on tack strength.

iii TEST CONDITIONS

To assess the influence of magnitude and rates of loading on tack strength.

- Loading rate and contact time.
- Separation rate.
- Contact Pressure.

iv PREPARATION

To assess the influence of sample preparation on tack strength.

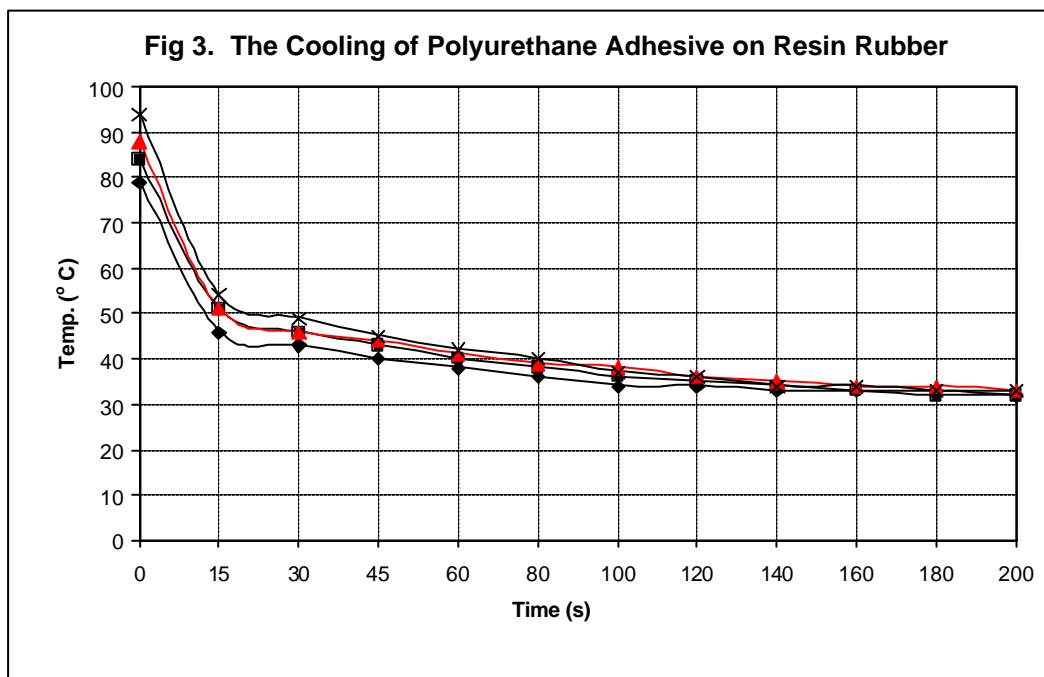
- Drying times.
- Coating Weight.

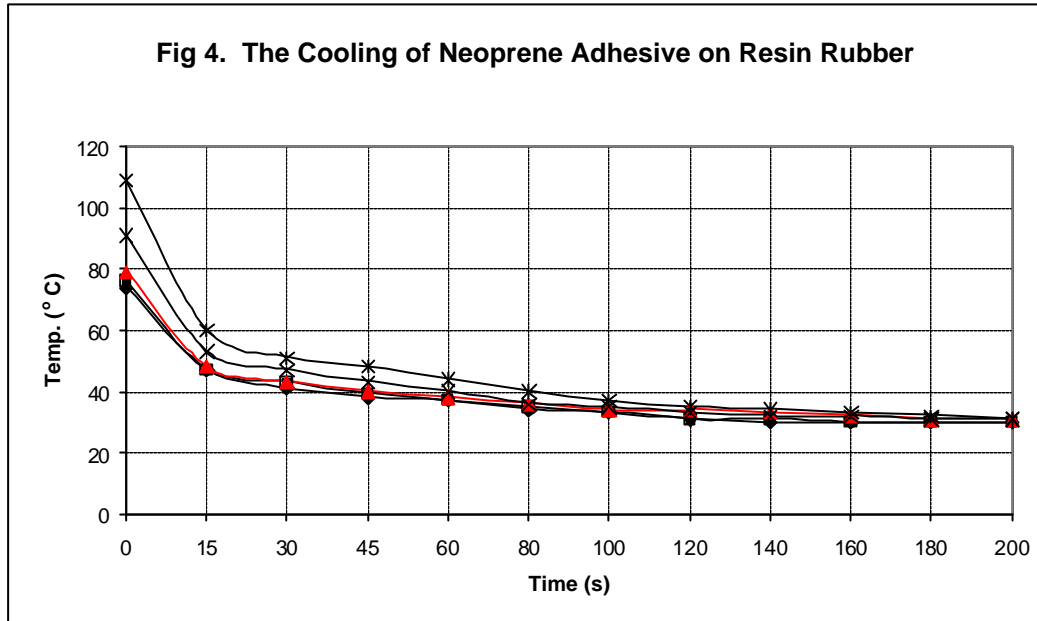
3. The Influence of Experimental Variables

3.1 THE INFLUENCE OF TEMPERATURE AND TIME

3.1.1 Examination of cooling rates

Fig. 3 shows the cooling rates for polyurethane adhesive on resin rubber and Fig. 4 shows the cooling rates for polychloroprene adhesive (Neoprene) also on resin rubber. The adhesive type makes no significant difference to the rate of cooling after activation. The measurements were taken using an infrared thermometer. It can be seen from these graphs that there is a relatively large surface temperature decrease in the first fifteen seconds after activation of the sample. Because of this sharp decrease, the repeatability of contact temperatures may be poor at elevated temperatures, reducing the accuracy of the tack strength results obtained. The significant cooling of the adhesive after activation emphasises the need for tightly specified measurement and manufacturing procedures to control tack.





3.1.2 Assessment of thermal effects on tack strength

a Minimised time between activation and adhered contact

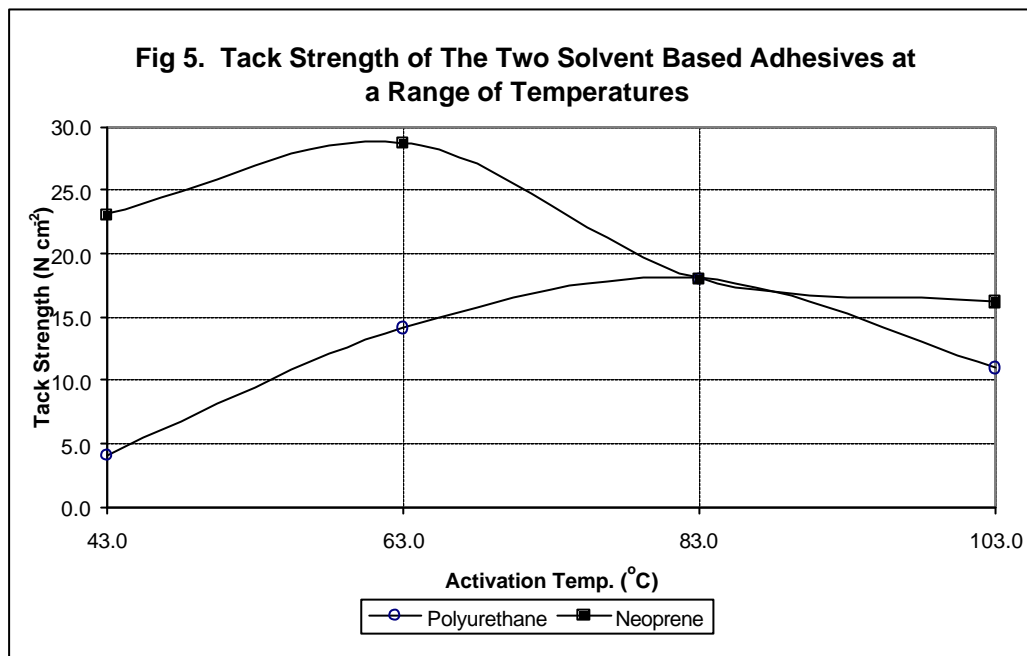
In the tests in this section the interval between activation and testing was minimised to maintain the temperature at the activation temperature. Tack strength was measured using standard resin rubber soling material and cotton upper material at a range of adhesive film temperatures between 43 and 103°C. The results are shown graphically in Fig. 5 and are summarised as follows (the figures in brackets are 95% confidence limits):

Temperature °C	Tack Strength N cm ⁻²	
	Polyurethane	Neoprene
43	4.0 (± 1.2)	23.0 (± 8.3)
63	14.1 (± 2.4)	28.7 (± 4.3)
83	18.0 (± 1.8)	18.0 (± 1.0)
103	10.9 (± 0.6)	16.2 (± 1.7)

It can be seen that the polyurethane adhesive gives a maximum tack strength, in these experiments, in the region of 83°C. The 43°C tack strength is very low, probably indicating that the adhesive has not been sufficiently activated. The Neoprene adhesive gives a maximum tack strength in the region of 63°C. The results for the Neoprene adhesive suggest that there may be some advantages in using a lower activation temperature for bonding.

For the experiments that followed, unless otherwise indicated, both adhesives were tested at 83°C as at this temperature they give similarly high tack. The loading and separation rates were set to the maximum speed (10 mm s⁻¹) with the machine

automatically reversing at a predetermined contact pressure. The contact pressure (unless otherwise indicated) was selected to correspond with the spotting pressure used in industry of 2 kg cm^{-2} (196 kPa or 19.6 N cm^{-2}). The results are based on four replicates unless otherwise stated.



b Activated to a higher temperature and allowed to cool to contact temperature.

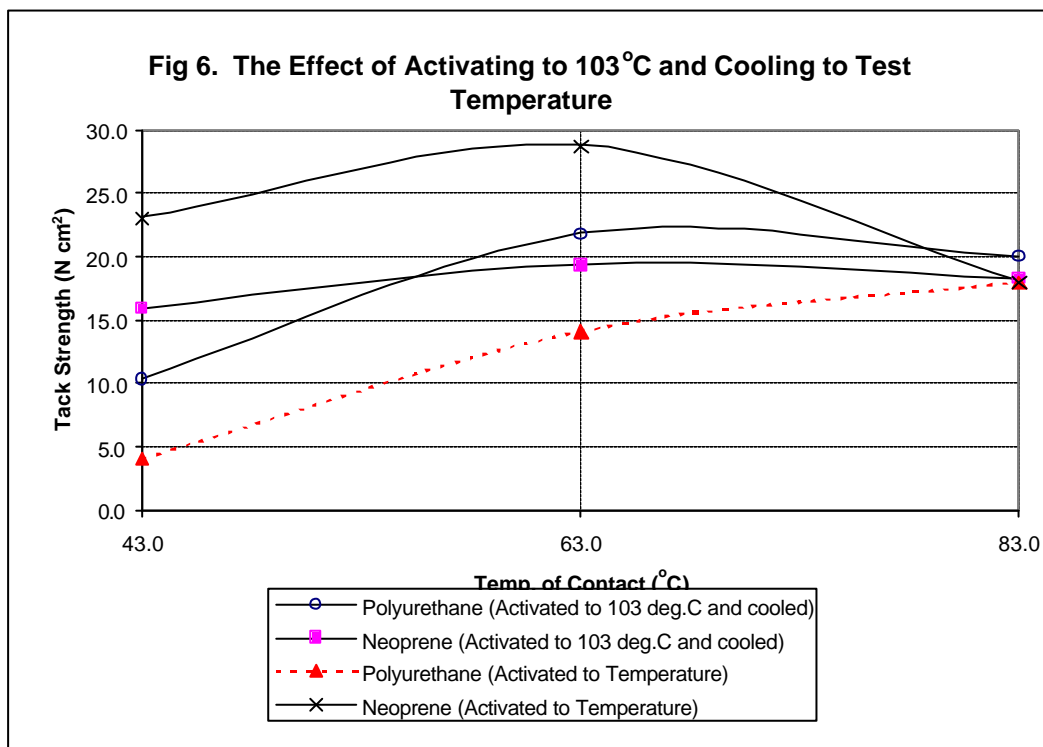
Tack strength was measured at a range of temperatures after heating to 103°C and allowing to cool to chosen temperature for adhered contact. The temperatures chosen for contact were 43, 63, and 83°C , so a comparison to the results of 4.1.2a could be made. The comparison is illustrated graphically in Fig. 6 and summarised as follows:

Temperature °C	Tack Strength N cm^{-2}			
	Polyurethane		Neoprene	
	4.1.2 a)	4.1.2 b)	4.1.2 a)	4.1.2 b)
43	4.0 (± 1.2)	10.3 (± 1.8)	23.0 (± 8.3)	15.9 (± 5.4)
63	14.1 (± 2.4)	21.7 (± 1.5)	28.7 (± 4.3)	19.3 (± 3.3)
83	18.0 (± 1.8)	19.9 (± 1.8)	18.0 (± 1.0)	18.2 (± 1.0)

It can be seen from the results, that different adhesives show different effects when heated and allowed to cool to the contact temperature. Polyurethane adhesive has an increased tack strength at all the temperatures tested, especially at 43 and

63°C. It is noteworthy that the best result for tack strength of polyurethane is given by heating the sample to 103°C and allowing to cool to 63°C. This may be due to the increased temperature below the surface of the adhesive when it has been heated to the increased temperature. The higher temperature allows the adhesive to flow more which gives a more uniform coverage before the pressure is applied. Where the actual activation temperature is lower, initially the adhesive will be more viscous and, hence, poorer at wetting the surface.

Neoprene adhesive however only shows a slight increase when heated to 103°C and cooled to 83°C. When the Neoprene is heated to 103°C and cooled to 43 or 63°C, there is a significant reduction in the tack strength observed. This may be due to an alteration in the crystalline structure of the adhesive formed on cooling from these temperatures. This shows that the tack test value is not simply a function of surface temperature, but is affected by the immediate thermal history.



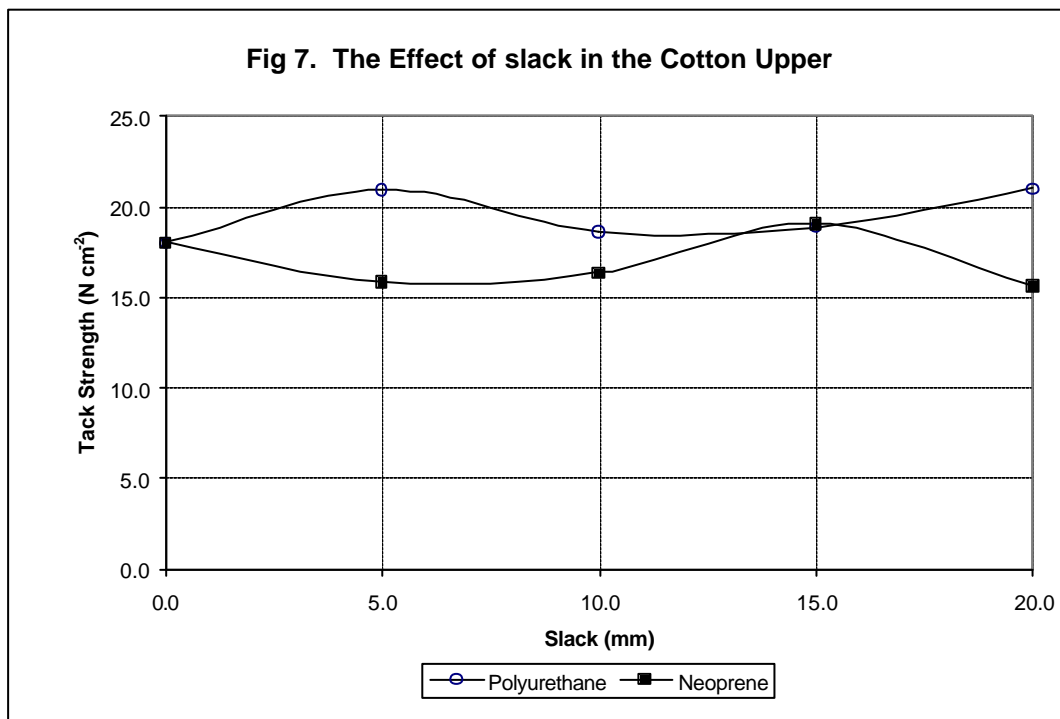
3.2 THE INFLUENCE OF COMPLIANCE OF SUBSTRATE MATERIALS

3.2.1 Assessment of the influence of upper compliance

An assessment of the influence of the upper compliance on tack measurements was made by allowing varying levels of slack in the upper cotton fabric. The normal test practice is zero slack in the upper. The results are shown graphically in Fig. 7 and are summarised in the table below:

Slack in Cotton Fabric (mm)	Tack Strength N cm ⁻²	
	Polyurethane	Neoprene
0	18.0 (± 1.8)	18.0 (± 1.0)
5	20.9 (± 4.5)	15.8 (± 3.3)
10	18.6 (± 2.2)	16.3 (± 1.3)
15	18.8 (± 2.8)	19.1 (± 3.5)
20	21.0 (± 2.5)	15.6 (± 3.0)

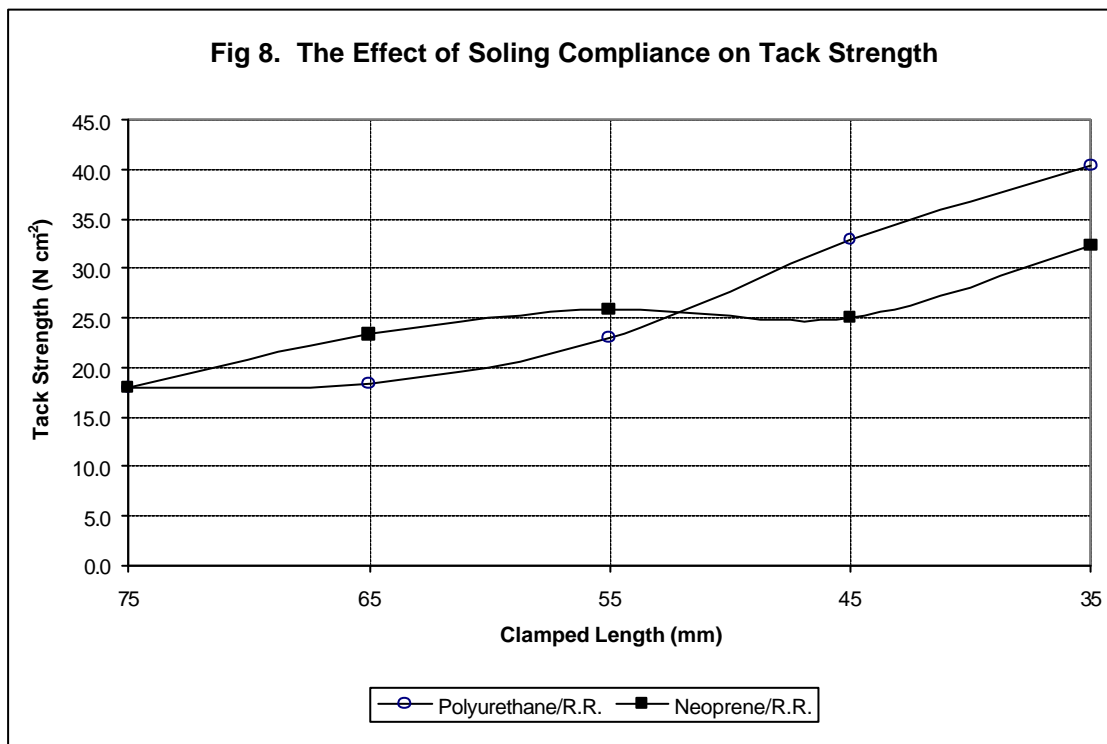
Any differences between the tack strengths for both adhesives are probably insignificant in comparison to the scatter in the data. Thus though slack in the upper fabric may effect the peel angle at the end of the bond, this factor seems to have little influence.



3.2.2 Assessment of the influence of soling compliance

To assess the influence of soling compliance, adjustments were made to the clamping positions of the sole material. Originally, only three positions were possible for testing (the normal setting of 75 mm clamp separation, plus 65 mm and 55 mm). After completion of the initial trials an adjustment was made to the upper clamp so that an extra two positions could be measured without the two clamps connecting. The results of these trials are shown graphically in Fig. 8 and summarised in the following table:

Clamped length (mm)	Tack Strength N cm ⁻²	
	Polyurethane/resin rubber	Neoprene/resin rubber
75	18.0 (± 1.8)	18.0 (± 1.0)
65	18.4 (± 3.2)	23.3 (± 2.1)
55	23.0 (± 2.1)	25.9 (± 3.1)
45	32.9 (± 2.1)	25.0 (± 2.1)
35	40.4 (± 7.7)	32.3 (± 3.3)



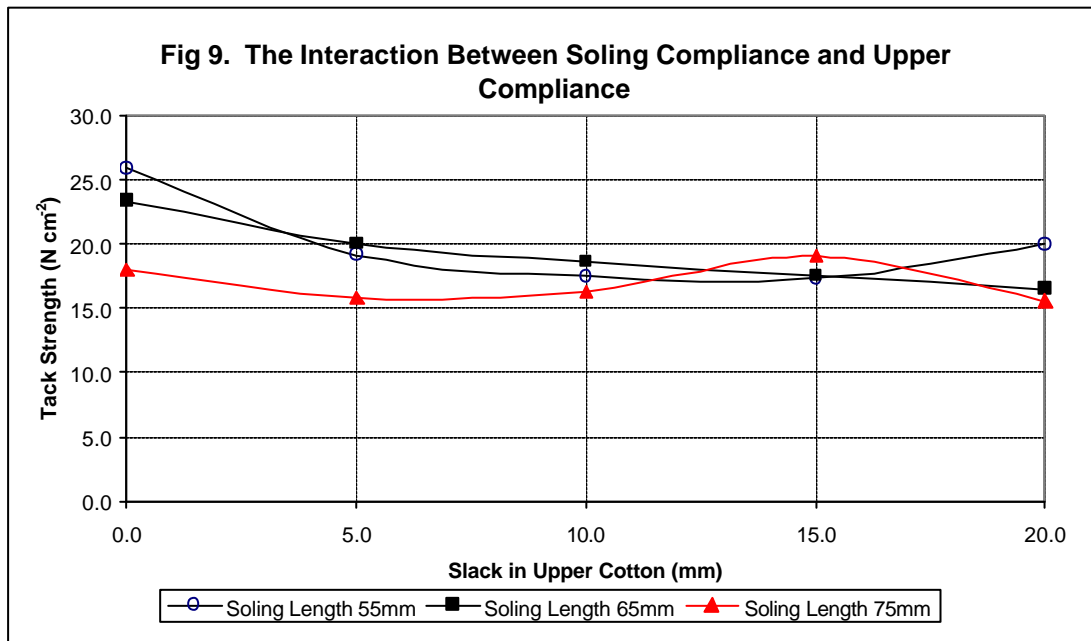
It can be seen from the results of the adhesives on resin rubber that as the clamps get closer together, reducing the compliance, the tack strength increases. This is shown clearly with the polyurethane adhesive, where the tack strength increased each time the clamps were moved closer together.

3.2.3 Investigation into the interaction of the two compliances

To investigate the interaction between the soling compliance and the upper compliance, a number of clamp positions and differing levels of slack in the cotton fabric were investigated. The results are all obtained using Neoprene adhesive on resin rubber soling and cotton upper. These can be seen graphically in Fig. 9 and are summarised in the table below:

As shown in previous experiments, there is a trend for the tack strength to increase as the clamps are moved closer together. There is also a slight trend for decreasing tack strength as more slack is introduced into the cotton that becomes more apparent as the soling compliance is reduced.

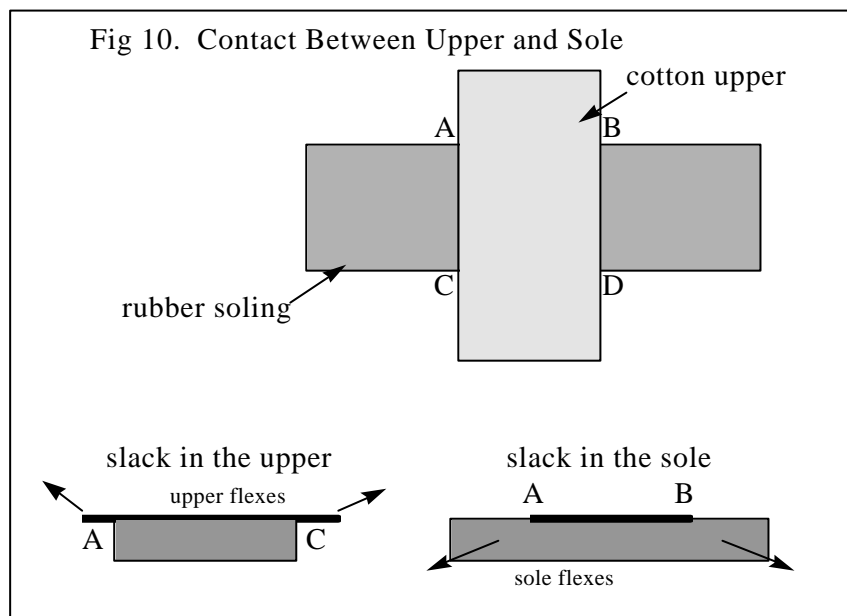
Amount of Slack in Cotton Fabric (mm)	Tack Strength N cm ⁻²		
	Clamped Length (mm)		
	55	65	75
0	25.9 (± 3.1)	23.3 (± 2.1)	18.0 (± 1.0)
5	19.2 (± 4.1)	20.0 (± 3.7)	15.8 (± 3.3)
10	17.4 (± 1.3)	18.6 (± 1.5)	16.3 (± 1.3)
15	17.3 (± 2.5)	17.5 (± 1.3)	19.1 (± 3.5)
20	19.9 (± 2.9)	16.5 (± 1.2)	15.6 (± 3.0)



The largest differences in tack strength can be seen between the measurements for 0 mm and 5 mm of slack in the cotton. This difference in the tack is thought to be due to a change in the peel angle as the slack in the cotton is increased. The effect is not large as, even at zero slack, the compliance of the cotton is sufficient to cause a non-zero peel angle at the ends of the cotton. With increasing

slack, there is more bending of the cotton upper. Therefore, there will be stress concentrations at the ends of the bond (along the lines AB and CD in Fig. 10). Thus, the ends disbond first leaving a progressively smaller bonded area less able to withstand the pull. Increasing the slack beyond 10 mm has little effect on the measured tack presumably because the peel angle changes little after this.

The main factor influencing these results is the soling clamp position. As the clamps are moved further apart, the resin rubber gains flexibility. It flexes upwards as the upper cotton is pulled away. This causes a stress concentration in the adhesive at the edges of the bond (AC and BD). This allows a peeling effect to take place instead of the preferred clean break over the whole contact area. This reduces the measured tack strength. When the clamps restrict the movement of the relatively rigid sole, the peel stress along AC and BD is removed. However, since the upper has a large compliance, the peel along AB and CD can never be eliminated.



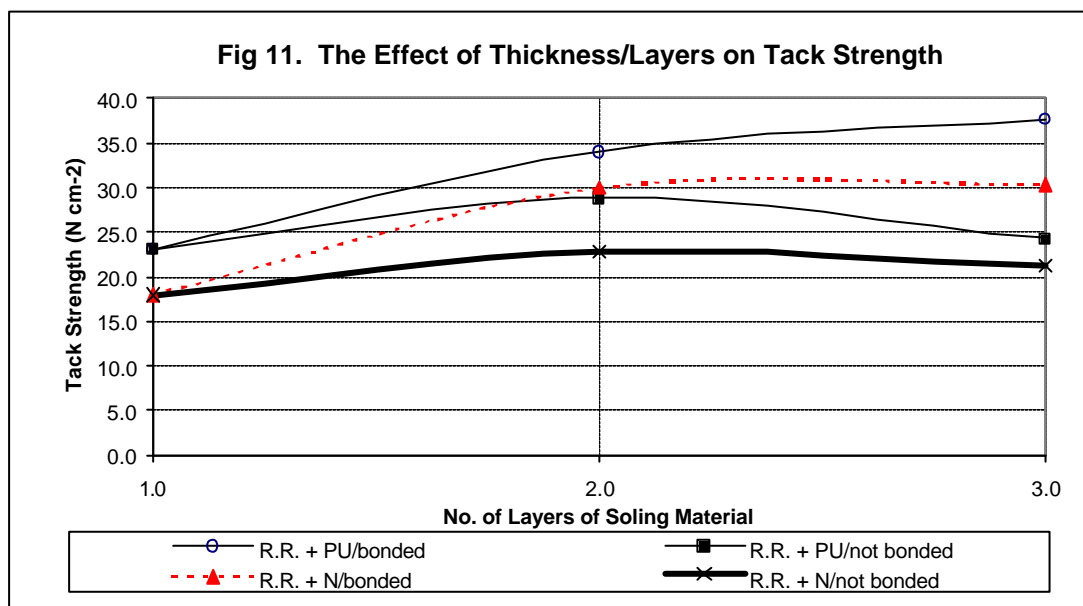
Where there is compliance in the measurement substrates, the stress will not be evenly distributed throughout the contact area. An increased proportion of the separation force will act on the extremes of the contact area. Depending on the compliances of the substrates, this peeling action can take place along just two or all four edges of the rectangular contact. This is a common feature in tack tests, such as the loop tack test for adhesive tapes, where the mechanical properties of the substrate influence the measured tack values.

3.2.4 Assessment of the influence of substrate compressibility and thickness on tack strength

A range of thickness of both PVC and resin rubber was investigated by building up the number of layers of the soling materials. To separate the influences of

compressibility and elastic properties, the tack strength was measured when the layers were bonded together and also without bonding. The results of these experiments are summarised in the table below and the results for the experiments with resin rubber can be seen graphically in Fig 11.

Number of Layers	Tack Strength N cm ⁻²					
	R.R.+C, Polyurethane		R.R.+C, Neoprene		P.V.C.+C, Polyurethane	
	Bonded	Unbonded	Bonded	Unbonded	Bonded	Unbonded
1	22.9 (± 5.2)	22.9 (± 5.2)	18.0 (± 1.0)	18.0 (± 1.0)	17.3 (± 2.7)	17.3 (± 2.7)
2	33.9 (± 4.2)	28.8 (± 1.7)	29.9 (± 3.0)	22.8 (± 1.9)	22.1 (± 2.8)	22.1 (± 1.9)
3	37.6 (± 5.1)	24.2 (± 3.9)	30.3 (± 2.0)	21.2 (± 1.5)	21.2 (± 1.7)	19.8 (± 1.45)

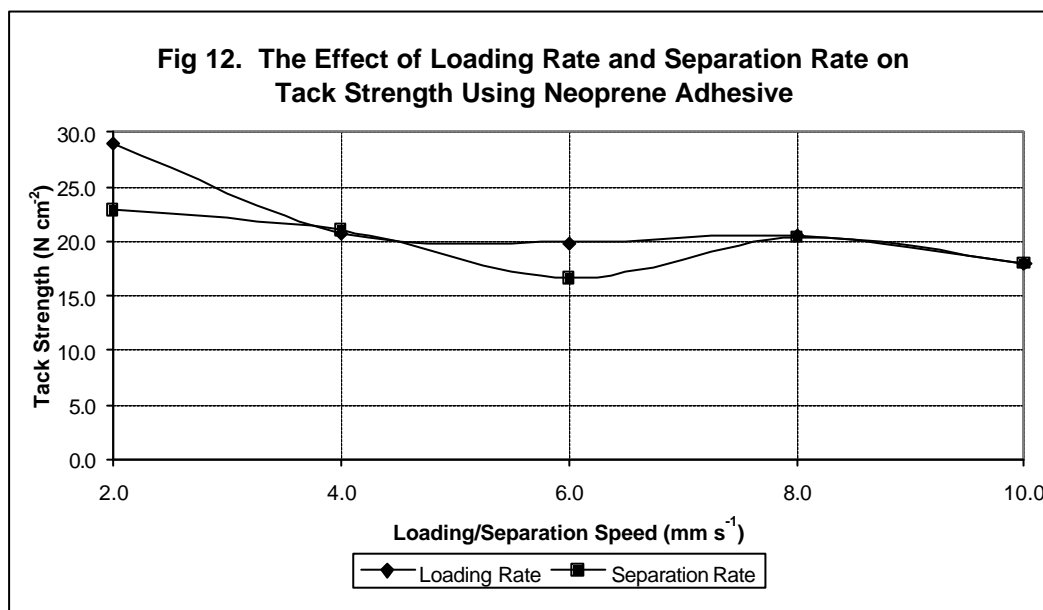


The results from these experiments show that the tack strength increases when more than one layer of resin rubber soling material is used. When the layers of soling material are bonded together, the thickness of the lower substrate and, hence, its rigidity increases. This prevents the upper layer from rising in the tension phase of the tack test which reduces the stress concentration along the bond edges. Significantly higher tack strengths are obtainable for the polyurethane and Neoprene adhesives. The shapes of the curves for the two adhesives are similar, with a relatively large increase between one and two layers, and then a levelling off or slight decrease. The slight decrease when built up to three layers may be due to compression within the sample, which forces the centre of the sample upwards when the clamps are being tightened at both ends. This upward

bow changes the contact made, giving a smaller test area and a non-uniform thickness of the adhesive bond. There will also be effects due to the increased angle of peel. These lead to a lower tack force.

Where more than one layer was used but not bonded together, the increase in measured tack is not as significant. As the layers are not bonded, the top layer is still able to flex as the upper is pulled off and the peel stress concentrations along the edges will still occur.

The same trends would be expected using standard PVC soling material. However, due to the relative softness of the standard PVC used, the results obtained were more variable and further testing is needed for more confidence in the results.



3.

3 THE INFLUENCE OF TEST CONDITIONS

3.3.1 Loading rate and contact time (incorporating separation rate)

To investigate the effect of adjusting the loading (downward) and separation (upward) rate, a range of speeds was selected on the test equipment. The resin rubber and Neoprene adhesive were tested. The results for loading rate and separation rate can be seen graphically in Fig. 12 and are summarised in the table below:

Speed Setting (mm s ⁻¹)	Tack Strength N cm ⁻²	
	Adjusted Loading Rate (Separation rate = 10 mm s ⁻¹)	Adjusted Separation Rate (Loading rate = 10 mm s ⁻¹)
2	29.0 (± 1.6)	22.8 (± 0.3)
4	20.7 (± 2.5)	20.9 (± 1.5)
6	19.9 (± 1.1)	16.6 (± 1.2)
8	20.5 (± 1.5)	20.3 (± 0.5)
10	18.0 (± 1.0)	18.0 (± 1.0)

In both the loading rate and the separation rate the same trend is visible. At the lowest speed the measured tack strength is the highest. As the speed increases there is a decline in the tack strength. This is most significant between the loading speeds of 2 and 4 mm s⁻¹, where it is thought that the differences in tack strength are due almost entirely to the increased contact time. As the contact time is significantly longer at the speed setting of 2 mm s⁻¹ the adhesive will have more time to spread. However, the measurements obtained may not be strictly measurements of initial tack strength.

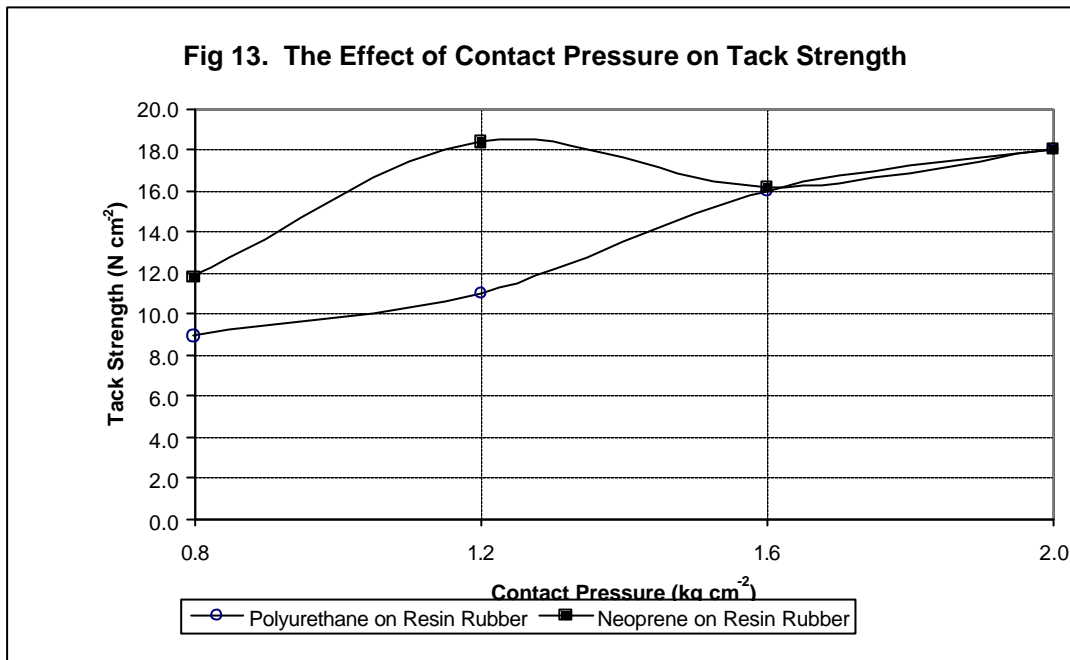
3.3.2 Separation rate

See section 3.3.1 above for results. The influence of separation rate appears less significant than the that of the loading rate. The range of rates used is probably too small to have much measurable effect on the visco-elastic properties of the adhesive.

3.3.3 Contact Pressure

To investigate the effect of adjusting the load per unit area of contact, a range of contact pressures was investigated from 0.8 kg cm⁻² to 2.0 kg cm⁻² at regular intervals. Unfortunately the equipment was unable to measure pressures much lower than 0.8 kg cm⁻² with this adherend combination due to the overshoot when working at full speed. The results are given graphically in Fig. 10 and are summarised in the table below:

Contact Pressure (kg cm ⁻²)	Tack Strength N cm ⁻²	
	R.R. + Polyurethane	R.R. + Neoprene
0.8	8.9 (± 1.5)	11.8 (± 1.6)
1.2	11.0 (± 1.6)	18.4 (± 1.4)
1.6	16.0 (± 2.9)	16.2 (± 2.1)
2.0	18.0 (± 1.8)	18.0 (± 1.0)



It can be seen from the results that, for the polyurethane adhesive, there is a trend towards increased tack strength with increased contact pressure as would be expected. This trend is less clear with the Neoprene adhesive. The initial increase in tack appears to level out (to within experimental error) after the contact pressure is increased above 1.2 kg cm⁻². The effect of increasing the contact pressure is to increase the spreading and wetting of the adhesive. Furthermore, an increased pressure is related to a longer contact time if the loading and separation speeds are kept constant. Where the adhesive flows easily, full spreading may be achieved with lower contact pressures thus a maximum tack force is seen. The Neoprene adhesive was shown in section 3.1 to give a good tack strength at lower activation temperatures. This suggests that it may have better flow properties than the polyurethane.

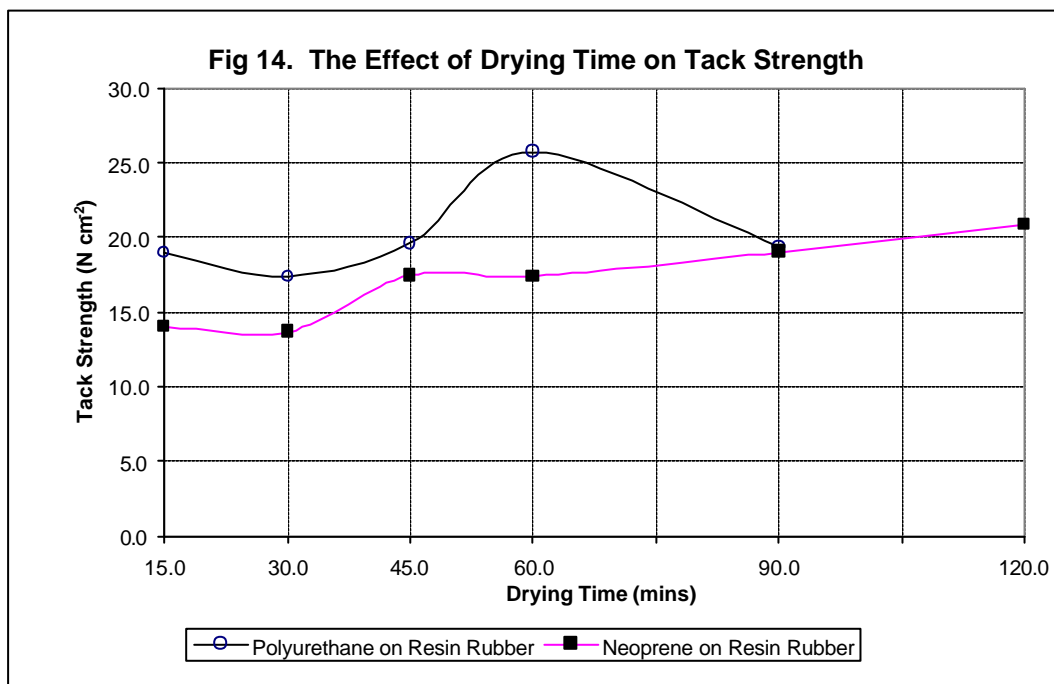
The 2.0 kg cm⁻² pressure is a fairly typical pressure used for spotting tack measurement in the footwear industry. There may be advantages to the tack strength in applying higher pressures to the polyurethane adhesive but these were not investigated here. At the standard pressure, the tack of the neoprene adhesive should be relatively insensitive to variations in pressure.

3.4 THE INFLUENCE OF PREPARATION

3.4.1 Drying times

The drying times of both the solvent based adhesives were accurately timed and tack tested at intervals up to 120 minutes. The results can be seen graphically in Fig. 14 and are summarised in the table below:

Drying Time (mins)	Tack Strength N cm ⁻²	
	R.R. + Polyurethane	R.R. + Neoprene
15	18.9 (± 4.4)	14.0 (± 4.0)
30	17.4 (± 7.2)	13.7 (± 2.4)
45	19.6 (± 3.1)	17.4 (± 2.4)
60	25.7 (± 4.5)	17.4 (± 2.4)
90	19.3 (± 4.4)	19.0 (± 3.0)
120	-	20.9 (± 1.2)



Due to the variability of the early results, the samples tested with a drying time of sixty minutes or less were an average of eight results rather than the usual four. It is thought that the 15 minute result is very variable, as neither of the adhesives are dry at this stage. The polyurethane results show an increase in tack strength as the drying time is increased, up to and including the 60 minute result. Following further testing, it was confirmed that the tack strength of polyurethane adhesive dropped off after 60 minutes. It was also found that the short timed results gave a similar higher reading for tack at 15 minutes in comparison to that at 30

minutes. However, the large scatter in the data prevents definite conclusions from being drawn.

An explanation for the effect of drying time on measured tack follows. When the film is activated, residual solvent is driven off and the temperature of the film depends on the heat used up as latent heat. At short drying times, some but not all solvent has evaporated. This leaves enough to ensure a tacky surface even though the temperature is not as high as normal. At slightly longer drying times, the heat drives off the remaining solvent. The temperature is still reduced, and the tack effect of the solvent is no longer present, so the tack strength drops. After further drying time, more solvent is evaporated. Therefore, there is less solvent left to evaporate and reduce the temperature on activation. Thus, the film temperature will be higher and, consequently, the tack strength. The fall in tack strength at longer times may occur because all the solvent has been lost prior to activation. The Neoprene adhesive shows continuously increasing tack strength up to two hours, suggesting that the solvent used evaporates less readily.

3.4.2 Coating Weight

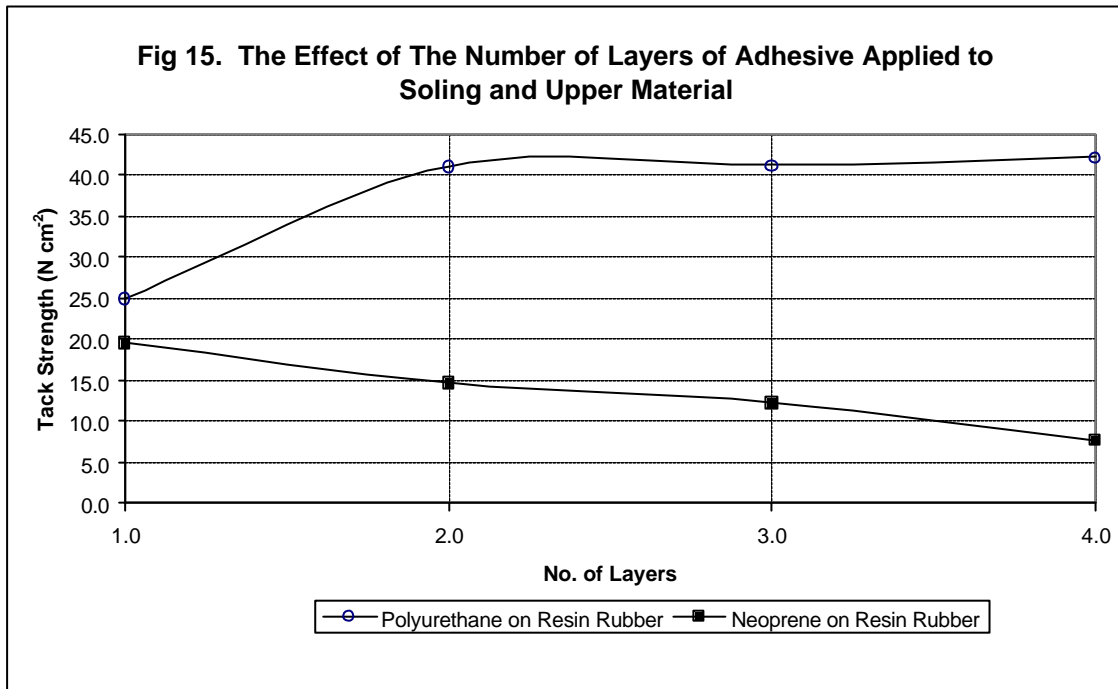
To investigate the effect of adhesive loading, to achieve a range of dry film thickness, a differing number of layers of adhesive were applied to the upper and soling materials at one hour intervals. The results can be seen graphically in Fig. 15 and are summarised in the table below:

No. of Layers of Adhesive Applied	Tack Strength N cm ⁻²	
	R.R. + Polyurethane	R.R. + Neoprene
1	24.8 (± 3.8)	19.4 (± 2.6)
2	40.9 (± 1.8)	14.6 (± 1.5)
3	41.0 (± 3.4)	12.2 (± 3.2)
4	42.1 (± 1.6)	7.7 (± 1.7)

The results obtained for the Neoprene adhesive may not be representative. The layers were applied at 60 minute intervals. Most of these tests were carried out with one to two hours drying time after application of the final layer. However, as shown earlier, the Neoprene adhesive takes much longer to dry. Thus the new layers are applied onto 'wet' adhesive, further slowing the drying of the bottom layer. Therefore, when the contact is made at a higher number of layers, the separation is very easy as the adhesive still shows fluid properties. A more reliable investigation of the influence of multi-layers of the Neoprene adhesive would need longer times between layer applications.

The resin rubber and polyurethane adhesive give more interesting results. With most of the solvent content being removed within 15 - 20 minutes, the layers of polyurethane adhesive were much easier to apply. The results show that when two layers of the polyurethane adhesive were applied, the tack strength increases from 24.8 to 40.9 N cm⁻². This is a relatively large increase that may be explained by the

greater amount of adhesive available for bonding on the upper material. When only one layer of polyurethane adhesive is applied to the upper cotton fabric, it has a tendency to soak into the body of the fabric. However, when another layer is applied to the cotton, it tends to bond more to the surface of the fabric and the previously applied adhesive rather than soaking into the body of the fabric. This makes a much better surface layer of adhesive available to bond with the soling material. More layers of adhesive seem to make no significant difference to the results obtained at two layers. Future work should investigate the effect of using only one layer of adhesive on the soling material but two layers on the upper cotton.



4. Findings on Improving Tack Strength

In order to obtain the higher tack values, various methods can be employed to increase the tack strength of an adhesive bond. Adherend properties have an influence on the measured tack and the effectiveness of the bonding methods will depend on the type of adhesive. The most effective of these methods are at the top of the list. These are then followed by lesser effects. However, any effect of more than 4.5 N cm^{-2} (roughly 25 % of the standard measured tack) is of interest for better bonding. To give the highest tack strength, a combination of the most positive results should be used according to adhesive and adherend types. It should be noted that not all these techniques can be readily introduced into a manufacturing environment.

- i The use of closer soling clamps to reduce compliance gave the largest improvement in tack strength. There is a gain of 22.5 N cm^{-2} (125 %) with polyurethane adhesive and 14 N cm^{-2} (80 %) with Neoprene adhesive, on reducing the distance between the clamps from 75mm to 35mm. This suggests that more clamping would lead to better bonding. However, this may not be cost effective on a production line.
- ii Another useful technique was using more than one application of polyurethane adhesive. It is thought that this is particularly important on absorbent materials. The use of two applications of polyurethane adhesive gave an increase in tack strength of 16 N cm^{-2} (65 %) although not much improvement was found by increasing the layers further. Using multiple layers of adhesive is likely to help solve some bonding problems but the greater use of materials and need for more preparation will increase costs. However, where there is insufficient drying time between layer applications, tack strength may be seriously compromised.
- iii Increases in tack strength can be obtained by control of the contact temperature. For example between 43°C and 83°C , the tack strength of the polyurethane adhesive increased by 14 N cm^{-2} (350 %). However, it must be noted that the tack at 43°C was extremely poor. Between 43°C and 63°C , the tack strength of the Neoprene adhesive increased by 6 N cm^{-2} (25 %). Improving control of the surface temperature during bonding is likely to lead to better bond formation during production.
- iv Substrate flexibility is also important. It was found that increasing the thickness of standard resin rubber from 3 to 6 mm increased the tack by 11 N cm^{-2} (50 %) with polyurethane adhesive and 12 N cm^{-2} (66 %) with Neoprene. A further increase in thickness to 9 mm gave another small increase in tack strength of 4 N cm^{-2} for the polyurethane adhesive. A similar but smaller effect was seen when more flexible PVC substrates and

polyurethane adhesive were used. When the thickness was increased from 3 to 6 mm, an increase in tack of 5 N cm^{-2} (30 %) was observed.

- v When loading and separation speeds were set at the slowest speed setting of 2 mm s^{-1} , an increase in tack was observed. This was 11 N cm^{-2} (60 %) with the slow loading rate, and 5 N cm^{-2} (25 %) with the slow separation rate (using Neoprene adhesive). This difference in measured tack strength is probably due to the increased time of contact at the higher pressure rather than a real effect of slower speeds.
- vi Drying time allowed for the adhesives is an important factor and should be controlled for maximum tack. This is particularly important with the polyurethane adhesive that shows marked increases between 45 and 60 minutes of 6 N cm^{-2} (30 %). A decrease then follows between 60 and 90 minutes of 7 N cm^{-2} (30 %). This demonstrates the importance of good control over the drying and open time of polyurethane adhesives. Neoprene adhesives continued to increase in tack strength over two hours. There is a relatively consistent increase in tack from 15 minutes to 120 minutes of about 7 N cm^{-2} (50 %).
- vii The effect of contact pressure is noticeable with both the polyurethane and the Neoprene adhesive. With polyurethane the tack gain is 9 N cm^{-2} (100 %) when the contact pressure is increased from 0.8 to 2.0 kg cm^{-2} . With Neoprene adhesive the tack increase is 6 N cm^{-2} (50 %). Using low contact pressures may have deleterious effects on bond integrity during manufacture.
- viii By activating the adhesive to a higher temperature (in this case 103°C), and allowing the sample to cool to the relevant temperature, differences were seen between the tack strengths of the two adhesives compared with the tack strength produced when the adhesives were heated to the contact temperature. With polyurethane adhesive, the tack strength at a contact temperature of 63°C showed an increase of 8 kg cm^{-2} (55 %) when the activation temperature is increased and the sample left to cool. Neoprene however shows the opposite effect at 43°C and 63°C . Decreases in tack strength of 7 or 8 N cm^{-2} (30 %) were recorded.

5. Conclusions

i TACK IS A COMPLEX PROPERTY DEPENDING ON A WIDE RANGE OF INFLUENCES

All of the variables studied - temperature, pressure, adherend compliance, adhesive coating thickness and state of dryness - have an effect on the measured tack force. The magnitude and significance of these factors vary between the two adhesive types considered, polychloroprene (Neoprene) and polyurethane. The SATRA/NPL tack tester has proved to be eminently suitable as a research tool for investigating the variables influencing tack, as well as a means of measuring tack for adhesive development and process control purposes.

ii TACK STRENGTH IS INFLUENCED BY THE THERMAL HISTORY OF THE ADHERENDS AND ADHESIVE

In a series of tests at intervals of 20°C, the highest level of tack strength with the polyurethane adhesive was achieved at an activation temperature of 83°C. This is the temperature normally recommended for solvent based adhesives in footwear manufacture. With the solvent based Neoprene (polychloroprene adhesive), the highest level was achieved at 63°C. The temperature/tack strength profiles with these adhesives suggest that increasing the temperature produces competing effects, for example, enhanced flow characteristics against reduced tensile strength for the adhesive.

The tack strength of the polyurethane adhesive increased when activated to a higher temperature and allowed to cool to an appropriate temperature before contact. Conversely, the tack strength of the Neoprene reduced for temperatures of 63 and 83°C when using this method. There is no real benefit, in terms of tack strength, in over heating Neoprene adhesives, but there is with polyurethane. However these results do not show if there is any effect on the internal structure of the adhesives concerned, and consequent strengthening or weakening of the final bond.

iii RESTRICTING MOVEMENT OF THE ADHERENDS HAS A SIGNIFICANT EFFECT ON THE MEASURED TACK STRENGTH

Moving the sole clamps closer together decreased the movement in the sole material. Consequently, the tack strength increased. However, allowing greater slack in the upper material only became influential as the soling became more constrained. Even then the effect was relatively minor. The highest values for tack were obtained when both the soling and upper materials are at their most constrained positions.

iv THE COMPLIANCE OF THE ADHERENDS HAS A SIGNIFICANT EFFECT ON THE TACK STRENGTH MEASUREMENT

Plying together a number of layers to increase the thickness of the soling material gives increased tack strength when the layers are bonded together. The effect was more marked with the stiffest material. An upward trend was also apparent when the layers were plied but not bonded together, however this was not as significant as when they were bonded. This suggests that the 'tensile compliance' of the more rigid adherend is of greater significance than its 'compressive compliance'.

v TACK STRENGTH IS CONTACT PRESSURE AND TEST SPEED DEPENDENT

Tack strength increases as contact pressure increases. This was particularly so with the polyurethane adhesive where a noticeable increase was observed in tack strength at each measurement level between 0.8 and 2.0 kg cm⁻². The behaviour of the Neoprene adhesive differed. There was a significant increase only at the lower pressures and a level tack strength at contact pressures of over 1.2 kg cm⁻². This would indicate that the polyurethane adhesive is very pressure dependent for tack strength. Above a certain minimum pressure, the tack strength of the Neoprene adhesive would appear to be independent of contact pressure. No further gains in tack strength can be achieved purely by increasing the contact pressure.

Increased test loading and separation rates tended to decrease the tack values measured. However, when the speeds are lower, the measured value may no longer be an accurate representation of **initial** tack strength, in view of the extended contact times of the adhesive interfaces.

vi DRYING TIME AND COATING WEIGHT MAY SIGNIFICANTLY AFFECT THE TACK VALUE OBTAINED

The tack strength of the Neoprene adhesive increased with increasing drying time up to 120 minutes, the longest time examined. The tack strength of polyurethane adhesive increased with up to one hour drying but fell off after this.

Undoubtedly the behaviour of the two adhesives has been influenced by a number of factors:

- the particular solvent blends used as carriers - that of the Neoprene was markedly slower to evaporate than that of the polyurethane;
- by the interaction between residual solvent and polymer solids, e.g. retained solvent increasing 'tackiness' of the film but reducing its tensile strength;
- and the impact of solvent lowering the temperature achieved in activation through absorption of latent heat of evaporation.

Tack strength of polyurethane adhesive increases with the number of layers of adhesive (coating weight) applied. Above two layers, the differences are relatively small. At this point, the surface is completely filled, after which there are no real advantages of adding more adhesive. It is likely that an equally strong tack strength could be achieved by applying only one layer of adhesive to the soling and

two layers to the absorbent upper fabric. The amount of adhesive on the cotton surface is thought to be the controlling factor in this combination.

Tack strength of Neoprene adhesive decreased in these experiments as the number of layers of adhesive increased. This reflects the low drying rate of the solvent blend of this adhesive. As the number of layers increases, the amount of solvent remaining in the adhesive layers also increases, and the tack therefore drops due to insufficient drying.

6.

Acknowledgements

The support of the DTI in funding this work through the Performance of Adhesive Joints Programme is gratefully acknowledged. Thanks are due to Abayomi Olusanya, Lewis Lay and Bruce Duncan of NPL and Richard Roberts of PIRA International for their guidance and helpful suggestions.