PRESSURE-SENSITIVE ADHESIVES FOR RE-SEALABLE, RE-MOVABLE, RE-USABLE SOLUTIONS

Nicolas E. Sajot, Ph.D, Material Technology Manager for Tape & Label Adhesive Business Development, Bostik, Paris-Saint-Denis, France

Introduction

Pressure-sensitive adhesive coatings have been used for many years in many applications. As an outcome of sophisticated innovative programs and from novel ideas and realizations, new applications are still appearing all the time. The innovation source resides often in the obvious question “Can we handle very tacky coatings in an easier, simpler, cheaper way?”, so that end-customers and consumers are so much happier.

One of the solutions could be to make “tacky coatings”… “non tacky”. This could be achieved through an additional step after coating, where radiation or heat is applied (1). Another solution could be to remove the essential, but ultimately useless release liner, and replace it by a surface which will not affect or be affected by the temporary contact with pressure-sensitive coating (2).

A particular example, “re-closable packaging” is one of those applications trying to fulfill the objectives of making everyday life easier. If pressure-sensitive adhesives have been used in the past, recent packaging designs made them more relevant to the consumers’ needs.

Water-based pressure-sensitive adhesive zone-lamination, associated with partly laser-cut converted films is a viable option in various package configurations. Water-based (3) and Hot-melt (4) pressure-sensitive adhesive labels are also well developed options to provide great features.

When it relates to industrial coating applications, there are many ways to bring several thin layers of only a few microns to surfaces, usually on flexible films. Coating techniques and key control parameters for hot melt adhesive technologies are published in literature by adhesive manufacturers (5) and equipment manufacturers (6).
Among those coating process options, there is another technology which is presented in this paper. It is about blow-extrusion films where a thermoplastic layer is made up of a hot-melt pressure-sensitive adhesive material.

Making things simple often requires overcoming difficulties, and this paper describes the basics of blown-film extrusion, the structure of such multilayer films and their use as lidding films (7), then demonstrating how well the hot melt pressure-sensitive materials are adapted to processing and use for the best benefits for consumers. Figure 1 represent the final application article.

All along the description of plastic transformation and processing, rheology is used to identify what is known, and what is unknown.

No strong background in rheology is needed to follow what is presented in this paper. Anyone can refer to good short course on viscoelastic behavior, for example the lecture “defining viscoelastic adhesive coordinates that matter” (8), which also teaches important points on both material science and optimization methodology. Rheology also serves to describe the behavior of adhesive during various steps of transformation. Relevant papers teach on final applications (9) or on specific conditions like instant cooling from molten stage (10).

**Blown co-extruded films**

Well known in the industry blown-film coextrusion leads to thin multi-layer structures. Each individual layer has some functional features and finds its purpose in either processing or final use. As a minimum such co-extruded films would have 3 layers, as illustrated in figure 2.

![Figure 2. Scheme of 3-layer blow extrusion process](image-url)
In figure 2, each of the 3 materials is extruded (push molten) through a co-centric die to flow in a circular / tube form as a molten layer. It will flow biaxially with the other layers. Blown air will stretch and expand and then cool down the resulting film in the vertical direction to the roof, so that the film can be wound on a roll for further converting.

The PSA material is the internal layer, made “invisible” during the whole process, providing easy handling of the film as it is surrounded by other co-extruded layers. One of those is a sealing layer on the internal side of what is called the “bubble” during the extrusion process, and will be a part of the inside surface of what will become a package.

In this process each extruder conveys the material forward, contributing to the heating, melting, homogenizing and mixing of the melt, and of delivering it to the die.

Design of the extrusion die is sophisticated and allows many combinations of co-extruded layers. Monolayer blown-film extrusion may still exist but is not used extensively anymore. Even three-layer equipment is becoming obsolete. It is common to have 5 to 9 to 11 layers in a well-designed film structure, allowing many benefits. Some of those benefits are direct impact of the hot melt pressure-adhesive material design as illustrated in Figure 3. Hot melt pressure-sensitive adhesive layer typically has a thickness of 15 micrometer.

![Figure 3. Potential benefits of multi-layer film structures](image)

Benefits are for example:
- Less lamination
- Down gauging
- Cheaper layers
- Easy sealing
- Easy processing
- Planarity
- Improved strength
- Organoleptics
- High barrier protection
- Recloseability

Although blown-film coextrusion is a well-known technology, typical issues may appear. Not all materials are extrusion friendly, considering a multitude of ways to produce poor quality films: holes, waves, or any kind of material flow irregularity may result in the wrong or inconsistent film features and functionalities, from mechanical properties to aesthetics to easy processing to barrier functions.

Many references treat specific issues with coextruded film irregularities, and a simple rule of thumb will dictate the combination of materials with similar viscosity profile at appropriate temperature and shear rate (11). Figure 4. illustrates this criteria, comparing hot melt pressure-sensitive adhesive materials with typical LDPE.
This first criteria may not be enough in some specific cases. Practice often demonstrates the need to set up more sophisticated theoretical models, if this is about better prediction. Without entering into formulation details, block copolymers are common and useful ingredients in hot melt adhesive products. It is known that the processing of those ingredients is sometimes tricky, and, again, rheology helps visualizing the situation. In a lab experiment, $G'$ (elastic modulus) and $G''$ (viscous modulus) values were collected while decreasing the applied shear rate (or stress frequency). The same experiment was done but this time with increasing shear rate values. As a result, figure 5 illustrates the ratio $G''/G'$, called tan delta, on both cases. One can easily see that tan delta is different whether shear rate goes low to high, or high to low. This is no time effect, as experiments can be repeated in any order, as many times as needed but giving same results.

Figure 4. Viscosity of various hot melt pressure-sensitive adhesive grades against shear rate, compared with typical LDPE

Figure 5. Tan delta of a typical hot melt adhesive material as a function of frequency
Considering the coextrusion process operations, one can see how uncontrolled variations in shear rate may affect the flow behavior of the adhesive material in this case. As well temperature variations - according to time-frequency equivalence – may induce uncontrolled issues.

This criteria is directly linked to the nature, composition and compatibility of materials involved. It shall be taken into consideration during first stages of material design, to avoid any issue with the behavior of such hot melt pressure-sensitive adhesive product during blown-film extrusion operations.

This is essential to the successful production of high quality films, and processing control then becomes easy.

**Film converting**

The co-extruded sealant film is then laminated to usually a reverse-printed, heat-resistant film, typically PET or Polyamide film, to form the lidding web. The structure of such lidding film is for example PET / Ink/ Laminating Adhesive/ Coextruded Sealable Film. The sealing layer, usually a specific LDPE composition, is obviously the outside layer of the lidding film, to become the internal surface of a packaging.

**Lidding films**

In a place where goods are packaged, the laminated lidding web is sealed, for example, to a thermoformed rigid tray. The tray has the right sealable material on its surface to ensure a strong protective bond with the sealing layer of the lidding film.

The sealed tray is then ready to use. After the consumer opens the package the first time, the coextruded sealant film breaks as a cohesive or adhesive failure of the hot melt layer, revealing a pressure-sensitive adhesive surface. In this way the lidding film is easily re-sealed to the tray structure as required. See illustration in figure 6, where only 3 layers are represented as the lidding film structure for the sake of simplification.

- **Lidding film = printed PET / laminating adhesive / Blown coextruded film**
- **PET tray with some sealable layer**

![Figure 6. Structure of blown coextruded film positioned and sealed onto a packaging tray.](image-url)
Opening, re-closing, re-opening

The sealing operation is the assurance for the consumer that the package has not been affected by external factors prior to opening. First opening, simulated by lab experiment of standardized peel force measurement (12) is represented in Figure 7 for various re-closable lidding film structures. Sophisticated film design; supported by computational models and material selection will allow both security and easiness of this first opening. Examples are multiple from re-closability to barrier efficiency to a myriad of other functionalities.

![First package opening force](image)

**Figure 7.** Peel force measurement simulating first opening of package, on several cases: basic re-closability, advanced re-closability, 9-layer barrier film, 11-layer barrier film

Obviously, another key feature of the re-closable package is the easiness to re-close and re-open several times, as represented in figure 8.

![Re-openings](image)

**Figure 8.** Peel force measurement simulating second to fifth openings of package, on several cases: basic re-closability, advanced re-closability, 9-layer barrier film, 11-layer barrier film

Peel consistency over re-opening history, level and smoothness of the peel behaviour is very important for the consumers’ best feeling. These criteria may be investigated through rheology. Figure 9 represents the typical temperature sweep of an extrudable hot melt pressure-sensitive adhesive material, measuring
G’ (elastic) and G’’ (viscous) moduli, and the resulting ratio tan delta (G’’/G’). Shaded rectangles represent areas with low interest for peel and tack considerations, and blue arrays represent some general material design rules for peel and tack.

\[ G' \text{ (elastic) and } G'' \text{ (viscous) moduli, and the resulting ratio tan delta (G’’/G’).} \]

Shaded rectangles represent areas with low interest for peel and tack considerations, and blue arrays represent some general material design rules for peel and tack.

**Figure 9.** Typical temperature sweep of an extrudable hot melt pressure-sensitive adhesive material, measuring G’ (elastic) and G’’ (viscous) moduli, and the resulting ratio tan delta (G’’/G’).

The rules to make the material tackier, higher peel, or in general with adapted “intrinsic cohesion” are quite well-known in the pressure-sensitive adhesive scientific field. Reading this curve will tell the adhesive formulating chemist which ingredient and which ingredient compatibility parameters to use for optimizing end-use applications.

Nevertheless, first conclusions from rheology expertise here may be overshadowed by some common sense observations. Said differently, the lab experiment is far from real conditions to obtain such a pressure-sensitive adhesive layer on a re-closable package. In reality, the pressure-sensitive surface is revealed by the first opening of a sealed area (film to tray). It may have a specific pattern, depending on how the breach has been created in term of speed for example. In reality also, the co-extrusion process conditions can greatly affect the interfaces between layers, affecting the nature of the first opening, and therefore the next ones. These very common observations could be further elaborated, as a lot of theoretical studies would deal with specific surface topologies (13) (14).

Thus, the rheology lab test on the adhesive “bulk” sample just become one of the criteria – and not the only one - to ensure the right tackiness and the right peel force of the pressure-sensitive adhesive surface in the end-use application.
**Extrudable hot melt pressure-sensitive adhesive material in pellets**

Pellet form is probably the easiest way to feed an extrusion equipment and generate material shearing, softening or melting, and flow along screw/barrel interspace.

When proceeding without caution, a pressure-sensitive adhesive pellet would obviously turn into a single block (solid or soft), and handling it would very difficult. A basic idea to avoid this blocking issue is to put a physical or chemical non-tacky barrier around each pellet to avoid intimate contact of those individual tacky surface areas with anything else around.

This principle is do-able, necessary, but typically not enough. If indeed this would allow to handle free-flowing pellets at the extrusion equipment, reality also resides in many other steps or conditions that those pellets shall go through. For example, extended storage time, before using the pellets, will result in a “cold flow” behavior, meaning that material of each pellet is slowing flowing, creeping outside of the physical or chemical non-tacky barrier. The gravity forces, and potentially the pressure created by heavy and tall pellet containers is the cause of this creep. This behavior would also be emphasized, and much faster, if local temperature raises. Figure 10 illustrates the shape of a single pellet over time or under pressure and high temperature.

![Figure 10. Scheme of a single thermoplastic pellet creeping over time and/or temperature](image)

Of course, this creep behavior is very typical of thermoplastic materials, and depends on their “cohesion”, or in other terms on the overall mobility of molecules.

Thus this is talking in our case about adhesive material design. Rheology, once again, explains and provides options to solve issues. Figure 11 compares two hot melt pressure-sensitive adhesive products, in a form of a temperature sweep.
Materials showed in figure 11 are obviously well different from each other, and a first observation is to keep the moduli levels high enough over a wide range of temperature.

Monitoring, during product development, those G’ and G” curves is key. It allows at the end the industrial realization of free-flowing pellets of pressure–sensitive adhesive products. Figure 12 shows a picture of those extrudable hot melt pressure-sensitive adhesive pellets.

**Conclusion**

Hot melt pressure-sensitive adhesive materials can be processed through blown-film extrusion.
This technology makes re-closable packaging possible, which provides a full array of possibilities and benefits to the industry and to consumers.

Rheology has been key to material design and formulation, and key as well to plastics transformation technologies. Rheology is an asset also applied to other re-closable packaging developments based on pressure-sensitive adhesive materials.

It encompasses various water-based and hot-melt coating processes.

Now, outside of packaging re-closability, what will be the next tape innovation based on pressure-sensitive adhesive blown-film extrusion?

Appendices

Lab test conditions for peel force measurement of sealed lidding films onto PET tray:

Lidding film: PET film / laminated adhesive / 50-micron blown co-extruded film
Tray: LDPE coated PET foil
Sealing conditions: 6 bar on flat jaws during 1 sec at 130°C, film to film
Peeling conditions: PSTC test method 101, Afera test method 5001 / F, ISO 29862, adapted: 300 mm/min speed, T-peel at 23°C, sample width 10 mm

Literature citations


(2) David R. Keely (2012), “Primerless Removable Adhesives”, PSTC Tech35 proceedings, Boston, USA


(5) Ian Grace, "HMPSA's and the application thereof", Afera Tapes College, April 2017, Brussels, Belgium


(9) Derek Bamborough “DMA predicts performances of book-binding adhesives”, Adhesive Age November 1990, p. 20-26


(12) PSTC test method 101, Afera test method 5001 / method F, ISO 29862, adapted – see appendices for details


**Acknowledgments**

Thanks a lot to colleagues who helped with this lecture:

Christophea.Robert@bostik.com

Jean-Francois.LeCam@bostik.com

Tyler.Derus@bostik.com

And also Fabrice Flores, Tom Ruppert, Ian Grace, Christophe Morel, Bob Haeger, Hal Story, Benoit Pollacchi.