November 30th 2022
December 2nd 2022
Hotel Academia,
Zagreb, Croatia

PAM Food Processing and Microbiology Research Unit

10th International Congress of Food Technologists, Biotechnologists and Nutritionists

Processing of biodegradable PHBV and chitosan films as a multilayer structure for food packaging application

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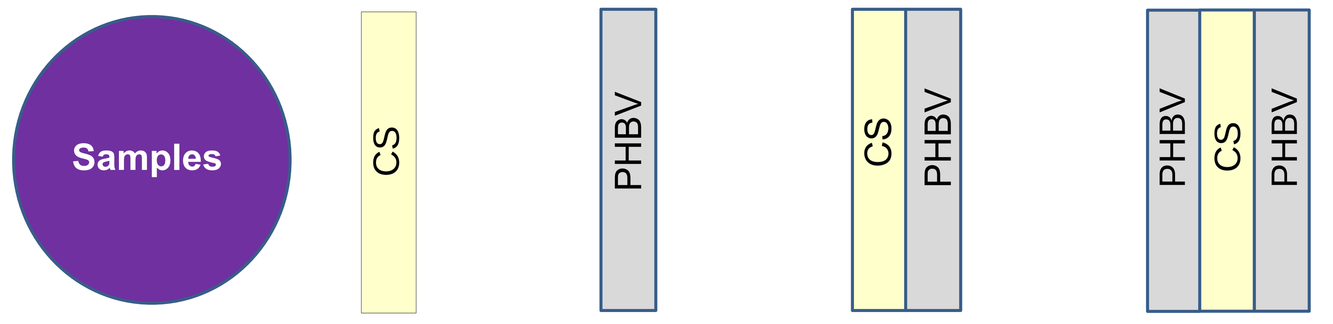
Introduction and objective

PHBV (3-hydroxybutyrate-co-3-hydroxyvalérate) is currently considered as one of the most promising substitutes of conventional plastics, with low environmental impact, especially for food packaging applications. Nevertheless, PHBV has disadvantages when compared to currently used thermoplastics. Indeed, high permeability to oxygen, high costs and thermal instability are still limiting its industrial applications[1] . The objective of this study was to improve the barrier performance of PHBV without compromising its sustainability (biodegradation, circularity,..). Chitosan, a by-product coming from seafood industries was used to produce multilayered PHBV-Chitosan-PHBV films with improved barrier and thermal efficiencies. This work deals with the assembling and shaping bi- and three-layer films (PHBV-Chitosan and PHBV-Chitosan-PHBV) and their impact on barrier (WVP), mechanical, thermal, surface and adhesion properties.

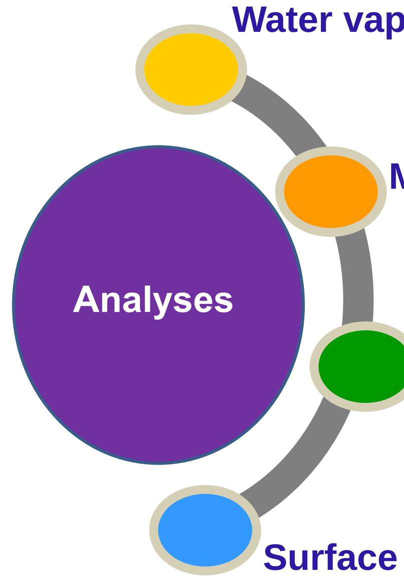
Materials and methods

mono, bi and three-layers Film preparation

- PHBV films were prepared by the solvent casting method (3% w/v in chloroform).
- Chitosan (CS) films were obtained from the wet casting method (2% w/v in an aqueous 1% (v/v) acetic acid solution plasticized with glycerol at 15% w/wt).
- Hot-press (HP) technique was used for assembling and shaping the bi- (PHBV-CS) and three-layers (PHBV-CS-PHBV) films and : PHBV-CS or PHBV-CS-PHBV were fixed between two plates and heated until 130°C at 10°C/min, isothermal at 130°C for 10min and then cooling at 15°C/min until 25°C. A constant pression at 400 bars was applied upon heating.



Physico-chemical properties



Water vapour permeability (WVP)
- Gravimetric method adapted for hydrophilic films [2], at 25°C
- ΔRH = 33-75%; 33-84% and 33-100%

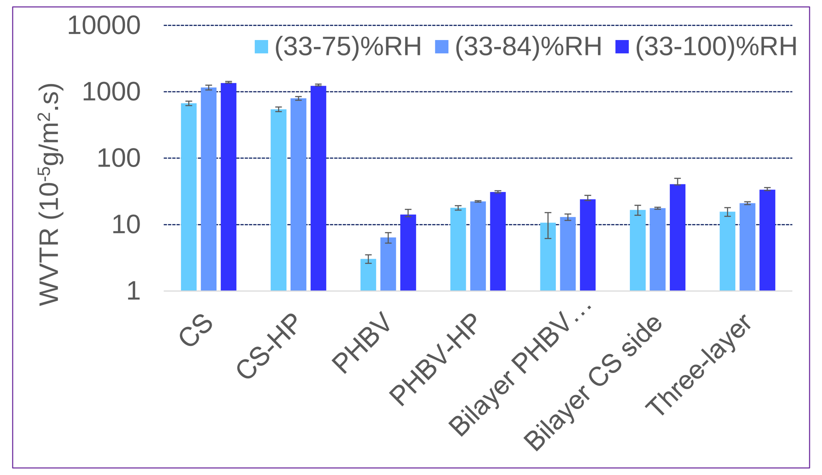
Mechanical properties
- NF EN ISO 527-1 [3]
- Tensile strength TS (MPa), Elongation at Break EAB (%)

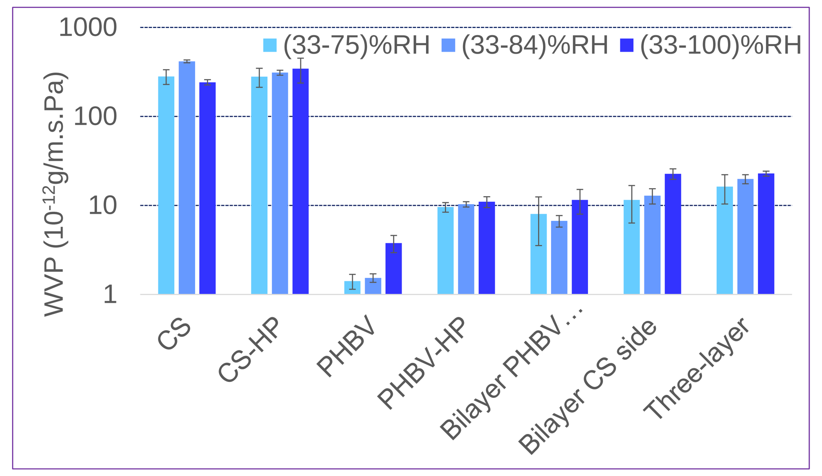
Thermal properties
- Thermal gravimetry TGA from 25 to 600 °C
- Thermal stability (from 25 to 500°C, 20°C/min)

Surface properties
- Work adhesion (mN/m)
- Water contact angle (°)
- Surface free energy SFE(mN/m)

Results

Water barrier properties

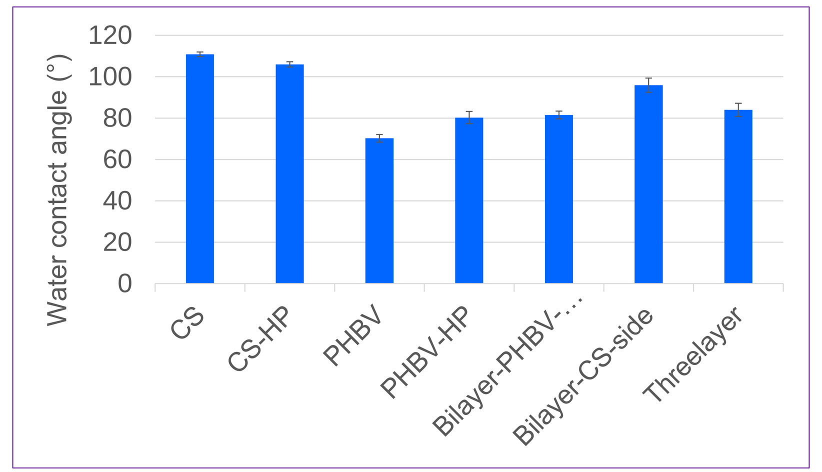


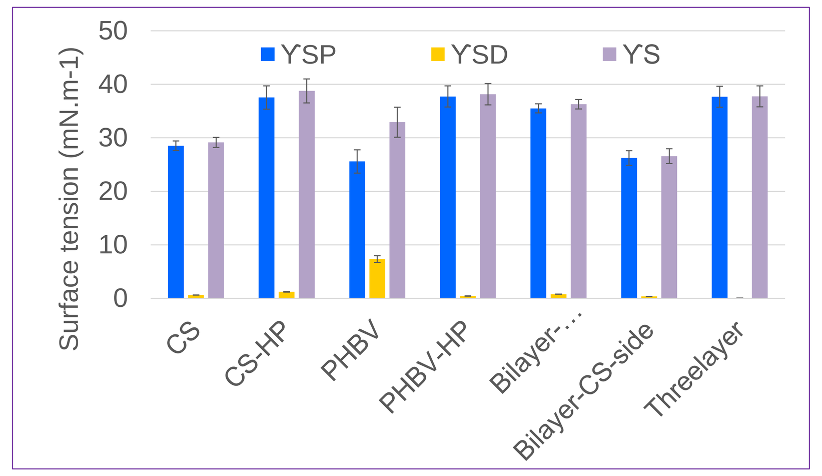


The HP increase significant the WVTR and the WVP for PHBV film without any significant effect on chitosan film compared to no-treated films. The WVTR of bilayer and three-layers films made by HP are between the CS-HP and PHBV-HP monolayer hot-press treated films for the three RH differentials conditions.

The WVP for a relative humidity differential of 33-75% decreases from 279±6 (10^{-12} g/m.s.Pa) and from 9.5±1.2 (10^{-12} g/m.s.Pa) for chitosan and PBHV monolayer films respectively, to 8±4.4 (10^{-12} g/m.s.Pa) for bilayer chitosan-PHBV. No significant difference was noticed between bi- and three-layers. The same behavior was observed for the other RH gradients tested (33-84 and 33-100%).

Surface properties

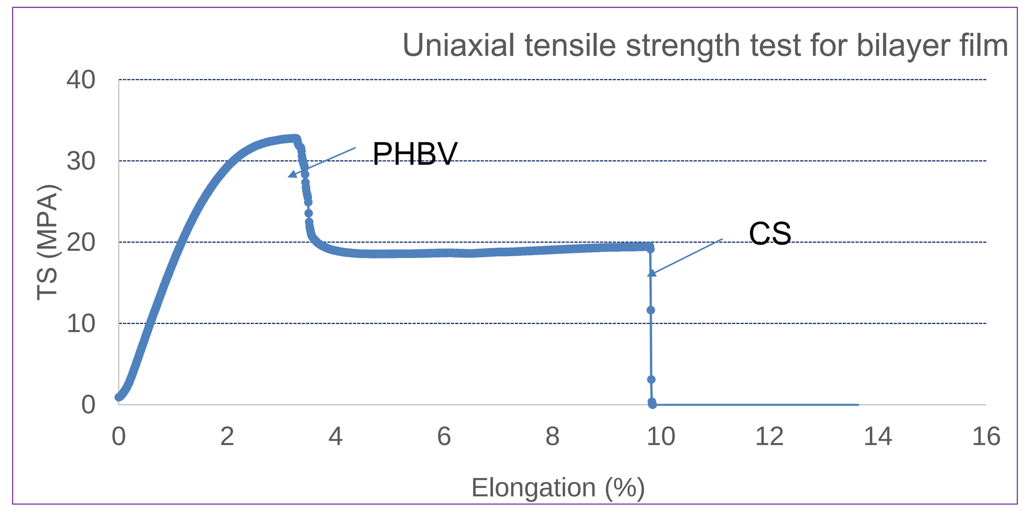


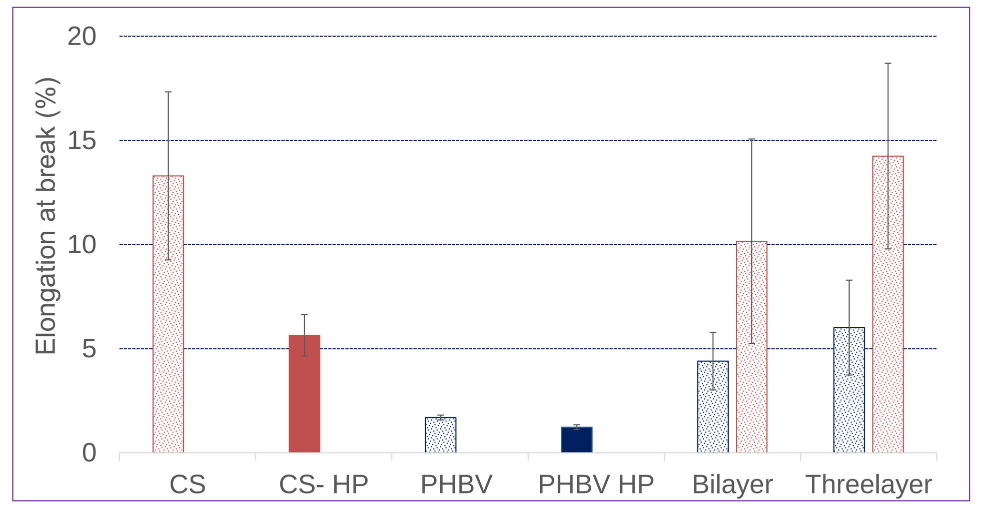


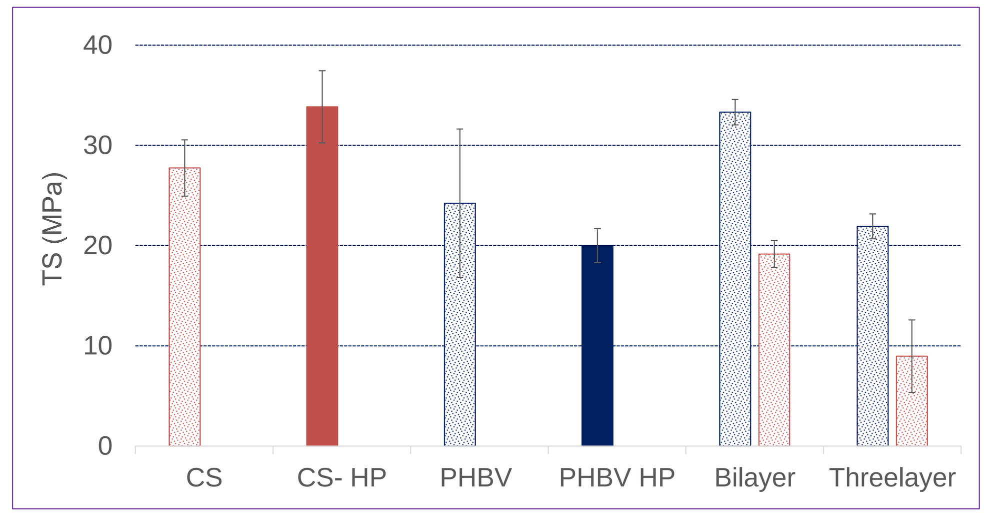
The hot-press process allowed good adhesion between the chitosan and PHBV layers. Indeed, the work of adhesion, determined from the contact angle and surface tension of each monolayer films, is about 58 $mN.m^{-1}$. This value indicates suitable adhesion for polymer assembly at industrial scale and well as a compatible surface energy for printing.

The water contact angle (°) of bi and three-layers films were between the monolayer CS and PHBV films.
The polar component of surface tension of film increase when bilayer and three-layers was shaped.

Mechanical properties







Firstly, PHBV film breaks more rapidly than CS which is more ductil. In a second time, we notice that the HP effect makes the films more brittle. Then, the assembly of the layers induced a higher TS for the multilayer films. The TS of chitosan and PHBV monolayers are respectively 27.7 and 24.2 MPa and rises up to 33.3 MPa for the bilayer films. As observed for the mechanical properties the thermal stability of multilayer films was also enhanced compared to monolayer films.

Conclusion

This study evidenced the potential of PHBV-Chitosan and of PHBV-Chitosan-PHBV complexes as a valid sustainable substitute of conventional plastics. The gas barrier properties (O_2, CO_2, N_2) will be assessed for MAP applications. It also could open an unexplored PHBV market opportunity. In addition, further investigations on PHBV-based laminates using other biopolymers valorized from agro-industrial waste or byproducts will be of great interest.

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With the support of the Région Bourgogne Franche-Comté (ANER ActiBioPack No., 2020-Y-12730) and EU-FEDER

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