Volume Two

PLASTIC FILM TECHNOLOGY

Extrusion of Plastic Film & Sheeting

Edited By

KIER M. FINLAYSON

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EDITOR'S NOTE: With the well received publication of *Plastic Film Technology, Volume One—High Barrier Plastic Films for Packaging,*[1] the publisher has decided to continue the series with *Volume Two—Extrusion of Plastic Film & Sheeting.*

This is a collection of papers dealing with the subject of plastic film extrusion that appeared during the three years 1989–1991 in the *Journal of Plastic Film & Sheeting*. The paper by S. J. Derezinski is an updated compilation of a number of papers that have appeared elsewhere.

The production of high quality blown film has been made possible by on-line film gauging, gravimetric feeding, innovation in film dies and wind-up equipment, and improved data handling. Attention to the uniformity of the process and property performance of the film resins will be necessary to obtain adequate cost control and effective material utilization.

[1] K. M. Finlayson, ed. 1989. Lancaster, PA: Technomic Publishing Co., Inc.

TABLE OF CONTENTS

NEW ETHYLENE-METHYL ACRYLATE COPOLYMERS FOR MULTILAYER FLEXIBLE PACKAGING APPLICATIONS

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ABSTRACT: Ethylene-Methyl Acrylate (E-MA) copolymers have been used for many years to improve the interlayer adhesion of tie layers in multilayered flexible packaging. A key property of E-MAs in this application is their excellent adhesion to many different types of polymer substrates. The E-MA blends with low density polyethylene (LDPE) commonly used in industry today sometimes exhibit marginal bond strengths and can also cause processing difficulties.

The first of a new series of E-MA resins has performed significantly better than the equivalent E-MA/LDPE blend in initial extrusion coating and laminating pilot tests. Nearly a 50% improvement in adhesion to OPP film was obtained and similar results were observed for other substrates. The new E-MA resin also demonstrated good extrusion coating processing making it an attractive option for the flexible packaging manufacturer.

KEY WORDS: Ethylene Methyl Acrylate copolymers, E-MA laminating, extrusion coating, interlayer adhesion, flexible packaging, bonding.

INTRODUCTION

HIGH PRESSURE AUTOCLAVE and tubular reactors have been used for over 30 years to produce a variety of ethylene copolymers. Among the comonomers used are vinyl acetate, methyl acrylate, other acrylic esters, and acrylic acids. These comonomers modify the base polyethylene properties resulting in special characteristics which are valuable in a wide variety of end uses ranging from bonding/heat seal layers in flexible packaging to thermoplastic olefin compounds and blown films. This paper will focus on one specific copolymer, Ethylene-Methyl Acrylate (E-MA), on how its special features have been utilized in flexible packaging, and on the development of a new E-MA which offers improvement to the current industry practice of using E-MA/LDPE blends for interlayer bonding.

BONDING APPLICATIONS IN FLEXIBLE PACKAGING

A large percentage of the flexible films used to package food today are multilayer structures, which permit the packaging engineer to economically combine the unique properties of several different, and many times incompatible, polymers to provide the desired package performance. Examples of this type of property combination would be the use of polyvinylidene chloride copolymer (PVDC) or ethylene vinyl alcohol (EVOH) as a barrier layer coupled with an oriented polypropylene (OPP) film for mechanical properties and a polyethylene copolymer as a heat seal layer. Multilayer structures are also used for package appearance, for example to allow for reverse printing.

These multilayer structures are made either by extrusion laminating films together with a bonding polymer (tie resin), extrusion coating an outer layer onto a film, coextruding several polymers or some combination of these three processes. This article focuses on the first two techniques which are similar in that they involve the extrusion of a polymer(s) onto or between polymer films and on the use of E-MA in these applications. A schematic of a typical extrusion coating/lamination line is shown in Figure 1. Normally the polymers being extruded function solely to bond the films together in

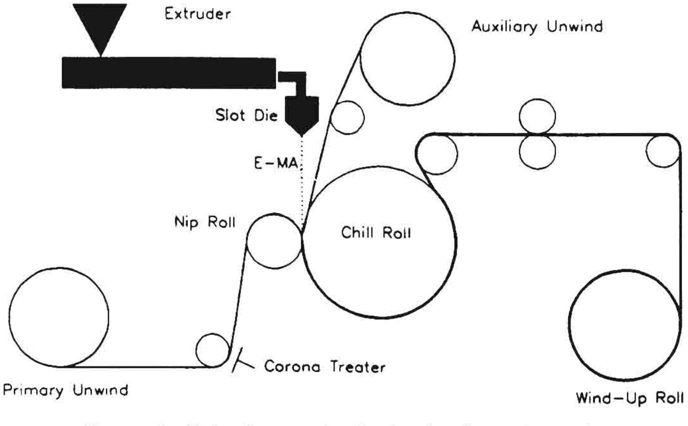

Figure 1. Extrusion coating/lamination line schematic.

Table 1. E-MA comparison to EVA, EAA, Ionomer.

Property	**E-MA**	**EVA**	**EAA**	**Ionomer**
Processing Range, °C	150–325	230 max	170–260	170–325
Thermal Stability	Excellent	Poor	Fair	Good
Corrosive	No	Yes	Yes	Yes
Moisture Sensitive	No	No	No	Yes
Adhesion to:				
Paper	Excellent	Good	Excellent	Excellent
OPP	Excellent	Poor	Poor	Poor
PVDC-Coated cello.	Excellent	Good	Poor	Poor
Al foil (unprimed)	Poor	Poor	Excellent	Excellent

the case of extrusion lamination or as a surface coating in the case of extrusion coating. E-MA's excellent adhesion to many different packaging substrates has led to it's inclusion in many of these laminations.

KEY E-MA PROPERTIES FOR BONDING APPLICATIONS

E-MAs have been used in a wide variety of applications for many years. The methyl acrylate comonomer adds polar functionality to polyethylene, lowering the copolymer's crystallinity and melting point, while still maintaining its excellent thermal stability. The polar functionality translates into a high level of compatibility with many different polymers and the ability to bond well to those materials. A low melting point gives E-MAs low heat seal initiation temperatures. A unique characteristic of E-MAs versus other copolymers are their excellent thermal stability and a wide range of extrusion processing temperatures. This gives packaging manufacturers the ability to optimize their process by tailoring operating conditions to the other materials being used.

Table 1 compares the extrusion processing and adhesion characteristics of E-MA to other ethylene copolymers commonly used in flexible packaging applications.

DEVELOPMENT OF A NEW E-MA FOR BONDING

Basis for Development Program

Because of its excellent adhesion to OPP films, a common use of E-MA in flexible snack food packaging is as the bonding layer in OPP/OPP structures. A current industry practice is to blend a low density polyethylene (LDPE) extrusion coating resin with an E-MA containing 20 wt% MA comonomer. Blends are employed to achieve the minimum thickness necessary for acceptable bond strengths and to reduce the tie layer cost; this was necessary in the past because E-MAs with different MA concentrations were not readily available. The blends gave the fabricator the ability to optimize the tie layer performance/cost balance; but in actual practice blend adjustments are seldom made because of the desire to maintain a consistent and reliable manufacturing operation.

The commonly used E-MA/LDPE blends have also been shown to have two significant weaknesses. First, it is well-known that adhesion in extrusion lamination is highly process dependent so, unless tight process control is maintained, bond strengths can vary significantly. The typical E-MA/LDPE blends (10–40 wt% E-MA) produce bonds which are marginally above the final package specifications so normal variations in the laminating process can easily result in off-spec product which is now a highly value-added multilayer structure. A second disadvantage concerns the processing characteristics of the blends. An E-MA containing 20 wt% methyl acrylate forms pellets which are fairly soft, rubbery, and slightly tacky. Dry blends of these with the harder LDPE pellets tend to segregate in the blend and feed hoppers of the extrusion laminator. This segregation yields variable blend levels which translate into additional bond strength variability. One possible solution to overcome these problems is to replace the blends with E-MAs having similar MA concentrations to the E-MA/LDPE mixtures. An easy approach to these "new" E-MAs would be to melt blend a 20% E-MA and LDPE, but this solution still wouldn't solve all the converters' bond strength problems.

Taking a more basic look at the bonding problem, a better solution was envisioned. The affinity of E-MA for OPP film is due to the MA functionality, so the LDPE in the E-MA/LDPE blend serves only as a diluent for the MA. If an E-MA copolymer with an MA content equal to that in the blend was made directly, all the polymer chains would theoretically contain the MA functionality, thereby increasing the E-MA's adhesive strength. This is the concept behind this portion of Exxon's current E-MA development program.

Development Plan and Experimental Methods

As an initial test of the above concept, a small batch of an E-MA, recently designated as TC-606, was produced during a standard E-MA production run. The TC-606 was made with a melt index of 6.0 and a methyl acrylate comonomer content of 6 wt%, approximately the MA content of a typical E-MA/LDPE blend used commercially. Two extrusion coating and laminating pilot line trials of the TC-606 were conducted in 1987 to compare its

Table 2.

	TC-110	TC-120	TC-606	E-MA/LDPE Blend
Melt Index	2.0	6.0	6.0	6.0
Methyl acrylate content, %	20	20	6	6

performance to that of the equivalent E-MA/LDPE blend and to the neat E-MA resins containing 20% MA. Neat E-MAs with melt indices of 2.0 (TC-110) and 6.0 (TC-120) were used. The E-MA/LDPE blend had a melt index of 6 (see Table 2).

Because the interlayer adhesion performance is a strong function of the coating and/or laminating conditions used, the experimental plans for both trials specified side-by-side comparisons to be made under identical process conditions. No attempt was made to optimize processing conditions to maximize the bond strengths or coating line speeds. Instead typical E-MA/LDPE coating conditions were employed for these initial investigations. Further pilot plant trials are now in progress to study the effect of processing on bond strength. Oriented polypropylene film was selected as the primary substrate for these initial trials because the most common flexible packaging use of E-MAs in bonding is in conjunction with OPP films. Several other films were laminated to an OPP primary substrate to compare the adhesion

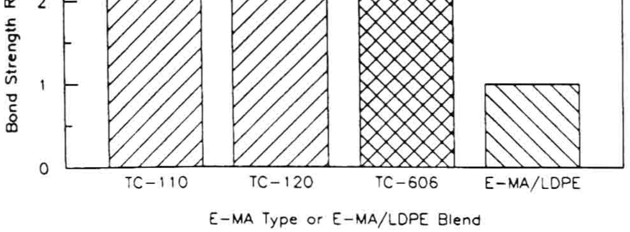

Figure 2. *E-MA adhesion comparison—Trial #1.*

of the E-MA/LDPE, the TC-120, and the TC-606 to other types of polymer substrates. A more extensive substrate scan is now in progress.

The interlayer bond strengths of the various coated/laminated structures were tested after aging for two weeks. Standard 90 degree T-peel tests were run on an Instron tensile test apparatus. Several different test methods were employed in an effort to eliminate the effect of the E-MA layer elongation on the bond strength testing. For all tests, side-by-side comparisons for each substrate were run. Since the absolute values of the bond strength varied from test method to test method, the results presented here were normalized within each test set to the values obtained with the control E-MA/LDPE blend.

Heat seal strength was also evaluated at a constant seal temperature, seal dwell time and sealing pressure. The sealing temperature used was the lowest temperature at which seals could be produced for all the structures tested.

EXTRUSION COATING/LAMINATING TRIAL RESULTS

Extrusion Coating Trial #1 Results

The first extrusion coating trial of TC-606 was run on a pilot line capable of high-speed operation and computer controlled. This line was a relatively large pilot line containing a 3½-inch extruder, a 2½-inch coextruder, chemical priming, corona treating, and normally operating at a 500 mm coating width. Maximum line speeds of 600 m/min were attainable. The four test materials, TC-110, TC-120, TC-606 and the E-MA/LDPE blend, were extrusion coated onto OPP film under conditions of constant temperature and coating thickness. The comparison of the aged bond strengths is presented in Figure 2. As expected the E-MAs produced significantly better bonds than the E-MA/LDPE blend. In fact the bond strengths were such that separation of the layers for testing was difficult. The blend equivalent, TC-606, was found to outperform the blend substantially and was nearly as good as the E-MAs having over three times the methyl acrylate content. This was in agreement with our supposition that spreading the MA functionality over all the polymer chains would improve the resin's adhesion capability. Also, Figure 2 indicates that the adhesive strength of the 20% E-MAs is significantly reduced when they are diluted with LDPE having a low bond strength to OPP film.

The coated OPP structures were heat sealed on a Packforsk heat seal/hot tack apparatus and tested at 100°C for heat seal strength after aging for 24 hours at room temperature. Those results are shown in Figure 3. Here the

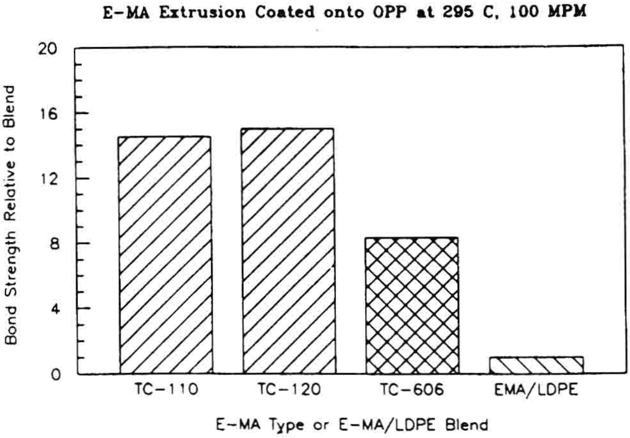

Figure 3. *E-MA heat seal comparison—Trial #1.*

performance of the TC-606 is again better than the blend but now the resins with a higher MA content produce significantly better heat seals. This was expected since the lower melting points of the TC-110 and TC-120 should give better heat seals at lower sealing temperatures. It appears that the melting point of the LDPE component of the blend, which is higher than that of TC-606, is the controlling factor in the heat seal capability of the blend. An initial estimate of TC-606's processing characteristics was obtained by measuring the maximum drawdown attainable under constant extrusion conditions. The results are shown in Figure 4. As can be seen the drawdown

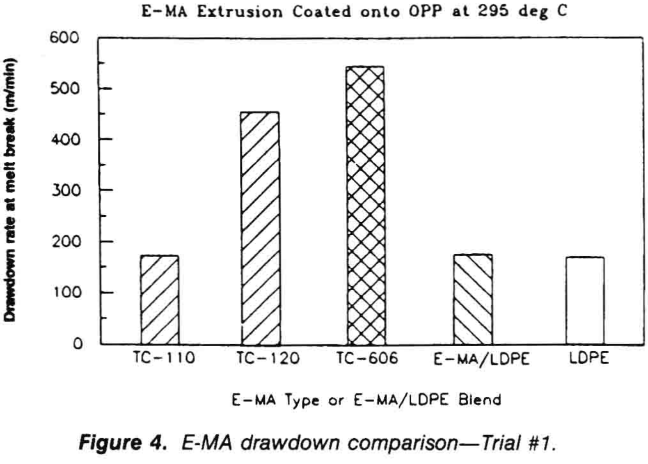

Figure 4. *E-MA drawdown comparison—Trial #1.*

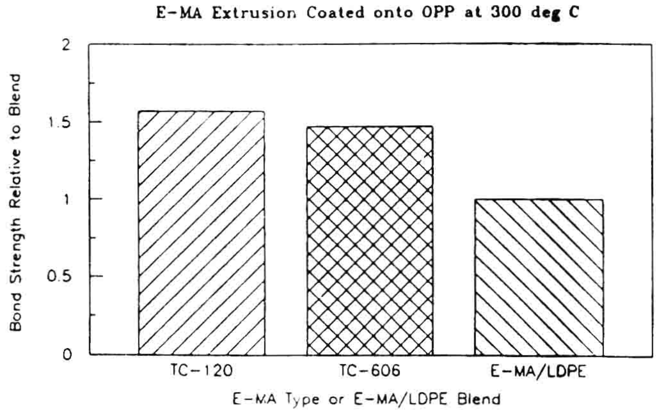

Figure 5. *E-MA adhesion comparison—Trial #2.*

capability of the TC-606 is similar to the drawdown performance of the other 6 melt index E-MA resin, TC-120.

Extrusion Coating/Laminating Trial #2

Based on the encouraging results obtained during the first extrusion coating testing of our new TC-606 E-MA, a second pilot line run was made to attempt to replicate the initial trial results and to evaluate TC-606's potential for bonding to substrates other than OPP. The second trial was conducted on a smaller, less instrumented extrusion coating and laminating pilot line. Slightly different processing conditions were used, so the data from the two pilot runs is not directly comparable, but again in trial #2 side-by-side tests on each substrate were run. Figure 5 shows the adhesion performance obtained for the extrusion coating onto OPP film (different type OPP film than in trial #1). The TC-606 E-MA again produced aged bond strengths that were almost equivalent to TC-120 which has a 20% MA content. The bonds were almost 1½ times as strong as those obtained with the E-MA/LDPE blend with the same MA content as TC-606.

E-MA's ability to adhere to films other than OPP was then investigated by laminating two different films, an oriented polyester (OPET) and Saran® HB PVDC, to the OPP primary substrate. The bond strengths between the OPET film and the E-MA are shown in Figure 6. For OPET the blend gave very poor bonds and peeled readily, whereas the E-MAs produced good

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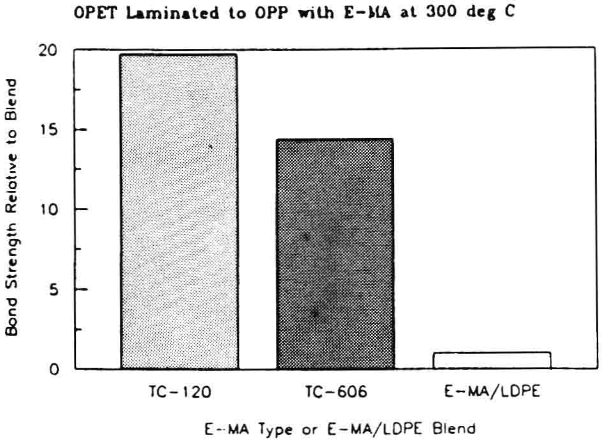

Figure 6. *E-MA adhesion to OPET film—Trial #2.*

bonds. The commercial significance of the bond improvement attainable through the use of an E-MA with a higher MA content will need to be evaluated in the actual application to determine whether the increase in bond strength is necessary. Figure 7 presents the performance obtained with Saran HB film. Here again the blend did not adhere to the Saran adequately and the OPP and Saran HB films were easily peeled apart. The difference between the TC-120 and TC-606 bond strengths is larger in this case but the lower strength bonds were still of very good quality.

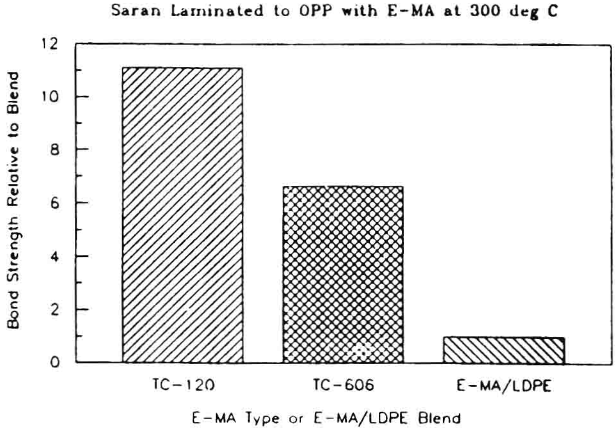

Figure 7. *E-MA adhesion to Saran HB—Trial #2.*

CONCLUSIONS

The initial extrusion coating and laminating trials of a new E-MA containing a lower MA content than is now normally available have shown that the adhesion performance of this resin is superior to that of a blend of E-MA and LDPE having an equivalent MA level. These results indicate that the presence of the methyl acrylate functionality in the polymer chains determines the E-MA adhesion performance and that the LDPE in the blend serves only as a diluent. The excellent adhesion and processing characteristics of this new TC-606 E-MA make it and other similar E-MAs attractive alternatives to the E-MA/LDPE blends currently used commercially.

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