The Use of Metallocene Polyethylene in Co-Extruded Lamination Film

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ABSTRACT

The value of metallocene LLDPE compared with other polyethylene's (e.g. LDPE, C4-LLDPE etc) has been examined in 3-layer co-extruded lamination film formulations, with particular focus on competitive sealant polymers such as C8-LLDPEs. The examination demonstrated that films formulated with metallocene LLDPE imparted significantly better sealing properties compared with other polymers, resulting in significant improvements in packaging line speed.

This presentation illustrates the comparative results and the conclusions drawn from them.

INTRODUCTION

Adhesive lamination is a converting technology used to make multilayer structures, often referred to as laminates. Laminates result from bonding together one or more substrates (e.g. polyamide, polyester or polypropylene films, aluminum foils, paper, etc.) and a polyolefin film (most often polyethylene) by means of an adhesive system (Figure 1).



The composite structure will retain some of the properties of the individual components, i.e.:

- Barrier properties, i.e. low or controlled permeability to gas and moisture, barrier to light and aroma retention;
- Sealing properties;
- Optical properties, gloss in particular;
- Mechanical properties;
- Organoleptic properties.

A laminate allows a much better compromise among these properties than a single film. Laminated structures therefore lend themselves naturally to applications in demanding end-uses of the flexible packaging industry, like food packaging with medium to long shelf-life.

Other technologies exist to form multilayer structures, such as extrusion lamination and co-extrusion. This paper will focus exclusively on adhesive lamination, although some of the findings may be applicable to other multilayer structures.

From a general point of view, a laminate performance should address both processing and packaging requirements. Among the processing requirements, one can include sealing properties (in particular, fast seal time, broad hot tack window), low and consistent coefficient of friction (COF) and low number of film defects (e.g. gels in the PE film). Packaging requirements are mainly set by the content of the package, depending on whether it is food or not, whether it should have a long or a short shelf life, etc. Among these requirements, barrier properties, package integrity (i.e. how tough is the package as a whole) and organoleptic properties are the most important.

Sealing properties are mainly determined by the formulation of the sealant film. This paper will focus on the sealing performance of laminates having different sealant films. The subject will be tackled from two different perspectives. First, different co-extruded film formulations will be compared with the objective of investigating the influence of each layer composition on sealing properties. Then, the results of a benchmark between metallocene C6-LLDPE (hereon: C6-mLLDPE) and higher alpha olefin LLDPE (hereon: HAO-LLDPE), conventional and metallocene, will be presented.

EXPERIMENTAL SET-UP

Ten films were prepared on a Windmöller & Hölscher co-extrusion blown film line under comparable processing conditions. All films are 3-layer with a layer ratio of 1/2/1 and a total thickness of 40 μ m. Film formulations are given in Table 1.

		1	1
	A - sealing side	B - middle	C - lamination side
1	90/10 C6-mLLDPE/LDPE	60/40 C6-mLLDPE/LDPE	90/10 C6-mLLDPE/LDPE
2	90/10 C6-mLLDPE/LDPE	60/40 C4-LLDPE/LDPE	90/10 C6-mLLDPE/LDPE
3	90/10 C6-mLLDPE/LDPE	60/40 C6-mLLDPE/LDPE	100 LDPE
4	90/10 C8-VLDPE/LDPE	60/40 C6-mLLDPE/LDPE	90/10 C6-mLLDPE/LDPE
5	90/10 C8-LLDPE/LDPE	60/40 C8-LLDPE/LDPE	90/10 <mark>C8-LLDPE</mark> /LDPE
6	90/10 C8-mLLDPE/LDPE	60/40 C8-mLLDPE/LDPE	90/10 C8-mLLDPE/LDPE
7	90/10 C6-mLLDPE/LDPE	70/30 C6-mLLDPE/HDPE	90/10 C6-mLLDPE/LDPE
8	90/10 C6-mLLDPE/LDPE	70/30 C4-LLDPE/HDPE	90/10 C6-mLLDPE/LDPE
9	90/10 C8-LLDPE/LDPE	70/30 C8-LLDPE/HDPE	90/10 C8-LLDPE/LDPE
10	90/10 C8-mLLDPE/LDPE	70/30 C8-mLLDPE/HDPE	90/10 C8-mLLDPE/LDPE
		Table 1	

Red text indicates changes in the formulation with respect to reference structure #1

The polymers used are listed below.

Polymer Type	M.I. 2.16kg/190°C (g/10')	Density (g/cm ³)	Slip Level (ppm)	Antiblock level (ppm)
LDPE	0.75	0.926	-	-
LDPE	0.75	0.926	550	1500
C ₄ -LLDPE	1.0	0.918	1000	2500
C ₄ -LLDPE	0.7	0.925	1250	750
C ₈ -LLDPE	1.1	0.919	800	1250
C ₈ -LLDPE	1.1	0.926	400	1000
C ₆ -mLLDPE *	1.0	0.918	750	2500
C ₆ -mLLDPE **	1.3	0.927	1250	750
C ₈ -mLLDPE	1.0	0.9175	1000	2500
C ₈ -mLLDPE	0.85	0.925	-	-
C ₈ -VLDPE	1.0	0.912	-	-
HDPE	0.7	0.961	-	-

Table 2

* ExxonMobil ExceedTM 1018EB

** ExxonMobil ExceedTM 1327ED

All films have been laminated against the same substrate and with the same adhesive in comparable conditions. The substrate used is oriented polyamide film with a thickness of 15 micron. The adhesive used is a 2-component solvent-less from Rohm & Haas, type Mor-Free 696A / C83. The adhesive was applied to a target coating weight of 1.6 g/m^2 .

Sealing properties were assessed in terms of hot tack and seal strength. All measures were done on the laminates.

INVESTIGATION ON CO-EXTRUDED SEALANT FILM DESIGN



The effect on hot tack of the composition of each layer of the sealant film is shown in Figure 2.

One can see that laminate # 1, whose sealant film contains C6-mLLDPE in all three layers, has the broadest hot tack curve. This translates into more robust packaging operations, with a potential of faster line speed and reduced leakage rate. The comparison shows that the composition of all three layers of the sealant film affects the sealing behavior of the laminate. Therefore, designing a sealant film requires a total system approach, and the composition of all layers should be carefully chosen.



The previous findings are confirmed by comparing the laminates shown in Figure 3.



Laminate #7 and laminate #8 differ only in the composition of the middle layer, where the C6-mLLDPE of #7 is replaced by C4-LLDPE in #8. Replacing C6-mLLDPE in the core layer makes the hot tack curve narrower, as previously observed. Additionally, by comparing laminate #1 with laminate #7 we see that increasing the stiffness of the core layer on a C6-mLLDPE based film does not negatively affect the breadth of the hot tack curve, as observed with other polymers (see Figure 4).

COMPETITIVE BENCHMARKING: C6-mLLDPE VS. HAO-LLDPE

Comparing stiff sealant films based on HDPE / LLDPE blends in the core layer (Figure 4) shows that the C6-mLLDPE based film (#7) delivers a broader hot tack curve after lamination compared to C8-LLDPE and C8-mLLDPE based films (#9 and #10 respectively).



Stiff sealant films impart higher stiffness to the laminate and are therefore a good solution for end-uses such as Stand-Up Pouches. Hence, we believe that a C6-mLLDPE / HDPE co-extruded sealant film structure would have significant advantages for this end-use compared to competitive formulations based on C8-(m)LLDPE / HDPE blends, in that it would provide stiffness and deliver at the same time better hot-tack than C8-LLDPE / HDPE formulations.

Together with hot-tack, a fundamental criterion for benchmarking sealing properties is sealing time. As illustrated in Figure 5 for a typical packaging cycle, in fact, decreasing sealing time by 20% can increase the packaging rate by 7%, leaving all other cycle times unchanged. This translated into "tangible" value for end-users, and represents a strong performance benefit.



Figure 5

ExxonMobil Chemical uses a procedure to benchmark the package integrity of different laminates as a function of the sealing conditions, expressed by sealing time and sealing temperature. This is accomplished by running packaging trials on a V-FFS machine in the following way. Empty pouches are made at different combinations of

sealing time and sealing temperature. The pouches are tested for rupture strength on the machine shown in Figure 6, called a SKYE tester.



Figure	6
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The SKYE tester consists of a needle to be inserted into the pouch and in a pressure measurement device. The pouch is inflated with air through the needle and the rupture pressure is measured. This is a measure of the rupture strength of the pouch at that sealing condition. By repeating the test for different sealing combinations, rupture strength can be plotted against sealing time at constant sealing temperature.

Figure 7 shows an example of the outcome of such a test for three laminates with a different sealant film composition. All laminates reach a similar plateau level of rupture strength above a certain sealing time. This indicates that in those sealing conditions the pouch will fail on the laminate and not at the seal, i.e. the highest mechanical rupture strength of the structure has been reached. Because all the laminates have comparable values of

rupture strengths, the plateau levels will be similar. Hence we can conclude that this plateau value of rupture strength represents the highest packaging integrity attainable. For the laminates under discussion, the highest package integrity has been set to 600 mbar, which is the closest value to the plateau level of all laminates. The faster the sealing time to reach the highest package integrity is, the better the sealing performance will be. In the case shown above, laminate #1, whose sealant film is based on C6-mLLDPE, has almost 10% faster sealing time than the laminate based on C8-LLDPE and 25% faster sealing time than the laminate based on C8-mLLDPE.

For a more complete evaluation of the sealing performance of these laminates, hot tack was assessed by conducting packaging trials on the same V-FFS line. To that end, filled pouches were made at different sealing conditions. The content of the pouches was 1.3 kg of a CaCO₃ compound. Soon after filling, the status of the bottom seal was assessed. Results are reported in the table in Figure 8.

Seal bar temperature 😅			1	20)°(С			130°C 140°C										150°C													
Structure id	150ms	200ms	250ms	300ms	350ms	400ms	450ms	500ms	150ms	200ms	250ms	300ms	350ms	400ms	450ms	500ms	150ms	200ms	250ms	300ms	350ms	400ms	450ms	500ms	150ms	200ms	250ms	300ms	350ms	400ms	450ms	500ms
1: C6-mLLDPE																																
5: C8-ZN-LLDPE																																
6: C8-mLLDPE																																

	Figure	8	
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As a legend, a colored box indicates that the seal was not deformed; a white box indicates that the seal opened under the weight of the filling and a cross-hatched box indicates that the seal underwent some deformation, but did not burst. In essence, the longer and less interrupted is the sequence of colored boxes, the better is the hot tack performance of the laminate. One can conclude that the laminate based on C6-mLLDPE provides the best hot tack performance. In particular, comparing the hot tack at the temperature chosen for package integrity, i.e. 140 °C, shows that C6-mLLDPE has a better hot tack at low seal times than C8-LLDPE and C8-mLLDPE.

Additionally, a lab measurement of hot tack was carried out on the three laminates considered to confirm the findings of the packaging trials (Figure 9)

As it appears from the chart, the C6-mLLDPE based laminate has a broader hot tack curve than the other two laminates. The lab measurement confirms the superior performance of the sealant film based on C6-mLLDPE vs. C8-LLDPE and C8-mLLDPE.

Finally, a comparison between a C6-mLLDPE based laminate and a C8-VLDPE based laminate was performed under the same conditions.

Figure 10

Figure 10 shows the rupture strength chart at 140 °C for the two laminates. It appears clearly that the laminate based on C6-mLLDPE has a considerably lower sealing time. This is remarkable, considering the difference in density of 0.006 g/cm^3 between the two products.

Also the comparison on hot tack is favorable to the C6-mLLDPE based laminate (Figure 11).

Seal bar temperature			1	20)°(2					1	30	°C	;					1	40)°C)					1	50)°()		
Structure id	150ms	200ms	250ms	300ms	350ms	400ms	450ms	500ms	150ms	200ms	250ms	300ms	350ms	400ms	450ms	500ms	150ms	200ms	250ms	300ms	350ms	400ms	450ms	500ms	150ms	200ms	250ms	300ms	350ms	400ms	450ms	500ms
1: C6-mLLDPE																																
4: C8-VLDPE																																
									-																							

Figure 11

This finding is confirmed by the lab measurement of hot tack shown in Figure 12, where the C6-mLLDPE based laminate demonstrates a broader hot tack curve than the C8-VLDPE based laminate.

Figure 12

CONCLUSIONS

A key performance criterion for an adhesive laminate is sealing properties. Better sealing properties translate into faster and more robust packaging operations on FFS lines, and therefore it is of great value for end-users.

We have examined and demonstrated that the composition of all layers in a co-extruded sealant film have the potential to affect the sealing performance of a laminate. In particular, when all layers of the sealant film are formulated with C6-mLLDPE, then the laminate will have superior sealing properties compared to sealant films formulated with C4-LLDPE or LDPE in any of the layers.

A competitive benchmark was also conducted to compare C6-mLLDPE performance against HAO-LLDPE. Laminates based on C6-mLLDPE demonstrated superior hot tack and lower sealing time compared to laminates based on C8-LLDPE, C8-mLLDPE and C8-VLDPE.