

Evaluation of Properties and Biodeterioration Potential of Polyethylene and Aliphatic Polyester Blends

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Abstract

Blends of high density polyethylene (HDPE) and biodegradable polymers - polylactic acid, PLA, poly(ϵ -caprolactone), PCL and Mater-Bi (thermoplastic starch (TPS) with PLA or PCL) - were prepared in a co-rotating twin-screw extruder, together with polyethylene modified with maleic anhydride (PE-g-MA) used as compatibiliser. The mechanical and rheological properties, morphology and potential for biodeterioration of polymeric materials were evaluated. Blends with PLA showed a reduced elongation at break but an increased Young's modulus while the addition of PCL led to materials with a greater elongation at break and a lower Young modulus. The rheological results evidenced that HDPE and the blend with the highest TPS level exhibited the highest viscosity. The microbial growth test carried out to evaluate the potential for biodeterioration of the blends, using a pure culture of *Pseudomonas fluorescens*, indicated that HDPE/PCL had a lower resistance to bacterial attack than the blend of HDPE/PLA. This was verified by a higher cell number on its surface after 10 weeks of incubation. The addition of 30% starch to the HDPE/PLA blend enhanced its biodeterioration potential, the same was not observed in the case of the HDPE/PCL blend containing just 18% starch.

Introduction

During the last decades, synthetic polymeric materials have progressively grown up to establish one of the most attractive categories of materials. This success is mainly related to their properties namely, low cost, aesthetic qualities and resistance to physical ageing and biological attacks (Vert, 2005).

The well known degradation difficulty of synthetic polymers, together with the growing environmental awareness and the new environmental regulations, are forcing industries to seek more ecologically friendly materials for their products, namely in applications where they are used for a short period

of time before becoming waste. The most desirable long-term solution to this problem is the use of biodegradable plastics, which undergo degradation through the action of living organisms, the most environmentally compatible pathway for solid waste disposal (Long Jiang, L. et al. 2006).

Biodegradable polymers may be derived from biosources (e. g. corn or wood cellulose) or from petroleum sources (Ray and Bousmina, 2005). The best known petroleum source-derived biodegradable polymers are aliphatic polyester and aliphatic-aromatic copolyesters. However, biodegradable polymers derived from renewable resources like polylactides (PLA) are attracting much more attention because of more eco-friendliness from their origin as contrast to the fully petroleum-based biodegradable polymers. Further, the complete biological degradability and compostability in the natural cycle and the protection of the climate through the reduction of carbon dioxide released are some of the other reasons why such materials have attracted the public interest (Mohantya, et al. 2000).

Although the properties of most biodegradable polymers are comparable to many non-biodegradable plastics, the high costs and performance limitations, such as high brittleness, moisture sensitivity, low heat distortion temperature and difficult processability, have restricted the adoption of these materials to relative small niches up to now (Mohantya, et al. 2000). Nowadays, the challenge in replacing conventional plastics by biodegradable materials is to design materials that exhibit structural and functional stability during storage and use, being susceptible to microbial and environmental degradation using adequate solid waste management disposal practices, without any adverse environmental impact (Mohantya, et al. 2000).

Blending biodegradable with non-biodegradable polymers is a method for reducing the overall production cost of the material, which offers simultaneously a scheme to modify both mechanical properties and biodegradation rates. The reasoning behind this approach is that, in principal, if the biodegradable component is present in sufficient amounts and if it is attacked or degraded by microorganisms in the waste disposal site, the plastic or film containing the non biodegradable component should lose its integrity and disintegrate (Chandra and Rustgi, 1998).

Among the common materials used in the packaging industry, the structure and properties of polyolefin-starch blends have been widely studied (Bikiaris et al., 1998; Breslin, 1993; Goheen and Wool., 1991; Rodriguez-Gonzalez et al., 2003; Swanson et al., 1993; St-Pierre et al., 1997; Thakore et al., 2001; Wang et al., 2005; Zuchowska et al., 1998). Blends of polyethylene with a minor amount of starch, where the former constitutes the continuous phase, can be melt processed to obtain products with polyethylene-like properties. Starch, either in its virgin form or chemically modified to increase its compatibility with the polymer matrix, has

been used to form this type of blends. It was found that the effective connectivity and accessibility of the starch, which is required for extensive enzymatic hydrolysis and removal, is achieved only if the starch content exceeds 30 % (Chandra and Rustgi, 1998; Zuchowska et al., 1998). However, increasing the amount of starch leads to a decrease in both tensile strength and elongation at break and the resulting materials have poor forming properties. These worsening properties arise from the different polar characteristics of starch from most of the synthetic polymers, which leads to poor interfacial adhesion. Therefore, a lot of research work has been done in order to improve the compatibility/adhesion between starch and polyethylene, such as the modification of starch (Abdul et al., 2001; Kiatkamjornwong et al., 2001; Thakore et al., 2001), modification of polymeric matrix (Chandra and Rustgi, 1997) and/or the introduction of compatibiliser (Bikiaris et al., 1998; 1999; Ik Yoo et al., 2002; Matzinos, et al., 2002; Nikazar et al., 2005; Swanson et al., 1993) into the blends of starch and PE. The compatibilisers include the ethylene-acrylic acid (EAA) copolymer, polyethylene grafted with maleic anhydride (PE-g-MA), ethylene-vinyl alcohol (EVA) copolymer, etc. EAA is one the most effective compatibiliser used so far, but it must be used in high amounts to achieve satisfactory mechanical properties. Unfortunately, EAA lowers the biodegradation rate of starch, while at the same time it accelerates the thermo-oxidative degradation of LDPE/starch blends when used in low amounts together with a pro-oxidant (Bikiaris et al., 1998). The results showed that using PE-g-MA as a compatibilizer, a much better dispersion of starch within the polyethylene matrix together with a significant reduction in the phase size is achieved (Bikiaris et al., 1998; Chandra and Rustgi, 1998; Nikazar et al., 2005). Concerning the biodegradability, it was observed that the compatibilised blends showed only a slightly lower biodegradation rate compared to their uncompatibilised materials (Bikiaris et al., 1998; 1999; Ik Yoo et al., 2002). Thus, even though the addition of a compatibilizer is a way to prepare biobased plastic materials, some drawbacks, such as decreases in the biodegradation rate and toxicity take place [Bikiaris et al., 1998; Ik Yoo et al., 2002]. In order to overcome these disadvantages and produce materials that could be safely applied in food packaging, other compatibilizers such as citric acid has been used (Ning et al., 2007).

Blending PE with other biopolymers, such as poly(lactic acid) and poly(ϵ -caprolactone), has been also studied (ContatRodrigo and Ribes Greus, 2002; Matinos et al., 2002), in order to increase the mechanical properties of the final materials or the biodegradability. Matzinos et al. (2002) found that the effect of poly(ϵ -caprolactone) on the mechanical properties of LDPE/TPS/PCL materials depends not only on its content but on the final obtained morphology as well. However, a systematic study on biodegradability of these materials is missing.

A biodegradation test of a polymeric material is based on the incubation of the sample in the presence of microorganisms or enzymes, under defined experimental conditions. The degree of polymer biodegradation can be measured according to the mass of carbon dioxide and/or methane evolved, degradation products (e. g., monomers) released, and polymer carbon converted into biomass (Agamuthu and Faizura, 2005; Massar-dier-Nageotte, et al., 2006; Pagga et al., 2001). Standard test methods have been proposed by several international organizations to assess biodegradability of polymeric materials (ASTM, ISO OECD; Pagga, 1997).

This work aims to prepare and evaluate the properties (mechanical and rheological) and the potential for biodeterioration of high density polyethylene (HDPE) blended with biodegradable polymers, such as polylactic acid (PLA), poly(ϵ -caprolactone) (PCL) and Mater-Bi (thermoplastic starch with PLA or PCL) in a co-rotating twin-screw extruder.

Experimental

Materials

High density polyethylene, HDPE 2008SN60 (MFI = 0.7 g/ 10 min at 190°C/2.16 kg), supplied by Total, polyethylene modified with maleic anhydride, PE-g-MA Lotader 3210 (MFI = 5 g/10 min at 190°C/2.16 kg, 3.1% weight (wt.) Maleic Anhydride content) supplied by Arkema, poly(ϵ -caprolactone), PCL CAPA FB100 (MFI = 0.4 g/10 min at 190°C/ 2.16 kg) kindly supplied by Solvay, and poly(lactic acid), PLA Polymer 2002D NatureWorks (MFI = 7 g/10 min at 210°C/2.16 kg) from Novamont were used throughout this work. The starch-based thermoplastic Mater-Bi , TPS, were supplied by Novamont, Novara, Italy. Thermoplastic MaterBi are blends of corn starch/poly(ϵ -caprolactone) (SPCL 70, 30/70 wt.%), corn starch/poly(lactic acid) (SPLA 70, 30/ 70wt. %) and corn starch/poly(L-lactic acid) (SPLA 50, 50/ 50wt. %). The compositions of the samples used in this work are shown in Table 1.

Compounding

All blends were tumble mixed and processed in a Leistritz LSM 30.34 laboratory modular co-rotating twin screw extruder

Sample	HDPE wt. %	PE-g-MA wt. %	PLA wt. %	PCL wt. %	Mater-Bi wt. %
1	100	0	0	0	0
2	30	10	60	0	0
3	30	10	0	60	0
4	30	10	0	0	60 ¹
5	30	10	0	0	60 ²
6	30	10	0	0	60 ³

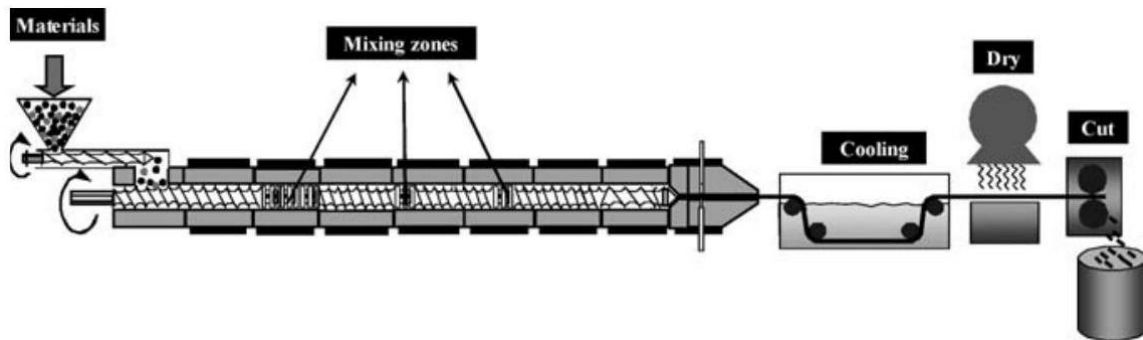


Fig. 1. Laboratory modular Leistritz LSM 30.34 using a barrel temperature set at 190°C, a screw speed of 100 min⁻¹ and a throughput of 3 kg/h. The extruded material was cooled, dried and cut in small pellets. The extruder

screws were design in order to contain a series of transport elements and three mixing zones improving an efficient mixing (Fig. 1).

Material Characterization

Mechanical Properties

The uniaxial tensile properties were evaluated in an Instron 4505 Tensile Machine at room temperature using a crosshead speed of 50 mm/min, relative humidity of 50% and temperature 23°C. The tests were performed using dumbbell specimens previously injection moulded. An extensometer was used to collect low-strain data to measure the Young's modulus. At least 10 specimens of each sample were tested. Prior to mechanical measurements, the samples were dried at 50°C during 24 h in a vacuum oven.

Scanning Electron Microscopy (SEM)

After fracture of the samples in liquid nitrogen and gold plating, the morphology of the blends was studied using a FEI Quanta 400 Scanning Electron Microscope.

Melt Flow Index

The Melt Flow Index (MFI) of HDPE and all blends produced in the above conditions was measured in a Ceast plastometer according to the ASTM standard D1238, at 190°C/2.16 kg.

Rheological Measurements

The samples were characterized in steady shear flow in an Advanced Capillary Rheometer Rosand/Bohlin Instruments RH10. The shear experiments were performed at 190°C in a range of shear rates from 100 s⁻¹ to nearly 2000 s⁻¹, which easily covered the typical extrusion shear rates. The tests were performed two times for each condition, being found an experimental error less than 10%.

Contact Angle

DataPhysics equipment was used to measure the contact angle of liquids with known surface tension on HDPE and polymer blends films, using a dynamic angle. The evolution of the droplet shape was recorded. A CCD video camera and image analysis software were used to determine the contact angle evolution. The polar liquids used in this study were water (surface tension of 73.05mN/m), ethylene glycol (surface tension of 48.40mN/m) and methylene iodide (surface tension of 50.76mN/m) (Kaye and Laby, 1992). The dynamic contact angle of each liquid was measured directly on the polymer film, 3 readings were averaged. The Owens method was used to determine the surface free energy of the polymer films from the contact angles of the three different liquids.

Microbial Growth Test

The potential for biodeterioration of the blends was assessed by the microbial growth test. The growth of a pure culture of *Pseudomonas fluorescens* was evaluated as a function of time with HDPE and all the polymer blends (Table 1) as sole carbon and energy sources.

Pseudomonas fluorescens, an obligate heterotrophic bacterium indigenous in soil, was chosen as the test specimen because it has been extensively used in biodegradation studies of aromatic compounds (Sgountzos et al., 2006) and polymers (Akmal et al., 2003; Kawai and Yamanaka, 1989; Tserki et al., 2006). The experimental procedure was adapted from ASTM G 22-76 (the essays were carried out in liquid phase instead of solid phase). The hypothesis underlying the method is that the growth of *Pseudomonas fluorescens* in a carbon-free media with polymer as the sole carbon source is due to polymer carbon converted into biomass. Each sample, shaped as a disc with 25 mm diameter and thickness of 0.25 mm, was decontaminated with ethanol 70%(v/v) and placed into sterilised conical shaped 100 mL Erlenmeyer flasks containing 40 mL of R2A carbon free medium at pH 7.0. Each flask contained one disk divided into two halves. The disk's density was lower than the water density. The flasks were closed with stoppers connected to air filters and spiked with the pure culture directly from an agar plate and incubated under static conditions at room temperature (22°C) during 10 weeks. Bacterial density on the surface of the polymer, forming a biofilm, was monitored by carrying out a series of total cell counts over a period of 10 weeks. Cells were enumerated by epifluorescence microscopy after DAPI staining (5 min, 2mg/L final concentration) at 1000 × magnification. Fifteen fields were counted per filter for a total of 400 microorganisms. Previously to cell enumeration, the biofilm was detached from the polymer surface in a ultrasound bath (15 min, 60 Hz) and homogenized (20 min) using a tissuemizer with SBS-dispensing tool. Each measurement was carried out in duplicate and a t-test was used where specific means were being compared. Acceptance or rejection of the null hypothesis was based on a α -level of 0.05 in all cases (Miller and Miller, 2005).

Results and Discussion

Blends Properties

Elongation at break, Young modulus and tensile strength results for all tested samples are depicted in Figs. 2 to 4, respectively. As expected, the mechanical properties depend on the blend composition. The addition of PLA (sample 2) or PLA and Mater-Bi (samples 4 and 5) to HDPE increased the stiffness, yielding a material with a higher modulus (1129, 825 and 804 MPa, respectively compared to 550 MPa of HDPE) and a lower elongation at break (6, 3 and 3.1 %). Conversely, the addition of PCL (sample 3) or PCL and Mater-Bi (sample 6) led to higher flexible materials, i. e., increased the elongation at break and decreased the Young modulus. Elongation

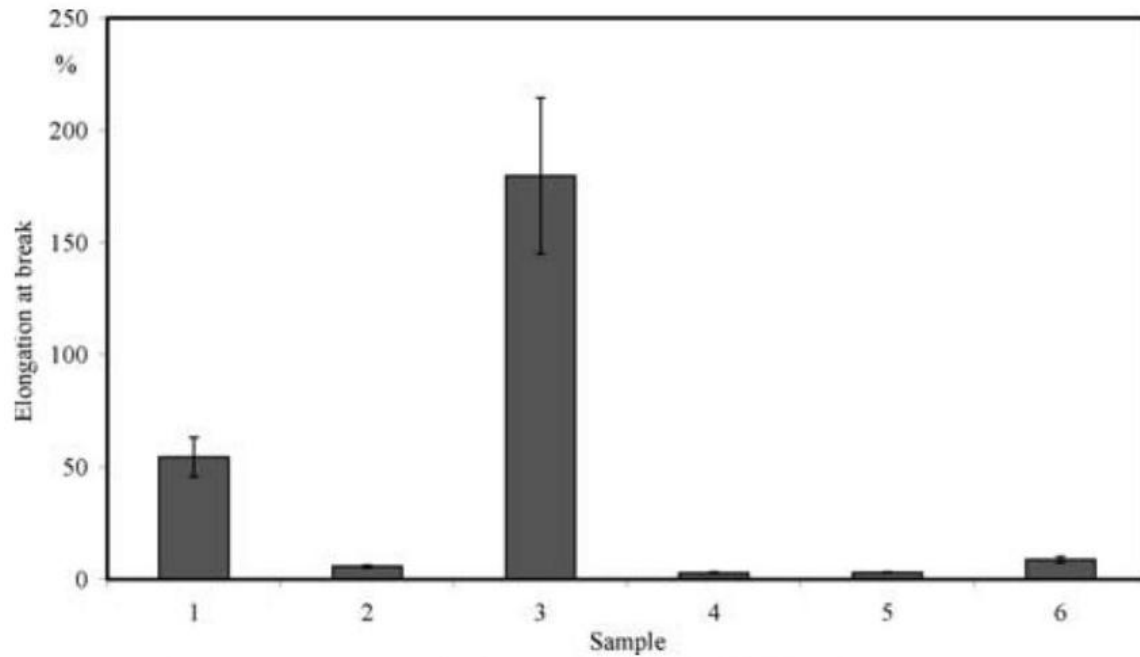


Fig. 2. Elongation at break for HDPE and all blends

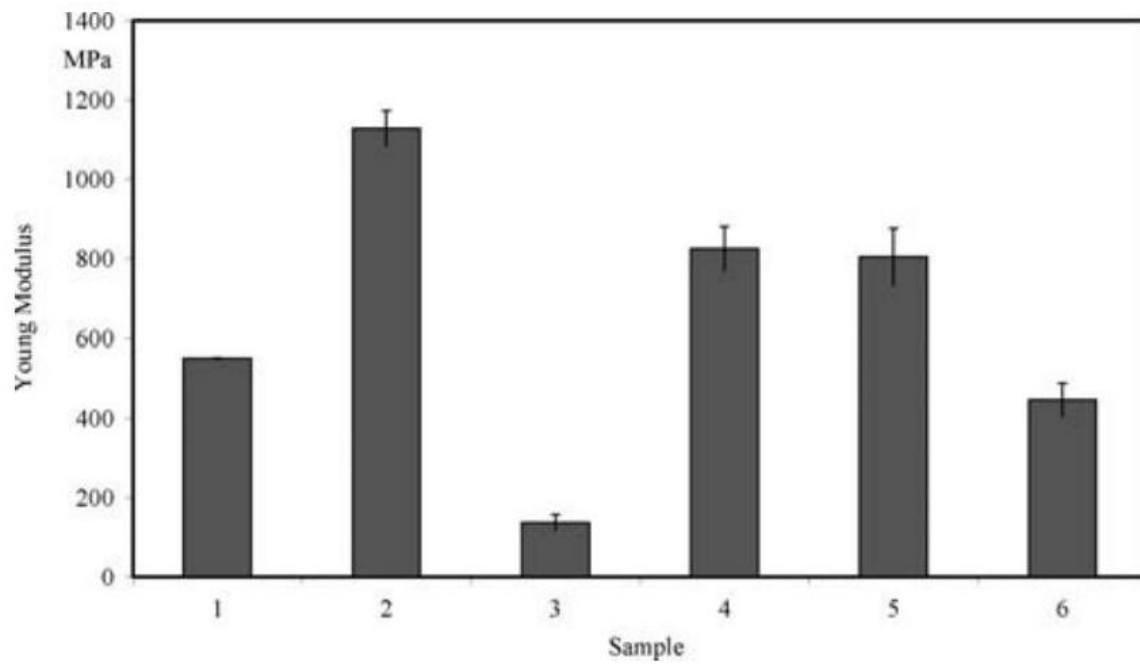


Fig. 3. Young modulus for HDPE and all blends

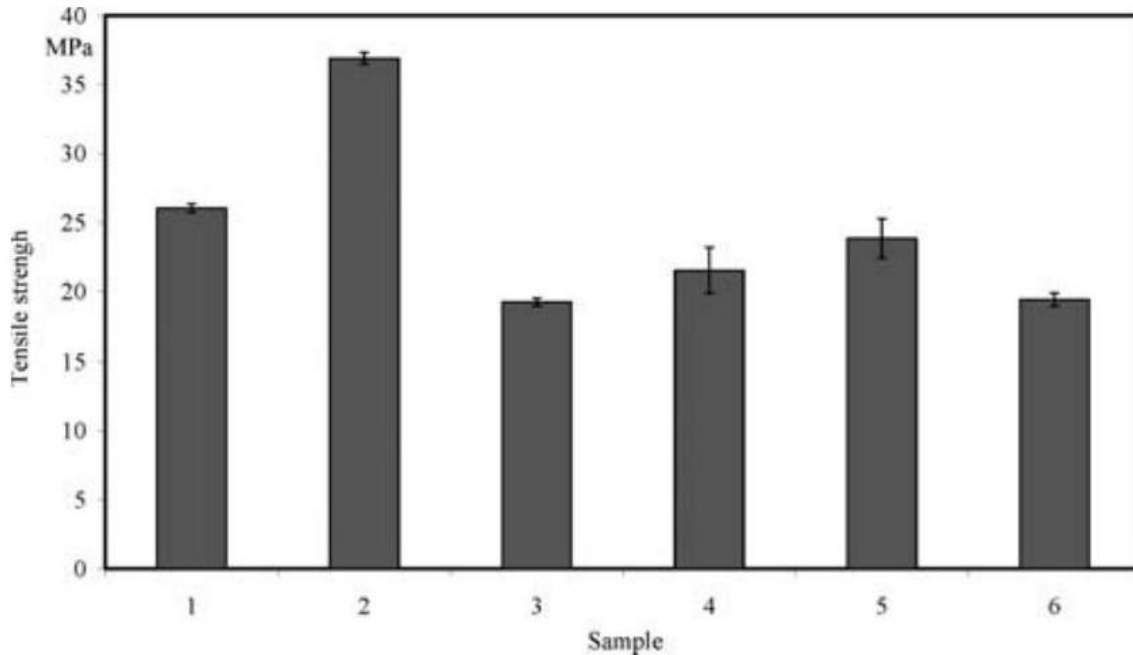


Fig. 4. Tensile strength for HDPE and all blends at break increased from 55 to 180% while Young modulus decreased from 550 to 137 MPa. This effect was however moderate if TPS is also added, as it occurred in sample 6.

Concerning the tensile strength results, the higher tensile strength was obtained for the materials containing PLA. A significant improvement of 42% was observed by the addition of PLA itself (sample 2), whereas for the other samples a lower value than the one obtained for HDPE was achieved. Samples containing PLC, i. e. samples 3 and 6, exhibited a decrease of 37% and 23%, respectively. Regarding the effect of the Mater-Bi, the results showed that for the same starch level (samples 5 and 6) the final mechanical properties were controlled by the amount of PLA and PCL. The addition of starch-based thermoplastic had a minor effect on mechanical properties.

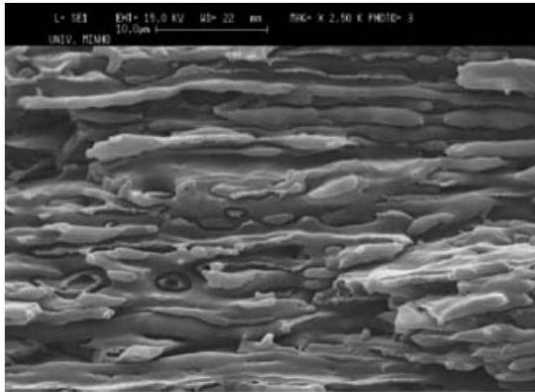
This behaviour can be explained by the mechanical properties of the individual components (HDPE, PLA, TPS and PCL) and by the morphology of the blends. As it is well known, HDPE has good mechanical properties, both elongation at break and modulus. Conversely, PLA and TPS are materials with high stiffness, high modulus and low elongation at break while PCL has low modulus and high elongation at break. The differences in mechanical properties of these polymers can be related with the chemical structure. When compared to PLA, PCL contains a major aliphatic chain having more mobility and flexibility, which explains the high elongation at break.

Since the mechanical properties depend on the morphology of the polymer blend it is crucial to study it. Usually, the minor component of the blend forms the dispersed phase, whereas the major component forms the continuous. The morphology of the blends is controlled by the processing conditions, composition, as well as the nature of the polymers (interfacial tension and viscosity ratio). Fig. 5 depict the morphology of all studied samples, it can be observed that the adhesion between PLA and HDPE is not good (Fig. 5A, sample 2), homogeneous elongated droplets of HDPE can be detected in the PLA matrix. Even with the addition of PE-g-MA, which acts as a compatibiliser by increasing the interfacial adhesion due to the chemical interaction between the hydroxyl groups of

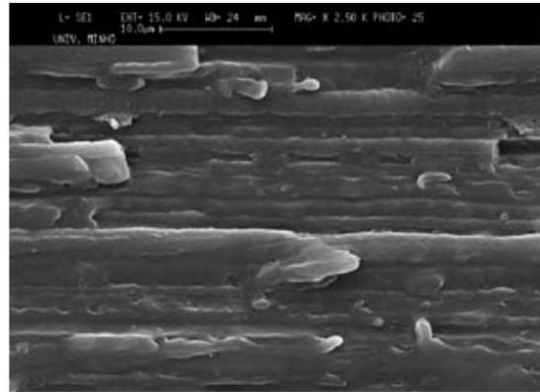
PLA and anhydride groups of PE-g-MA, the adhesion is not good enough to improve the elongation at break. In fact, the elongation at break of sample 2 is similar to the value observed for PLA. In Fig. 5B (sample 3) it is not easy to distinguish both polymers, indicating a good adhesion between PCL and HDPE. The differences observed in morphology when either PLA or PCL are added, can be due to their chemical structures. As it is known, the ratio between ester links/aliphatic chain is higher for PLA than for PCL affecting the polarity (Gross and Kalra, 2002). Thus, the polarity of PLA is higher than that of PCL and consequently the compatibility of PCL with HDPE is better.

Sample	1	2	3	4	5	6
MFI (g/10 min)	0.79 ±0.00	7.61 ±0.16	3.16 ±0.04	0.50 ±0.01	1.81 ±0.01	1.47 ±0.01 36.5
Surface Energy (mN/m)	31.9	40.2	34.9	40.0	41.3	

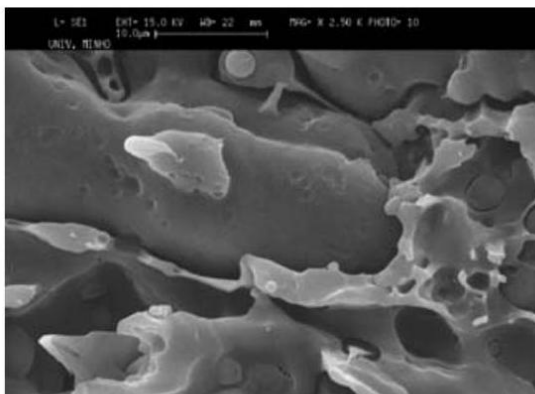
Table 2. Melt Flow Index at 190°C and surface energy of all samples



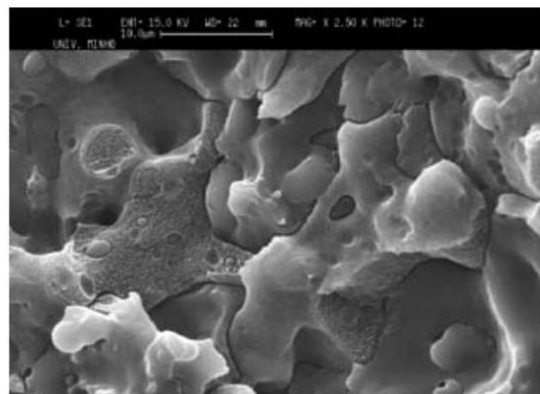
A)



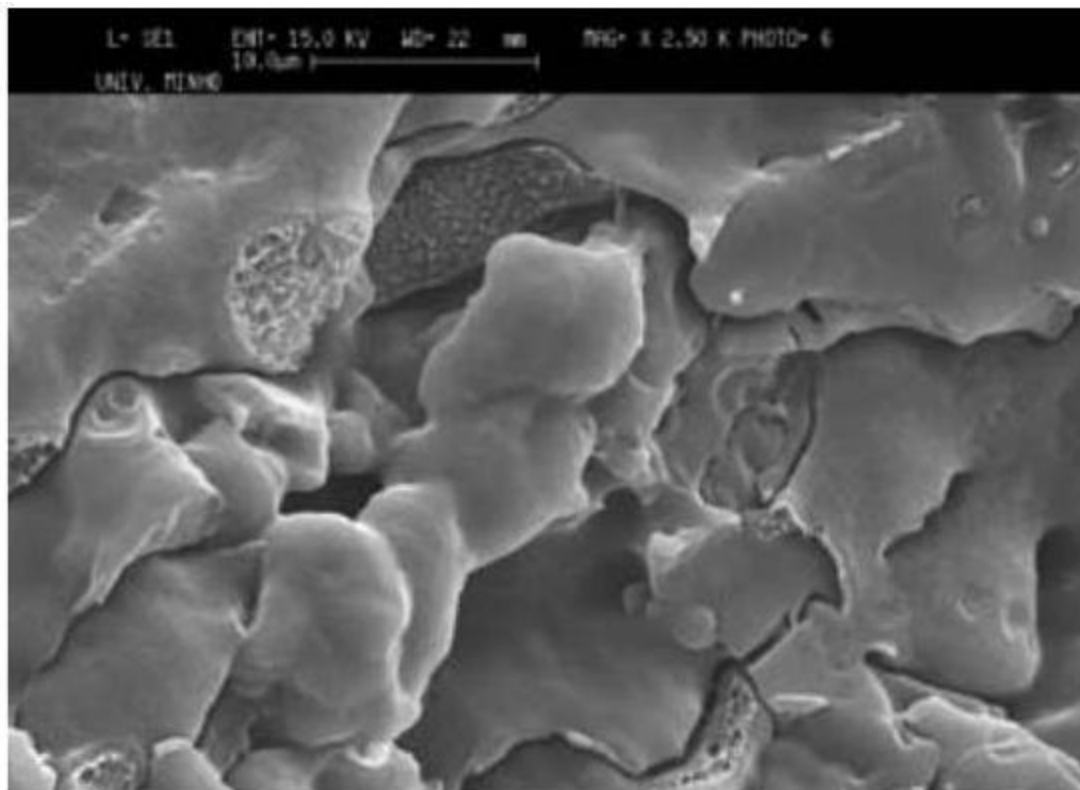
B)



D)



E)



C)

Fig. 5. SEM micrographs of blends with PLA (A), PCL (B), SPLA 50 (C), SPLA 70 (D) and with SPCL (E)

The morphology of all HDPE/Mater-Bi blends (Figs. 5C to E, samples 4, 5 and 6) is similar, the individual components of the blends can be detected, which can be associated with the interfacial tension among the components. As in the case of the PLA, the addition of PE-g-MA was not efficient enough to promote a good adhesion between the blend components.

MFI and shear steady viscosity at 190°C in the shear rate range between 100 s⁻¹ and 2000 s⁻¹ for polyethylene and all produced blends are presented in Table 2 and Fig. 6, respectively, in order to evaluate its processability. As it can be seen, the final MFI of the samples ranged between the MFI of each individual component, keeping in a general way the value of the polymer present in higher amount (in the case of PLA and PCL). Concerning the effect of the Mater-Bi addition, the results showed that increasing the starch-based thermoplastic quantity led to a material with a higher MFI value.

Shear viscosity measurements showed that HDPE and sample 4 exhibited the highest values, as anticipated from the above MFI results. The effect of the addition of a biodegradable polymer to HDPE is also in accordance with the MFI results, being the lower viscosity observed for blends with PLA and PCL (samples 2 and 3) and slightly higher for Mater-Bi. Furthermore, it can be observed that the blend with higher starch-based thermoplastic content (sample 4) showed a higher viscosity. This behaviour, i.e., an increase in viscosity for a rising level of TPS, was also observed in previous work (Rosa et

al., 2007; Shin et al., 2004). For processing purposes, the above mentioned results show that blending HDPE with biopolymers leads to materials which can be extruded more easily. However, in order to overcome some potential processing difficulties, care must be taken concerning to the set extrusion temperature profile.

Surface tension can be defined as the amount of work necessary to form a new part of surface (against air or vapour) (Brady et al., 2000). Since the aim of this work is to evaluate the

biodegradation potential through bacterial growth attached to the polymer surface, measurements of surface tension can be very useful to correlate with the microorganisms' adhesion onto the polymer surface. It is known, that the lower the surface energy the worse the adhesion achieved between the microorganisms and the polymer surface (Tsibouklis et al., 1999). The data presented in Table 2 shows the results obtained for all the samples, which exhibit surface energy values in the range 31.0 to 41.3mN/m.

All samples have higher surface energy than HDPE (sample 1). The latter has a highly hydrophobic macromolecular chain explaining the low surface energy value obtained (Tsibouklis et al., 1999). It was observed that the addition of PLA increased the surface tension of ca. 26 % while PCL contributed only to an increase of 9.4%. On the other hand, for the samples with Mater-Bi, the ones with PLA (samples 4 and 5) have similar results to sample 2. The same behaviour was observed for samples 3 and 6. These values indicated that the addition of starch-based thermoplastic had an insignificant effect on the surface tension.

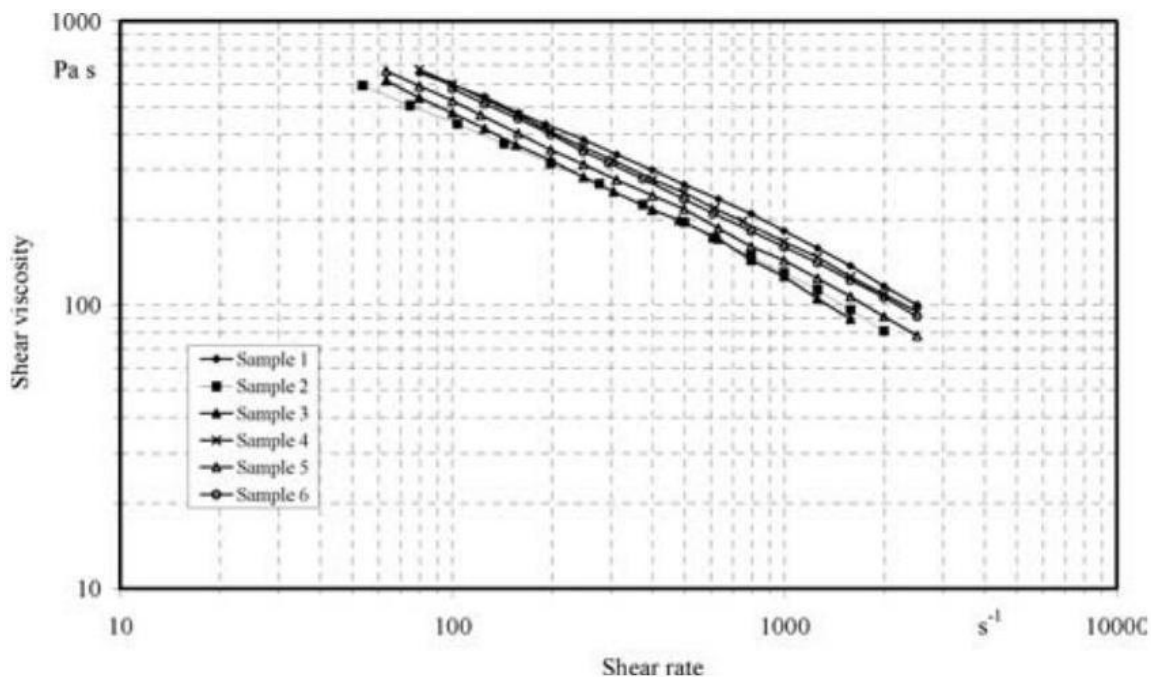


Fig. 6. Shear viscosity for all HDPE blends at 190°C

Biodeterioration Potential of Blends

Blends were incubated in the presence of *Pseudomonas fluorescens*, under defined experimental conditions, and an increase of cell counts in time was observed in the biofilm

formed on the surfaces of samples 2 to 6, as depicted in Fig. 7. HDPE (sample 1) shows the lowest cell count while HDPE/SPLA 50 (sample 4) exhibits the highest. This result might be related with both surface energy, being lower for HDPE thus unfavourable to cell adhesion (Tsibouklis et al., 1999) and resistance to microbial attack, which is lower for HDPE/SPLA 50 due to the presence of starch and PLA (Gross and Kalra, 2002). The ratio between bacterial counts in the biofilm obtained after ten and two weeks for samples 1, 2 and 3 (Table 3), presents the highest value in the case of HDPE/PCL blend (sample 3), suggesting that PCL is less resistant to bacterial attack than PLA. Massar-dier-Nageotte et al. (2006) reported that PCL and a TPS/PCL blend seemed to be rather biodegradable compared to PLA, in a biodegradability study carried out under aerobic conditions and in liquid phase.

The effect of increasing amounts of starch-based thermoplastic (0%, 18% and 30%) on the biodeterioration potential of PLA blends are presented in Table 3. The present study indicates that the ratio between bacterial counts obtained after ten and two weeks of experiment lifetime is not significantly different in the cases of the blends containing 0% and 18% of starch but increases significantly in the case of 30% (t-test).

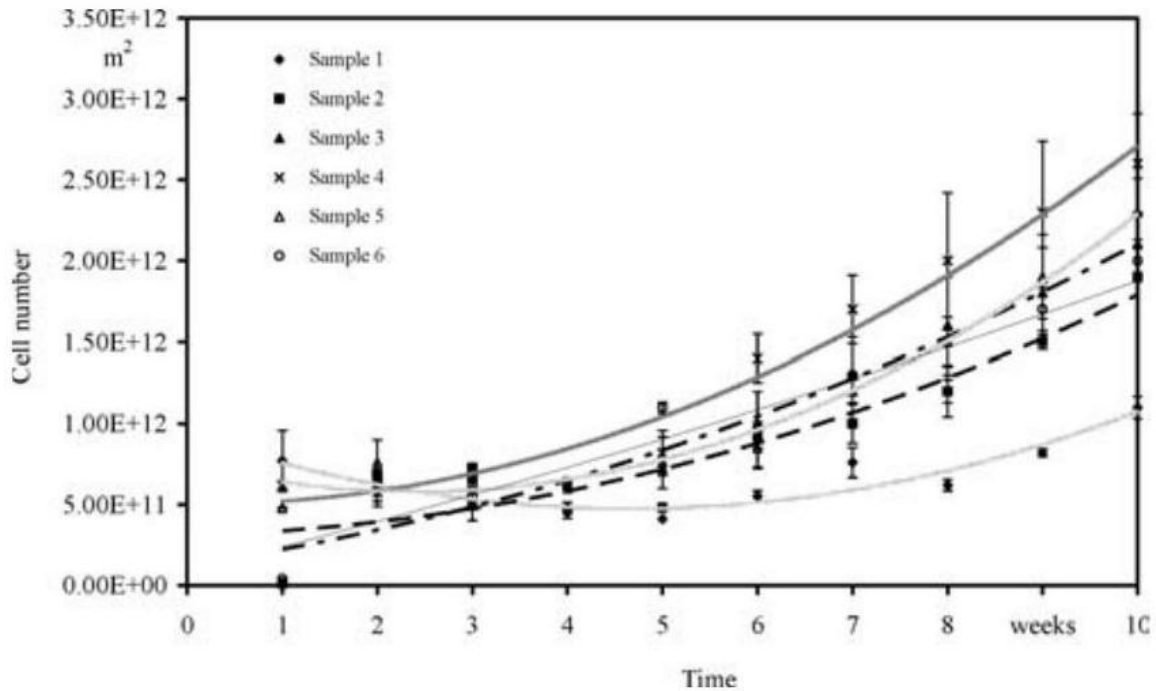


Fig. 7. Growth curves of *Pseudomonas fluorescens* in the biofilm formed on the surface of polymeric materials as determined by bacterial counts

Sample number	Cell number ratio (10 th week/ 2 nd week)
1	1.75 ± 0.14
2	2.71 ± 0.31
3	3.49 ± 0.80

4	4.59 ± 1.21
5	2.98 ± 0.83
6	3.29 ± 0.27

Table 3. Ratio between the biofilm bacteria number on polymeric samples quantified in the 10th week and in the 2nd week of experiment lifetime. Values listed in the table are the average \pm 95% confidence interval

The results suggest that the amount of starch might have been too low or simply not available at the polymer surface for bacterial growth for the blend containing 18% starch. At 30%, starch decreased the resistance of the blend to bacterial attack decreased and promoted microbial growth. This result might be attributed to crystallinity and hydrophobicity of starch (Thakore et al., 2001). Usually, the biodegradation occurs preferably in the amorphous regions because of the higher mobility of the chains and their higher accessibility to the microorganisms. Starch, being less crystalline compared to PLA, is more prone to microbial attack. Additionally, its hydrophilic nature characterized by a higher number of hydroxyl groups in structure compared to the one present in PLA promotes swelling in the culture medium enhancing biodeterioration. The effect of 18 % starch-based thermoplastic on the biodeterioration potential of a PCL blend (sample 6) was not significant (Table 3), as in the case of PLA and the same explanation for the observed result might be feasible. However, more detailed knowledge on the kinetics of the biological reactions and its products needs to be gained and further work is required.

Conclusions

The purpose of this work was to prepare and evaluate the properties of blends of non-biodegradable polymers (HDPE) and biodegradable polymers, such as polylactic acid, poly(ϵ -caprolactone) and Mater-Bi (including PLA/TPS or PCL/TPS). Samples were prepared in a co-rotating twin-screw extruder in order to improve mixing.

The uniaxial tensile tests showed that the addition of PLA leads to materials with a higher modulus and a lower elongation at break than HDPE. Conversely, the addition of PCL increases the elongation at break and decreases as expected, Young's modulus. The addition of Mater-Bi has a minor effect on mechanical properties. The rheological results showed that the shear viscosity of all samples is lower than that measured for HDPE, indicating that the blends are easily processed.

In considering the biodeterioration potential of the blends, it was found that HDPE/PCL has a lower resistance to bacterial attack than HDPE/PLA verified by a higher cell number on its surface after 10 weeks of incubation. The addition of 30% starch to the HDPE/PLA blend enhanced its biodeterioration potential, the same was not observed in the case of the HDPE/ PCL blend containing only 18% starch.

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1	SPLA	50	(50	wt.%	TPS	+	50	wt.%	PLA)
2	SPLA	70	(30wt.%	TPS		+70wt.%		PLA)
3	SPCL	70	(30wt.%	TPS	+	70wt.%		PCL)

Table 1. Samples composition