Development of multilayer films from biobased and biodegradable polymers and food waste.

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The massive use of plastics and the incorrect management of their waste after their useful life is a major problem. In 2019, plastics production in Europe almost reached 58 million tonnes, of which approximately 23 million tonnes were used for packaging, especially for fresh food products (PlasticsEurope, 2020).

Bioplastics are being developed to address some of the problems arising from the use and waste management issues of conventional petroleum-based plastics. There are a large number of bioplastics, developed from biobased and/or biodegradable polymers (Jost, 2018), but only some of them are marketed in appreciable quantities. One of them is poly(lactic acid) (PLA), which is currently used in demanding applications such as medicine, 3D printing or packaging, among many others.

One of the main drawbacks for the expansion of bioplastics is their relatively poor properties, in comparison with conventional plastics. For instance, they can show high permeability (low barrier) to O_2 and H_2O, which limits their application in food packaging (Beltrán et al., 2020). Among the alternatives to obtain films of biodegradable materials that show a sufficient barrier to be used in food packaging, an interesting possibility is the design of multilayer films from different biobased and biodegradable polymers (Meléndez-Rodríguez et al., 2021). The barrier properties can be further improved by using additional barrier layers obtained from food waste.

The materials selected in this work for obtaining multilayer films are PLA and poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), two of the most accessible and important bio-based and biodegradable polymers nowadays. In order to improve the gas barrier properties of the multilayer films, other biobased layer can be incorporated. In this work, an inner layer of thermoplastic chitosan, obtained from chitin, a food waste, has been selected as barrier layer. The method of obtention of thermoplastic chitosan film is based on the work of Dou et al. (2020). The chitosan film can be treated to improve its adhesion to PLA and PHBV layers.

Commercial grades of PLA (Ingeo Biopolymer 2003D) and PHBV (Ercros PH016) were used. Before treatment, the PLA and PHBV were dried in an oven at 40ºC, under vacuum conditions, overnight. The PHBV and PLA were melt compounded using a Rondol Microlab twin-screw extruder with L/D = 20 and a screw speed of 60 rpm. The applied temperature profile, from the hopper to the die, was 130ºC, 170ºC, 190ºC, 190ºC, 180ºC for PHBV; and 130ºC, 170ºC, 190ºC, 190ºC, 180ºC for PLA. The obtained materials were then compression moulded into 100 µm films using an IQAP-LAP hot-plate press at 185ºC. Multilayer films were obtained in the same press at 130 °C.

Confocal Raman microspectroscopy was used to study the dimensions, structure and chemical composition of the multilayer films. Raman spectra were obtained with 32 scans between 100 and 3400 cm^{-1}, using a 532 nm laser. The Raman spectra were measured at different points in a cross-section of each multilayer.

Figure 1 shows a bilayer film with a thickness of approximately 210 µm. Raman microspectroscopy allows to measure not only the spectra of both layers but also the spectra of both sides of the interphase (Figure 2).

The spectra recorded at the PLA side of the interphase (Interphase 1) are slightly different from the spectra recorded at the centre of the PLA layer. The small differences can be explained as a result of differences in crystallization, due to the interaction with PHBV in the interphase.

Figure 1. Micrograph of a PLA-PHBV bilayer

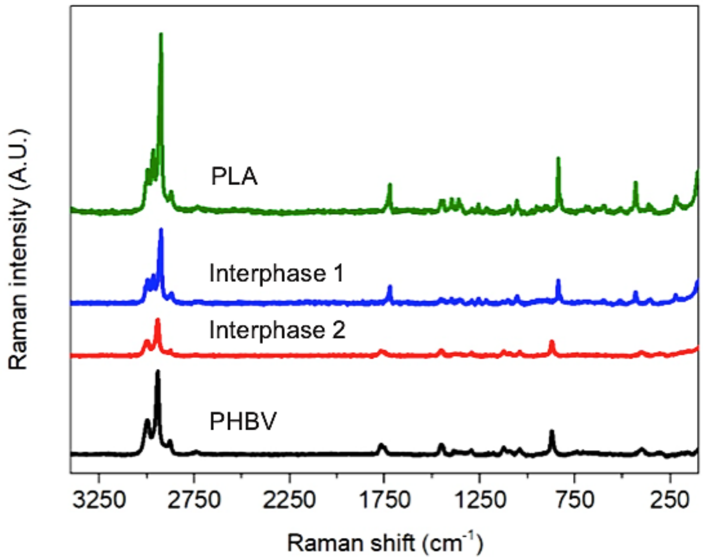

Figure 2. Raman spectra corresponding to the bilayer of Figure 1.

The gas permeability of O_2 was measured using a homemade permeation cell (ISO 2782 standard), with a gas pressure of 1 bar and at a temperature of 30 °C. The results indicate that multilayer films with acceptable barrier properties can be obtained from biobased and biodegradable polymers and food waste.

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