## Development of rosin-containing, Biobased thermoplastic blends for use as hotmelt adhesives (HMA)



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Modifications of thermomechanical properties of rosin for use in HMA were studied in the present work, with a focus on biopolymer blendings. 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.

#### Introduction

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 in the literature and marketed products. The main laboratory work of this study was to synthesize a novel rosin-containing material compatible with PLA.

#### Strategy

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 analyzed 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.

#### A reasonable yet efficient approach

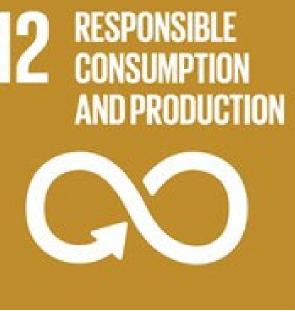
Innovative strategies for replacing or improving daily-use products is very challenging. Fossil resources provide low-cost materials with good purity, which eases any further processing. For bio-based materials on the contrary, the frequent diversity of constituents and supply's variations doesn't enable to easily build robust and affordable processes which could compete with conventional ones.

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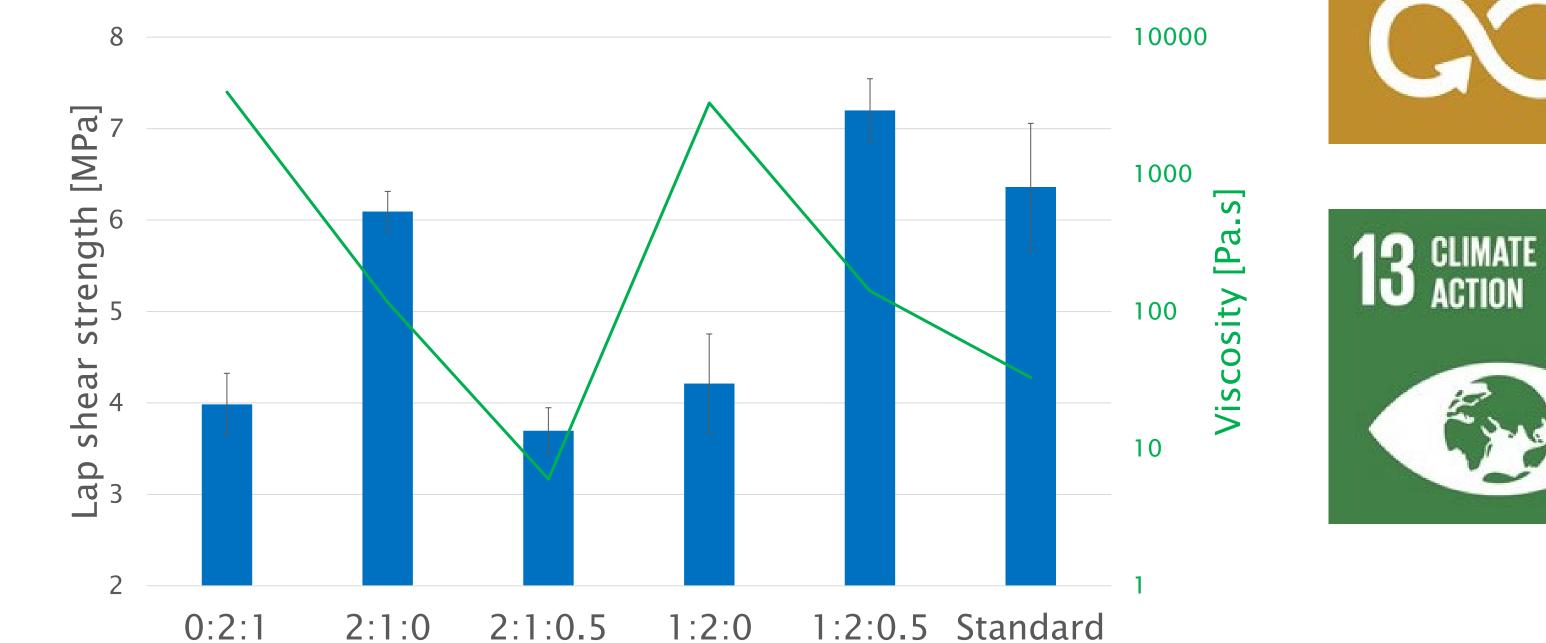






### Innovative Hot Melt Adhesive formulations

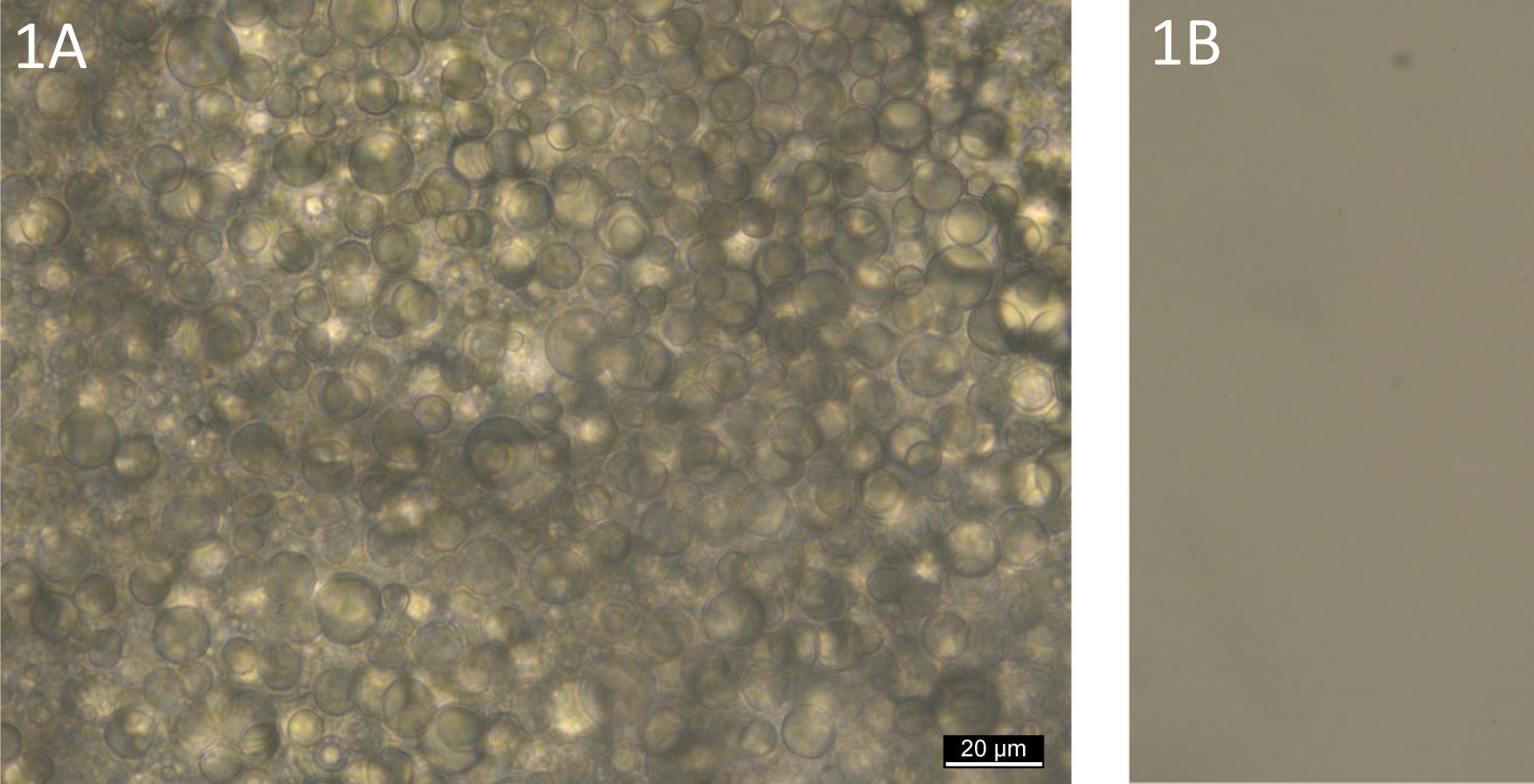
The here-developed chemical processes involve only industrially available materials and short reaction times (1 hour), while showing the use of iron oxide (rust) as an efficient catalyst for chemical reaction. Hence, processes to compatibilize rosin and PLA via simple chemistry and readily available materials were developed, paving the way for the development of efficient, totally biobased HMA formulations.



#### Materials testing

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-toelastic behavior trade-off has not yet been found for pressure sensitive hot melt formulations, hence requires further attention.

Figure 2: Lap shear strength (blue columns) and viscosity (green line) values for the chosen formulations R-PLA:PLA:OLAS.







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