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Assessment of the Correlation Between Tack and Visco-Elasticity

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Performance of Adhesive Joints
Project PAJ1: Failure Criteria and their Application to Visco-Elastic/Visco-Plastic Materials

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Summary

Tack is the property of an adhesive that enables it to form a bond with a surface on immediate contact under light pressure. Tack performance is thought to be related to the visco-elastic properties of the polymeric adhesive. Hence, there is the possibility of using visco-elastic characteristics to predict tack in different bonding applications. Dynamic measurement methods, such as oscillatory shear, have been used to characterise the visco-elastic properties of double sided pressure sensitive adhesive tapes. There are some correlations between these properties and the tack performance of the tapes. However, the correlations break down under certain conditions. Tack strength may be controlled by the tensile extension and cohesive rupture of fibril structures. These are not characterised by these dynamic techniques but could possibly be studied by extensional rheological techniques.

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1. INTRODUCTION

Tack is the property of an adhesive that enables it to form a bond with a surface on immediate contact under light pressure. Hence, tack is important where the joint is required to sustain loads as soon as it is made. Tack performance is thought to be related to the visco-elastic properties of the polymeric adhesive[1]. Visco-elasticity characterises the material properties in terms of viscous properties and elastic properties. The viscous (or flow) properties enable the adhesive to spread over the surface and form intimate contact. The elastic properties give the bond strength. These properties are rate and temperature dependent. This report presents findings of research aimed at assessing the relationship between visco-elastic properties and tack.

Tack can be measured using many different methods[2]. However, the results produced by different techniques are not always comparable[3]. It is thought that some of these differences may be due to the visco-elastic response of the adhesive since dwell times and separation rates vary between test methods. Visco-elastic properties can be characterised through dynamic or oscillatory techniques[4]. Rheological techniques have been shown to have applications in characterising low modulus solids[4]. This report describes the application of these techniques to pressure sensitive adhesive tapes.

Visco-elastic characterisation of the adhesive could supply a method for correlating tack between different measurement techniques or bonding applications. Ultimately, it may be possible to predict tack in a bonding process simply from the visco-elastic properties of the adhesive and substrates. As part of a study of tack measurement performed in Project PAJ1 of the DTI Performance of Adhesive Joints programme, relationships between tack properties and visco-elastic characteristics have been investigated.

2. EXPERIMENTAL

2.1 MATERIALS

In this study a number of double sided tapes have been investigated. Double sided tapes can be built into multi-layers for dynamic mechanical thermal analysis (DMTA) or bonded between the parallel plates of the rheometer. Thus, both tack and visco-elastic measurements can be made. The tapes used are listed in Table 1. The same general type of adhesive (acrylic) was used on all three tapes although it was not known whether the exact adhesive formulation was the same in each case. In addition to the double sided tapes, an uncured paste adhesive (PPG 3289Y5000) was studied.

Table 1: Adhesive Tapes

Tape Designation	Backing Material	Adhesive
Tesa 4965	Polyester	acrylic adhesive
Tesa 4970	PVC	acrylic adhesive
Tesa 51968	Polypropylene	acrylic adhesive

2.2 TACK MEASUREMENTS

Probe tack measurements[5] were performed using a commercial probe tack tester supplied by TMI, shown in Figure 1. The tapes being tested were attached to annular weights. Paste adhesives were coated onto a tape attached to the annular weight. The probe was raised to make contact with the adhesive. The contact pressure was controlled through using different annular weights. Contact was maintained for the set dwell time. Then the probe was separated from the surface at the set test speed. The tack was characterised by the maximum in the separation force that is recorded by the instrument.

The tack performance of pressure sensitive adhesives may depend on more than simply the maximum tack force. Zosel[6] has demonstrated that good tack performance is characterised by the formation of fibrils. These enable the bond to sustain large deformations before rupture and, hence, the failure energy is likely to be higher. A number of tack tests were performed to characterise the force-deflection behaviour of the samples. The output from the probe tack tester's force transducer was logged to a PC for further analysis.

Tests could be carried out using different test speeds, dwell times and contact pressures in order to characterise the tack properties of the adhesives. Temperature and humidity will also influence the results. However, control of these last two conditions was not available. Temperature and humidity measurements were made regularly during series of tests. Tests were carried out under approximately the same environmental conditions.

2.3 VISCO-ELASTIC PROPERTIES

2.3.1 Dynamic Mechanical Thermal Analysis (DMTA)

DMTA measurements were performed on a commercial instrument supplied by Polymer Laboratories. DMTA samples were prepared by laminating several layers of the double sided tape to form a bar approximately 4 mm thick (25 mm long by 10 mm wide). These samples were prepared to give stiffer test sample. The DMTA has relatively poor sensitivity when the sample stiffness is low[4]. The backing tape was removed from the test sample except at the clamped ends. The samples prepared from the tapes were extremely soft and 'barrelled' when the clamps were tightened. There was also some evidence that the samples buckled during tests that scanned large temperature ranges owing to thermal expansion. Both these cause problems with the measurements.

2.3.2 Shear Oscillation

Shear oscillation measurements were performed in a TA Instruments Carrimed CSL500 rheometer. This instrument and modifications made to it have been described elsewhere[4, 7]. Double sided tapes, trimmed to shape, were tested between 10 mm diameter parallel plates. The gap was set slightly smaller than the tape thickness

(typically 100 μm - 200 μm) to ensure good contact but avoid over-compressing the tape. Figure 2 shows that the shear modulus measured can depend on the compression of the tape. The paste adhesive was tested using the same plates. The gap was set to 500 μm.

Variable oscillation frequency and variable oscillation torque tests were carried out at 20 °C (comparable to the temperature during tack tests). Temperature in these tests was controlled using the Peltier plate system. Variable temperature tests (using the extended temperature module) were carried out at constant frequencies and oscillation amplitudes. In all cases, torque amplitude rather than displacement amplitude was used to control the instrument.

3. RESULTS

Tack data for the three double sided tapes and the PPGY5000 adhesive at different test speeds are shown in Figure 3. The tack of tapes 4965 and 4970 is similar. This probably reflects the similarities of the pressure sensitive adhesives. The tack of tape 51968 is larger at low separation speeds but lower at high separation speeds. These data show that tack is rate dependent. However, the rate dependence is not simple. From the lowest speeds, the tack increases with the separation speed. However, tack ceases to increase at higher separation speeds. The data even suggest that further increasing speed leads to lower tack results. This sensitivity to rate is also evident at different dwell times, DT (Figure 4) and contact pressures, P (Figure 5).

Dynamic moduli for the Tesa double sided tapes 4965, 4970 and 51968 measured by DMTA and rheometry are shown in Figures 6-8 respectively. The data were measured at constant frequencies whilst increasing the test temperature. Thermal expansion or contraction of the samples caused problems in both measurement techniques. In the rheometer, thermal contraction (and poor low temperature tack) resulted in poor adhesion between the parallel plates. Hence, some low temperature results were unreliable. In the DMTA, thermal expansion behaviour of the specimens is thought to impose off-axis forces to the drive shaft. This may be the cause of the ‘peaks’ in the elastic modulus, E’, seen in Figures 6 and 7. These problems sometimes prevented the DMTA from sweeping the entire temperature range. Additional experiments using a higher initial temperature were required to cover the range of interest (e.g. Figure 6).

The dynamic data in Figures 6-8 indicate that the shear moduli measured by the rheometer are between 2.5 and 4 orders of magnitude lower than the tensile moduli determined from DMTA. Earlier work has shown that the compliance of the rheometer can lead to significant under-estimates of the shear moduli of stiffer samples[4]. Compliance corrections can be applied to calculate the true moduli but these can be a major source of measurement uncertainty. However, compliance corrections are not thought to be significant here as the dynamic moduli are low (less than 1 MPa). Compliance corrections would be of the order of a few percent of the measured values and certainly insufficient to account for the differences.

The differences between the DMTA and rheometry data are more likely to be due to the physical nature of the adhesive tape specimens. Double sided adhesive tapes consist of a carrier tape coated on both sides with a pressure sensitive adhesive. The

carrier tape imparts tensile strength to the adhesive allowing it to be handled and applied. In DMTA experiments the double sided tape is flexed and the resistance to flexure ('stiffness' of the sample) is measured. The strain in different layers of the sample will be roughly the same (ignoring through thickness distributions). Therefore, DMTA tests will contain a stiffness contribution from the carrier tape that may be much more significant than the compliant adhesive coating. The DMTA data are similar in form to those obtained on solid polymer specimens. In shear, the deformations of the different layers are de-coupled. Therefore, the stiffness of the carrier tape will have less significance. The response will be influenced more by the adhesive coating. These differences between the techniques are analogous to the differences between electrical resistance in series (flexure) and parallel (shear) circuits.

Tensile or flexure techniques on the tape are not likely to characterise accurately the visco-elastic properties of the pressure sensitive adhesive that control the tackiness of the tape. However, they may be useful in measuring the stiffness of the whole tape (including backing where appropriate) that influences the results obtained in tack tests such as the loop tack test[3]. Shear measurement techniques are more likely to characterise the visco-elastic response of only the pressure sensitive adhesive.

The tack results indicate that tack is a rate sensitive phenomenon. Therefore, a series of frequency sweep experiments at constant temperature and oscillation torque was performed to investigate the rate dependence of the visco-elastic properties. The test temperature was 20 °C, close to the temperature during the tack tests. Rheometry data were only obtained at this temperature as tack results were not available at other temperatures. The test results showed that the moduli were relatively insensitive to oscillation torque between 100 μNm and 10000 μNm. Figure 9 shows frequency sweep results for the three tapes (torque = 1000 μNm) and the PPGY5000 adhesive (torque = 100 μNm).

The moduli for the tapes differ. The values of the elastic and viscous components of modulus in each tape are similar. The elastic and viscous modulus components for each tape cross-over at some characteristic frequency. This cross-over frequency is similar for all three tapes. This behaviour is also reflected in the temperature sweep measurements around room temperature (e.g. Figure 6). The point of cross-over of the elastic and viscous moduli is often used to characterise polymer melts. However, in the case of polymer melts, elastic characteristics are more significant at high rates whereas here the viscous characteristics become more important at higher rates. It is possible that the frequency (and hence strain rate) of this cross-over corresponds to the separation speeds where tack strength appears to decrease.

In comparison the dynamic moduli of PPGY5000 are approximately two orders of magnitude lower than the tapes. The viscous modulus dominates the behaviour of the PPGY5000 paste adhesive (Figure 9). This is reflected in the significantly lower tack measurements shown in Figure 3.

4. CORRELATION OF TACK AND VISCO-ELASTICITY

Fundamental to the tack strength is the adhesion between the adhesive layer and the substrates. This is determined by the surface chemistry of the adhesive, substrates and any contaminants. This is typically thought not to be influenced by visco-elastic properties. As an aside, it is known that interfacial tensions can be rate dependent. In surfactant solutions, for instance, the interfacial tension increases with the rate of formation of interface. This rate dependence is due to the diffusion of surface active species to the interface taking finite times. If interfaces are formed rapidly in relation to diffusion timescales then the surfactant concentration at the interface will be lower and the resulting interfacial tension higher. Similar effects may happen in adhesives when wetting timescales are comparable with diffusion rates or molecular re-arrangement timescales. However, in tack tests contact times are typically in excess of 1 s. It is likely that equilibrium conditions will have been reached at the interface. The effects of processing rates on interfacial adhesion were outside the scope of this programme. However, if adhesion strengths were sensitive to processing rate then this would be a further variable in the tack behaviour.

Although surface adhesion is fundamental to good tack strength, it is more likely that tack strength relates to cohesive failure of the adhesive. The cohesive strength of the polymeric adhesives may be linked to their visco-elastic properties. The dynamic elastic and viscous moduli of the pressure sensitive adhesives increase with oscillation frequency (proportional to shear strain rate). This is consistent with the increase in tack strength with separation speed seen at separation rates between 0.01 cm/s and 1 cm/s. However, this increase in tack does not continue as the separation speed is further increased. This is not consistent with the visco-elastic properties measured in shear.

Tape 51968 has the highest dynamic shear moduli of all the tapes, Figure 9. However, this tape shows the greatest tack at low separation speeds but the lowest tack strength at high separation speeds. High visco-elastic moduli may not necessarily lead to good tack strengths. For example, if the moduli are too high then the adhesive may not spread easily. Intimate contact with the surface, particularly one that is rough, may be patchy. As the contact and separation speeds of the probe tack tester are the same, the adhesive may not spread as easily at a high speed setting as at a low speed setting. Thus, the greater stiffness of the adhesive on separation may be counteracted by poorer surface contact and, potentially, a smaller bonded area resulting in a lower tack strength.

To investigate the rate dependence of tack further, a series of tests was performed with the output of the probe tack force transducer logged to a PC. This provided a time-dependent trace of separation force. Force-displacement plots could be determined from the separation speed. Typical force-displacement plots are shown in Figures 10 to 12. The stiffness of the system (K), calculated from the slopes of the rising portions of the curves, ignoring the initial transients, and typical displacements at rupture are shown in Table 2. It is noticeable that the stiffness increases with increasing speed but the displacement at rupture falls. This envelope of rupture displacement and stiffness

with rate begins to explain the failure of the tack strength to increase at higher separation speeds.

The displacements to rupture are typically in excess of twice the adhesive layer thickness (0.1 to 0.2 mm). Much of this displacement is likely to be deformation of the backing tape rather than extension of the adhesive. A check on the separation during the tests, using a displacement transducer, indicated that the separation speed when the probe tack tester is set to 5 cm/s is actually closer to 2.5 cm/s. This reduced separation speed makes it less likely that any increases in tack will be apparent on changing the test speed from 1 cm/s to 5 cm/s.

Table 2: Probe Tack Properties

	Tape 4965		**Tape 4970**		**Tape 51968**	
speed set (cm/s)	stiffness (g/mm)	rupture displacement (mm)	stiffness (g/mm)	rupture displacement (mm)	stiffness (g/mm)	rupture displacement (mm)
0.01	540	0.92	490	1.17	435	0.97
0.1	1170	0.75	1040	1.00	790	0.77
1.0	2400	0.46	1940	0.78	1860	0.60
5.0	2740	0.50	2100	0.52	2500	0.53

The rate dependence of the stiffness is shown in Figure 13. Nominal strain rates were calculated from the separation speed divided by the nominal tape thickness (0.2 mm). The actual test speed of 2.5 cm/s was used to determine the strain rate at the highest speed setting. Since the separation displacement also includes deformation of the tape these strain rates are likely to be overestimates. Elastic modulus (G') data determined using the rheometer are shown for comparison. Maximum shear strain rates (ω) were calculated from the oscillation frequency (f), oscillation amplitude (θ), plate radius (R, 5 mm) and gap (d, nominally 0.2 mm):

$$\dot{w} = 2pf\frac{Rq}{d}$$

The value of G' increases by a factor of ca. 10 per decade of strain rate whereas the value of K only increases by a factor of ca. 2 per decade. The stiffness rankings in the tack tests do not correlate with the rankings of the dynamic shear moduli. Tape 51968 which has the highest shear modulus shows the lowest stiffness (although the data suggest that it may have the highest stiffness at higher rates) and the lowest tack.

The cross-over of the elastic and viscous properties (Figure 9) described earlier may relate to the speed where tack strength ceases to increase. (Figure 3). The scatter in the tack data makes the accurate identification of a 'critical' test speed, where the tack is at a maximum, difficult. However, this is clearly close to 1 cm/s. The estimated strain rate at this speed is 50 s^{-1}. The crossover frequencies for tapes 4965, 4970 and 51968 were approximately 1.6 Hz, 3 Hz and 1.2 Hz respectively. These correspond to shear strain rates between 30 s^{-1} and 70 s^{-1}. The strain rates at the cross-over frequency and the 'critical' speed are of similar magnitudes.

Research, such as that undertaken by Zosel[6], indicates that the tack strengths of pressure sensitive adhesives are enhanced if the adhesive forms fibril (or bridging) structures during separation of the surfaces (Figure 14). These allow a relatively long-range interaction between the surfaces. The deformation of these fibrils is a tensile (or extensional) process. It is known that, in polymer melts, the extensional properties can differ considerably from the shear properties[8]. McKinley[9] has suggested that tack is determined by the visco-elastic properties of the polymeric adhesive in extension. Tackiness is, thus, controlled by the extensional growth and failure of the fibrils formed.

Time-temperature superposition can be used to relate visco-elastic modulus values at different rates and frequencies[10]. If the tack can be related to the visco-elastic properties then it may be possible to use these principles to predict tack at different rates and temperatures. However, the shear visco-elasticity measurements carried out do not correlate particularly well with the tack measurements. Thus, time-temperature superposition is unlikely to be accurate for these systems. Some tests performed on tapes that had been pre-cooled in a freezer to -18 °C showed that the low temperature tack was very poor. The tack performance improved as the sample warmed following removal from the freezer. Under time-temperature superposition, low temperatures correspond to higher rates. Qualitatively, this agrees with the trend towards reduction of tack strength (at room temperature) at the highest test speeds available from the probe tack tester. It should be noted that condensation of moisture on the cold surfaces may also have contributed to lower tack.

The properties measured in shear may not accurately predict the extensional behaviour of the adhesive. Extensional measurements on tacky polymer layers are likely to be difficult to perform. Extensional visco-elastic properties for polymer melts can be characterised through converging flow or uniaxial stretching methods[11]. Converging flow methods are not likely to be applicable. Uniaxial stretching methods have more promise but the small thickness of the material in tapes would cause many testing difficulties. Such methods could be used if the pressure sensitive adhesive was available in bulk form. Further research would be required to develop these methods and relate the results to tack performance.

5. CONCLUSIONS

Dynamic measurement methods can be used to determine the visco-elastic characteristics of tacky adhesives. Oscillatory shear methods are suitable for film, double-sided tape and paste adhesives. For tapes or films, there will be less contribution from the carrier tape than in tensile/flexural methods. Thus, shear methods give more information on the properties of the adhesive likely to be relevant to tack.

The correlation between the shear visco-elastic properties and the tack results appears relatively poor. However, the differences between the visco-elastic properties of the three tapes and the paste adhesive are reflected in the tack results. The tapes have significantly greater visco-elastic moduli and tack strengths than the paste adhesive. However, there is less discrimination between similar materials. The three tapes have

similar properties but the rankings of the visco-elastic and tack properties do not correlate well.

The phenomenon of tack does not simply depend on the low strain moduli of the adhesive but on the extension to failure. Extension of the adhesive may lead to the formation, growth and rupture of fibril structures. These are not characterised by the shear measurement. Extensional rheological measurements exist which would be more capable of characterising material properties relating to tack.

6. ACKNOWLEDGEMENTS

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Figure 1: TMI Probe Tack Tester

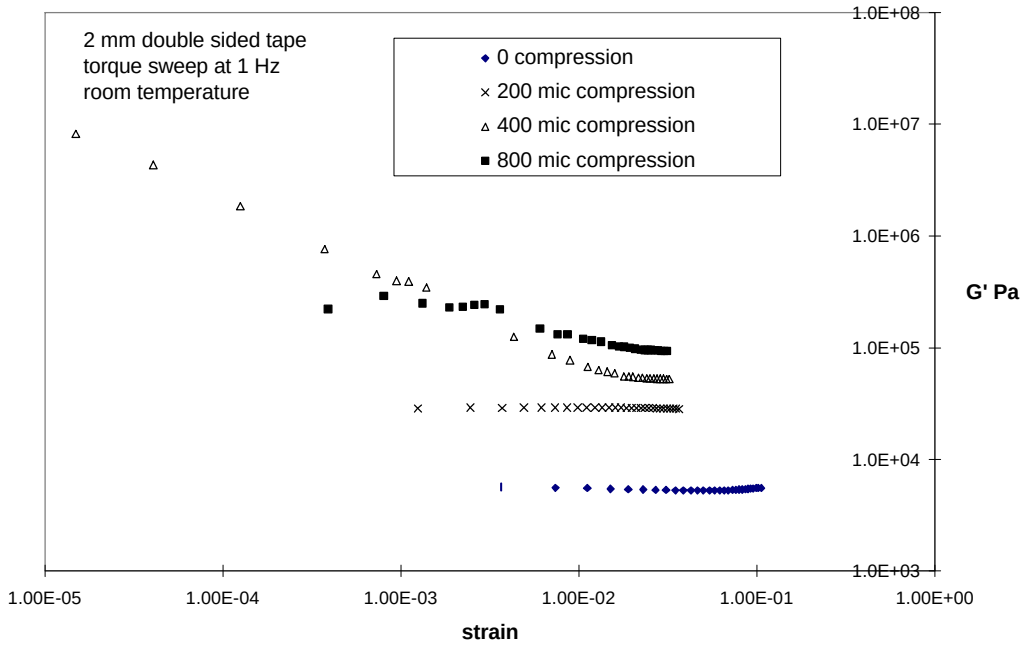

Figure 2: Effect of Tape Compression on Measured Shear Modulus G'

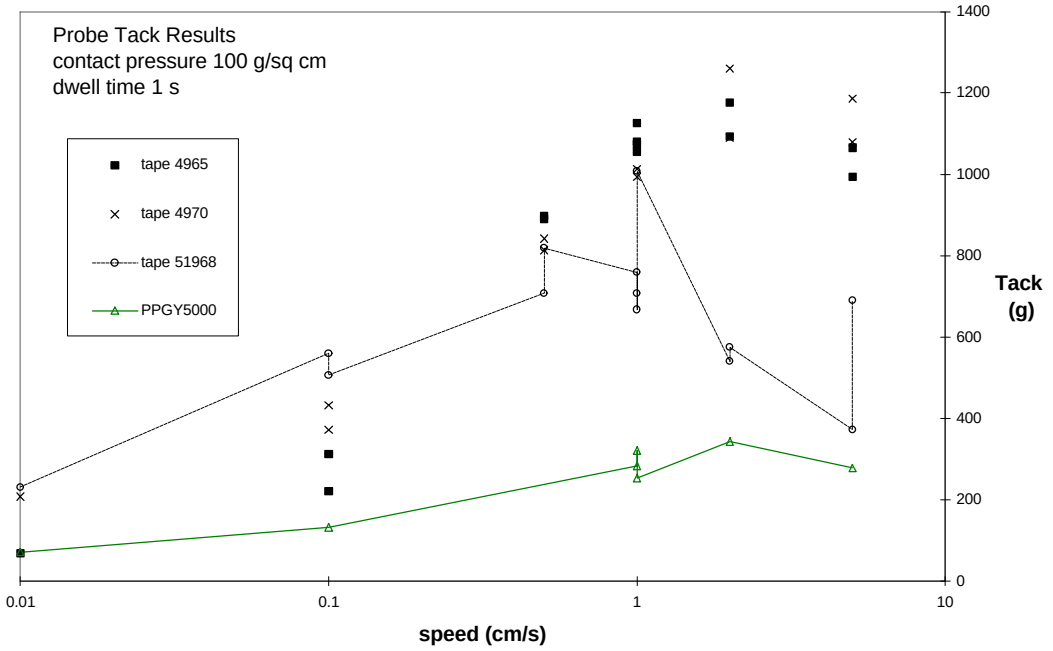

Figure 3: Summary of Tack Properties

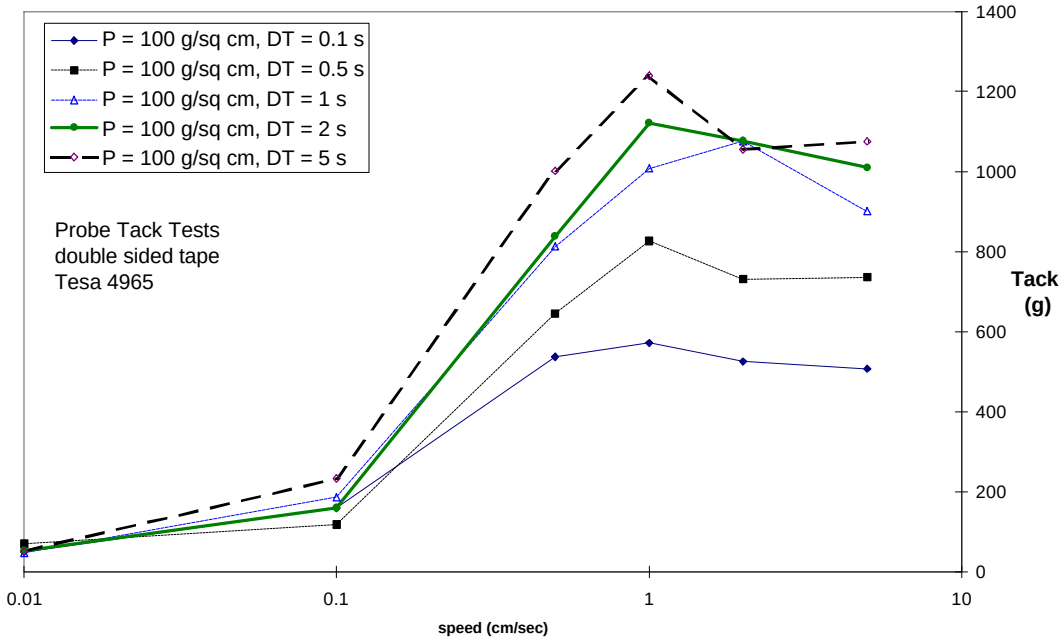

Figure 4: Tape 4965, Tack at Different Dwell Times (DT)

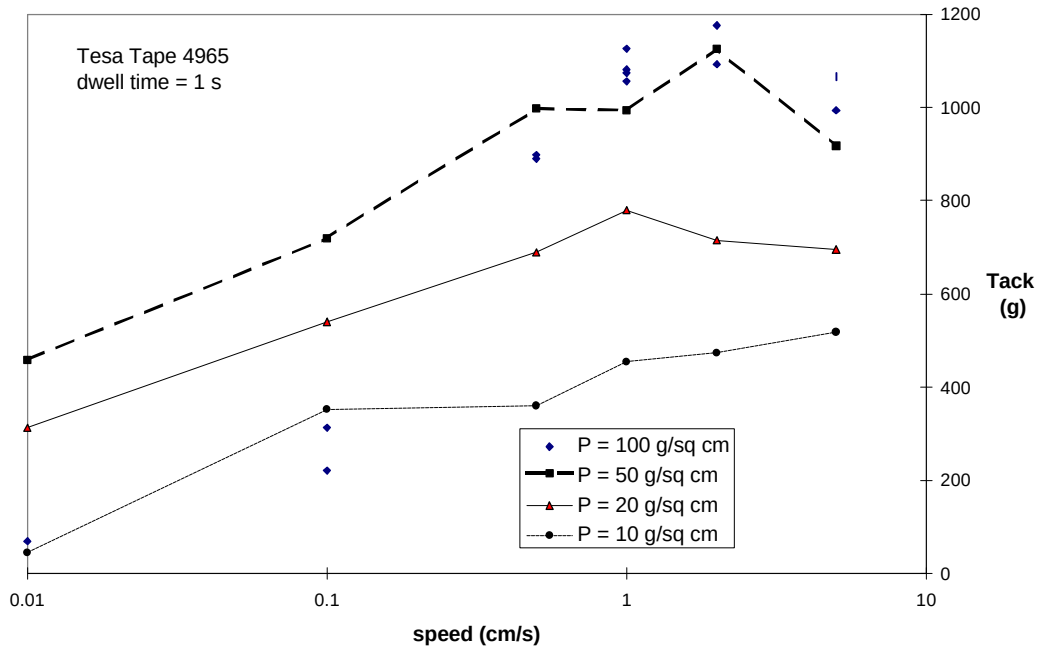

Figure 5: Tape 4965, Tack at Different Contact Pressures (P)

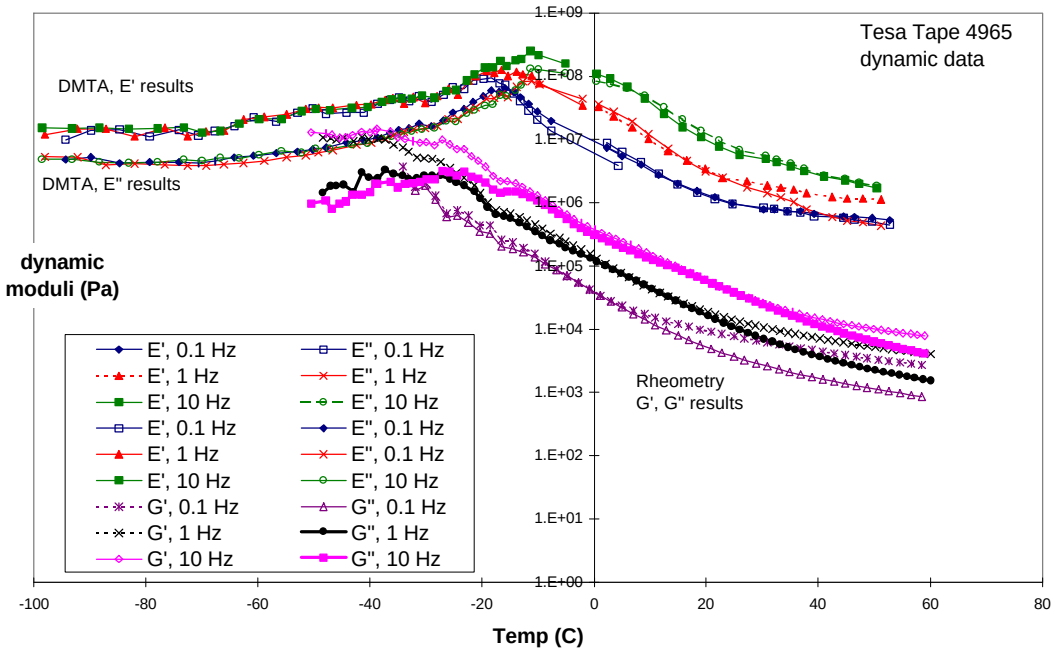

Figure 6: Dynamic Properties of Tape 4965

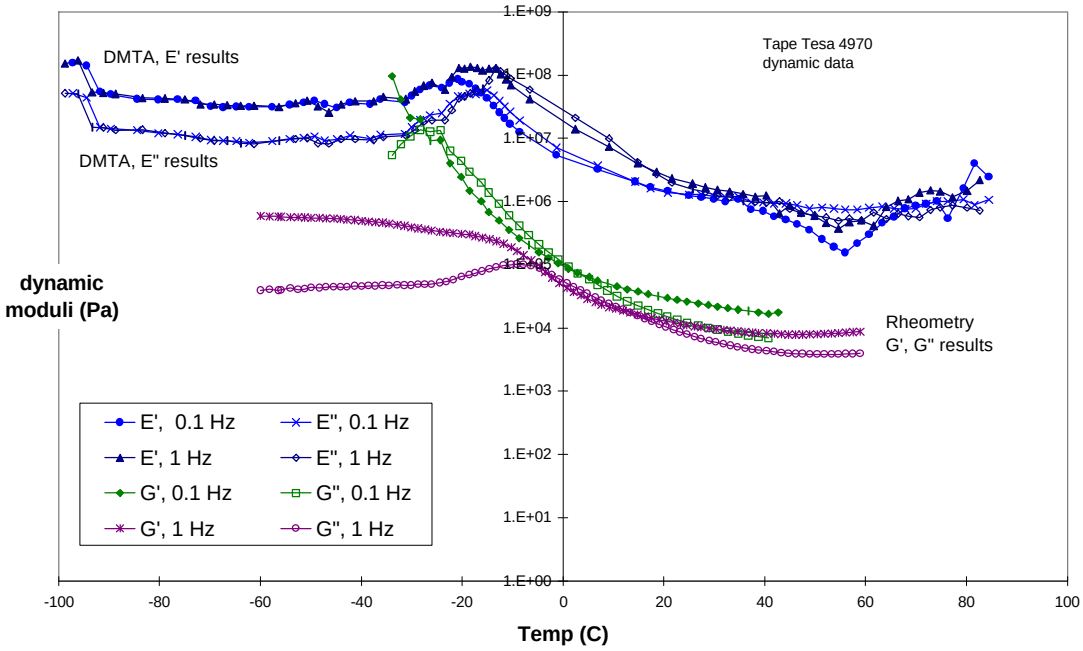

Figure 7: Dynamic Properties of Tape 4970

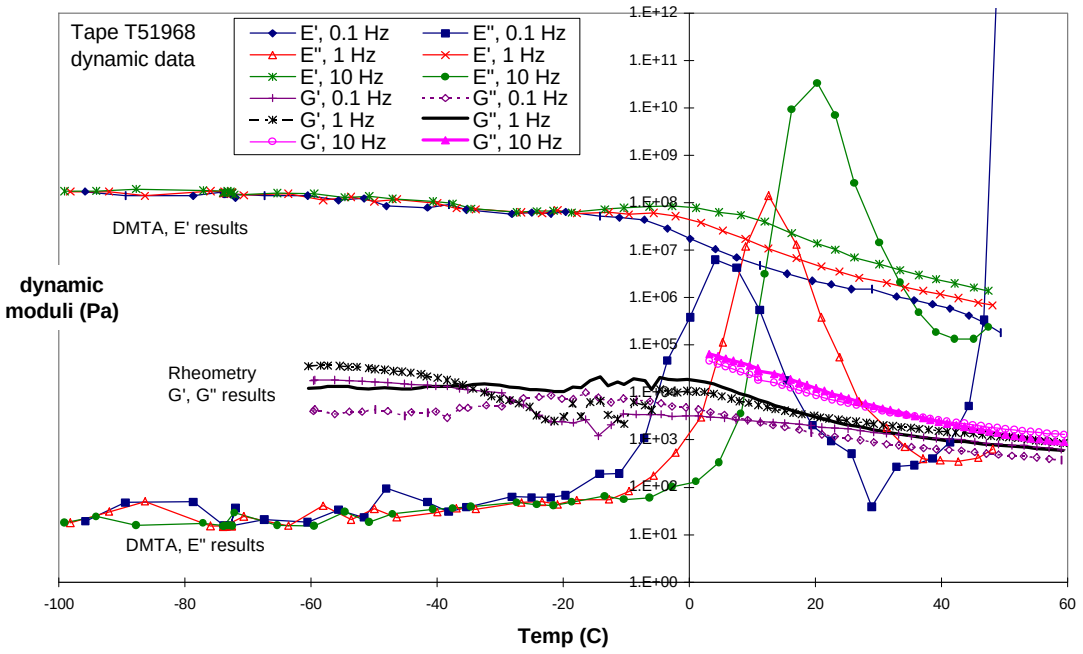

Figure 8: Dynamic Properties of Tape 51968

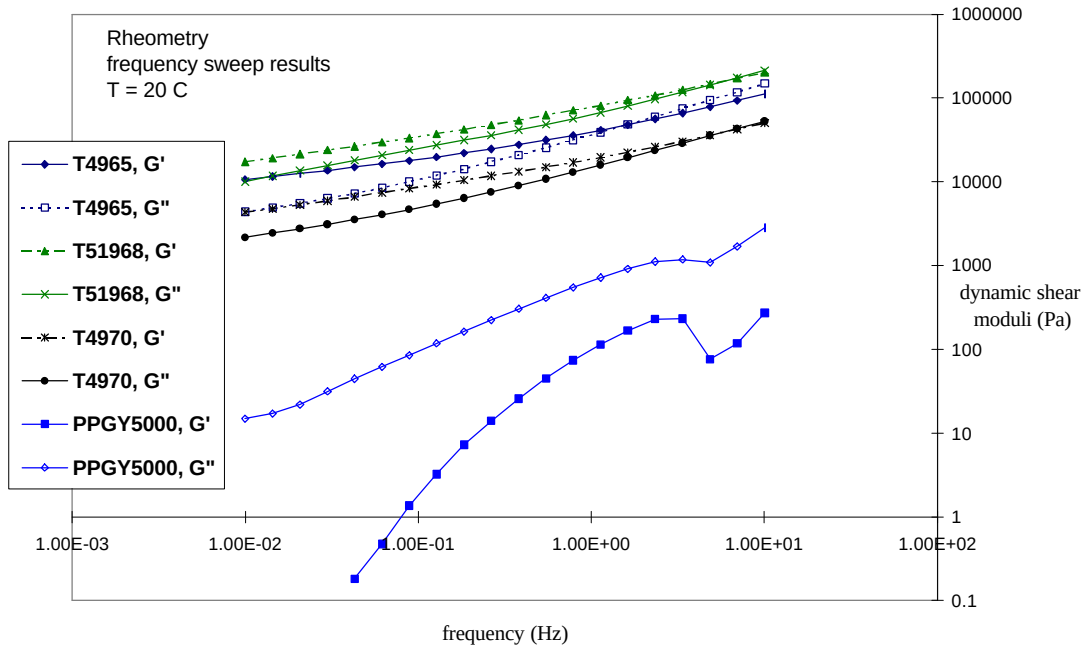

Figure 9: Frequency Dependence of Visco-Elastic Properties

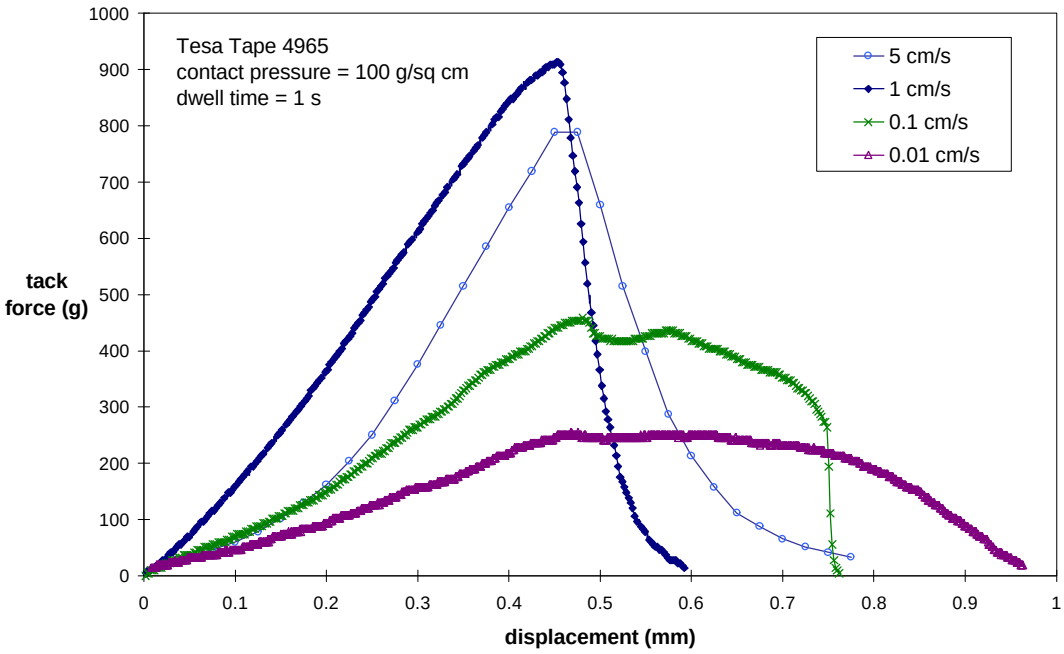

Figure 10: Tape 4965, Tack Force-Deformation at Different Set Separation Speeds

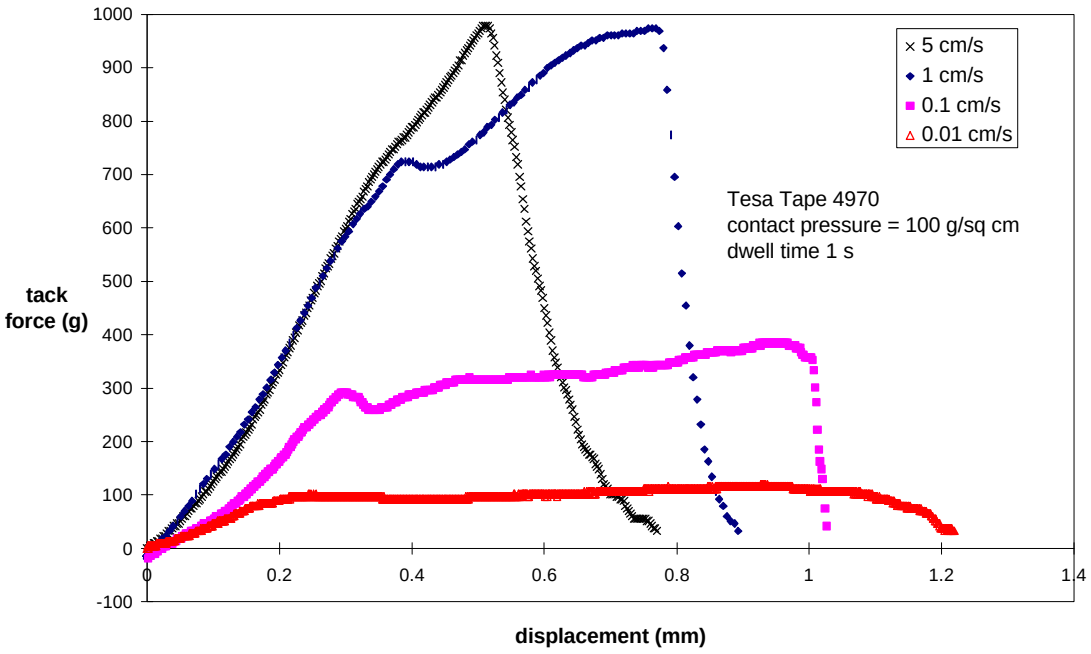

Figure 11: Tape 4970, Tack Force-Deformation at Different Set Separation Speeds

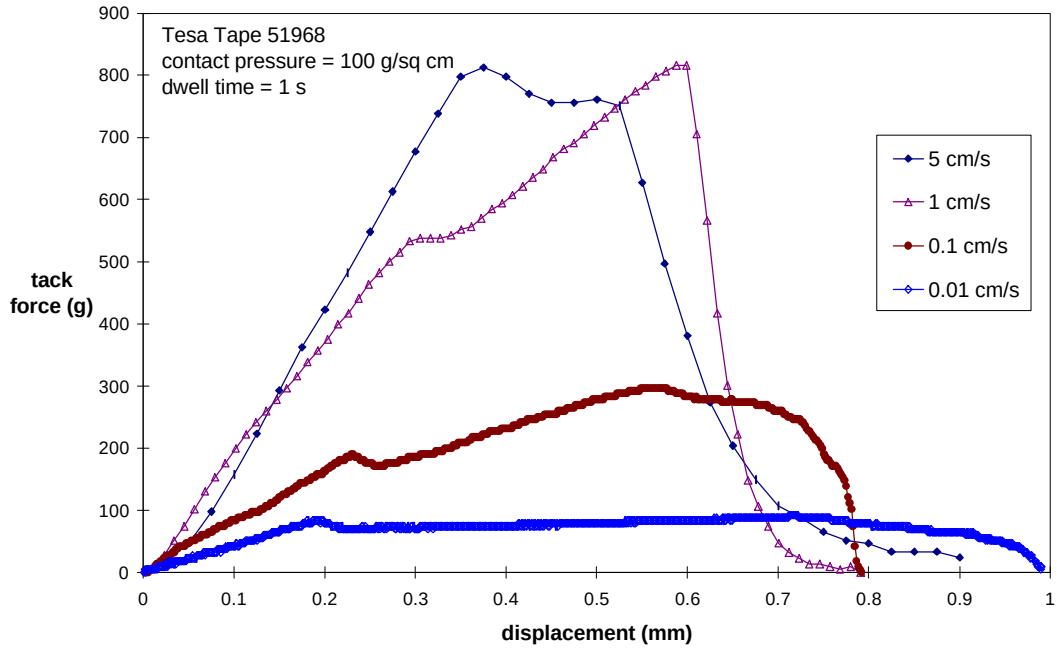

Figure 12: Tape 51968, Tack Force-Deformation at Different Set Separation Speeds

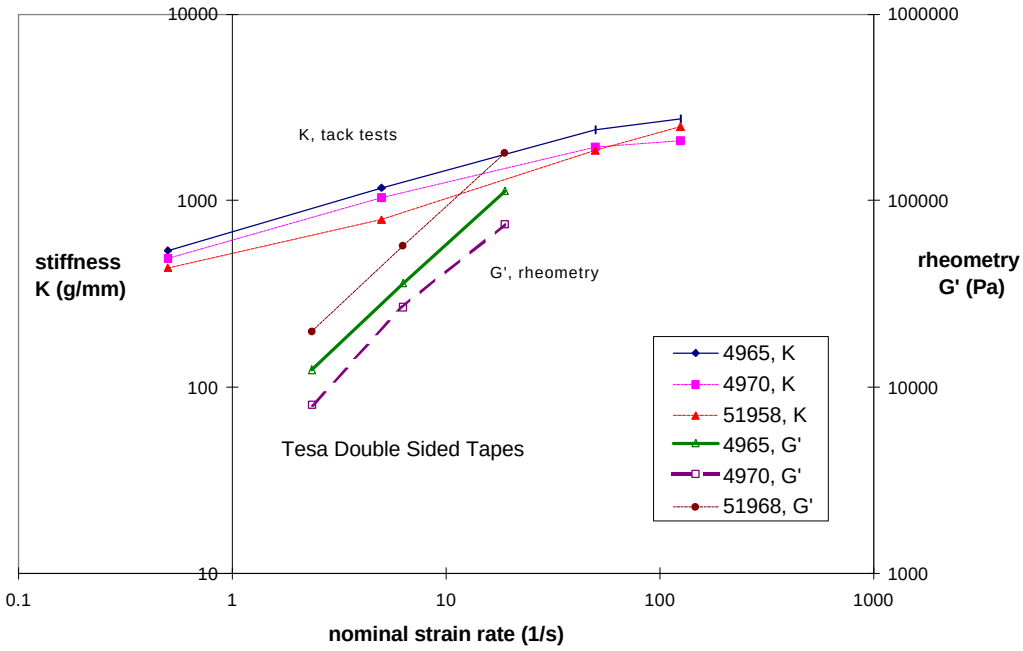

Figure 13: Rate Dependence of Tack Stiffness and Elastic Shear Modulus

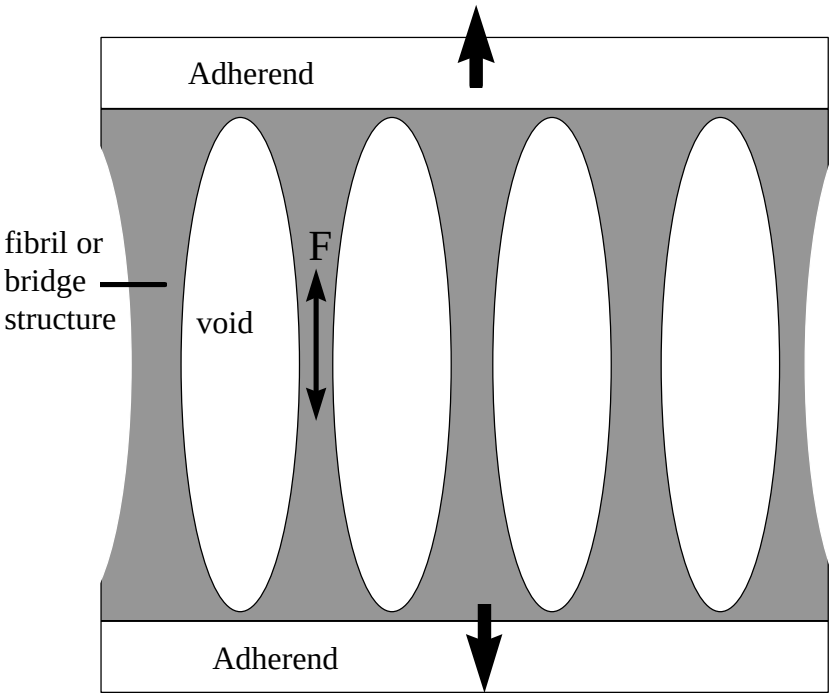

Figure 14: Schematic of Fibril Structures