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Controlling interfacial instabilities in PP/EVOH coextruded multilayer films through the surface density of interfacial copolymers

Stéphanie Vuong, Liliane Léger, Frédéric Restagno

Abstract

Coextruded polypropylene/tie/ethylene vinyl alcohol/tie/polypropylene (PP/tie/EVOH/tie/PP) films often exhibit optical defects which appear as randomly distributed scattering objects, in the submillimeter range. These defects may strongly affect the film transparency and prevent their practical use in packaging. Based on an objective optical test aimed at quantifying the film transparency, and on a systematic analysis, through optical microscopy, of transverse cuts of films obtained in various coextrusion conditions, the nature of the defects could be identified as resulting from a modulation of the thickness of the inner ethylene vinyl alcohol (EVOH) layer, with no variation in the overall thickness of the multilayer films. Thanks to a recently developed method to dose the surface density of interfacial copolymers, a clear correlation between the amplitude of the thickness modulation of the inner EVOH layer and the density of copolymer molecules formed in situ at the EVOH/tie layer interface during the coextrusion process was established. These results open the way to a better design of tie layers composition to avoid these kinds of defects.

1 | INTRODUCTION

Nowadays, industries often need to design composite materials offering specific complementary properties. Due to their lightweight, polymers are one of the most used materials to answer this type of demand, in particular through the possibility of forming multilayer films. The successful association of different polymers in order to benefit from their different properties (eg, mechanical, dielectric, or optical properties)^[1-4] within a unique material is however still a challenge because most of the polymer pairs are not miscible, and develop very weak adhesion when forced into contact.^[5] The fabrication of

multilayered polymer materials is not simple, as the interfaces need to be reinforced mechanically. Such necessary adhesion enhancement is usually produced by diblock copolymers that are carefully designed, so that each block is compatible with one of the materials in contact. The copolymers tend to assemble at the interface, and they stabilize it.^[6-12] For practical applications, these copolymers are created in situ during the mixing process by an interfacial chemical reaction between reactive polymers initially mixed within each of the joined polymers.

Food packaging often uses multilayer films made of polypropylene/ethylene vinyl alcohol (PP/EVOH), to form transparent films, allowing one to see what is inside

the package, and displaying good mechanical and oxygen barrier properties. In such PP/EVOH multilayer films, the interface is stabilized by ethylene vinyl alcohol-copolypropylene grafted maleic anhydride (EVOH-co-PP-g-MA) copolymers formed at the interface during the coextrusion process (Figure 1). To achieve this goal, a tie layer is inserted between polypropylene (PP) and ethylene vinyl alcohol (EVOH) layers. The tie layer is a mixture of a small quantity of maleic acid grafted PP (PP-g-MA) with unmodified PP (that can differ from the external PP, but is fully miscible with it). The cohesion between the tie layer and the PP layer is ensured by interdiffusion of PP chains through the PP-tie layer interface when both are into contact and heated above the melting transition temperature of PP. The EVOH-co-PP-g-MA copolymers are synthesized in situ during the coextrusion process by the chemical reaction of PP-g-MA chains on EVOH chains, as schematically shown in Figure 1. The final five layers polypropylene/tie/ethylene vinyl alcohol/tie/polypropylene (PP/tie/EVOH/tie/PP) films present good mechanical and barrier qualities. However, these

coextruded films often show optical defects, which affect the film transparency. Surprisingly enough, these defects are not observed with other quite similar associations, like for example polyamid6 (PA6)/polypropylene. These defects lead to a grossly frosted glass aspect, identified in this article as *granity defects*. The associated loss of transparency is not acceptable for a number of food packaging applications. A typical example of *granity defects* is displayed in Figure 2, where two multilayer assemblies with different transparency qualities are shown. Both tested films are placed at 20 mm from the book entitled “*Physical Polymer Science*.”^[13] The title can (a) easily be read through film and (b) with difficulties through film.

The goal of the present article is to present a systematic investigation of multilayer PP/tie/EVOH/tie/PP films, aimed at identifying the mechanisms that can explain the nature and the origin of transparency defects in coextruded films. As these defects reveal themselves by scattering light, they could be due either to local variations of the optical properties of one or several of the constituting polymers layers, or to interfacial instabilities

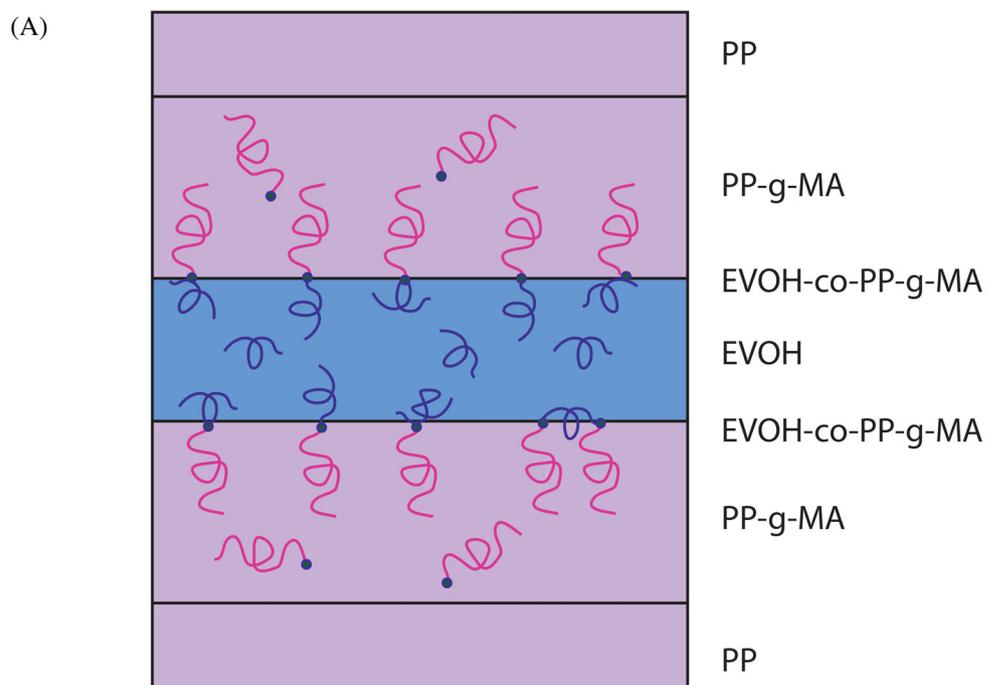


FIGURE 1 A, Schematic of a five layers film: PP/tie/EVOH/tie/PP. EVOH-co-PP-g-MA copolymer is created at the interface. B, Chemical reaction of the copolymer formation. EVOH-co-PP-g-MA, ethylene vinyl alcohol-copolypropylene grafted maleic anhydride; PP/tie/EVOH/tie/PP, polypropylene/tie/ethylene vinyl alcohol/tie/polypropylene [Color figure can be viewed at wileyonlinelibrary.com]

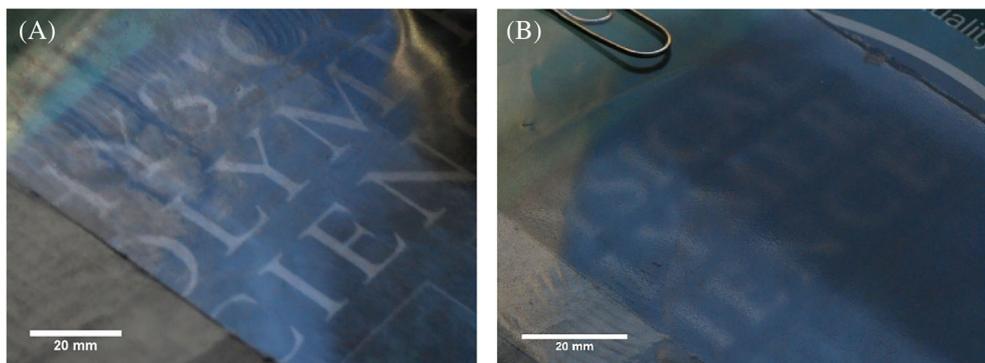


FIGURE 2 Coextruded polypropylene/tye/ethylene vinyl alcohol/tye/polypropylene (PP/tye/EVOH/tye/PP) films with different degrees of severity of the *gravity defects*. One should notice that the zoom of the picture A is slightly higher than the picture B [Color figure can be viewed at wileyonlinelibrary.com]

that would modulate the total thickness of the film or the thickness of the internal layers.

Interfacial instabilities in coextrusion process have been widely studied in the case of nonreactive polymers. In some experimental approaches, based on the analysis of extrudates formed in industrial configurations, stability charts function of process parameters, and/or relative rheological parameters of the polymers were proposed.^[14-17] Those studies showed that stable/unstable transitions were characterized by the development of wavy interfaces between polymers. In further experimental studies, the development of instabilities within the flow was investigated.^[18-22] Main results show that viscoelastic stresses stratification across the interface is the dominant factor in the instability.^[23] However, apart for simple asymptotic cases (extremely long or short waves), the exact physical mechanisms at the origin of the instability are still unknown. Moreover, it was recently shown that interfacial chemical reactions can drastically affect the stability of an interface. The impact of interfacial reaction in coextrusion still needs to be understood.^[24,25]

Several studies point out clear links between the contrast in rheological properties of joined polymers and the interfacial instability. However, rheology alone cannot explain the observed different sensitivities to *gravity defects* between PP/tye/PA6/tye/PP and PP/tye/EVOH/tye/PP. Other mechanisms should contribute that need to be identified. Recently, in close collaboration with the results presented in the present article, Bondon et al^[26] have analyzed, through electronic microscopy, the interphase formed between EVOH and tie layer (PP-g-MA plus PP) for lab-formed assemblies. They have evidenced a roughening of the interface at nanometric scale when the interfacial reaction between EVOH and PP-g-MA was developing. This roughening of the interface was found to be associated to an abnormal mechanic relaxation of the interphase as measured by rheometry.^[24,25]

In order to fill the gap between lab formed and extruded films, we present below the results of systematic experiments aimed at identifying the nature of the optical

defects responsible for the appearance of *gravity defects*, and trying to better pinpoint the role of interfacial copolymers. To do so, we have analyzed in a systematic manner these *gravity defects*, proposing an objective test to quantify the loss of transparency. In order to identify more precisely the origin of that loss of transparency, we have analyzed in detail optically thin cuts of multilayer films, to try to answer the following questions: which microscopic parameters cause this macroscopic optical defect? Why, using the same materials, but varying the coextrusion parameters, the optical quality of the films can be so different? What are the parameters controlling the appearance of *gravity defects*? We present here the main results of this investigation.

2 | MATERIALS

The studied coextruded films are five layers multi-materials PP/tye/EVOH/tye/PP (50/10/8/10/50 μm). They were kindly provided by ARKEMA. Tie layers are mixtures of PP-g-MA in PP. Nine samples were investigated. They differ from the process conditions and formulations of PP-g-MA (PPg1 or PPg2 or PPg3). PPg2 was formulated in reactive extrusion and PPg3 in solution. Except for average maleic anhydride (MA) per chain, the precise characteristics of the different polymers cannot be divulged for confidentiality reasons. Average MA/chain is <1 for PPg1, ~ 1 for PPg2, and 3-6 for PPg3. EVOH contains 32% ethylene and 68% enol. The properties of the nine samples used are summarized in Table 1. The type of formulation together with the process conditions (PP processing temperature, PPg processing temperature, EVOH processing temperature, and residence time (ie, the time during which the materials are inside the feed-block and in the air gap before crystallization) are identified for each sample. The residence time is the time during which the tie layers and EVOH are molten and into contact, that is, the time during which the interfacial copolymers can be formed. PPg1, PPg2, and PPg3 were

TABLE 1 Formulation and process conditions of the samples

Samples	Tie layers	T_{PP} (°C)	T_{PPg} (°C)	T_{EVOH} (°C)	Residence time (s)
1	PPg1	220	225	190	33
2	PPg2	220	225	190	33
3	PPg3	220	225	190	33
4	PPg1	200	200	190	31
5	PPg2	200	200	190	31
6	PPg3	200	200	190	31
7	PPg3	225	225	210	7
8	PPg2	225	225	210	7
9	PPg1	225	225	210	7

Abbreviations: EVOH, ethylene vinyl alcohol; PP, polypropylene; PPg, grafted polypropylene.

synthesized by ARKEMA, PP was PP RD204CF from BOREALIS (Total Petrochemicals) and EVOH was EVASIN 3251FS provided by ARKEMA.

In order to quantify the surface density of the interfacial EVOH-co-PP-g-MA copolymers in the EVOH/PP multilayers, a selective dissolution of the EVOH part of the coextruded films was needed. This step was achieved by several solvent and reagents: dichloromethane amylene stabilized (CarloErba Reagent), acetyl chloride (AC) (Sigma Aldrich) reagent grade, 98%. These reagents were purchased and used without any further treatment.

3 | FILM TRANSPARENCY AND INTERNAL LAYER THICKNESS

3.1 | Method

3.1.1 | Classification of the films

Several practical tests to characterize the ability of an object to transmit light and to quantify its transparency exist. These tests are separated into two categories: clarity meter and haze meter.^[27,28] A loss of transparency means that light is deviated by the object, due to scattering, diffraction, or refraction, depending on the nature and on the size of the heterogeneities responsible for local changes in the light/object interactions. The length scale of these optical heterogeneities fixes the cone angle inside which light is deviated. The optical aspect of a coextruded film presenting defects depends on the angle at which the light is deviated when crossing the film: small angles (less than 2.5°) refer to clarity and wider angles refer to haze. The first one affects the resolution when observing an object through the film, and the second one has an impact on the contrast. We decided not to perform these existing tests, because the *granity defects* impact both the resolution and the contrast. A test directly relevant for

the application has thus been developed, that is, quantifying the ability to clearly see an object located at a given distance from the film, when observing this object through the film.

The method is detailed in the following. A sheet (Figure 3) with letters of different sizes is observed through the film. The letters are sorted by size so that each line contains one size of letters. Each line (each size) corresponds to a mark, going from 1 to 11 (1 for the line with the biggest letters and 11 for the line with the smallest letters). When looking to the sheet through a given sample, the smallest letters the observer is able to distinguish will specify the mark affected to the sample. The more transparent is the sample, the smaller are the letters, which can be distinguished by the operator and the higher is the mark attributed to the sample. Three operators were involved in the present study. The respective distances between the sheet and the film and between the film and the eyes of the observer were fixed to respectively, 100 and 500 mm. These distances were kept constant during all series of experiments in order to compare samples meaningfully. One advantage of this setup is its low space-consuming aspect, so that it can be settled directly on-line at the exit of a process extrusion machine. It should be pointed out that it is crucial to fix the same distances between the film and the letters and the film and the operator for all experiments for the sake of comparison. The absolute mark can slightly differ according to the operator, but the raking between samples remains the same. Statistical data analysis of the marks from different operators has not been done.

3.1.2 | Microscopic characterization

As already mentioned in the introduction, a rapid and easy examination of the samples showed that the studied defects are not due to a degradation of the external

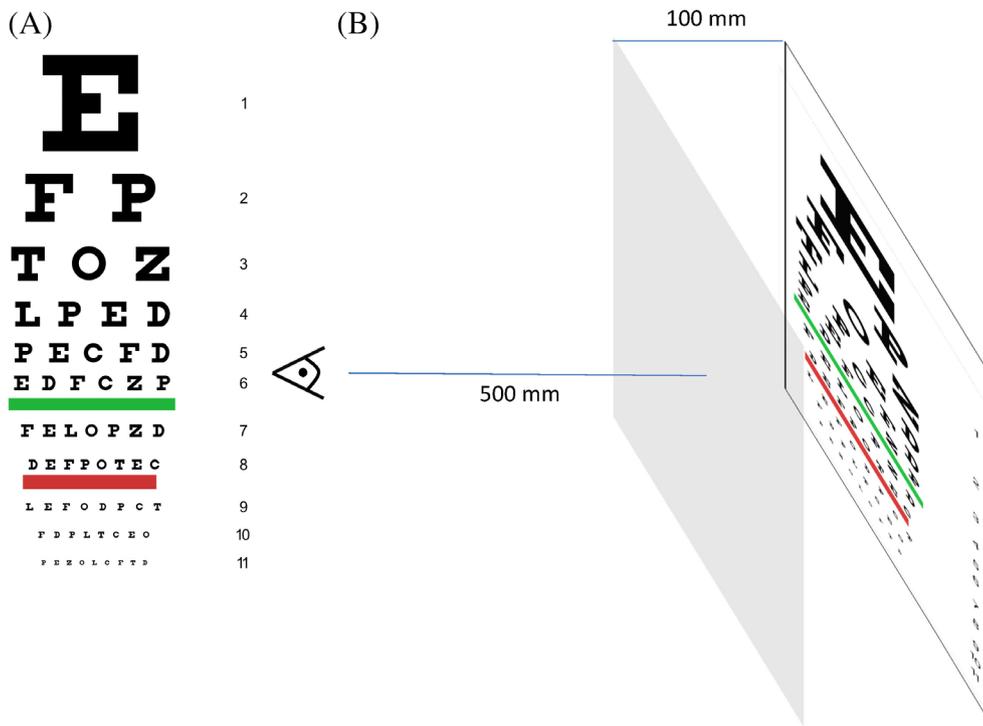


FIGURE 3 A, Pattern used to classify the quality of the samples. B, Testing setup [Color figure can be viewed at wileyonlinelibrary.com]

Sample	Mark	$\frac{\Delta E}{E}$	$\frac{\Delta E}{E}$	$\frac{\Delta E}{E}$	λ (μm)	λ (μm)	λ (μm)
		A	L	T	A	L	T
8	5	0.1	0.3	0.2	54.0	40.9	/
9	5	0.2	0.3	0.0	84.2	/	/
1	3	0.3	0.5	0.4	6.9	86.6	98.2
2	3	0.4	0.4	0.3	110.5	61.7	98.2
4	3	0.0	0.3	0.1	/	/	76.0
5	4	0.3	0.2	0.2	77.6	41.8	85.1
7I	4	0.3	0.3	0.1	107.3	/	/
3	1	1.2	1.4	1.0	/	/	/
6	1	0.8	0.7	0.6	/	/	111.7
7E	1	1.2	1.1	0.8	/	/	/

TABLE 2 Marks for all samples in regards with the amplitude of the undulation of the internal layer thickness. The samples containing spherulites are highlighted in gray

surfaces quality of the samples (roughness, thickness of the overall multilayer assembly...). Then, in order to identify the origin of the loss of transparency, systematic observations of transverse sections of the films were performed by optical microscopy.

For each sample, sections were cut to obtain around 10 μm thick chips. These cuts were performed at CNRS Imagif in Gif-sur-Yvette, with a cryogenic microtome LEICA, working at -80°C with a glass knife. These chips were immersed in a drop of an immersion oil (refractive index 1.518), sandwiched between two glass slides and observed with an optical microscope (Olympus BS51) with $\times 20$ or $\times 50$ magnifications. A particular attention

was paid to the orientation of these chips with respect to the direction of extrusion and are labeled in the following as: transversal (T), longitudinal (L), and random orientation (A). For each coextruded film, 12 chips, four of each orientation, were investigated.

3.2 | Results

3.2.1 | Marks

All samples were assigned a mark according to the optical quality test described above. These marks are

summarized in the second column of the Table 2. In this table, samples are organized from the mark 5 (the most transparent samples) at top, down to the mark 1 (the less transparent samples) at the bottom of the table. Only marks 5, 4, 3, and 1 were attributed, samples corresponding to the other quality mark were not in the series of film available. Regarding samples 5 and 7I and for all operators, some letters could be read through the film but not all of the letters in this line corresponding to mark 4. This heterogeneity of the results obviously came from the heterogeneity of optical quality within the same film. It has been decided to gather samples 5 and 7I (mark 4) with the group with mark 3. Regarding marks 5 and 1, appreciations of all operators were consistent. All examined coextruded films can thus be organized in three groups of different optical qualities. Finally, sample 7 exhibits obviously two areas with two different qualities: the lateral extremities of the film (7E) and the center (7I). These two parts were analyzed separately.

3.2.2 | Microscopic observation

Two most representative pictures of the chips, for each group of optical quality, are displayed in Figure 4.

4 | FILM TRANSPARENCY AND COPOLYMER GRAFTING DENSITY

Even if not suitable for any application, as the different layers spontaneously delaminate, multilayer films made

of pure EVOH and PP, where no interfacial copolymers can be formed during the coextrusion, do not show instabilities of the inner EVOH layer thickness. It is thus natural to search for a possible correlation between the amount of interfacial copolymer and the severity of the optical *grainy defects*. In order to do so, it is necessary to quantify the amount of interfacial copolymers actually present at the tie/EVOH interface. We briefly describe below the principle of the dosage method we have developed,^[29] and then correlate the analysis of the thickness variation of the inner EVOH layer to the surface density of copolymers formed at the tie/EVOH interface during the coextrusion process.

4.1 | Selective dissolution

To access the surface density of EVOH-co-PP-g-MA copolymer molecules present at the end of the coextrusion process at the buried tie/EVOH interface, parts of the multilayer films should be selectively dissolved, in order to exhibit the copolymer layer, and then to dose it through spectroscopic techniques. To do so, small pieces of a coextruded multilayer films were brought inside a container with AC vapor for a one-night reaction. The reaction modifies the EVOH layer, transforming alcohol functions into ester functions. This esterification contributes to weaken the hydrogen bonds between EVOH chains so that each five layers PP/tie/EVOH/tie/PP film can be split into two PP/tie/EVOH parts. Then, all EVOH chains not chemically linked to PPg were removed by immersions of three successive

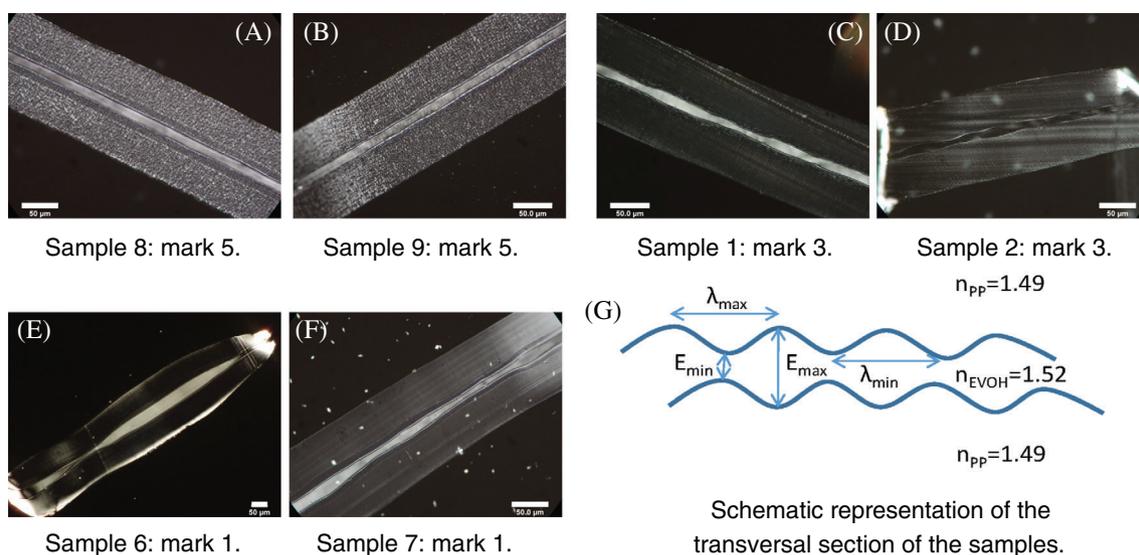


FIGURE 4 Typical pictures of microscopic observations: A, B, The films in the first line were attributed mark 5. C, D, The films in the second line were attributed mark of 3. E, F, The films in the third line were attributed mark 1. G, Schematic representation of the transversal section of the samples [Color figure can be viewed at wileyonlinelibrary.com]

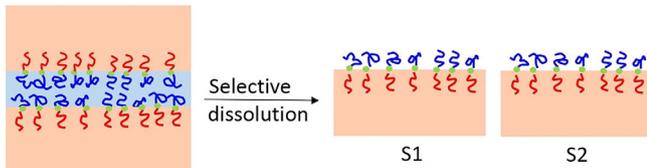


FIGURE 5 After EVOH selective dissolution of coextruded films, two sheets S1 and S2 are obtained [Color figure can be viewed at wileyonlinelibrary.com]

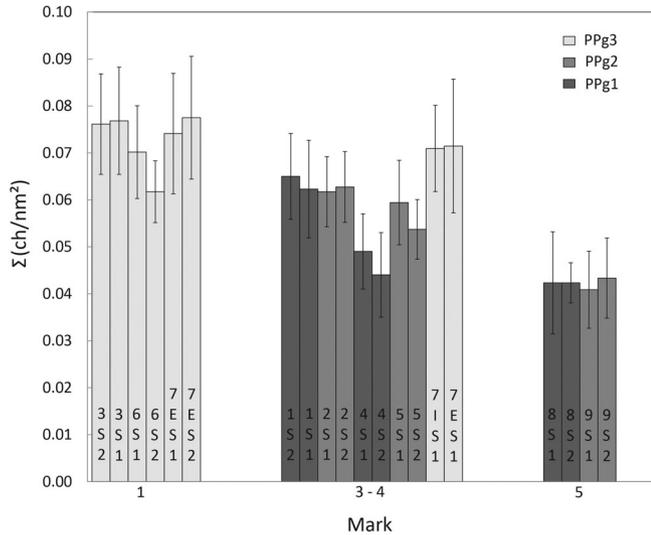


FIGURE 6 Surface density vs marks

stirred baths of dichloromethane. At the end of the selective dissolution process, two sheets of PP films (S1 and S2 in Figure 5) are obtained with esterified EVOH-co-PP-g-MA interfacial copolymers on one of their surfaces. The ester groups corresponding to this nanolayer of EVOH-co-PP-g-MA copolymers are then quantified by Fourier Transform Infra Red Spectroscopy (FTIR). Knowing the average molecular weight of the EVOH used, it is then possible to convert the infrared height peak ratio into surface density of EVOH-co-PP-g-MA. The precise protocol has been described by Vuong et al.^[29]

4.2 | Grafting density measurement by FTIR quantification of EVOH-co-PP-g-MA

To enhance the sensitivity of the analysis and meaningfully quantify the surface density of copolymers, the S1 or S2 films obtained after the selective dissolution were folded several times and conditioned in disks of 20 mm diameter and 202 μm thickness. These disks were analyzed in transmission and the ratio of the height peaks at

1738 cm⁻¹ (from ester signal) over that at 900 cm⁻¹ (from PP signal) X was determined. These peak ratios were converted into surface density Σ through the following relation:

$$X = \frac{\rho_{\text{PA6}} \Sigma N a^3}{\rho_{\text{PP}} h}, \quad (1)$$

where h the thickness of the film.

The surface density of copolymers vs the mark of the samples are displayed in the histogram displayed in Figure 6.

5 | DISCUSSION

5.1 | Mark and microscopic observations

The figure displayed in Figure 4 reveal two phenomena possibly responsible for a loss of transparency: spherulites appearing in the PP layer of some of the samples and thickness irregularities of the inner EVOH layer. Indeed, the transverse sections of the samples can be schematically drawn as in Figure 4G. As the refractive index of PP and EVOH are different, the thickness variation of the EVOH layer would locally transform the film in successions of converging and diverging lenses, which will deviate light in all directions, altering the imaging capacities of the film. Both the amplitude and the wavelength of these thickness variations of the inner layer should affect the degree of light deviation, making the film more or less hazy.

In order to identify which phenomena was mostly responsible for the loss of transparency of the films, the presence or the absence of spherulites has been spotted for each sample and the waviness of the inner layer has been quantified, measuring the wavelength and the amplitude of the thickness variations of the inner EVOH layer. The results are summarized in Table 2. The wavelength, λ , reported for each orientation is the average of λ_{min} and λ_{max} both defined in Figure 4G. For some samples, the wavelength could not be measured either because it is bigger than the size of the chip or because it could not be determined owing to the accuracy of the pictures or also because the number of observable thickness oscillations was too small to allow for an estimated average. These cases are symbolized with a slash in Table 2.

The amplitude of the thickness variation of the inner layer has been defined as follows:

$$\frac{\Delta E}{E} = \frac{E_{\text{max}} - E_{\text{min}}}{\bar{E}}, \quad (2)$$

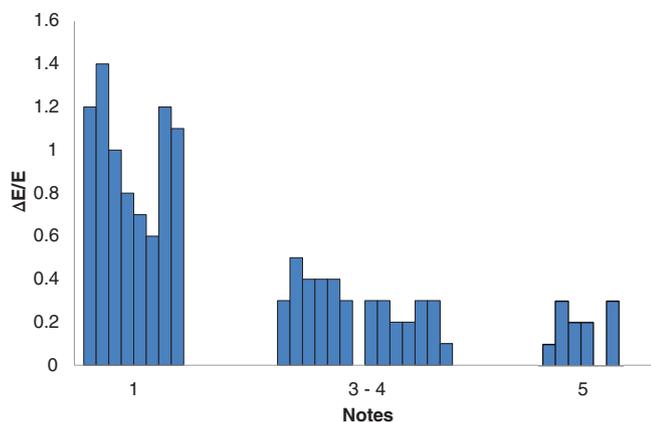


FIGURE 7 Amplitude of variation of the inner layer thickness vs notes of all the samples studied in this article [Color figure can be viewed at wileyonlinelibrary.com]

where $\overline{E_{\max}}$ and $\overline{E_{\min}}$ respectively the average of E_{\max} and E_{\min} as defined in Figure 4G and \overline{E} the average of all thickness measured.

The $\frac{\Delta E}{E}$ reported for all orientations is the average of $\frac{\Delta E}{E}$ in all directions. The lines corresponding to samples containing visible spherulites at the scale of observation are filled in gray in Table 2, while lines for which spherulites were not observable are unfilled.

Samples 8 and 9 with a mark 5 and sample 7 with a mark 3 (for a cut located at the center of the sample: 7I) or 1 (for a cut located at the extremity of the sample: 7E) display spherulites. Spherulites can thus be found whatever the optical quality, meaning that they are not responsible for the loss of transparency of the films, at least at first order.

Concerning the wavelength of the thickness oscillations of the inner EVOH layer, the quality of the data (number of thickness oscillations) collected in this study is not sufficient to distinguish a possible correlation between the value of this wavelength and the lack of transparency. The number of sections per sample should be increased to reasonably be able to conclude on this point.

The most relevant parameter which emerges from the analysis of the chips appears to be the amplitude of the thickness variation of the inner EVOH layer: whatever the orientation, this amplitude is observed to be in the range 0.0 to 0.3 for all samples with the mark 5; 0.1 to 0.5 for the samples with the marks 3 to 4 and 0.6 to 1.4 for the samples with the mark 1. There is thus a clear correlation between the amplitude of the thickness variation of the inner EVOH layer and the optical quality of the films. This correlation appears even clearer in the histogram shown in Figure 7, which gathers all the collected data. The larger is the amplitude of thickness variation, the less transparent is the film.

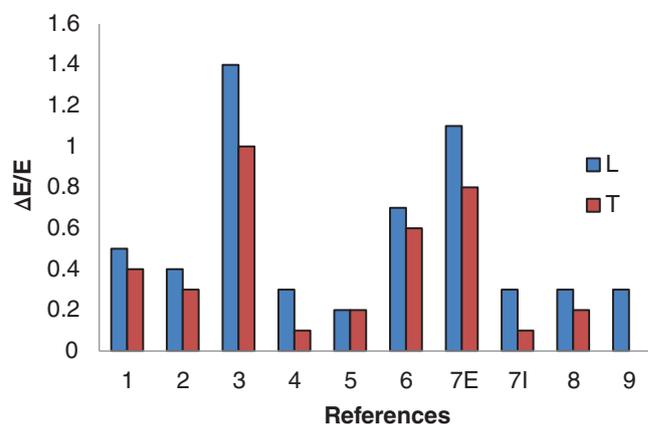


FIGURE 8 Amplitude of variation of the inner layer thickness for all samples, longitudinal orientation in blue and transversal orientation in red [Color figure can be viewed at wileyonlinelibrary.com]

One can notice that the amplitude of the thickness oscillations of the inner EVOH film observed in real coextruded film is much larger than that observed on the same polymer sandwiches but made with no shear.^[26]

Another interesting result is revealed by the histogram in (Figure 8). This histogram keeps tracks of the orientation L and T, width amplitude of the variations of the inner layer thickness reported in blue for the longitudinal orientation and in red for the transverse orientation. In the production line, films were immediately rolled, and the orientation of the roll was conserved during all the study allowing to be able to characterize the thickness variations of the inner layer in both directions. The amplitude of the thickness variation appears significantly larger for the longitudinal than for the transverse orientation. There is thus an anisotropy in the destabilization of the thickness of the inner EVOH Layer, with an enhanced destabilization in the direction of extrusion. We address in more details this phenomenon below.

5.2 | Link between the mark and the grafting density

A clear correlation between the surface density of copolymer and the optical quality of the films is visible in Figure 6: increasing the surface density of copolymers decreases the optical quality of the films (except for sample 7, which we shall discuss separately in the next paragraph). One can also notice that all the worst films (with the mark 1) correspond exclusively to one kind of tie: PPg3. We can conclude that the formulation of the tie is a key parameter if one wants to process films with good optical quality. But the formulation alone is not enough

to determine the optical quality of the films: some of the films formulated with PPg1 have obtained the mark 5 (sample 8) or the mark 3 to 4 (sample 1 and 4), a result also visible for PPg2 (mark 5 for sample 9 and mark 3 to 4 for samples 2 and 5). Anyhow, for each type of PPg, the best films are those made with the smallest surface density of copolymers, as shown in Table 1: all best films with mark 5 have been made with a small residence time (7 seconds) inside the coextrusion line, a condition which corresponds to a short open time for the grafting reaction between PP-g-MA and EVOH, and ensures a small surface density of copolymers.

The sample 7 demonstrates that the surface density of interfacial copolymers is not the only parameter influencing the optical quality of the final film. Indeed, within the same sample, the surface density of interfacial copolymers is significantly homogeneous, while for sample 7, the optical quality was depending on the area investigated. Differences in stress/flow history in the various zones of a film could be responsible for this additional dependence of the optical quality on the location in the whole coextruded film. We have at present no more precise analysis of the influence of the flow conditions on the final optical quality of the films. However, we have pointed out the importance of these flow conditions, through two observations: variation of the optical quality with the location within the film, and evidence of an anisotropy in the amplitude of the modulation of the inner layer thickness, with an enhanced instability in the direction of the main flow along the coextrusion line.

Finally, a still opened question remains the precise characterization of the precise molecular organization of the interfacial layer, which cannot be measured with the techniques developed in the present article.

6 | CONCLUSION

One category of optical defects observed in coextruded multilayer films used in food packaging has been investigated. These *granity defects*, responsible for a loss of transparency of the film, develop on the specific association PP/EVOH materials without any variation of the overall thickness of the film.

Optical microscopic observations of chips of films cut perpendicularly to the surface of the film reveal that the presence of visible spherulites inside the PP part of the films is not the main factor at the origin of the loss of transparency. Instead of that, fluctuations of the thickness of the inner EVOH layer were identified as the mean cause of the loss of transparency. We have proposed a simple method allowing one to objectively compare the optical quality of films formed with various coextrusion

parameters, and various kinds of reactive PPg chains, in order to be able to search for possible correlations between the structure, the surface density of copolymers, the coextrusion process parameters, and the optical quality of the films. We have thus put into evidence a general rule: the larger the surface density of interfacial copolymers EVOH-co-PP-g-MA is, the worst the optical quality is.

Finally, we demonstrated that the interfacial copolymers surface density is not the only parameter governing the optical quality of the multilayer films. This shows up through two different results: the optical quality depends on the position on the overall film, with a better quality in the central part of the film than along the lateral edges of the film, while both parts of the film contain the same surface density of EVOH-co-PP-g-MA copolymers. This suggests that stress and flow history during the coextrusion process are also important parameters. This sensitivity to the exact local flow conditions during the coextrusion process reflects in the fact, that we have put into evidence, that the amplitude of variation of the inner layer thickness is larger for chips cut along the coextrusion line (longitudinal section) than for chips cut transversally (transverse section).

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DATA AVAILABILITY

The raw/processed data required to reproduce these findings cannot be shared at this time due to intellectual properties limitations.

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