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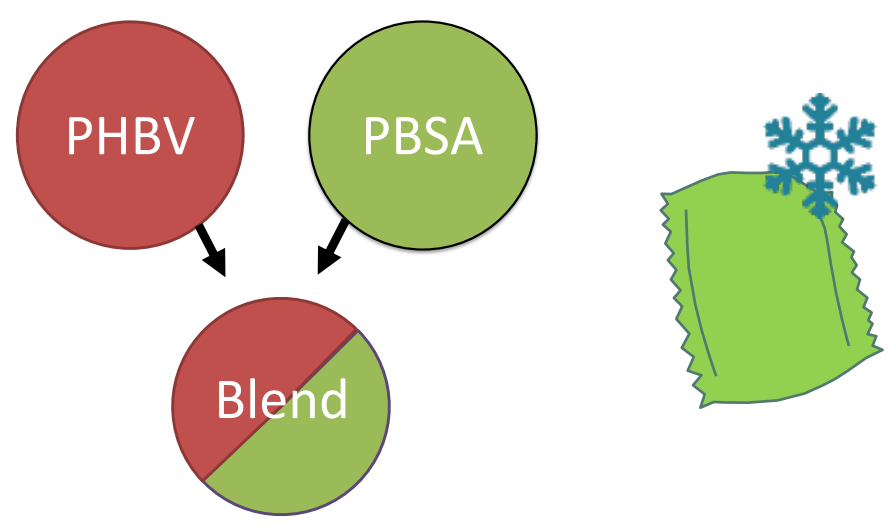
Film blowing of biodegradable poly(hydroxybutyrate-co-hydroxyvalerate) and poly(butylene succinate-co-adipate) blends

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Introduction

Objective: Formulation of frozen food packaging using biobased, locally sourced and biodegradable polymer blends



Polyhydroxyalkanoates (PHAs) are thermoplastic polyesters produced as carbon and energy storage compounds by microorganisms under nutritional stress. Short-chain-length PHAs, mainly represented by poly(3-hydroxybutyrate) P(3HB) and the copolymer poly(3-hydroxybutyrate-co-3-hydroxyvalerate)P(3HB-co-3HV) are already commercially available. PHBV can be produced using carbon sources from waste or by-products of agriculture and food industries. This would further promote circular economy because local food waste could be used to make food containers. PHBV is a stiff, brittle, highly crystalline polymer with low thermal stability and a narrow processing window. One strategy that can be adopted to overcome these drawbacks is based on physical melt-blending. *In-situ* compatibilization can be used to improve the mechanical properties thanks to the enhancement of interfacial adhesion between both phases in binary blends. We developed PHBV/PBSA blends compatibilized with dicumyl peroxide (DCP) and investigated the morphological rheological, thermal, mechanical characteristics, and processability. The formulation, optimized using melt blending, could be processed using film-blowing extrusion. Blown films were then tested in a service situation, corresponding to frozen food packaging.

Blend processing

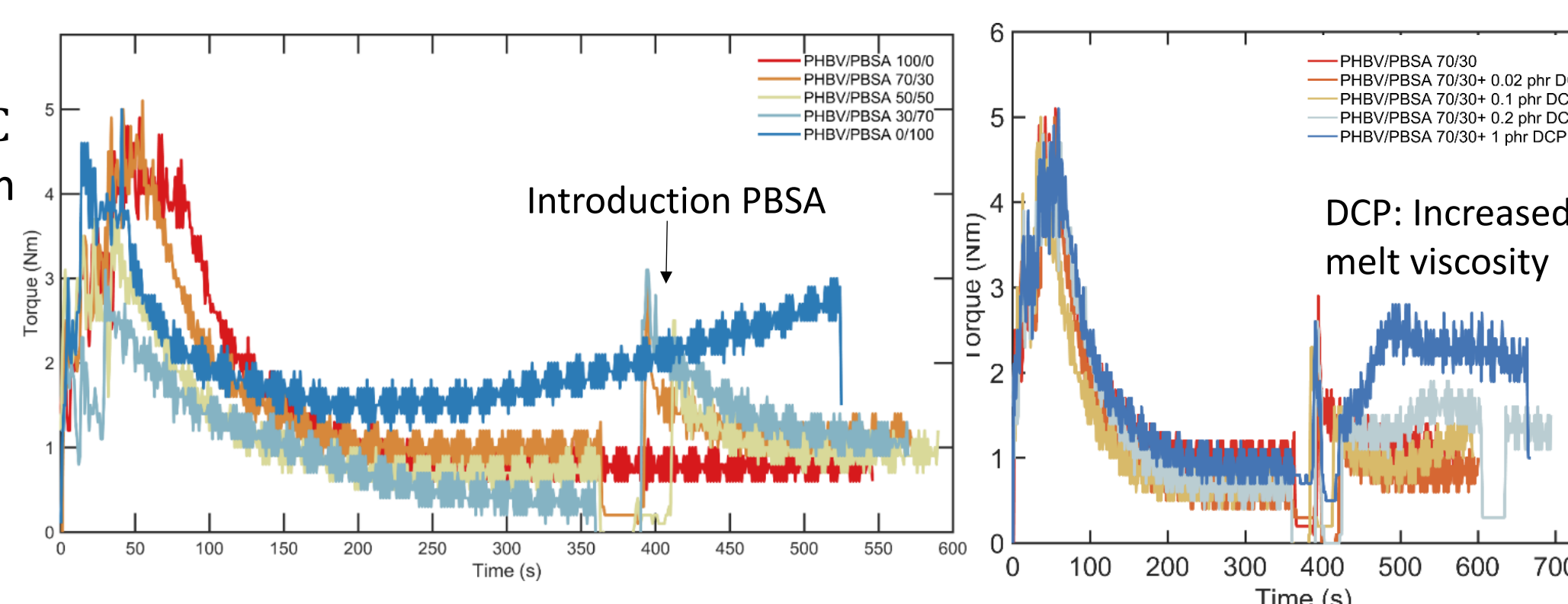
Development of the compatibilized blend using an internal mixer



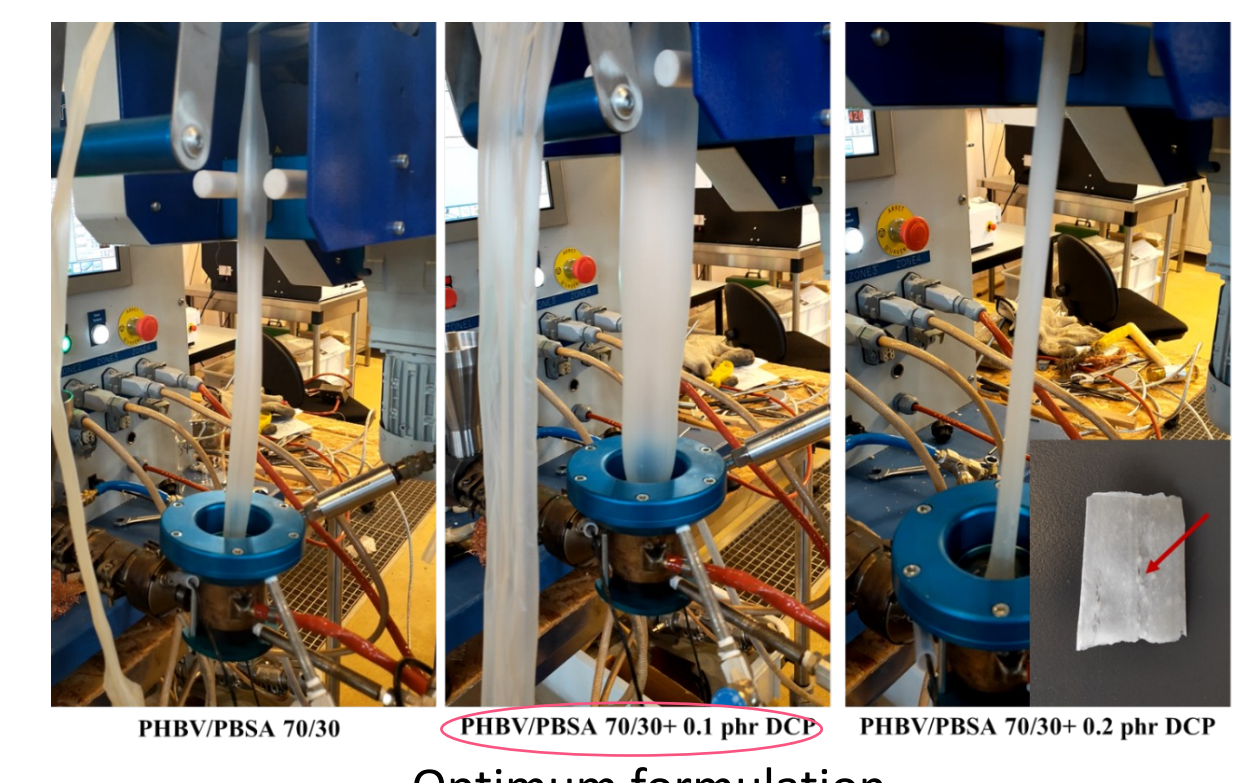
Blending:

- Temperature: 190°C
- Screw speed: 90rpm
- Time: 9'

Sample film fabrication:
Thermo-compression at 180 °C, 150 bar



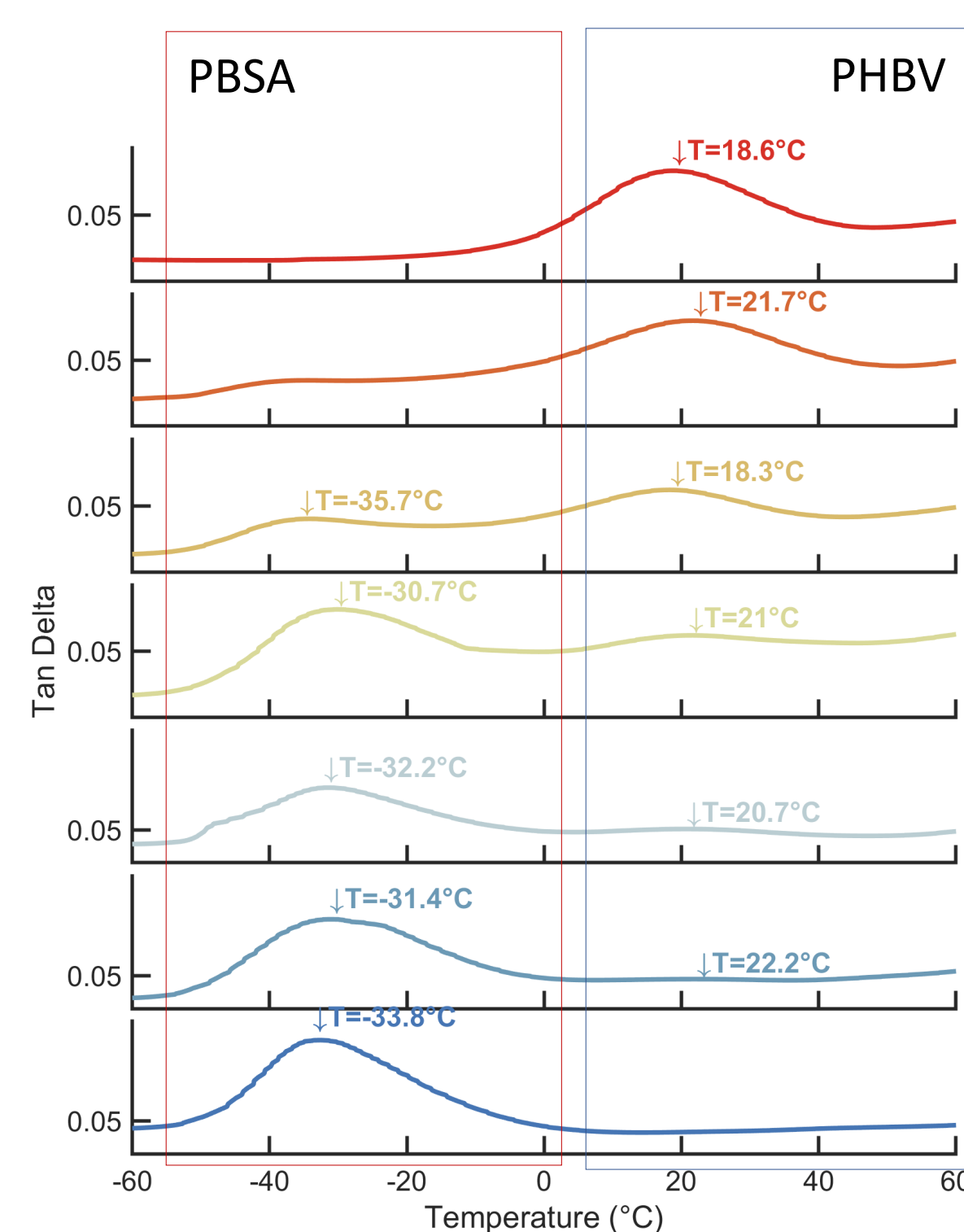
Impact of DCP on film blowing



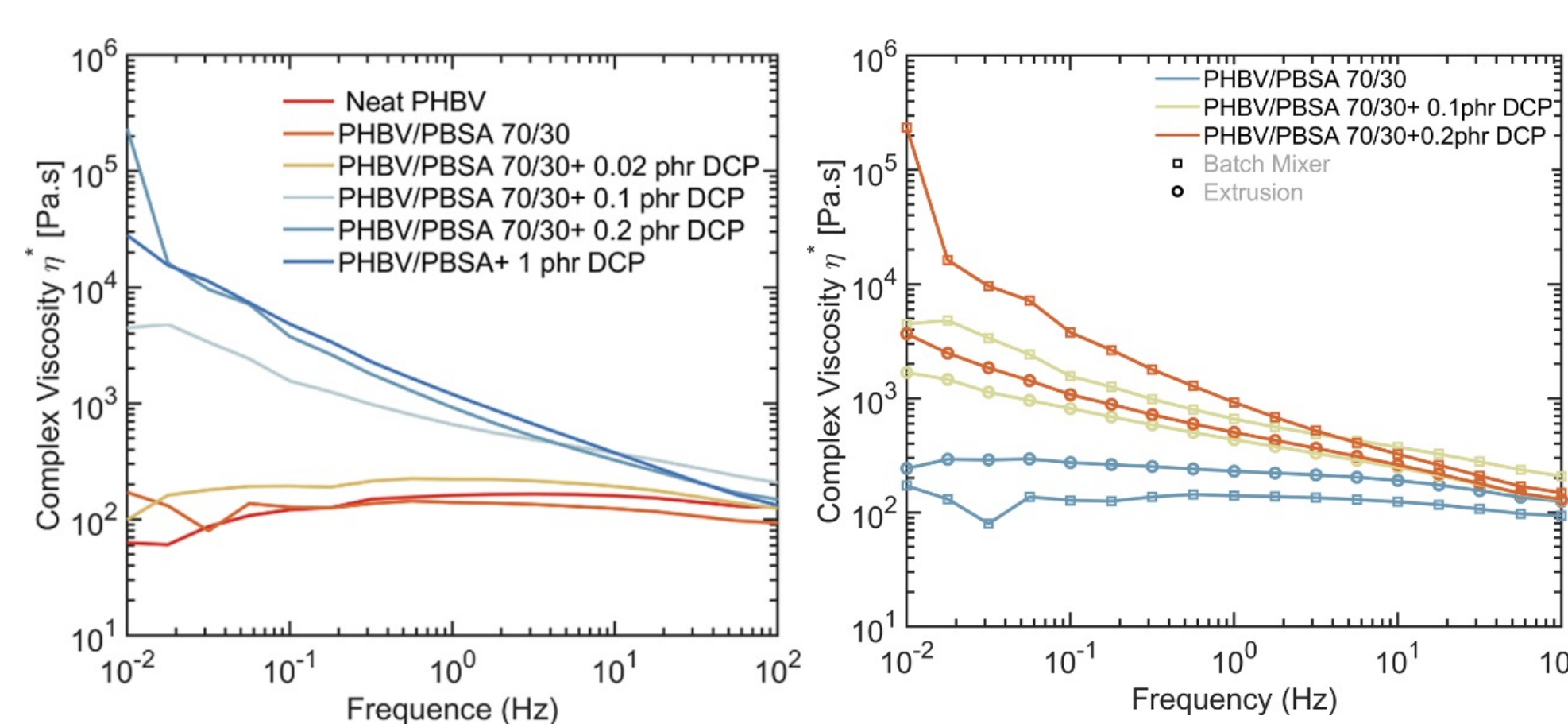
Optimum formulation

Results

DMA analysis of PHBV/PBSA



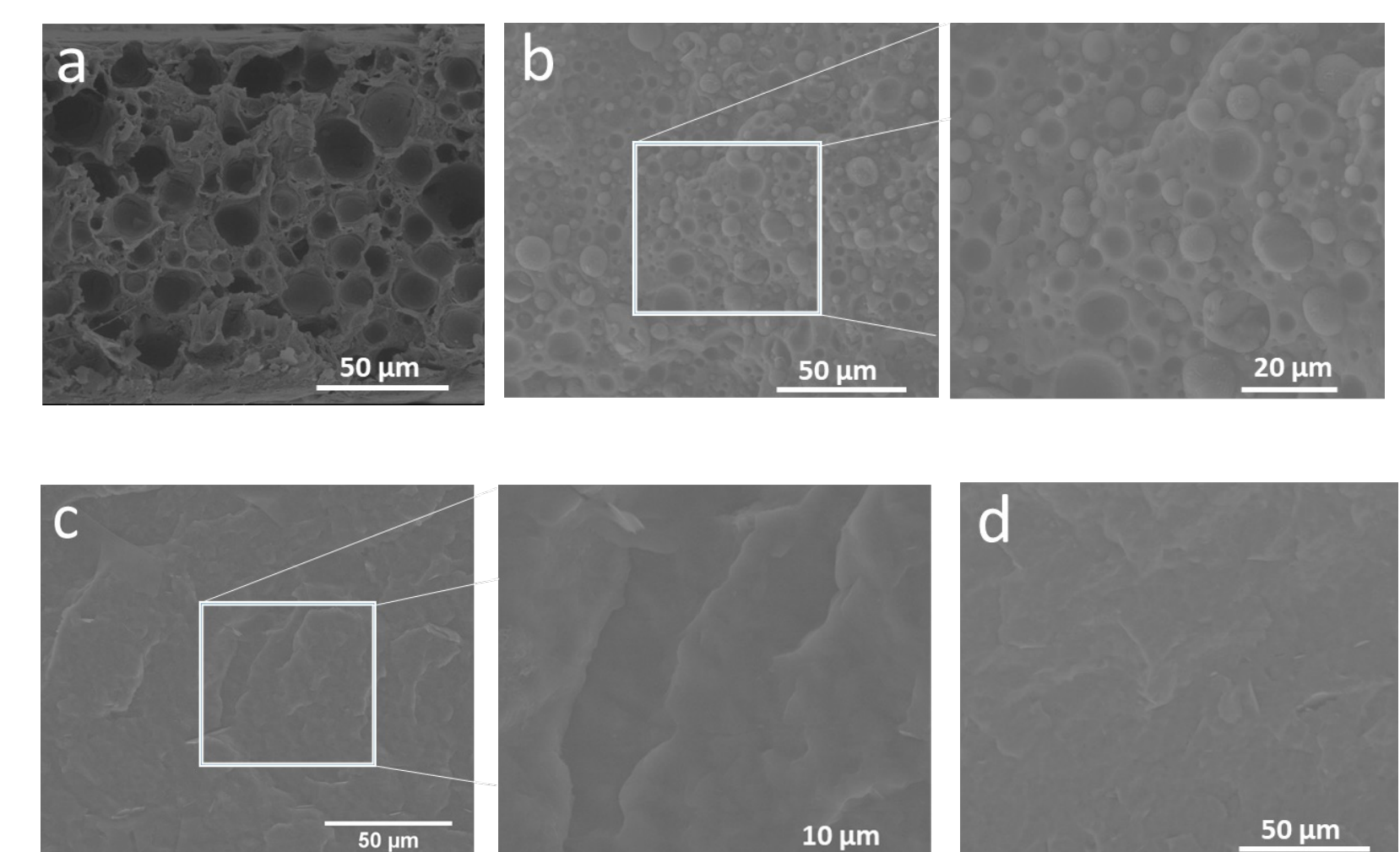
Melt viscosity of PHBV/PBSA/DCP blends



• DCP increased melt viscosity -> better processability in film blowing.

- Two distinct glass transition temperatures: PBSA and PHBV.
- Remain almost unchanged among blend ratio.
- PHBV and PBSA are immiscible.

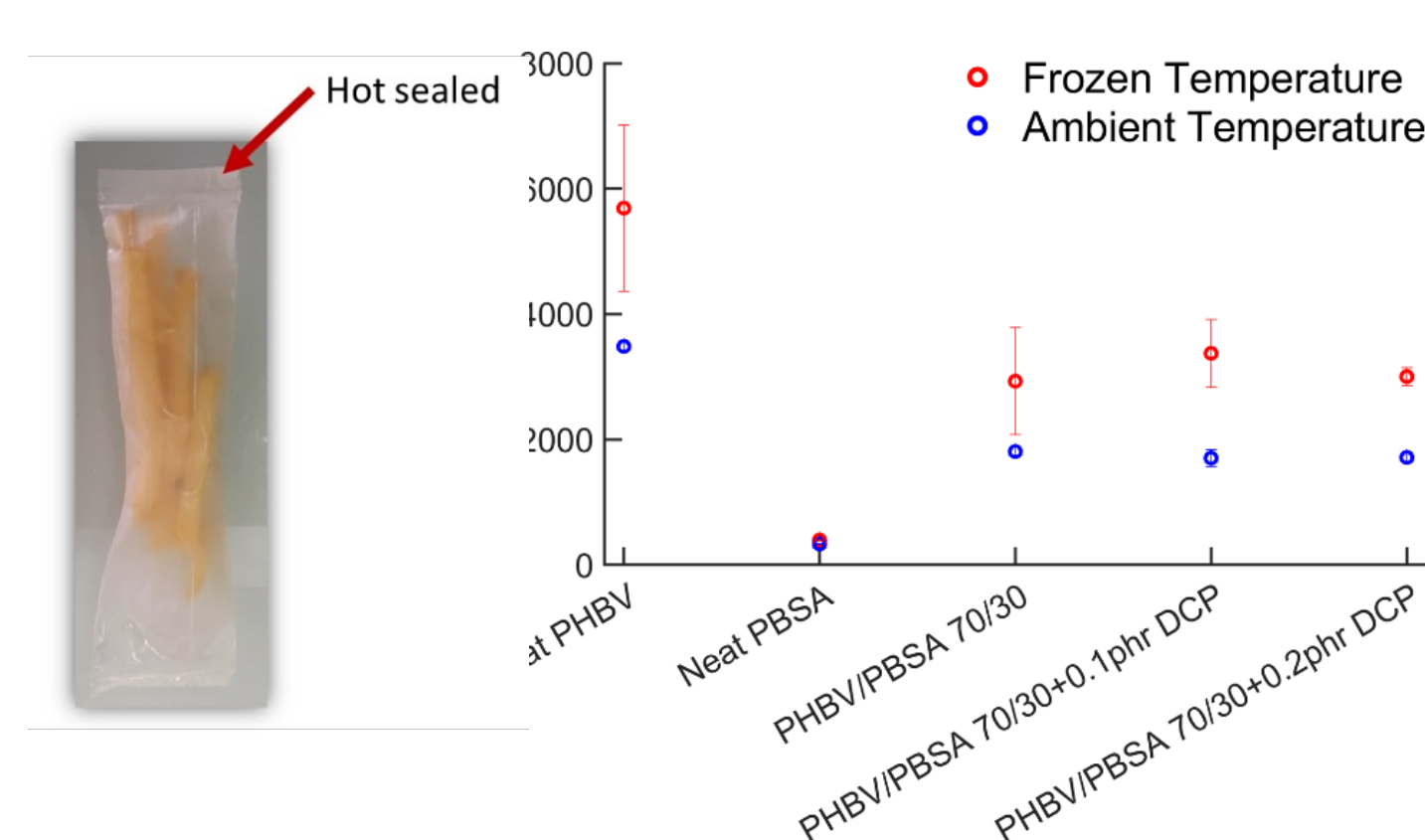
Morphology of PHBV/PBSA/DCP blends



SEM images of cryo-fractured surfaces of compatibilized PHBV/PBSA blends (batch mixer) with different DCP content, (a) 0 phr, (b) 0.02 phr, (c) 0.1 phr and (d) 0.2 phr.

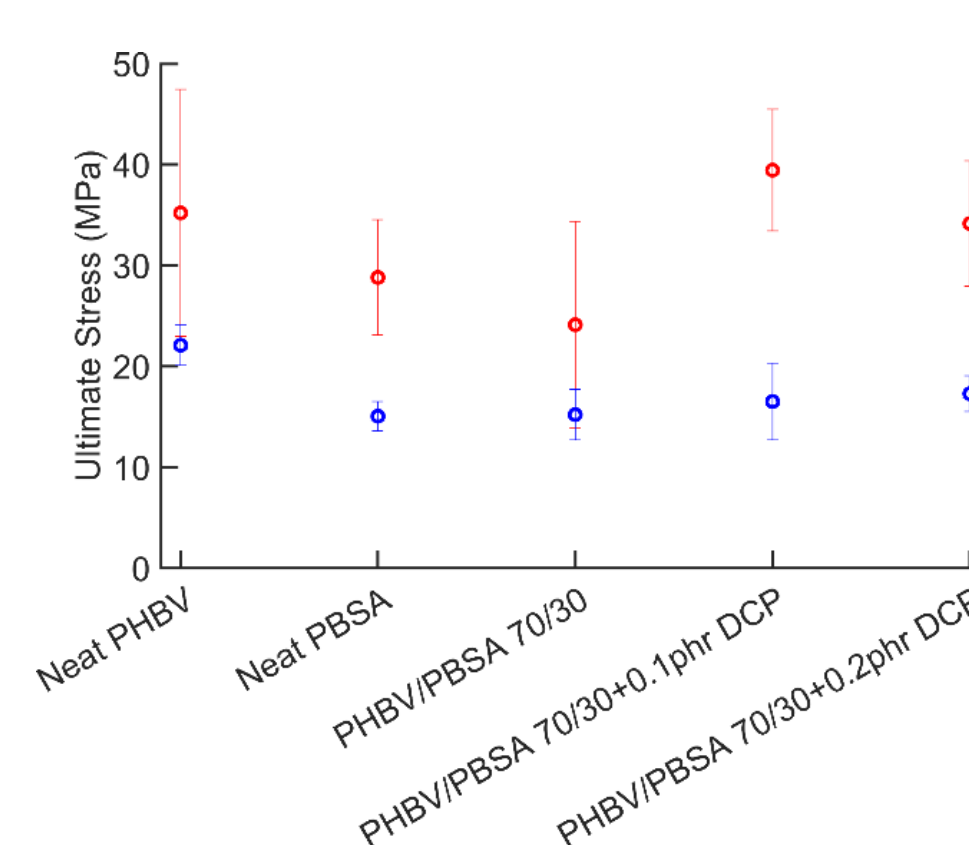
→ DCP decreased size of PBSA nodules in PHBV.

Mechanical properties of the films in service situation



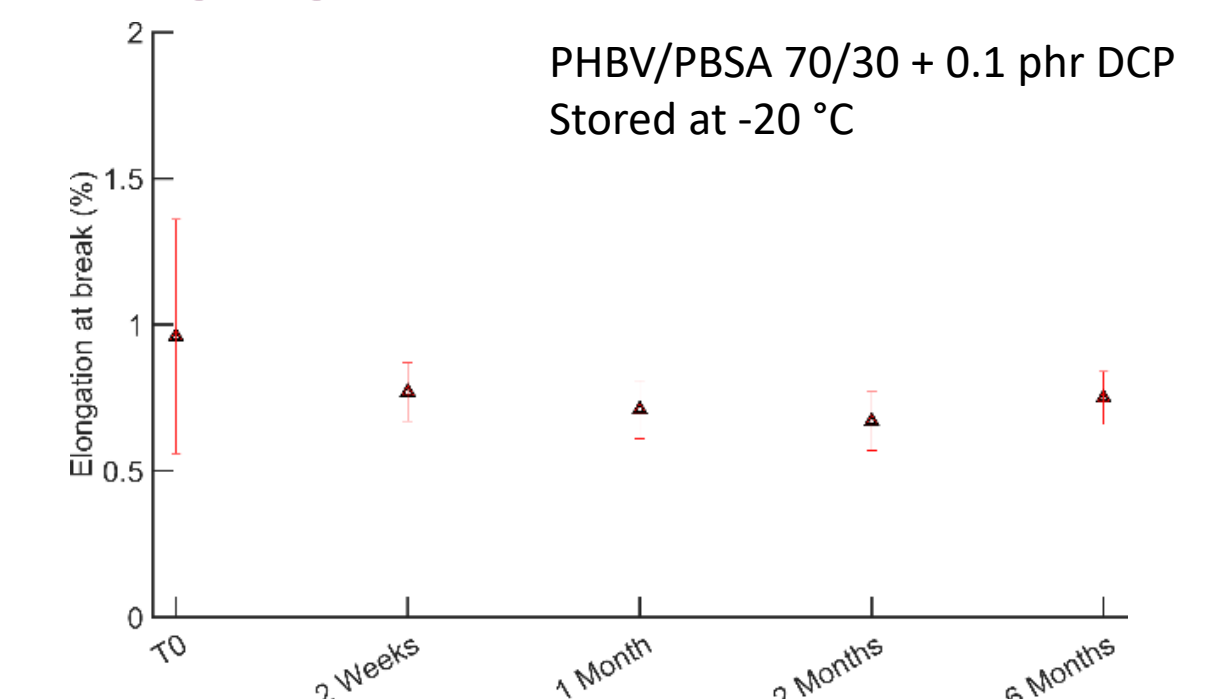
→ Blends can be sealed.

- Elastic modulus of blended films lower than neat PHBV.
- DCP increased elastic modulus.
- Ultimate stress of DCP blends similar to neat PHBV.



- Elongation at break slightly improved.
- Only small differences between ambient (23 °C) and freezer temperatures (-20 °C).

Aging in service conditions



- No evolution of properties during 6 months in the freezer in presence of food.

Conclusions

This study investigated the *in-situ* compatibilization of PHBV/PBSA blends at a weight ratio of 70:30 with introduction of dicumyl peroxide (DCP) in reactive extrusion. Batch mixing was used for the optimization of the formulation. Film blowing trials were successful and demonstrated the ability of the blend to be blown at a PHBV content of 70 wt%. Addition of DCP induced reaction between PHBV and PBSA leading to the formation of a cross-linked PHBV-*g*-PBSA structure. This resulted in higher melt viscosity of the blends. The results of tensile testing showed that the elastic modulus was increased with addition of DCP, but no evolution of the ultimate stress was found. DCP led to limited improved ductility of the blends. The analysis of the mechanical properties at ambient and freezer temperature showed close behavior. During storage of food in freezer conditions, no evolution of the mechanical properties was observed.

Compatibilized PHBV/PBSA blends could therefore be a performing biobased and biodegradable food packaging for frozen foods integrated in a concept of valorization of local resources for the production of PHBV.

REFERENCES

- Le Delliou, B., Vitrac, O., Castro, M., Bruzard, S., Domenek, S. *J. Appl. Polym. Sci.* **2022**, *139*, 52124.
- Le Delliou, B., Vitrac, O., Domenek, S., "Agricultural, Forestry and Bioindustry Biotechnology and Biodiscovery" Chong, P. A., Newman, D. J., Steinmacher, D. A., Eds. Springer International Publishing: Cham, **2020**, 227.