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Melt processing of dialcohol cellulose fibres into bulk re-processable materials

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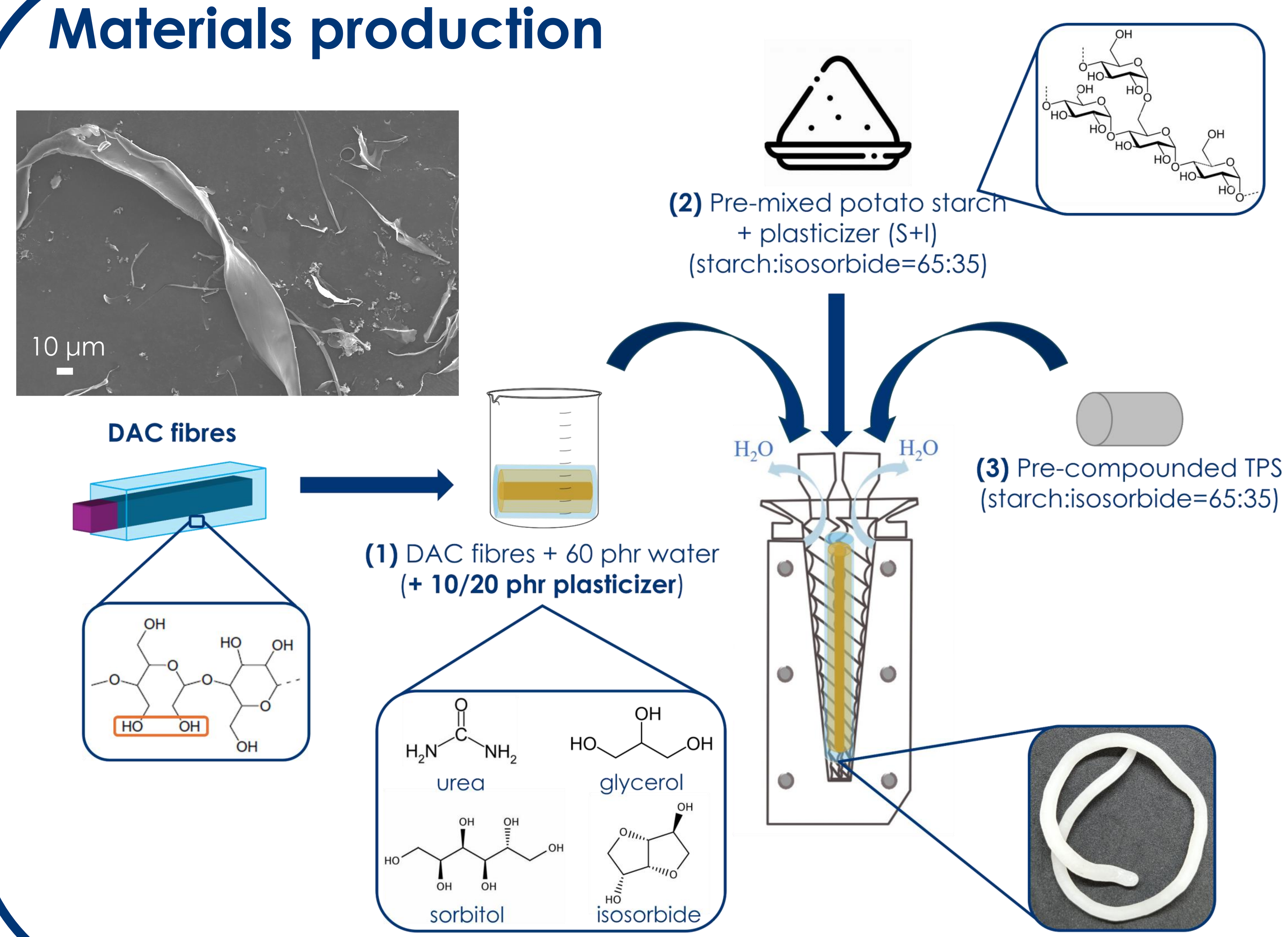
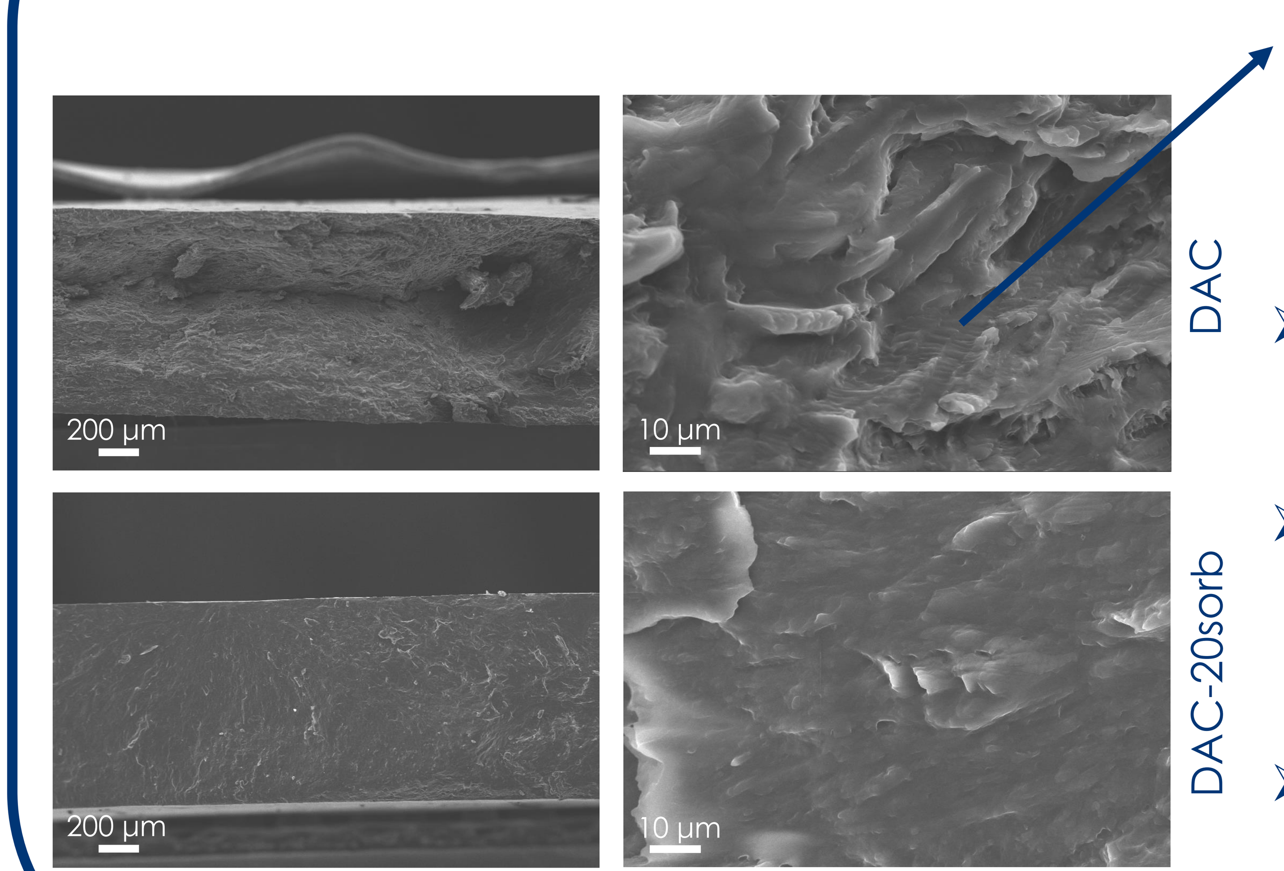
Introduction

Cellulosic materials are good alternatives to conventional plastics in packaging applications. However, cellulose degrades before reaching the molten state, limiting the possibility of exploiting the methods used in the production of conventional plastics, such as melt processing. Cellulose chemical modification into dialcohol cellulose (DAC) fibres reduces cellulose crystallinity, creating a processability window between the decreased glass transition and the degradation temperature. Previous studies demonstrated that DAC can be melt-processed using water in relatively large amounts as a processing aid[1], yielding a quite rigid material with moisture-sensitive mechanical properties. Aiming at tailoring the thermomechanical and viscoelastic properties of DAC, in this study, a set of less volatile plasticizers (i.e. urea, glycerol[2], sorbitol, isosorbide) were exploited. Results showed enhancements in the processability and reduction of DAC-based materials inherent sensitivity to external conditions. To further expand the range of thermo-physical properties, DAC was blended with starch, an abundant bio-based and biodegradable polymer, melt-processable in the presence of suitable plasticisers.

Materials production



Morphology

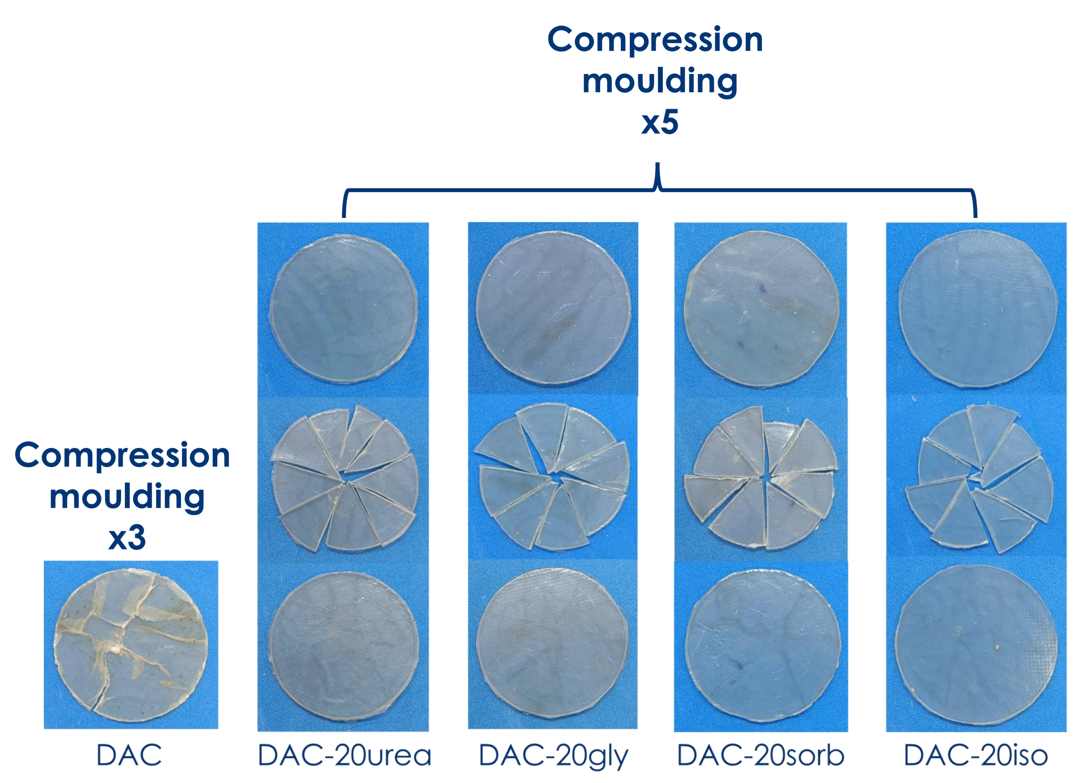


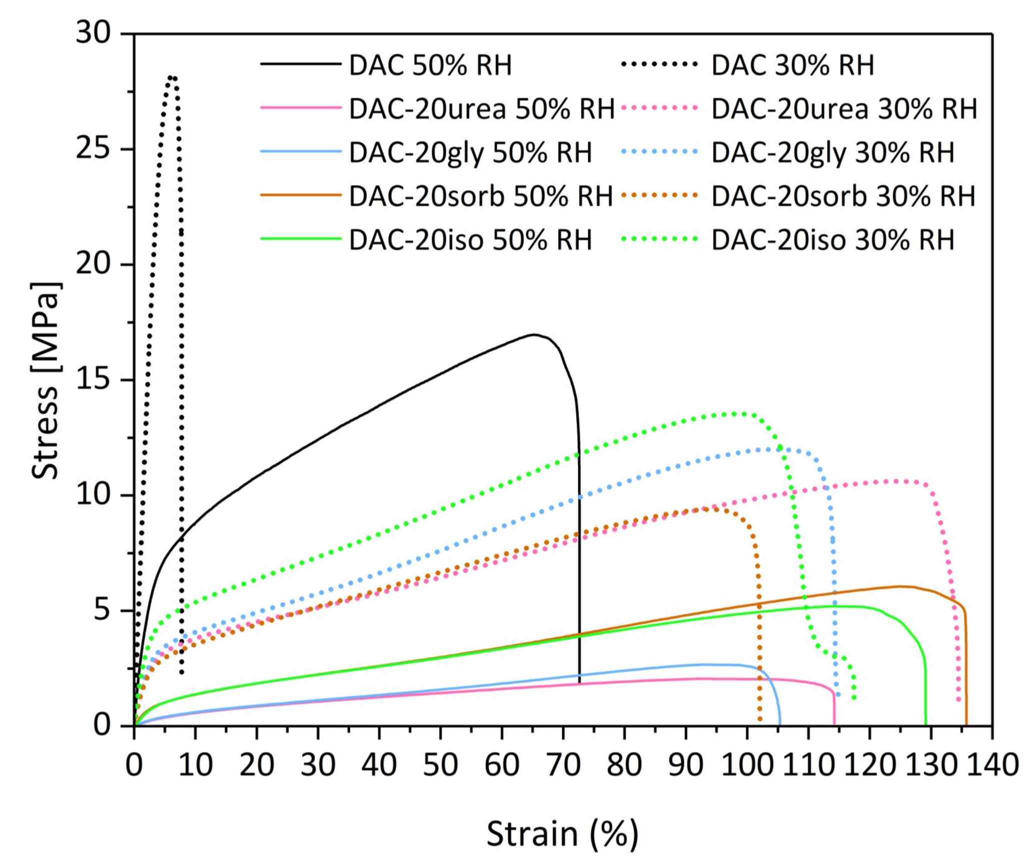
Textured surface due to fracture mechanism → **it breaks where there is a fibre**

- No individual fibres visible → **densified system**
- **Improved densification** by adding a **plasticizer**
- **No plasticizer phase separation**

Plasticizer addition

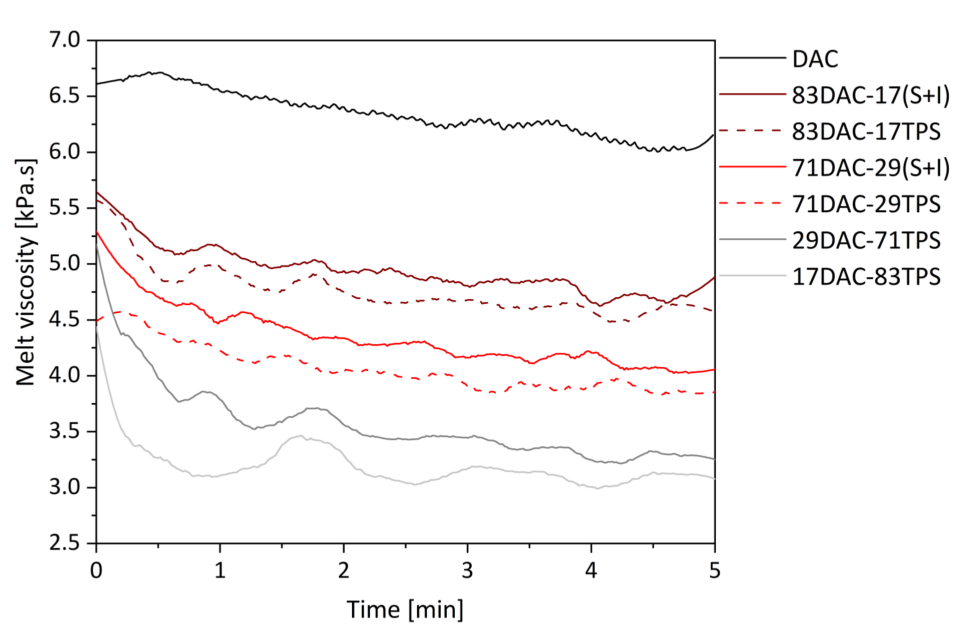
Sample	In-line melt viscosity [kPa*s]	T_g [°C]
DAC	5.5 ± 0.5	30
DAC-10urea	4.6 ± 0.7	20
DAC-20urea	4.0 ± 0.4	14
DAC-10gly	4.5 ± 0.4	17
DAC-20gly	4.0 ± 0.3	1
DAC-10sorb	4.7 ± 0.3	21
DAC-20sorb	4.2 ± 0.3	12
DAC-10iso	4.8 ± 0.4	20
DAC-20iso	4.0 ± 0.4	10

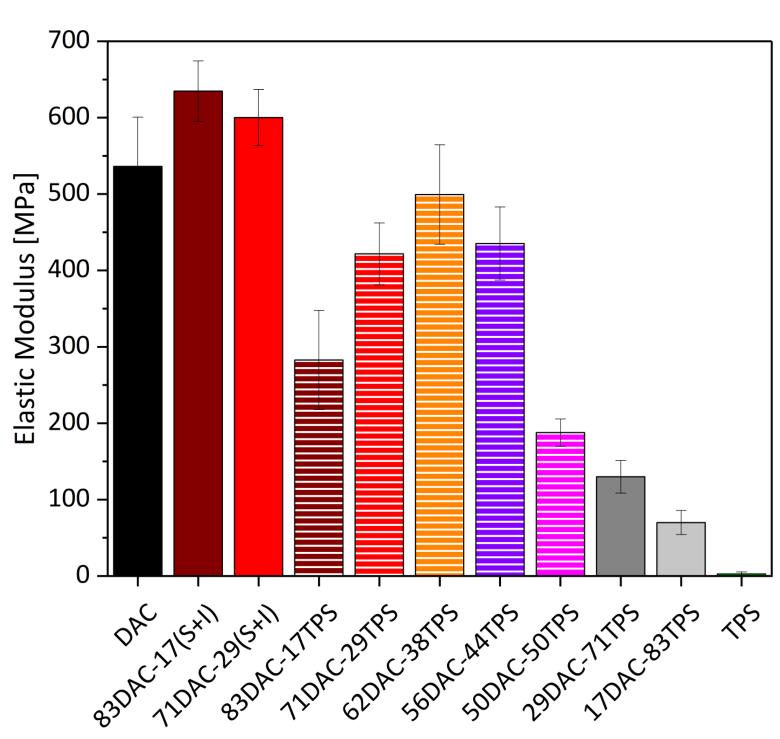


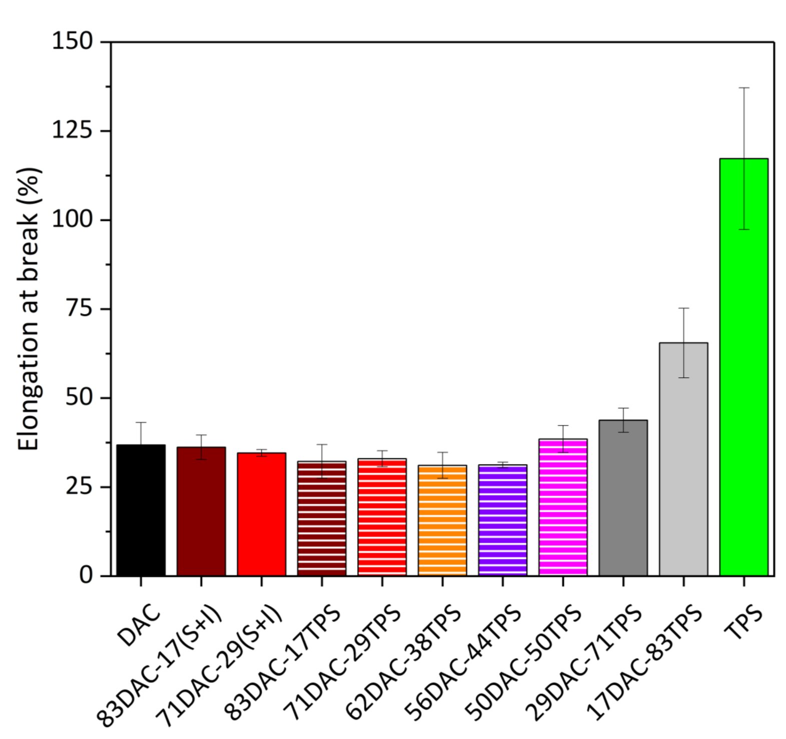


- **Improved melt-processability** (melt-compounding and moulding)
- **Wider processability window** (decrease in T_g)
- **Improved deformability** (up to a water + plasticizer saturation content)
- **Reduced** dialcohol cellulose based materials **dependence on environmental conditions**

DAC/starch blends







Sample	DAC [wt.%]	(S+I)/TPS [wt.%]
DAC	100	0
83DAC-17(S+I)	83	17
71DAC-29(S+I)	71	29
83DAC-17TPS	83	17
71DAC-29TPS	71	29
62DAC-38TPS	62	38
56DAC-44TPS	56	44
50DAC-50TPS	50	50
29DAC-71TPS	29	71
17DAC-83TPS	17	83
TPS	0	100

Co-plasticization

- **Higher** in-line melt viscosities
- Not plasticized starch acts as a **reinforcement**

Pre-compounded TPS

- **Improved melt-processability** (decrease in in-line melt viscosities)
- Possible **phase inversion** at **DAC:TPS=62:38**

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Future perspective

- Investigation on macromolecular mobility of the system
- Evaluation of the interactions between the blend components (DAC and starch)
- Modification of DAC into a **covalent adaptable network**

[1]Pellegrino, E., Al-Rudainy, B., Larsson, P. A., Fina, A., & Lo Re, G. (2025). Impact of water plasticization on dialcohol cellulose fibres melt processing-structure-properties relationship. *Carbohydrate Polymer Technologies and Applications*, 9, 100642.
[2]Pellegrino, E., Jonasson, K., Fina, A., & Lo Re, G. (2025). Plasticization of dialcohol cellulose and effect on the thermomechanical properties. Polymer Degradation and Stability, 111259.