COLD SEAL ADHESIVES

PEER REVIEWED

# Performance and characterization of hot melt applied cold seal adhesives

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## ABSTRACT

*Natural rubber latex formulations have wide use in cold seal and self-seal adhesive applications because they form films that exhibit auto adhesion and blocking resistance. Their potential for skin sensitization, discoloration, and bond decrease on aging have prompted the industry to seek alternatives. Novel cold seal adhesives applied as hot melts provide adhesive performance comparable to that of natural latex without its inherent disadvantages. Data obtained through dynamic mechanical analysis correlate with sealing and blocking resistance measurements and can provide an operating window for cold seal applications.*

***Application:***

*Hot melt applied cold seal adhesives offer advantages over natural rubber latex products.*

The adhesives industry uses the term cold seal to describe an adhesive that will bond to itself under pressure at ambient temperature but does not adhere to other materials. A cold seal adhesive differs from a pressure sensitive adhesive. A pressure sensitive adhesive bonds to many substances with only light pressure. Pressure sensitive adhesives require release liners or release coatings to prevent unwanted adhesion before use. Systems using natural rubber latex (NL) have long dominated the cold seal adhesive market for applications such as medical packaging, food packaging, and self-seal envelopes.

Dispersions of NL based systems exhibit excellent cold seal properties. The NL particles coalesce and form continuous films on application. These films can provide auto adhesion under pressure. They can also exhibit excellent blocking resistance that allows self winding without use of release systems.

NL has several deficiencies that have prompted consideration of other systems for cold seal applications. NL comes from the rubber tree (Hevea brasiliensis). The chemical composition is cis-polyisoprene with a high molecular weight and low glass transition temperature. These two properties provide stable performance over a wide temperature range. Natural proteins occurring with the isoprene rubber can cause allergic reactions in humans *(1)*. The medical industry has therefore sought alternate adhesive systems. NL also has several performance disadvantages. Since it is an unsaturated polymer, exposure to irradiation during sterilization will change final bond properties. NL is also susceptible to oxidation with a concomitant undesirable color change.

Hot melt applied cold seals provide an alternative to NL-based technology. Formulated systems can avoid bond property changes from oxidation or sterilization. The hot melt raw materials do not contain the natural proteins present in rubber latex products. Using rheological studies of cold seal adhesives can provide a window of utility for certain polymer bases to define a balance of sealing and blocking properties for cold seal applications.

An understanding of the factors involved in adhesion is useful in the development of cold seal adhesives. The categories are interfacial, bonding, and debonding energy contributions according to the following equation *(2)*:

$$P = P_0 DB \quad (1)$$

where
P = adhesion
$P_0$ = interfacial term
D = debonding term
B = bonding term.

The interfacial term includes the effects of surface energy and any migration or orientation of material across the interface or inter-phase region. The debonding term includes the dissipation of energy when the bond breaks whether the failure is along the interface (adhesive) or within the adhesive (cohesive). The bonding term is the ability of the adhesive to deform and wet the substrate.

When the bonding term is the limiting factor in adhesion, dynamic mechanical analysis (DMA) is a valuable tool in adhesive characterization. Several studies of pressure sensitive adhesives have shown correlations between DMA measurements and bonding performance or tack *(2-4)*. These correlations have helped provide an understanding about individual components and their interactive influence on adhesive performance.

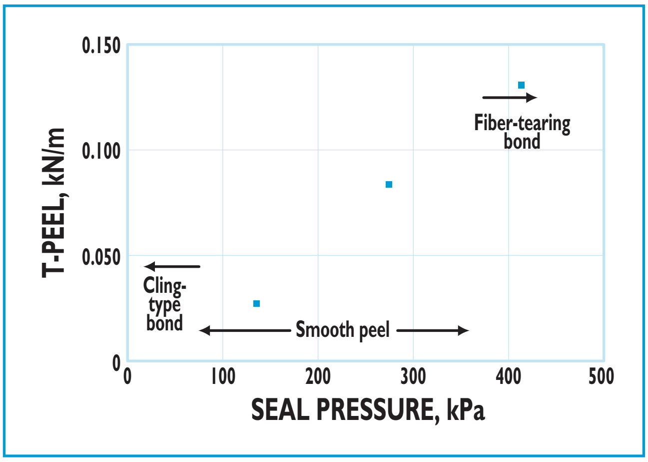
Dahlquist was among the first to show the dependence of adhesion on rheology *(5)*. He tested the tensile compliance of several pressure sensitive adhesives. For the adhesives to wet out and form a bond, he concluded that their tensile

| | A | B | C |
|---|---|---|---|
| Cold seal testing: | | | |
| Seal pressure, kPa | 689 | 413 | 138 |
| Dwell time, s | 1 | 1 | 1 |
| Seal temperature, °C | 25 | 25 | 25 |
| Bond failure mode | No adhesion | Smooth peel | Seal with fiber tear |
| Peel value, kN/m | - | 0.13 | - |
| Press blocking tests: | | | |
| Blocking pressure, kPa | 689 | 689 | 345 |
| Blocking surface | Release coated paper | Release coated paper | Release coated paper |
| Dwell time, s | 1 | 1 | 1 |
| Blocking temperature, °C | 25 | 25 | 25 |
| Bond failure mode | Cling | Cling | Blocking |

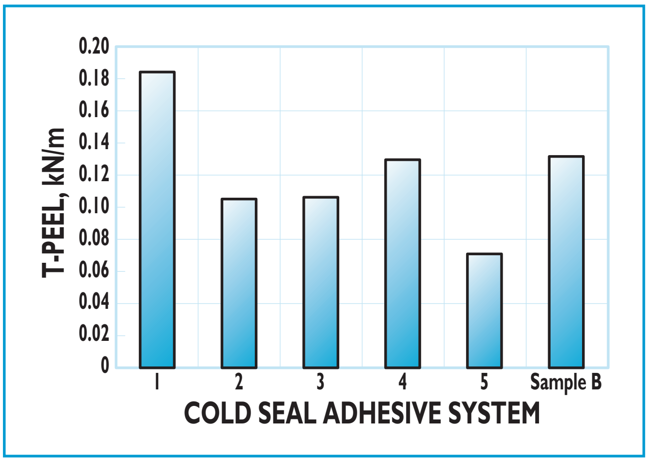
*I. Typical performance of pattern coated cold seal adhesives*

| | D | E |
|---|---|---|
| Cold seal testing: | | |
| Seal pressure, kPa | 551 | 551 |
| Dwell time, s | 1 | 1 |
| Seal temperature, °C | 25 | 25 |
| Bond failure mode | Smooth peel | Smooth peel |
| Peel value, kN/m | 0.16 | 0.02 |
| Commercial unit blocking tests: | | |
| Blocking pressure, kPa | 345 | 345 |
| Blocking surface | 75 g/m² white paper | 75 g/m² white paper |
| Dwell time, h | 72 | 72 |
| Blocking temperature, °C | 49 | 49 |
| Bond failure mode | Cling | Cling |

*II. Typical performance of fully coated cold seal adhesives*




*1. Effect of seal pressure on bond properties for cold seal adhesive sample B*




*2. Comparison of T-peel strength of sample B with various cold seal adhesive systems*

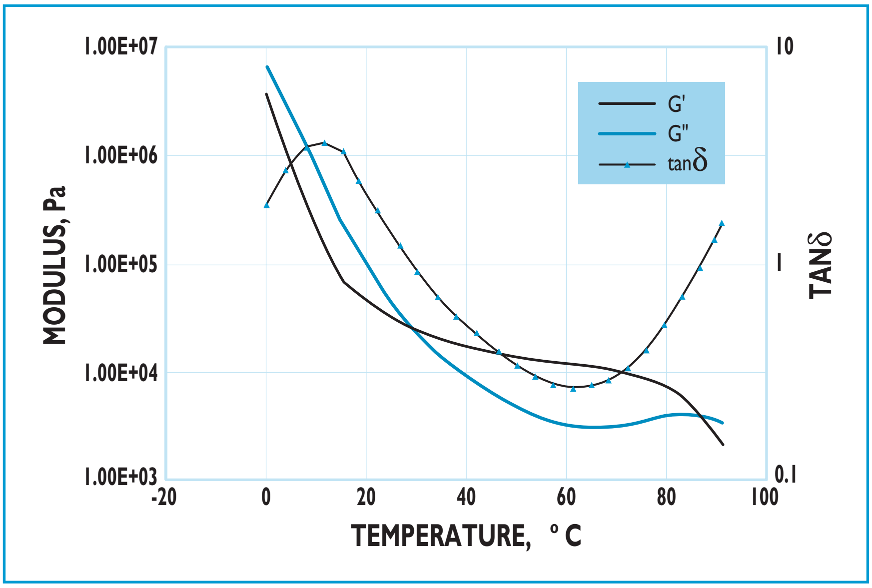
compliance must exceed $10^{-6}$ Pa. With the development of DMA in the 1970s, extension of Dahlquist's observation provided a threshold value of the shear storage modulus, G'. This value is usually below 3 x $10^5$Pa *(4)*. G' is the resistance to a small deformation for which the energy is stored. This approach for pressure sensitive adhesives that must wet under little stress and provide tack can extend to cold seal materials that must wet to themselves only under significant stress and have little or no tack.

The objective of this work is to show that hot melt applied cold seal adhesives have the potential to replace NL-based systems. The use of DMA is an approach to produce an operating window for product development and applications.

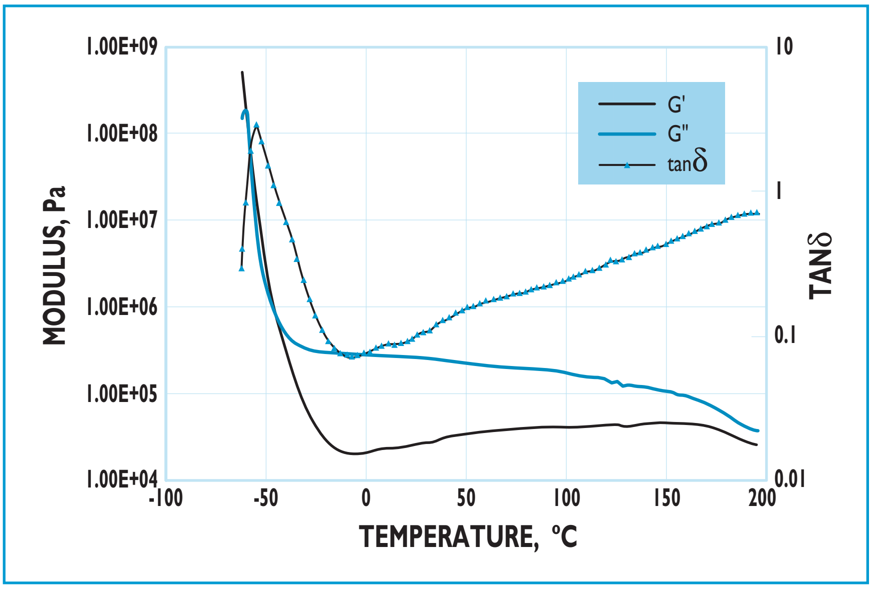
## EXPERIMENTAL

The ingredients were premixed and equilibrated at 176°C in a convection oven. The molten formula was stirred in an upright mixer maintained at 176°C with a heating mantle for 15 min to ensure a uniform composition. Preparation of adhesive compositions used a variety of methods. A hand proofer with an 85Q gravure pattern applied the adhesive. Complete coating sample application used a draw-down adhesive apparatus to apply 0.025 mm thickness. The paper chosen was a white 75g/m² stock with a brightness rating of 84. Coated sheets of paper substrate were sealed adhesive to adhesive in a press at 25°C with a 1 s dwell and variable pressure of 69–690 kPa (10–100 psi).

The bond was characterized by its failure mode during a T-peel test. Blocking evaluation used two methods. One involved using a press to seal adhesive coated stock against paper stock coated with release lacquer. The lacquer application of 8.4 g/m² used a wire wound rod. The adhesive coating used the technique specified above. Blocking tests also used a commercially available testing unit at a specified pressure for a given dwell time at a certain temperature. T-peel samples were made by sealing samples prepared as described above and sealed using a press at a desired pressure with 1 s dwell. Peel rate was 5 cm/min using a commercially available tensile peel tester. The average tensile peel and type of failure were recorded.




*3. DMA temperature plot for typical hot melt label pressure sensitive adhesive*




*4. DMA temperature plot for natural rubber latex cold seal adhesive*

A commercially available unit provided a strain controlled, 10 radians/s temperature ramp or step test. The strain amplitude was adjusted during the tests to remain within the linear viscoelastic region. Parameters measured as a function of temperature were the following:

- G': Shear storage modulus—elastic resistance to deformation
- G": Shear loss modulus—viscous resistance to deformation
- Tanδ: G"/G'—ratio of energy dissipation to energy storage.

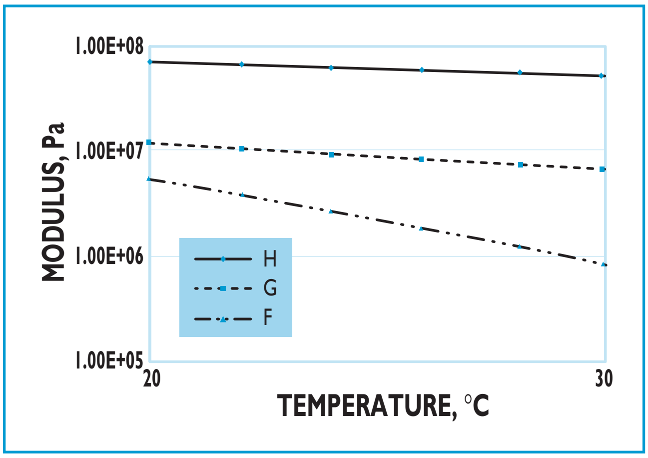
## RESULTS AND DISCUSSION

**Table I** shows representative sealing and blocking properties for two hot melt applied formulations. These low molecular weight versions require a release system to provide blocking resistance. **Table II** shows similar data for continuous hot melt applied cold seals. For these higher molecular weight systems, no release coating is necessary to obtain excellent blocking resistance.

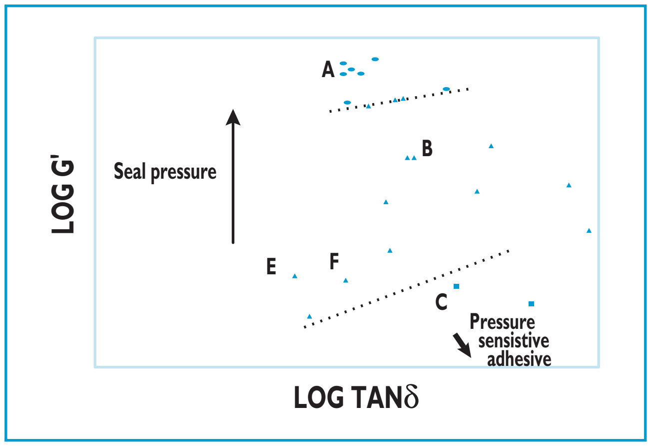
The average peel strength and failure mechanism for these cold seals depended on sealing pressure. **Figure 1** shows this effect for sample B. A seal pressure of 100 kPa (15 psi) or lower gave a bond that produced a weak, cling type peel. Seal pressures between 100 kPa (15 psi) and 380 kPa (55 psi) produced bonds with smooth, controlled peel performance. Seal pressures above 380 kPa gave bonds with sufficient strength to produce substrate failure upon peel. The dependence of peel value on seal pressure suggests that the bonding term is the limiting factor in adhesion. This dependence can also provide the bond strength necessary for a particular application. **Figure 2** compares the T-peel strength for sample B when sealed at 413 kPa (60 psi) to several adhesives used in the medical packaging market. The hot melt applied example falls well within the range of products shown.

The dependence of adhesion on seal pressure suggests that adhesion is bonding limited and that direct correlations with DMA data should be possible. **Figures 3 and 4** show DMA temperature plots of a hot melt pressure sensitive adhesive and a NL dry film, respectively. As expected, marked differences between the two sets of DMA curves are evident for these two very different materials. Comparison of theses curves helps to provide insight into the interpretation of DMA features and how they can relate to cold seal performance.

A measured G' gives a measure of the cohesive stiffness or resilience of a material. The level of G' though the plateau region is significantly higher and flatter for a NL latex. Chu *(3)* reported that the G' value at the appropriate temperature and time scale should be below approximately 3 x $10^5$ Pa to have pressure sensitive adhesive performance. The pressure sensitive adhesive readily meets this constraint, but the NL latex does not even at elevated temperatures.




*5. Effect of shear storage modulus on sealing behavior of cold seal adhesives*

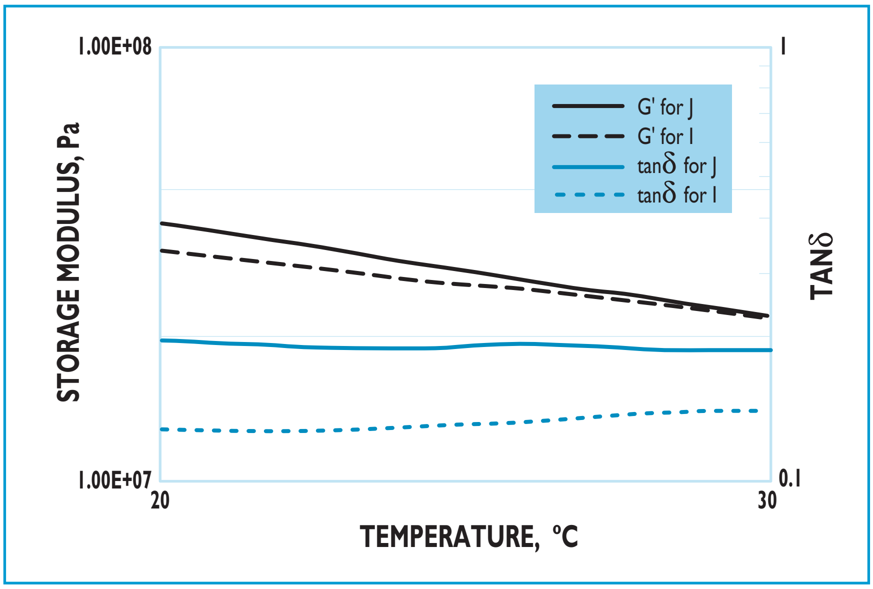



*7. Rheology window for cold seal adhesive performance*

The temperature would need to be at the point of latex degradation before the storage modulus decreased sufficiently to allow for pressure sensitive tack.

Our experiments confirmed the significance of G' in determining cold seal performance. **Figure 5** is a plot of the G' values of sample formulations as a function of temperature. Sample F seals but has poor blocking resistance. Sample H will not seal at a pressure of 690 kPa. Sample G seals at elevated pressure and shows good blocking resistance.

G' alone is an inadequate predictor of cold seal performance. Another key parameter obtained in DMA is tanδ. Tanδ is the ratio of energy dissipated (viscous flow) to energy stored (elastic) for a small deformation. The tanδ level for the latex is significantly lower than for a pressure sensitive adhesive. With the higher level of G'', this also accounts for the absence of tack with the latex film. Some amount of viscous flow of the adhesive is necessary to wet the microscopically rough surface of a substrate. This intimate contact between adhesive and substrate is necessary to enable the interfacial effects to contribute to the bond strength fully. If all the adhesive deformation is elastic, the adhesive will attempt to return to its original shape upon removal of the external stress, and the resulting bond will be weak.




*6. Significance of loss tangent in sealing performance of cold seal adhesives*

**Figure 6** shows G' and tanδ values for two hot melt formulations designated I and J. Although the G' values for these two hot melt formulations are not significantly different, J provided a measurable bond at a seal pressure of 690 kPa, but sample I did not. The comparison of their tanδ values explains their different seal performance. The higher tanδ of sample J indicates a greater fraction of viscous deformation. This enhancement of the adhesive to wet accounts for its ability to seal to itself under pressure.

**Figure 7** shows the significance of comparison of G' and tanδ. Values selected from DMA measurements for the plot used 25°C and 10 radians/s. These conditions approximate those of the seal tests. The NL-based cold seal falls within the window. As long as the adhesive performance is bond limited, the window developed is independent of the chemistry of the system.

Various combinations of hot melt components were tested to establish a more clearly defined window for cold seal development using rheology. In Fig. 7, diamonds indicate formulations that would not seal at 690 kPa. Triangles designate samples that were good potential candidates for cold seal—sealed and exhibited little tack. Circles indicate samples that had substantial tack as indicated by their ability to form a bond to paper when pressed at 690 kPa for 1 s.

As G' increases toward the upper end of the scale, greater pressure is necessary to form a bond. Seal pressures beyond 690 kPa would be necessary to obtain cold seal performance. On the upper margin of this window, the tanδ value acquires added significance for the harder, marginally sealing formulations. A higher tanδ value is necessary to form a cold seal. As G' decreases to approach the value of a pressure sensitive adhesive, a lower sealing pressure is necessary. Poor blocking resistance is also evident at lower values of G' and increased levels of tanδ.

## CONCLUSIONS

Hot melt applied cold seal adhesives have the potential to replace NL-based products. A correlation window ties their sealing characteristics and block resistance properties to DMA measured parameters. These correlations are possible because the bonding term of the model equation limits adhesive performance. The small deformation bulk rheology of the adhesive determines the ability to bond. When this is the case, the correlation window should be valid for a wide variety of chemistries. **TJ**

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*Acknowledgments: The authors thank Matt Inwards and Joe-Lahai Sormana for conducting some performance characterization tests and Tina Sylvia for performing some DMA measurements. We also thank the H. B. Fuller Company for allowing us to present this work.*

*Received for review March 31, 1998.*
*Accepted July 3, 1998.*

*Presented at the TAPPI 1998 Hot Melt Symposium.*

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