



Neat EVOH and EVOH/LDPE blend centered three-layer co-extruded blown film without tie layers



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ABSTRACT

Ethylene–Vinyl Alcohol (EVOH) Copolymer incorporated multi-layer co-extruded film usually requires tie layers to bond EVOH to Low Density Polyethylene (LDPE). This study investigated various combinations of neat EVOH and EVOH/LDPE centered three layer co-extruded blown film without tie layers. LLDPE-g-MAH used as the compatibilizer, was blended in different layer locations. Oxygen transmission rate, peel strength between layers, tensile and dart impact strength of the films were measured to assess the appropriateness for food packaging. The study found that the effect of a compatibilizer on a three-layer film's properties varies when the EVOH concentration, layer combinations, and locations of the EVOH and LLDPE-g-MAH are changed. The study identified the 15 wt% EVOH/LDPE blend centered film as a possible film structure for future investigation due to its trade-off between barrier and layer peel strength.

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1. Introduction

Low Density Polyethylene (LDPE) is a widely used packaging film in food packaging. It can be used as blown film and as the sealing layer of a laminate. LDPE film is good because of its ability as a moisture barrier, processing ability, heat sealing properties, and has a reasonably good tensile strength. However, LDPE acts as poor oxygen and aromatic barrier. Ethylene–Vinyl Alcohol (EVOH) Copolymer is known to behave as an excellent oxygen barrier and is now increasingly seen as a better barrier layer than aluminum foil and metallized film due to its recyclability and transparent nature (Barlow & Morgan, 2013; Franklin Associates, 2014). However, EVOH tends to absorb moisture because of the presence of hydrophilic hydroxyl groups in its polymer structure, which limits its application mostly to the center layer in laminates or multi-layer co-extruded film. Combining LDPE and EVOH into one structure makes an ideal film for food packaging. The LDPE film, as an outer skin layer slows down the diffusion of water molecules into the EVOH layer in the center, and at the same time acts as a sealing layer and print layer. EVOH layer blocks the diffusion of oxygen molecules into the inner LDPE layer.

EVOH resins have very poor adhesion to most polymers except nylons. A tie layer is usually needed to combine the LDPE and EVOH

together in a lamination or co-extrusion process. A typical commercial LDPE/EVOH structure is a five-layer structure, namely LDPE/tie/EVOH/tie/LDPE. The tie resins used for EVOH and LDPE adhesion are based on graft polymers such as Plexar™ (Equistar Plastics), Bynel™ (E. I. du Pont de Nemours Co.), and Modic™ (Mitsubishi Petrochemical Industries, Ltd.). Applying a tie layer into a five-layer co-extrusion process is a complex and expensive process. A number of research projects based on a single extruder and three-layer co-extruded film were conducted; starting during the late 90s to incorporate the Linear Low Density Polyethylene (LLDPE) grafted Maleic Anhydride (LLDPE-g-MAH) as a compatibilizer into LDPE/EVOH film composites.

Kim and Chun (1999) blended 90% wt.LDPE and 10% wt. EVOH into a blown film with aa with LLDPE-g-MAH. They found that the oxygen barrier properties reached optimal values in the presence of 4 wt% LLDPE-g-MAH. Lee and Kim (1997, 1998) developed LDPE/EVOH/LLDPE-g-MAH blown film using Admer™ as a compatibilizer based on a single screw extruder and annular blown film die. The crystallization and melting behavior of the LDPE were significantly affected by the presence of Linear Low Density Polyethylene (LLDPE)-g-MAH. Huang, Wu, and Huang (2003) applied Modic™ to LDPE+EVOH blend and produced a blown film based on a single extruder. Morphological, thermal, and mechanical properties of the LDPE/EVOH with different concentrations of LLDPE-g-MAH were studied. The oxygen barrier properties of the biaxially oriented blown films decreased upon increasing the amount of the compatibilizer. Mechanical

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Table 1
Barrier and mechanical testing method.

Test	Units	Method	Equipment
OTR	cm ³ /m ² day atm	ASTMD 3985	Moccon OXTran [®] 2/20
WVTR	g/m ² day atm	ASTM F 1249	MOCON PERMATRAN- W [®] 3/33.
Tensile strength	Mpa	ASTM D 882	INSTRON 4301
Peel strength	grams	ASTM D 1876	INSTRON 4301
Dart impact test	grams/mm	ASTM D 1709	Dart impact tester (Testing Machines Inc)

measurements of the produced LDPE/EVOH film showed that there exists an optimal amount of LLDPE-g-MAh for maximizing both the tensile and tear properties in both the machine and transverse directions. The barrier, mechanical and thermal properties of LDPE/neat EVOH/LDPE blown film with various EVOH concentrations were produced (Ge, Lei, & Aldi, 2015). The study found that a trade-off between the peel strength and barrier properties appears to be the film combination of 7.5–10 wt% EVOH center layer and 2 wt% LLDPE-g-MAh blended outer LDPE layers, which retains some peel strength without compromising the barrier properties.

Villalpando-Olmos, Sánchez-Valdes, and Yáñez-Flores (1999) produced LDPE/EVOH/LDPE cast film with 17 wt% EVOH in the center layer and 83% LDPE layer with LLDPE-g-MAh in the outer layers. They found that when the amount of the g-MAh reached 0.08 wt%, the adhesion between film layers increased significantly, and there was no delamination found. The oxygen and moisture barriers remained unchanged. Huang, Wu, and Huang (2004) blended the LLDPE-g-MAh into an outer skin layer and produced a three layer LDPE+ LLDPE-g-MAh/EVOH/LDPE+ LLDPE-g-MAh blown film. The study found that the tensile strength of the LLDPE-g-MAh added film did not change. The peel strength increased sharply when the amount of LLDPE-g-MAh was greater than 12.5 wt% due to interactions between EVOH and LLDPE-g-MAh.

While the above studies reflect the efforts of blending LDPE and EVOH layers together without tie layers to develop sustainable and cost effective film, only two studies dealt with the multi-layer co-extruded film: the three-layer co-extruded blown film (Huang et al., 2004) and the three-layer co-extruded cast film from Olmos's study (Villalpando-Olmos et al., 1999). The LLDPE-g-MAh in both studies was added into the outer skin LDPE layer only. Some properties derived from the studies, such as tensile strength did not reflect the EVOH concentration. In addition, the concentrations of LLDPE-g-MAh between the two studies are also not comparable. No studies were found to address scenarios where the LDPE + EVOH blend act as the center layer in the three-layer co-extruded film, and what impact the location of the LLDPE-g-MAh has when positioned in the center layer instead of the outer layers. In addition, two important packaging film's characteristics are not found in any of the aforementioned studies: Dart impact value and crystallinity. Therefore, a systematic study is needed to investigate the effect of the compatibilizer on properties in packaging applications with

variables in EVOH concentration, combination of the center layer, as well as the location of the compatibilizer in a three-layer co-extruded blown film.

This study aims to investigate three basic combinations of three-layer co-extruded blown film LDPE/EVOH/LDPE without tie layers. They are: a) neat EVOH as a center layer sandwiched by LDPE blended with the LLDPE-g-MAh. i.e. LDPE + LLDPE-g-MAh/EVOH/LDPE + LLDPE-g-MAh, b) LDPE + EVOH blend as a center layer sandwiched by LDPE blended with the LLDPE-g-MAh; i.e. LDPE + LLDPE-g- MAh/EVOH + LDPE/LDPE + LLDPE-g-MAh, and c) LDPE + EVOH blend with LLDPE-g-MAh as a center layer sandwiched by LDPE; i.e. LDPE/EVOH + LDPE + LLDPE-g-MAh/LDPE. The EVOH concentrations in the film were set to 5 wt% and 15 wt%. The compatibilizer LLDPE-g-MAh resins were set as 0.5 wt%, 1 wt%, and 2 wt% as a higher loading of the LLDPE-g-MAh causes the reduction of the barrier properties (Huang et al., 2004). The purpose of this study is to find a film combination that could be an alternative to LDPE/tie/EVOH/tie/LDPE, which barrier and peel strength are acceptable by industries.

2. Experimental

2.1. Materials

The Petrothene[™] NA960 LDPE was acquired from LyondellBasell, USA and is a blown film grade ($\rho = 0.92 \text{ g/cm}^3$). Eval[™] E171B EVOH ($\rho = 1.14 \text{ g/cm}^3$) was supplied from Kuraray, USA and possesses 44% ethylene mol. content. The OTR of extrusion grade EVOH Eval[™] E171B is $1.5 \text{ cm}^3/\text{m}^2 \text{ day atm}$. This value was measured with an environment of 20 °C, 65%RH, and the film thickness was 20 μm . The modified LLDPE grafted with Maleic Anhydrid Bynel[™] (MAh) 41E687 from Dupont, USA was used as a compatibilizer to improve layer-to-layer adhesion of the coextruded film ($\rho = 0.91 \text{ g/cm}^3$). Each film layer contains 5 wt% and 15 wt% of EVOH as a center layer to LDPE of the total mass of the film. LLDPE-g-MAh resins with 0.5 wt%, 1 wt%, and 2 wt% were added to the total percent of the individual outer layers.

The different weight percentages of EVOH and LDPE were realized by setting the extruders to the appropriate rotational speed ratios assuming that the screws had the same volumetric flow rate. Both EVOH and compatibilizer loading were percentage of the total blend mass added to the center layer and outer layers. The amount of the LLDPE-g-MAh concentration was added to the pelletized LDPE blend prior to tumbling, drying and processing.

Table 2
Film thickness.

	LDPE/EVOH/LDPE- neat EVOH centered		LDPE/EVOH + LDPE/LDPE- EVOH/LDPE blend centered	
	5 wt% EVOH	15 wt% EVOH	5 wt% EVOH + LDPE	15 wt% EVOH + LDPE
Measured film thickness (μm)	35.6 ± 0.082	35.6 ± 0.041	35.6 ± 0.052	35.6 ± 0.121
Layer thickness (μm)	17.0/1.8/17.0	14.0/7.62/14.0	12.8/10.0/12.8	12.8/10.0/12.8

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