Development of rosin-containing, biobased thermoplastic blends for use as hotmelt adhesives.

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Objectives discussed in the present work are: (1) Modifications of thermo-mechanical properties of rosin for use in HMA; (2) Review of biopolymer blending candidates for rosin-based products; (3) Development of a novel material for biobased HMA assessment. An innovative tackifier based on rosin and compatible with PLA has been developped, and formulations to produce biobased HMA were characterized to assess their technological viability.

Hotmelt adhesive (HMA) is intensively used for assembly purposes. After reviewing the suitable polymers and products, **Polylactic acid (PLA) and rosin**, both biobased and industrially available, were chosen for alternative HMA formulation. However, they are intrinsically incompatible (cf. Fig. 1A), and no trace of compatibilization could be found. The main laboratory work of this study was to synthesize a novel rosin-containing material compatible with PLA.

To this end, rosin has been modified through two strategies. **Alcoholysis of PLA chains** by a potentially biobased diol for PLA oligomer synthesis (OLAS), followed by the **introduction of rosin molecules by esterification** has been attempted (R-OLAS). Single-step **acidolysis of PLA by rosin** was also performed (R-PLA). Synthesized products were homogeneous (cf. Fig. 1B for R-PLA) and analysed mainly with dynamic scanning calorimetry, gel permeation chromatography, dynamic rheology and optical microscopy. R-OLAS was showing partial compatibility, while R-PLA was showing compatibility when blended with PLA.

Commercial **all-purpose and pressure sensitive HMA properties** were chosen to compare the formulations made of a **base polymer (PLA), a tackifier (R-PLA) and a plasticizer (OLAS)**. The individual ratios were varied and resulting products tested in terms of mechanical, rheological and phase separation properties. Formulated HMAs were showing decent mechanical and rheological properties compared to commercial HMA. Lap shear strength of 7.2 MPa was measured for the best formulation (1:2:0.5 R-PLA:PLA:OLAS), compared to 6.4 MPa for the commercial all-purpose HMA (cf. Fig. 2). An optimal viscous-to-elastic behavior trade-off has not yet been found for pressure sensitive hot melt formulations, hence requires further attention.

Processes to compatibilize rosin and PLA via simple chemistry and readily available materials were therefore developed, paving the way for the development of **efficient, totally biobased HMA formulations**.

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Management of Processes and Innovation

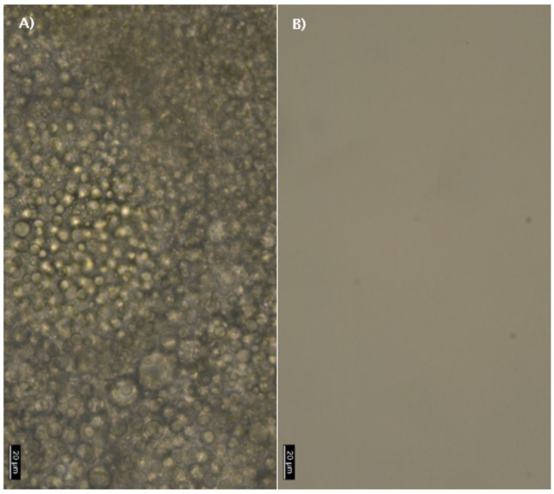

Fig. 1: Micrographs of (left) rosin blended with PLA showing coarse microstructure; (right) homogeneous R-PLA tackifier.

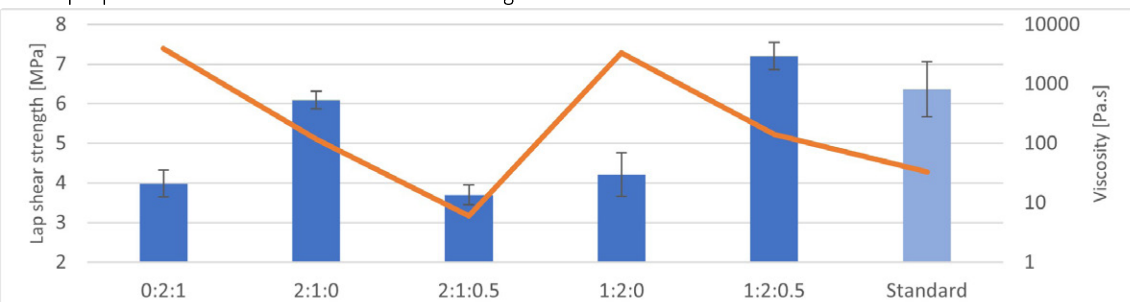

Fig. 2: Lap shear strength (blue) and viscosity at 120 °C (orange) of developed formulations. X:X:X is R-PLA:PLA:OLAS ratio.