"In-Line Processing of Multilayer Food Packaging Laminating Structures with EB Adhesives"

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Abstract:

Multi-layer laminated structures are commonly used in food packaging applications for combinations of different barrier properties and heat sealability. These printed laminated structures are often prepared in multiple passes instead of a single step mainly due to the long drying oven in the case of solvent and water borne systems, or due to low immediate bond development in the case of solventless systems. Now, with electron beam (EB) curable laminating adhesives, a single pass (in-line) converting process of these laminates is possible with environmental and cost benefits. Examples of the proposed structures and their performance attributes are demonstrated along with FDA compliance test results.

Background

As converters strive to improve the cost structure of packaging materials, increase production efficiency, enhance product performance, reduce carrying inventory, lower VOC emissions, reach for the goal of on-demand manufacturing or create new packaging designs, they always search for the latest available technologies or chemistries. More often than not, the solutions to their wants and needs have been pointing towards EB/UV curable chemistry for obvious reasons. EB/UV curable technology has been known for many benefits including environment friendliness (e.g., zero-VOC), production speed enhancement, overall cost reduction, and process enabling capability. Recently, with the help of comprehensive and FDA-compliant migration testing protocols [1-3], EB/UV curable chemistry has emerged as one of the mainstream chemistries for food packaging end-uses.

Following the successful and mature application of EB curable chemistry (inks & topcoat) in the folding carton market with packaged products like juice, dairy products, TV dinners, ice cream, pizza, beer cans,…etc, the Radtech industry has started a new trend within the last 2-4 years in the flexible packaging market with the realization of EB topcoats replacing laminates [4-6]. Our lab was the first to demonstrate and commercialize this "mono-web" concept and achieve the following benefits [6]:

- Reduced package design complexity.
- Realization of a 10-20% cost reduction, depending on the complexity of the multi-layer structures.
- Achieving FDA compliance status with the most stringent detection method.
- Enabling more in-line and on-demand manufacturing/processing.
- Maintaining or improving package appeal.

However, there are still other flexible food packagings that will require the use of multilayer laminated structures for various reasons including specific end-use requirements, better barrier properties, and tougher product resistance. Even though the Radtech industry has been pushing hard with EB laminating adhesives [7-11] for food packaging for the last 4 years to compete with the current chemistries (solvent borne polyurethanes (PU), water borne PU, solventless PU, and two new rivals, water borne acrylics and water borne acrylate/PU hybrids), the adoption of this EB alternative has been slow despite the obvious advantages such as:

- 100% reactive and no VOC concern.
- One part system and at least 6 months shelf life.
- Immediate cure with full bond or nearly full bond development.
- Lower energy consumption.
- No aromatic amines/ isocyanates.

The main reasons/challenges for this slower than expected progress, even with a focused effort among major formulators, converters and equipment suppliers, are:

- The higher cost of EB laminating adhesives despite process advantages and inventory reduction benefits (Higher dry pound cost than solventless PU, but only slightly higher than those of solvent and WB systems).
- Not as versatile over as many substrate combinations per single adhesive.
- More sensitive to moisture compared to PU systems but similar to acrylic systems.
- Adhesion to various inks remains challenging.
- FDA certification using EB laminating adhesives.

In-line Processing with EB Laminating Adhesives

Besides addressing the bond performance over the most commonly used substrate combinations for food packaging uses with our latest FDA approved EB laminating adhesives, we are also proposing a new concept of producing multi-layer structures, specifically triplex constructions, in one single step utilizing at least one layer of an EB laminating adhesive. Normally, these types of triplex constructions are accomplished in two or more steps depending on the complexity of the structure or press limitation. This single step, or so-called “in-line”, manufacturing of triplex structures is the key to bigger cost savings for converters despite the higher dry pound cost of current EB adhesives compared to those of the existing chemistries. Two of the many possible in-line process arrangements are outlined Fig. 1.

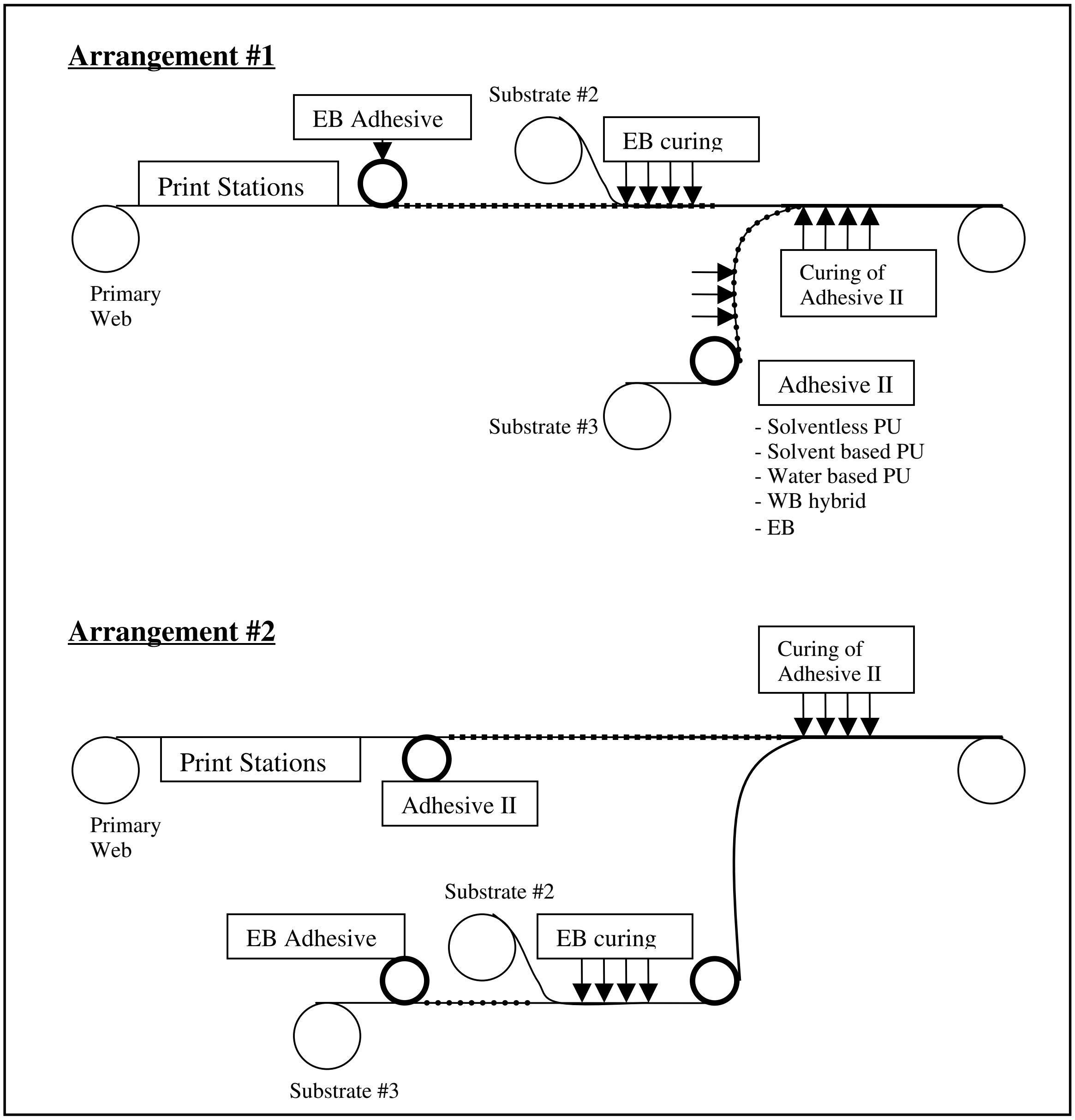

Fig. 1. Two possible arrangements of in-line processing of triplex structures.

The second adhesive in the description can be any of the above mentioned laminating adhesives including solventless PU, solvent borne PU, water borne PU, water borne hybrid systems, and EB laminating adhesive. However, the best choice from the environmental and cost standpoints would be the solventless PU adhesive system.

Experimental:

Substrates commonly employed in the packaging industry (48 gauge PET, 40 gauge Metallized PET, 40 gauge OPP, 70 gauge metallized and sealable OPP, 1 mil aluminum foil, 2 mil LLDPE, 2 mil PE) were used in the study. They are specified in the test results and were supplied by our customers. Corona treatment was applied on all nonmetallized films in-line or off-line immediately before use. The adhesives were applied in the lab with a direct gravure lab coater and cured with an ESI (Energy Sciences, Inc.) lab curing unit (model: Electrocurtain CB175) at specified Kilovolts (KV) and Mrads or at ESI with the pilot coater line at 110KV, 3 Mrads, or with the customer's commercial coater and EB curing unit at higher line speeds. EB adhesives were applied at a coating weight of 0.9 to 1.5 lb per 3000 ft^2 (0.9-1.5 lb/ream) as specified in the results for all laminates. The 2-part solvent based PU laminating adhesive (PB6300/PB610 from Sovereign Specialty Chemicals, Inc) was applied with a dry weight of 1.3-1.6 lb/ream and dried before making laminates.

Results and Discussion:

Performance over various film combinations:

Table 1 shows the immediate bond strengths of some of the most commonly used film combinations for duplex structures in food packaging applications.

Table 1: Immediate Bond Strength of Various Duplex Laminates

	48ga.PET 2mil LLDPE	**48ga. MPET 2 mil LLDPE**	**40ga.OPP 2 mil LLDPE**	**48ga.white PET 70ga. MOPP**
Formula #1	350-370 gli FT $^{\text{Condition 1}}$	140-200gli Some metal transfer $^{\text{Condition 1}}$	230-250 gli FT $^{\text{Condition 1}}$	150-200 gli some metal transfer $^{\text{Condition 2}}$
Formula #2	170 gli FT $^{\text{Condition 3}}$	30-40 gli Slight metal Transfer $^{\text{Condition 1}}$	170 gli FT	230 gli FT $^{\text{Condition 3}}$
Condition 1: @ESI, 110KV, 3Mrad, 50fpm; 1.1 lb/ream Condition 2: 110KV, 3Mrad, 500 fpm; 1.5 lb/ream Condition 3: 110 KV, 3 Mrad, Lab EB unit, 1.1 lb/ream				

The immediate bond strength development of an EB laminating adhesive, Formula #1, is shown in Table 2 for two common triplex laminates between PET and a center substrate (outer layer). A solvent borne, 2-part PU laminating adhesive (PB6300/PB610 from Sovereign Specialty Chemicals, Inc.) was used between the center substrate and the sealant layer (inner layer).

Table 2: Immediate Bond Strength of Two Triplex Laminates.

	PET//Foil//LLDPE	**PET//MOPP//LLDPE**
Formula #1 in one layer. SB PU (PB6300/PB610) in inside layer.	150 gli, Slight delayed film tear No delamination after heat seal	100 gli, 90% Metal transfer No delamination after heat seal
Note: Lab EB unit, 110 KV, 3 Mrad, 1.0-1.1 lb/ream.		

As one can see from the examples in Table 1 and table 2, these new formulas give adequate bond development and film tearing or metal transfer bonds over many commonly used film combinations.

FDA Compliance Study

When considering the FDA compliance of any EB laminating adhesive, a migration (extraction) test must be performed according to the FDA testing protocol. The simulants and extraction conditions for various food applications are outlined in the FDA guidelines[12] and are best summarized in our previous publications.[1-3] The detection and determination of EB and UV curable ingredients are unique and are drastically different from those used for polymeric resins. Studies[1-3] have shown that liquid chromatography with a mass selective detector (LC-MS-MS) is best suited to detect all such EB/UV curable ingredients while gas chromatography with a mass selective detector (GC-MS) is only suited in limited situations where all ingredients must be volatile to be detected.

To demonstrate the difference in barrier properties of various films towards the migration of ingredients in EB laminating adhesives (especially the inner layer film with which the food is in contact) with two examples are illustrated below to show the importance of proper film selection.

The first example, see Table 3, uses an industrial EB laminating adhesive to show some important and interesting facts of a migration study with various sealant films. (Note that the extraction was performed through the sealant film side and the result is not expected to be affected by the outside film.) We can draw these conclusions:

- Metallized film provides a better barrier than does the plain LLDPE.
- 95% Alcohol (fatty food simulant) is more aggressive than 10% alcohol (aqueous food simulant)
- 40 ^{O}C is more aggressive than room temperature (RT) accelerated testing (40^{O}C simulates room temperature food storage and RT simulates refrigerated storage).
- 2 mil thick PE (higher molecular weight) film is a better barrier than LLDPE.
- Foil is excellent and the best barrier.

Table 3. Extraction Results of an Industrial EB Laminating Adhesive, Formula#3, through various sealant films.

Construction		**Simulant**	**Extraction Temperature & Time**	**Detection Method**	**Total Extractables**
Outside	Food side				
OPP-40ga.	**LLDPE**-2 mils	10% Alcohol	RT-10 days	GC LC	1822 ppb 5822 ppb
PET-48ga.	**MOPP-**70 ga. (sealable)	10% Alcohol	RT-10 days	GC LC	ND ND
		10% Alcohol	40^{O}C-10 days	GC LC	206 ppb ---------
		95% Alcohol	40^{O}C-10 days	GC LC	1276 ppb --------
PET-Foil 48ga.-813type	**PE-**2 mils	10% Alcohol	RT-10days	GC LC	ND ND
PET-48ga.	**Foil-sealant** laminate	95% Alcohol	40^{O}C-10 days	GC LC	ND ND

ND: not detected. Detection limit = 50 ppb. (ppb: parts per billion based on 10 grams of food contact per square inch area).
Note: curing conditions for EB laminating adhesive are 115 KV, 3 Mrad using lab EB curing unit.
Coating weight: 1.0-1.1 lb/ream.

Table 4 shows the extraction results of the previously mentioned EB laminating adhesive, Formula #2, with both duplex and triplex structures which qualify it for indirect FDA applications (CFR 175.105, all food types, with condition of use E) in the construction as listed.

Table 4. Extraction Results of EB Laminating Adhesive, Formula#2, through various sealant films.

Construction		Simulant	Extraction Temperature & Time	Detection Method	Total Extractables
Outside	Food side				
PET-48ga.	**MOPP-**70 ga. (sealable)	10% Alcohol	40^{O}C-10 days	GC LC	ND ND
		95% Alcohol	40^{O}C-10 days	GC LC	ND ND
PET-48ga.	**Foil/PU/Sealant film**	10% Alcohol	40^{O}C-10 days	GC LC	ND ND
		95% Alcohol	40^{O}C-10 days	GC LC	ND ND
PET-48ga.	**MOPP/PU/sealant film**	10% Alcohol	40^{O}C-10 days	GC LC	ND ND
		95% Alcohol	40^{O}C-10 days	GC LC	ND ND

PU: PB6300/PB610, a solvent borne 2K PU laminating adhesive from Sovereign.
ND: not detected. Detection limit = 50 ppb. (ppb: parts per billion based on 10 grams of food contact per square inch area).
Note: curing conditions for EB laminating adhesive are 115 KV, 3 Mrad using lab EB curing unit.
Coating weight: 1.0-1.1 lb/ream

Conclusions:

The paper has addressed many of the hurdles facing EB laminating adhesives for flexible food packaging applications. A one step, in-line processing of triplex laminate structures is made possible and demonstrated here using at least one layer of an EB laminating adhesive with environmental advantages and more cost saving benefits than have been achieved before. The examples shown here hopefully will encourage converters to re-evaluate the cost benefits and coater manufacturers to design or retrofit with an EB curing option. Performance of the new generation of EB laminating adhesives has been demonstrated to provide adequate or equal film tearing bonds for many of the common film combinations, both duplex and triplex laminates, with FDA compliant certification. The immediate cure with full bond or close to full bond development characteristics should give the converters an added competitive edge.

Moisture sensitivity for EB laminating adhesives in a water soaking test for some applications, however, continues to be a challenge. Adhesion over various inks may require continuous improvement, although one can for now specify the inks that give good adhesion performance. As the RadTech industry continues to devote more effort to realize the full potential of EB laminating adhesives, these issues and special requirements are expected to be resolved or met in the very near future.

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