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Research article

Assessment of ultrasonic treatment on multilayer packaging composed of polyethylene and ethylene-vinyl alcohol copolymer

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ABSTRACT

While there is an increase in the non-conventional processing of foods, the impact of emerging technologies on packaging materials is scarce, limiting their application. This work evaluated the ultrasound (US) technology, applied alone or in combination with thermal processing (thermosonication), on the structure, properties and functionality of PE/EVOH38/PE multilayer flexible packaging films. Samples were processed using US (25 kHz, 9.74 W/L) alone (25 °C) or in combination with mild thermal processing (60 °C) for up to 120 min. Packaging structure, properties and functionalities were evaluated through FTIR, DSC, XRD, SEM, mechanical and barrier properties, as well as overall migration. US treatment induced variations in relative crystallinity (XRD and DSC), but without changing the molecular or microscope structure (FTIR, SEM) of the films. As a consequence, the packaging functionality was not affected, preserving the mechanical properties (tensile test at both machine and transversal direction, and seal strength assay), water barrier (WVTR) and food-contact regulations (overall migration). As a consequence of the plasticizing effect of water on the EVOH layer, oxygen barrier (OTR) was increased, but keeping the barrier performance by remained within the expected range. Overall, the results indicate that the US-assisted treatment conditions here evaluated can be applied to PE/EVOH/PE multilayer packaging without compromising its performance.

1. Introduction

Monolayer or monomaterial packaging cannot simultaneously fulfill all essential food packaging requirements, including containment, protection, preservation, and adequate mechanical performance. In addition, packaging materials must also provide promotion and convenience, remain cost-effective, and comply fully with food safety regulations [1–3]. For this reason, multilayer and multimaterial flexible packaging systems have been widely developed. These structures typically consist of low-thickness structures composed of multiple polymeric layers, sometimes combined with non-polymeric components, designed to combine and optimize key properties such as barrier performance, mechanical strength, flexibility, and functionality [4–6].

A wide range of polymers can be used in the manufacture of these structures, selected according to their complementary properties, sealability, and processing performance. Commonly used materials include polyolefins such as polypropylene (PP) and polyethylene (PE), polyesters such as polyethylene terephthalate (PET), polyamides (PA), and ethylene vinyl alcohol copolymer (EVOH) [6–8]. To further enhance characteristics such as rigidity, printability, and barrier properties, non-polymeric materials, e.g., paper, cardboard, or metallic layers may also be incorporated into the multilayer structure [8,9].

Among these systems, multilayer flexible packages based on PE and EVOH (e.g., PE/EVOH/PE) has gained particular relevance due to the complementary properties of these polymers. PE provides flexibility, sealability, and moisture resistance, while EVOH offers excellent oxygen barrier performance. The combination of these materials results in structures with balanced mechanical, barrier, and processing properties,

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making them widely used in food, pharmaceutical, and industrial packaging applications [10–12]. Additionally, when properly designed and proportioned, PE/EVOH multilayer structures can be combined to produce recyclable materials suitable for PE recycling streams [13,14].

In parallel with the evolution of packaging materials, the development of novel food processing technologies aims to enhance product safety, quality, and efficiency while reducing environmental impact. Among emerging technologies, ultrasound (US) has gained increasing attention as a green physical processing technique capable of inducing desirable structural and functional changes in foods through the action of high-frequency acoustic waves [15]. Depending on the applied intensity and power, US may be used either for non-destructive monitoring or to induce physicochemical modifications in products and processes [16].

US has also been investigated as a processing technology for packaged foods. However, the acoustic cavitation phenomena associated with US may interact not only with the food matrix but also with the packaging material itself, potentially affecting the integrity and performance of polymeric structures.

Previous studies have reported that US treatment can alter the permeability, thermal stability, and surface morphology of polymeric films, as well as influence the migration of compounds from multilayer materials into food or food simulants [17–20]. However, the reported effects vary substantially according to the packaging composition and architecture, and limited information is available for polyolefin/EVOH-based multilayer structures. Recent studies illustrate that the effects of US on packaging are strongly dependent on the composition and architecture of the material. In multilayer PE/PA/PE and PE/PA/PE/PA/PE films, for example, US treatment did not significantly affect overall migration or the migration of hexamethylenediamine; however, the migration of ε-caprolactam depended on the packaging structure, food simulant, and exposure time, with a slight increase observed after 60 min in contact with the fatty simulant [21]. In metallized PET_{met}/PE and PET_{met}/PP structures, US under comparable conditions resulted in more pronounced structural effects, including reductions in crystallinity and discontinuities in the metallized layer, which were associated with increases in water vapor and oxygen transmission rates, particularly after treatment at 25 °C for 60 min [22]. In contrast, studies on PET/Al/PE and PET/Al/PP laminates showed that exposure for up to 30 min caused no significant changes in chemical composition, thermal stability, seal strength, or barrier properties, although longer exposure resulted in structural damage to the aluminum foil; no significant migration of aluminum or antimony was detected [20].

Collectively, these findings indicate that US does not produce a uniform response across multilayer packaging systems: its effects may range from limited changes in migration or bulk polymer properties to localized damage of highly sensitive barrier components. In this context, the PE/EVOH system presents a distinct case, as its oxygen barrier is provided by a thin EVOH layer embedded between PE layers, rather than by PA, metallization, or aluminum foil. Although PE and PA are widely used in packaging for their flexibility and strength, EVOH is notable for its superior oxygen-barrier properties. However, its hydrophilicity can cause those properties to vary with environmental and processing conditions. Consequently, US treatment of EVOH films may pose specific challenges, such as altered barrier efficiency, morphology, or interlayer adhesion, that warrant careful evaluation.

Considering the growing interest in US-assisted treatment technologies for packaged foods, it is essential to assess how these treatments may affect packaging materials commonly used for food contact. Therefore, this study aimed to evaluate the effects of US treatment on multilayer PE/EVOH/PE films, examining how different exposure conditions influence their structural, mechanical, and barrier properties. By investigating the interaction between US treatment and this widely used packaging structure, this work seeks to contribute to a better understanding of the food–package–processing relationship and to support the safe application of emerging processing technologies in packaged food systems.

2. Experimental section

2.1. Material

A multilayer PE/EVOH38/PE flexible film supplied by [Valgroup, Itamonte, MG, Brazil], was used in this study. The film consisted of three layers with nominal thicknesses of 46/17/49 μm (PE/EVOH/PE). The EVOH38 layer corresponds to an ethylene vinyl alcohol copolymer containing approximately 38 mol% ethylene, a composition commonly employed in packaging materials due to its high oxygen barrier performance. The outer and inner layers were composed of PE, providing mechanical strength, flexibility, and sealability.

These multilayer structures are widely used to package oxygen-sensitive foods. For testing, a food simulant of 10% (v/v) ethanol was selected to represent non-acidic aqueous foods (pH > 5). Although such packaging serves aqueous and fatty products, such as dairy beverages, cheeses, meats, and vegetable puree the aqueous simulant was chosen because, given EVOH's hydrophilicity, it promotes stronger interactions with the packaging and therefore represents the most critical exposure scenario. In addition, the aforementioned products are also suitable candidates for conventional processing or for processes enhanced by emerging technologies, such as US-assisted treatments performed at moderate temperatures (e.g., up to 60 °C), aiming to improve process efficiency while preserving product quality.

2.2. Ultrasound processing

Rectangular packages (120 mm × 150 mm) were created by sealing two identical films with a sealing width of 3 mm. Each package was filled with 100 mL of the prepared aqueous food simulant (10% v/v ethanol solution) and sealed without headspace, ensuring a horizontal sealing position at the top of the bag. An ultrasonic water bath (Ultronique, Model Q 13/25 A CR, Indaiatuba, Brazil) was used to treat the packages. The system operated at a nominal frequency of 25 kHz and a volumetric power of 9.74 W/L, determined by the calorimetric method [22,23]. Water was used as the coupling medium for ultrasound propagation. Before each treatment, the water level and sample positioning were standardized, and the packages were immersed at a consistent depth and location within the bath to ensure comparable ultrasound exposure. The water bath was allowed to reach the predetermined operating conditions before each treatment, and the same setup and procedure were followed for all replicates to ensure repeatability. Two temperatures were applied to consider both US processing (25 °C) and US-assisted processing, or thermosonication (60 °C). For that, an integrated heat exchanger coupled to an external water bath was used.

Initially, the processing was conducted in intervals of 30 min, extending up to 120 min, to establish foundational conditions. Processing times of 60 and 90 min were selected based on mechanical property results. These conditions were chosen to explore a range range applied to low acid foods such as meat emulsions [24–26] or chicken breast [27,28].

All US treatments were performed in duplicate. Following the treatments, the samples were evaluated using various techniques, including TS analysis, seal strength assessment, water vapor transmission rate (WVTR) measurement, oxygen transmission rate (OTR) measurement, overall migration analysis, scanning electron microscopy (SEM), Fourier transform infrared (FTIR) spectroscopy, differential scanning calorimetry (DSC), and X-ray diffraction (XRD).

2.3. Characterization of packaging films

2.3.1. Fourier transform infrared (FTIR) spectroscopy

The Fourier transform infrared (FTIR) spectroscopy analysis was

conducted on a Spectrum 100 spectrometer (PerkinElmer, Waltham, MA, USA) operating in attenuated total reflectance (ATR) mode with a zinc selenide (ZnSe) crystal. Using the PerkinElmer Spectrum software (v. 10.4.00), spectra were recorded in the range of 4000–650 cm^{-1} at a resolution of 4 cm^{-1} , and 4 scans for each measurement. To account for material heterogeneity, three spectra per layer were obtained from distinct locations.

2.3.2. Differential scanning calorimetry (DSC)

The thermal behavior of the samples was investigated by Differential Scanning Calorimetry (DSC) using a DSC 250 calorimeter (TA Instruments, New Castle, USA). The measurements were performed at a heating rate of 10 °C/min over a temperature range of 40–300 °C. Thermal analysis was performed on each sample (~5 mg) under a continuous nitrogen purge. The resulting data were used to determine the melting temperature (T_m) and melting enthalpy (ΔH) and all values reported are the average of three replicate measurements.

2.3.3. X-ray diffraction (XRD)

X-ray diffraction (XRD) analysis was conducted to assess the crystallinity of the films using an X'Pert-MPD diffractometer (Philips, Almelo, Netherlands). The system utilized Cu Kα radiation (λ = 1.54 Å) and was operated at 40 kV and 40 mA. Scans were recorded over the 2θ range of 10° to 30° with a step size of 0.04° and a dwell time of 1.2 s per step. The crystallinity index (X_c) was estimated by Gaussian peak deconvolution using the Fityk software, in which the crystalline reflections were fitted with Gaussian functions after baseline correction, allowing separation of crystalline and amorphous contributions. The degree of crystallinity was calculated as the ratio between the integrated area of the crystalline peaks and the total scattered area (crystalline plus amorphous) within the analyzed 2θ range and is therefore reported as a relative crystallinity index.

2.3.4. Scanning electron microscopy (SEM)

The inner surface and the cross section of the films were analyzed in a scanning electron microscope (SEM, VEGA LMU, Tescan, Czech Republic) equipped with an energy dispersive X-ray spectrometer (EDS; AZTEC, Oxford, UK). The analysis was conducted at an accelerating voltage of 5 kV and work distance of 15 mm. The samples were prepared on metal stubs and sputter-coated with a thin gold layer (approximately 200 Å) using a SCD 050 coater (Balzer, Liechtenstein). SEM images were acquired at magnifications of 200 × and 1000 × [29].

2.3.5. Mechanical properties

The mechanical properties, including tensile strength (TS), elongation at break (EB), and seal strength (SS), were evaluated under standardized conditions of 23 ± 2 °C temperature and 50 ± 5% relative humidity, following a conditioning period of at least 48 h in the same environment. Rectangular specimens, 25.4 mm wide and 120 mm long, were precisely cut using a cutter (JDC Twing Albert). All tests were performed with a universal testing system (Instron 5966-E2, Norwood, USA), with five replicates per sample. For TS and EB assessments, a load cell of 1 kN was used, applying a constant extension speed of 50 mm/min, with the clamps spaced 50 mm apart [30]. SS was measured using a 100 N load cell, at a faster pull rate of 300 mm/min, with a 25 mm clamp gap [31]. Testing was conducted in both the machine direction (MD) and the transverse direction (TD).

2.3.6. Barrier properties

The water vapor transmission rate (WVTR) was assessed using a PERMATRAN device (3/34 G, MOCON, Minneapolis, USA) equipped with an infrared sensor, conducted at a temperature of 38 °C and 90% relative humidity [32]. For oxygen transmission rate (OTR) measurements, an OXTRAN apparatus (2/20H, MOCON, Minneapolis, USA) was employed, with the sample's external surface exposed to pure oxygen (100% O_2) [33]. The measurements were calibrated to a standard permeant gas pressure differential of 1 atm and performed at 23 °C under dry conditions. The effective permeation area of each sample was used to calculate the respective rates.

2.3.7. Overall migration

The overall migration of substances from the packaging to the food simulant was assessed immediately after US treatment and again after a 10-day storage period at 40 °C. These conditions were selected to represent accelerated and worst-case contact scenarios for aqueous, non-acid foods, such as milk, dairy-based beverages, and isotonic drinks, which may be subjected to extended storage and elevated temperatures during processing, transport, or distribution. The migration tests adhered to the relevant standards [34,35], including RDC No. 51/2010 (MERCOSUR/GMC/RES. no. 32/2010) [36], RDC No. 589/2021 (MERCOSUR/GMC/RES. no. 19/2021, no. 2020/2021 and no. 2021/2021) [37] and Regulation (EU) No. 10/2011 [38], ensuring compliance with regulatory guidelines [39,40]. After the processing and storage phases, migration levels were quantified gravimetrically; the food simulant was evaporated on a heating plate (TE038, Tecnal), and the resulting residue was weighed using an analytical balance (MSA225P-1CE-DA, Sartorius) with a sensitivity of 0.00001 g.

2.3.8. Statistical analysis

The quantitative data were presented as means accompanied by their standard deviations. These results were subjected to statistical analysis using analysis of variance (ANOVA), followed by Tukey's post-hoc test for pairwise comparisons. Differences were considered statistically significant at $p < 0.05$, with distinct superscript letters indicating significant variations between groups.

3. Results and discussion

Different treatment conditions were initially evaluated (30, 60, 90, and 120 min). The 30 min treatment was considered as an initial screening condition and did not result in significant changes in key mechanical properties, such as tensile strength (TS) and elongation at break (EB) (Section 3.5). Based on these results, the 30 min condition was discontinued from the subsequent experimental characterization. The subsequent analyses therefore focused on the 60 and 90 min treatments, which showed more pronounced effects and were selected for further structural characterization (XRD, DSC, FTIR, and SEM), as well as for the evaluation of material properties and migration behavior.

3.1. Fourier transform infrared (FTIR) spectroscopy

Fourier-transform infrared (FTIR) was used to evaluate potential chemical changes in the multilayer films after US treatment. This technique enables the identification of polymeric components and the detection of structural modifications by monitoring characteristic functional groups, making it a valuable tool for assessing changes in packaging materials.

Characteristic FTIR bands of PE are associated with its C-H stretching (2916, 2848 cm^{-1}), bending (1462, 1473 cm^{-1}), and "rocking" (720, 710 cm^{-1}). EVOH and other multilayer components exhibit distinct peaks, enabling layer-specific analysis. The structure of PE/EVOH/PE was confirmed by FTIR analysis. The FTIR spectrum in Fig. 1a demonstrated aliphatic hydrocarbons: ν_{as} CH_2: 2950 cm^{-1}, stretching vibration at 2925 cm^{-1} and 2851 cm^{-1} (C–H), ν_s CH_2: 2825 cm^{-1}, around 1480 and 1380 cm^{-1} are ascribed the deformation of $–CH_2–$ and $–CH_3–$ groups, δas C–H in CH_2 groups: 1467 cm^{-1}, δCH_2 in (CH_2) n (n > 3): 717 cm^{-1} [41–43]. The FTIR spectra of EVOH are characterized by distinct absorption peaks, notably for C–H asymmetric stretching (around 2950 cm^{-1} and 2825 cm^{-1}), O–H deformation around 1480 and 1400 cm^{-1}, C–O stretching vibrations 1200–1000 cm^{-1} , and skeleton vibration around 720 and 710 cm^{-1}. The relative intensity of the O–H band is commonly used as an indicator of the degree of hydrolysis in

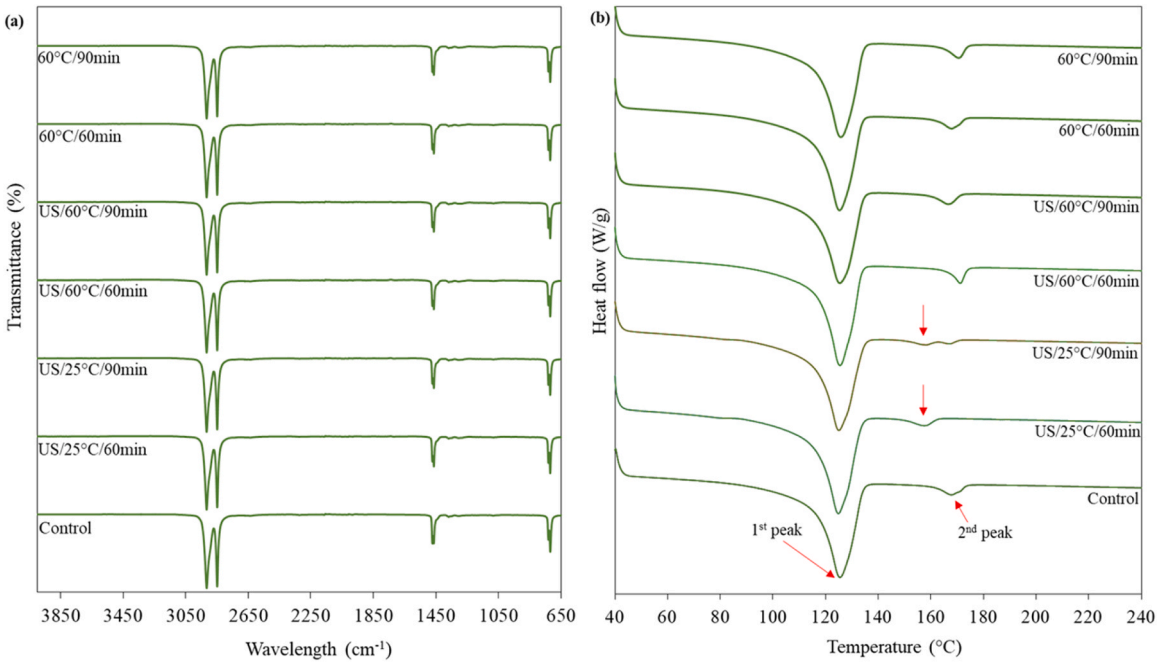


Fig. 1. FTIR spectra (a) and differential scanning calorimetry (DSC) curves (b) of PE/EVOH38/PE flexible packaging films before and after ultrasound (US) treatment.

EVOH materials.

The investigation into various US treatment parameters and their effects revealed that there was no significant alteration in the surface spectra of the multilayer packaging polymers. This indicates that the chemical composition of the materials remained intact following US treatment, regardless of the temperatures applied. Similar observations were made with samples consisting of PE and PA films [44], PET/Al/PP, PET/Al/PE [20], as well as PET_{met}/PE and PET_{met}/PP [22], all subjected to US. The stability of the chemical properties across these different materials suggests that the US treatments did not compromise the structural or functional integrity of the polymers, demonstrating the durability of these materials when exposed to US treatment.

3.2. Differential scanning calorimetry (DSC)

Differential scanning calorimetry (DSC) was used to assess the thermal behavior of the PE/EVOH multilayer films before and after US treatment. The thermograms (Fig. 1b and Table 1) showed two distinct melting peaks, corresponding to the PE layers and the EVOH layer, respectively, reflecting the multilayer structure of the material [45,46]. In addition to the individual peaks, the total enthalpy of fusion (ΔH) was determined by integrating the entire melting endotherm, encompassing the contributions from both PE and EVOH phases.

The melting behavior of the multilayer films revealed distinct responses for the PE and EVOH layers under US treatment. The first melting peak (T_m), corresponding to the PE layer, showed minimal variation among samples, ranging from 124.9 °C to 126.0 °C, indicating that US treatment did not significantly affect the crystalline phase of PE. In agreement with this observation, the ΔH, which is largely dominated by the PE phase due to its higher proportion in the multilayer structure and its more intense melting peak, also showed no significant differences among the samples.

In contrast, the second melting peak, associated with the EVOH layer, exhibited a pronounced decrease under US at 25 °C, with Tm values of 158.1 °C (US/25 °C/60 min) and 161.1 °C (US/25 °C/90 min) compared to 168.7 °C for the control. Samples processed at 60 °C, either with or without US, presented Tm values close to the control (167.1–171.2 °C), suggesting that higher processing temperatures may mitigate the effects induced by US cavitation.

Table 1
Differential scanning calorimetry (DSC) results of PE/EVOH38/PE flexible packaging films before and after ultrasound (US) treatment.

Treatment condition	T_m (°C) 1st peak - PE	T_m (°C) 2nd peak - EVOH	ΔH* (J/g)
Control	125.5 ± 0.3^{ab}	168.7 ± 2.1^{a}	117.7 ± 7.3^{a}
US/25 °C/60 min	124.9 ± 0.2^{b}	158.1 ± 0.5^{b}	112.9 ± 0.6^{a}
US/25 °C/90 min	125.1 ± 0.2^{ab}	161.1 ± 5.2^{bc}	115.8 ± 5.1^{a}
US/60 °C/60 min	125.6 ± 0.4^{ab}	171.2 ± 0.2^{a}	121.6 ± 1.4^{a}
US/60 °C/90 min	125.5 ± 0.6^{ab}	167.1 ± 1.5^{ac}	120.5 ± 1.0^{a}
60 °C/60 min	125.3 ± 0.1^{ab}	168.6 ± 1.9^{a}	117.5 ± 3.9^{a}
60 °C/90 min	126.0 ± 0.6^{a}	170.5 ± 1.1^{a}	120.3 ± 2.0^{a}

T_m: melting temperature, ΔH*: total enthalpy of melting, PE: polyethylene and EVOH: ethylene-vinyl alcohol copolymer.
Values are expressed as mean ± standard deviation (n = 3). Different lowercase letters within the same column indicate statistically *significant differences between treatments (p < 0.05).*

In semicrystalline polymers such as PE and EVOH, the Tm and ΔH describe distinct, although interrelated, aspects of crystalline morphology. The Tm is directly related to lamellar thickness and crystalline perfection, as described by the Gibbs–Thomson equation [47]. According to this relationship, thinner or less perfect lamellae exhibit lower melting temperatures. In contrast, the ΔH reflects the overall crystalline fraction of the material rather than the structural quality of individual crystals [48].

The decrease in Tm observed for EVOH samples after US treatment at ambient temperature (25 °C) suggests structural modifications within the crystalline phase. These changes may include a reduction in lamellar thickness and/or a decreased crystalline perfection, as discussed above. Such effects are consistent with US cavitation phenomena, which

generate localized shear forces that can promote molecular disorganization. It is important to note that a reduction in Tm does not necessarily imply a decrease in crystalline fraction, as structural reorganization can occur without significant changes in ΔH.

Furthermore, the distinct responses of EVOH and PE to US treatment can be attributed to their physicochemical differences and their position within the multilayer structure. EVOH is a polar polymer containing hydroxyl (–OH) groups capable of forming intermolecular hydrogen bonds, making it more sensitive to thermal and mechanical perturbations, as well as to environmental factors such as moisture. In contrast, PE is a nonpolar polymer with weaker intermolecular interactions, which confers greater resistance to such effects.

3.3. X-ray diffraction (XRD)

X-ray diffraction (XRD) analysis provided complementary insight into the crystalline organization of the PE/EVOH38/PE multilayer films (Fig. 2). The film consisted of three layers (46/17/49 μm, PE/EVOH/PE), with PE accounting for ~84% of the total thickness, thus dominating the diffraction response. Accordingly, all diffractograms exhibited the characteristic reflections of PE, with intense peaks at ~21° and ~24° (2θ), corresponding to the (110) and (200) planes of orthorhombic PE crystals, respectively [49]. In contrast, EVOH38 typically presents a main diffraction peak around 20.4° [50]. However, in the multilayer structure, this reflection overlaps with the more intense PE peaks, hindering clear resolution of the individual contributions. Nevertheless, the presence of EVOH can still be inferred from subtle peaks in the diffractograms. No shifts in the 2θ peak positions were observed after US treatment, suggesting that the orthorhombic crystalline structure was preserved for all samples (interplanar spacing of the crystalline regions remained unchanged, according to Bragg's law) [51].

Quantitative evaluation by Gaussian peak deconvolution revealed relative crystallinity indices (X_c) between 53% and 64%, with the control film exhibiting the highest crystallinity. US-treated samples showed a moderate decrease in crystallinity, particularly under longer exposure times, while samples subjected to thermal treatment at 60 °C displayed crystallinity values closer to the control. These variations are consistent with structural rearrangements typically induced by US, where localized cavitation promotes chain mobility, partial lamellar distortion, and an increase in amorphous content without complete disruption of the crystalline framework [22,44].

The absence of peak shifts in the 2θ positions suggests that US treatment did not alter the unit cell dimensions or the orthorhombic packing of PE chains. In this context, the preservation of peak positions combined with the reduction in X_c supports the hypothesis that US promotes partial disruption of the crystalline phase without inducing polymorphic transitions [52]. The decrease in X_c can be attributed to localized cavitation effects, which enhance chain mobility and promote the transformation of ordered regions into amorphous phases. At the same time, the remaining crystalline domains retain their orthorhombic structure, as evidenced by the unchanged diffraction peak positions.

When considered together with DSC results, a more comprehensive understanding of the structural changes can be established. While XRD indicates a reduction in crystalline order and/or fraction, DSC analysis showed no significant changes in enthalpy for the PE phase and only variations in melting temperature for EVOH. This suggests that US primarily affects crystalline organization and perfection rather than the total crystalline content of the multilayer system. In particular, the decrease in melting temperature observed for EVOH can be associated with lamellar thinning or reduced crystalline perfection, whereas the PE phase exhibits greater structural stability, with changes mainly reflected in crystalline order rather than thermodynamic properties. Overall, these results indicate that US treatment induces a form of "crystalline erosion", characterized by a reduction in structural order without altering the fundamental crystal structure.

From a functional perspective, these subtle changes in crystallinity are highly relevant to both oxygen barrier and mechanical performance. An increase in amorphous regions within the PE layers enhances chain mobility and free volume, which can facilitate gas diffusion pathways, thereby contributing to the variations observed in oxygen transmission rate (further discussed). Conversely, higher crystallinity levels are associated with reduced permeability due to the impermeable nature of crystalline domains and their role as tortuous barriers to gas transport [53,54].

Similarly, the mechanical properties of the films are directly influenced by the balance between crystalline and amorphous phases in the PE layers. The moderate reduction in crystallinity observed after US treatment is consistent with slight decreases in stiffness and yield-related parameters, while preserving sufficient crystalline content to maintain tensile strength and structural stability. In contrast, samples treated at elevated temperature show minimal reduction in crystallinity, which may contribute to improved stress transfer and dimensional stability. These observations align well with reported literature on PE-based multilayer films [55,56], where small variations in crystallinity are often linked to changes in ductility, elastic modulus, and resistance to mechanical deformation rather than to catastrophic loss of

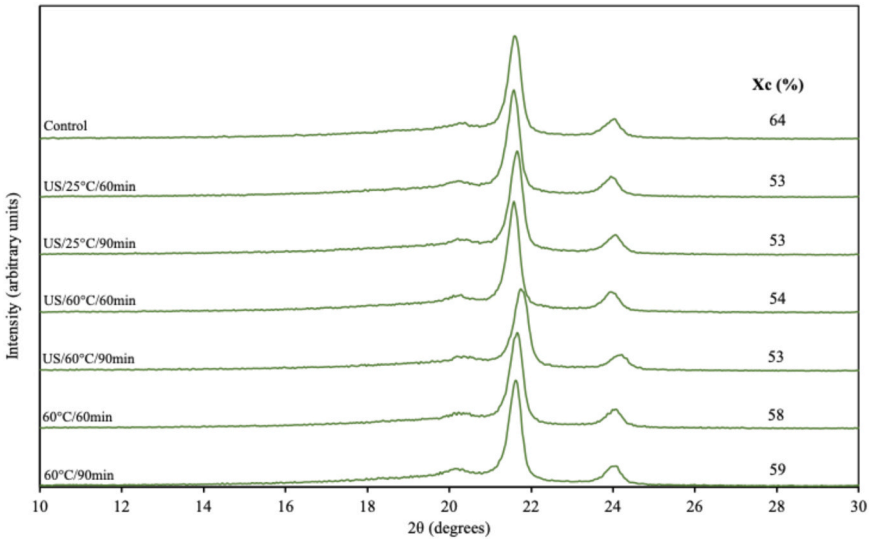


Fig. 2. X-ray diffraction (XRD) patterns of PE/EVOH38/PE flexible packaging films before and after ultrasound (US) treatment.

performance.

3.4. Scanning electron microscopy (SEM)

As shown in Fig. 3, SEM images of the outer layer surfaces of the analyzed films revealed the presence of irregularities, such as spots and scratches, consistently across all experimental conditions, including the control sample. This behavior suggests that such features are inherent to the film itself and may stem from its manufacturing process, rather than necessarily being related to the US treatment. However, in some specimens subjected to ultrasonic treatment at 60 °C for 60 min, an intensification of these irregularities was observed, characterized by larger regions and/or a greater number of them. This behavior may be linked to cavitation effects generated by the liquid during US treatment, although this hypothesis cannot be confirmed solely by the SEM images. Cross-sectional images showed no significant internal alterations or defects, indicating that the PE/EVOH/PE films maintained their morphological integrity under the evaluated treatment conditions. Thus, if the ultrasound process did induce any morphological change, the results suggest that its effect was predominantly confined to the outer surface, with no evidence of significant alterations to the material's internal structure or intermediate layers. Overall, the results indicate good morphological stability of the films under the treatment conditions evaluated, as no significant internal structural changes were observed.

3.5. Tensile strength (TS) and elongation at break (EB)

The results of the mechanical properties presented in Fig. 4 provide a thorough analysis of the different processing conditions, both with and without the use of US, and their impact on the mechanical characteristics of the PE/EVOH38/PE films. The primary focus is on TS and EB, evaluated in both the machine direction (MD) and the transverse direction (TD).

The variations in TS observed across processing conditions are apparent, but they should be interpreted with caution. While the data suggest that treatments combining US and higher temperatures can increase TS in the MD (Fig. 4a), this effect is small and becomes non-significant when additional sources of variability are considered (e.g., non-uniform stress distribution and fracture location). This increase can be attributed to a structural reorganization of the material, facilitated by the rise in temperature combined with acoustic cavitation. Fig. 4a and c reveal that applying US at 25 ºC for 60 min resulted in a significant decrease in the TS of the films. This phenomenon can be explained by the water absorption promoted by the hydrophilic polymer (EVOH) under low-temperature conditions, which compromises its structural integrity. However, this behavior did not show a consistent trend with the increase in exposure time.

US treatment increases EB in the MD, indicating greater film extensibility (Fig. 4(b)). This enhanced ductility can be explained by thermal and structural changes in the material: a lowered melting temperature and reduced crystallinity increase polymer chain mobility, decreasing matrix stiffness and facilitating molecular rearrangement under deformation. Thus, alongside cavitation and mass-transfer mechanisms, the combined effect of reduced melting temperature and crystallinity directly contributes to the increased elongation prior to fracture.

As discussed by Almeida et al., the application of energy through treatments involving high temperatures and/or acoustic cavitation tends to intensify molecular movement, promoting either an increase or

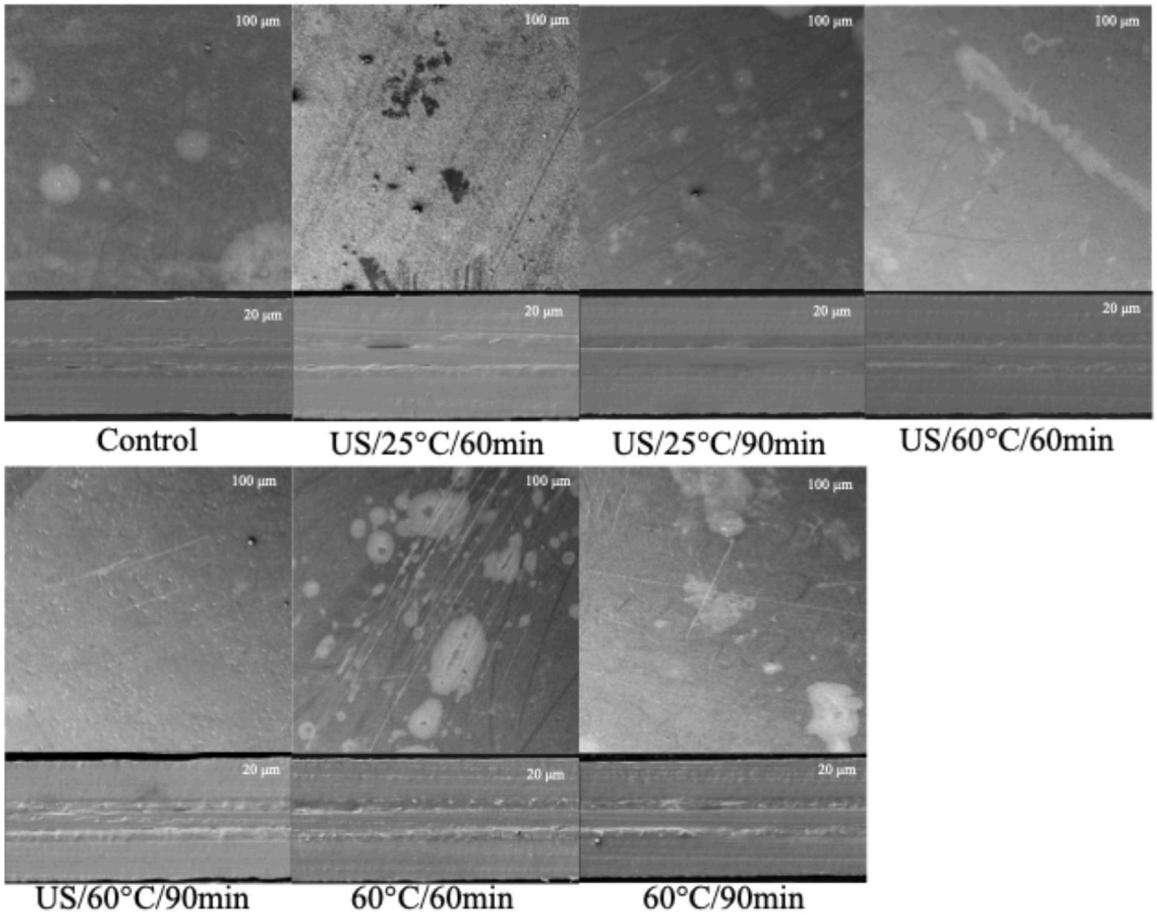


Fig. 3. Scanning electron microscopy (SEM) images of the outer-layer surface and cross-sections of PE/EVOH38/PE flexible packaging films before and after ultrasound (US) treatment.

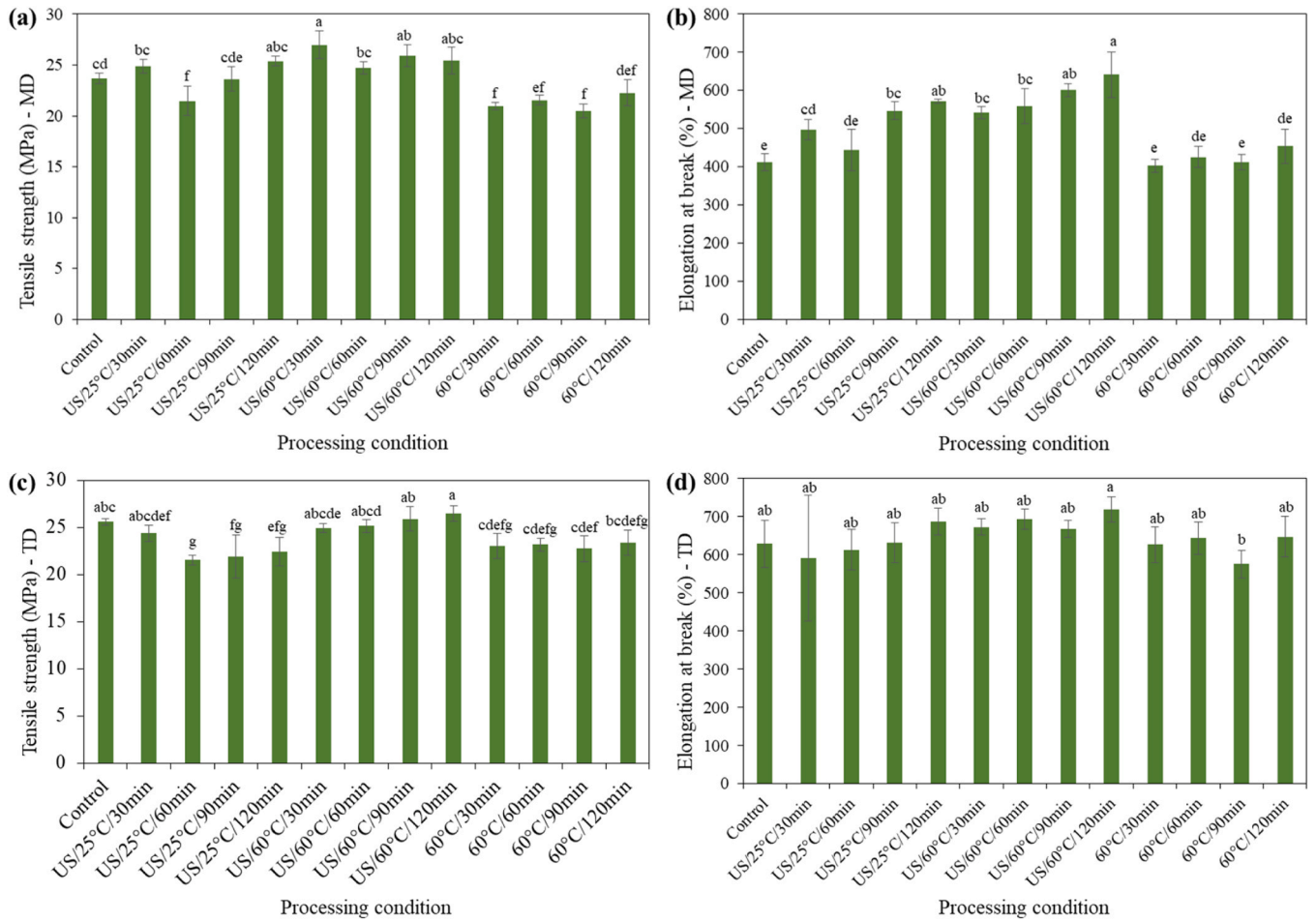


Fig. 4. Tensile strength (TS, MPa) and elongation at break (EB, %) of PE/EVOH38/PE flexible packaging films before and after ultrasound (US) treatment, measured in the machine direction (MD) and transverse direction (TD). Values are expressed as mean ± standard deviation (n = 5). Different lowercase letters within the same column indicate significant differences between treatments ($p < 0.05$).

compaction in the interstitial space between the polymer units [20]. This rearrangement leads to significant changes in the organization of the molecular network and in the local crystallinity of the material [20, 57]. The resulting reorganization modifies the displacement of the polymer chains, leading to changes in the mechanical properties. Such modifications have substantial implications for the performance of the materials, directly impacting their flexibility and strength.

Finally, considering that in various situations the 30-min processing time did not show a significant impact on TS compared to the control, subsequent analyses were conducted with samples processed under

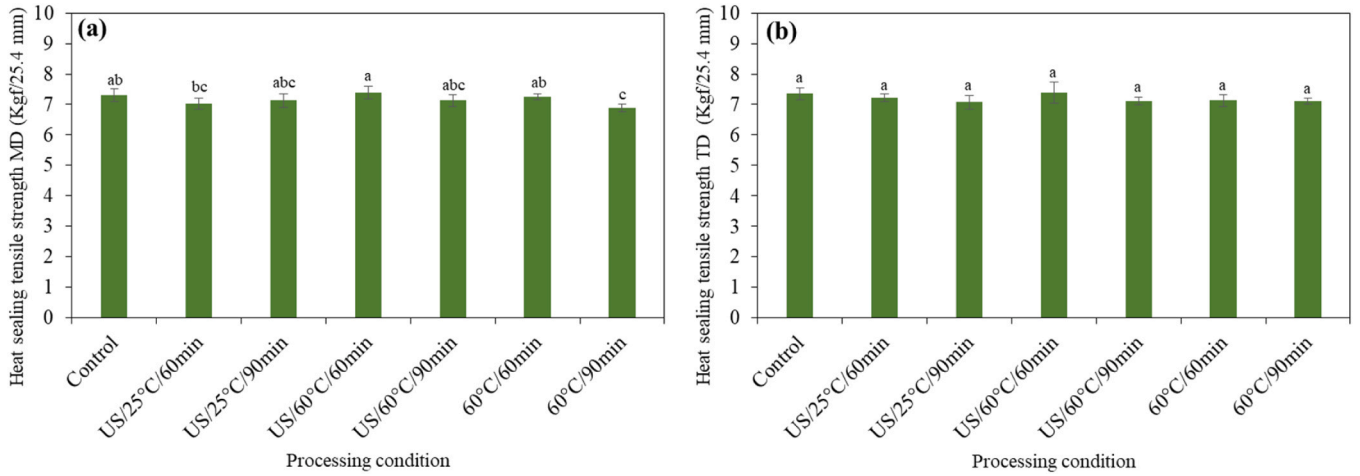


Fig. 5. Heat-sealing tensile strength (kgf/25.4 mm) of PE/EVOH38/PE flexible packaging films before and after ultrasound (US) treatment in the machine direction (MD) (a) and transverse direction (TD) (b). Values are expressed as mean ± standard deviation (n = 5). Different lowercase letters within the same column indicate significant differences between treatments ($p < 0.05$).

intermediate conditions, specifically 60 and 90 min, as detailed in the following results.

3.6. Seal strength

The results of the sealing strength of the PE/EVOH38/PE films, before and after the application of different treatment conditions, in both the machine direction (MD) and the transverse direction (TD), are illustrated in Fig. 5. The data reveal that the sealing strength values for all tested conditions remained around 7 kgf/25.4 mm, aligning with values reported in the literature for films containing PE in the sealing layer [58,59]. It is important to emphasize that the various treatment conditions involving the use of US did not result in significant differences in sealing strength when compared to the control. This finding is considered positive, as it ensures the maintenance of robust sealing during the stages of processing, storage, and transportation of the products.

This phenomenon can be explained by the adequacy of the sealing conditions employed in the films, where the intensity of acoustic cavitation was not sufficiently high to compromise the intermolecular bonds between the sealing layers, specifically the PE-PE interactions [20]. Such dynamics ensure that the integrity of the seal remains intact, even under intensified processing conditions.

3.7. Barrier properties – WVTR and OTR

The results of the water vapor transmission rate (WVTR) and the oxygen transmission rate (OTR) of the PE/EVOH38/PE films, before and after the application of different treatment conditions, are presented in Table 2. The WVTR values ranged from 1.22 to 1.32 g water.m^{-2}.day^{-1}, reflecting a significant barrier to water vapor, attributed to the PE layers in the multilayer structure. The absence of statistically significant variations in WVTR indicates that neither US nor thermal exposure altered the integrity or continuity of the PE layers, which is consistent with the XRD results showing only moderate changes in the crystalline organization of PE layer, and with the DSC results that indicated no significant difference in the crystalline fraction of PE. The preservation of a relatively high crystalline fraction in the PE layers explains the stability of the water vapor barrier, as crystalline domains act as impermeable regions that limit moisture diffusion.

Regarding the OTR, the measured values fluctuated between 1.19 and 2.91 mL(STP).m^{-2}.day^{-1}. This oxygen barrier is primarily attributed to the EVOH layer, and the values found are consistent with existing literature for films that use EVOH as an oxygen barrier layer [14,60].

The increase in OTR observed after US treatment may be associated with structural modifications within the multilayer system. XRD analysis (Fig. 2) revealed a moderate reduction in the overall relative crystallinity, indicating partial disruption of ordered domains and an increase in amorphous regions. These structural changes are known to enhance chain mobility and increase free volume, facilitating oxygen diffusion through the material. The EVOH layer, responsible for the barrier properties, may also be affected by US through changes in crystalline organization, as suggested by DSC results. Even in the absence of significant changes in crystallinity, reductions in crystalline perfection or lamellar thickness can decrease barrier efficiency by increasing segmental mobility within the polymer matrix.

In addition, US-induced cavitation can generate localized shear forces, promoting molecular rearrangement and the formation of microstructural heterogeneities, such as defects at the crystalline–amorphous interface. These features can act as preferential pathways for gas transport, contributing to the observed increase in permeability.

Table 2
Overall migration in aqueous food simulant, water vapor transmission rate (WVTR), and oxygen transmission rate (OTR) of PE/EVOH38/PE flexible packaging films before and after ultrasound (US) treatment.

Treatment condition	Overall migration (mg dm^{-2}) - After 40 °C/10 days	WVTR (g water.m^{-2}.day^{-1}) 38 °C/90% RH	OTR (mL(STP).m^{-2}.day^{-1}) 23 °C
Control	≤ 3.00*	1.24 ± 0.02^{a}	1.19 ± 0.11^{d}
US/25 °C/60 min	≤ 3.00*	1.23 ± 0.11^{a}	1.99 ± 0.27^{bc}
US/25 °C/90 min	≤ 3.00*	1.29 ± 0.02^{a}	1.95 ± 0.07^{b}
US/60 °C/60 min	≤ 3.00*	1.27 ± 0.13^{a}	2.44 ± 0.35^{ab}
US/60 °C/90 min	≤ 3.00*	1.22 ± 0.20^{a}	2.73 ± 0.38^{ac}
60 °C/60 min	≤ 3.00*	1.24 ± 0.07^{a}	2.34 ± 0.15^{ab}
60 °C/90 min	≤ 3.00*	1.32 ± 0.06^{a}	2.91 ± 0.37^{a}

Values are expressed as mean ± standard deviation (n = 3). Different lowercase letters within the same column indicate significant differences between treatments ($p < 0.05$). The applicable overall migration limit is 10 mg dm^{-2} of plastic material, according to ANVISA Resolution No. 589/2021. * indicates the limit of quantification of the method under the analytical conditions used. STP: standard temperature and pressure.

3.8. Overall migration

The overall migration results obtained for the control and under treatment condition samples are shown in Table 1. According to the results obtained, the samples analyzed according to the requirements described in Resolution RDC No. 51/10 of the Brazilian Health Regulatory Agency – Anvisa [61] meets the limits of overall migration established in Resolution RDC No. 589/21 of Anvisa and Regulation (EU) No. 10/2011 [62], for direct contact with aqueous food simulant - 10% v/v ethanol solution (pH > 4.5) for packaging at maximum temperatures of 60 °C for 90 min, with and without US treatment, followed by prolonged use at temperatures up to 40 °C. Therefore, it can be stated that the migration values obtained after different processing and storage conditions meet the limits established by European Union and Brazilian legislation. These results are consistent with the overall migration values reported in the literature for PE/PA/PE, PE/PA/PE/PA/PE, PET$_{met}$/PE, PET$_{met}$/PP PET/Al/PP, and PET/Al/PE films processed with US and tested in contact with acidic and aqueous food simulants [20–22], where US treatment did not influence the overall migration of the films into the different food simulants.

4. Conclusions

This study demonstrated that US-assisted treatment, combined with mild thermal conditions, promotes subtle modifications in the properties of PE/EVOH38/PE multilayer flexible packaging films, without impacting its application under the conditions investigated in this study. Mechanical results showed that US and temperature slightly affects TS in the machine direction, indicating improved ductility without compromising structural integrity. Barrier properties were largely preserved after treatment. WVTR values remained stable, confirming the effectiveness of the PE layers as moisture barriers, while OTR increased moderately following US treatment. This increase may be associated with the structural changes observed in the multilayer system, particularly the reduction in relative crystallinity, which may facilitate oxygen diffusion through increased chain mobility and free volume. Despite this increase, oxygen barrier performance remained within the expected range for EVOH-based multilayer systems. Structural analyses supported these findings. XRD revealed moderate variations in relative crystallinity, with US treatments inducing slight reductions associated with increased amorphous content. DSC results confirmed that these changes were related to crystal rearrangement rather than a loss of crystalline content. SEM and FTIR analyses further confirmed the morphological

and chemical stability of the multilayer films, and overall migration results demonstrated compliance with current Brazilian and European food-contact regulations. Overall, the results indicate that the US-assisted treatment conditions here evaluated can be applied to PE/EVOH/PE multilayer packaging without compromising mechanical performance, barrier functionality, or regulatory compliance, supporting its potential use in food processing and packaging applications.

CRediT authorship contribution statement

Nanci Castanha: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Amanda Alves Domingos Maia:** Writing – original draft, Data curation. **Paula Fernanda Janetti Bócoli:** Writing – original draft, Formal analysis. **Danielle Ito:** Writing – review & editing, Formal analysis. **Fiorella Balardin Hellmeister Dantas:** Writing – original draft, Formal analysis. **Roniérik Pioli Vieira:** Writing – review & editing, Formal analysis, Data curation. **Pedro Esteves Duarte Augusto:** Writing – review & editing, Writing – original draft, Formal analysis, Conceptualization. **Luís Marangoni Júnior:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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