Morphological and Rheological Evaluation of of Polyhydroxyalkanoates (PHAs) through Constitutive Equation Modelling

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The processing of biobased, biodegradable polymers inherits rheological challenges due to the thermal instability of their molecular structure. Polyhydroxyalkanoates (PHA) is one of these biobased, biodegradable polymers that is emerging in different applications. However, there is not a comprehensive study investigating the influence of the structural changes, which are inevitable during most processing, on the rheological properties and final product performance. In this study, the rheological behaviour of PHA, in pure forms and hybrid forms with polylactic Acid (PLA), has been investigated. Dynamic, shear and extensional rheology were employed to analyze the rheological properties in a wide range of stress fields with different levels of deformation. The generalized Maxwell model, a combination of spring and dashpot elements, was used to represent the relaxation modulus of the studied resins. Time-temperature superposition and constitutive modelling were applied to model the obtained rheological data. Also, different damping functions were used to examine the nonlinearity in viscoelastic response as a function of deformation levels. Our results showed that the rheological properties of PHA are very susceptible to temperature and residence time at high temperatures. We found that even short residence times of two minutes at the temperature of 200 °C have substantially influenced the morphology and rheological responses of PHA and PLAs containing 10 wt.% of PHAs hybrid materials. The rheological data at different temperatures indicated that PHA decreases PLA/PHA's viscosity to values lower than the volumetric average values, implying PHA's contribution in changing the chain interactions within PLA morphology. However, the presence of PHA did not influence the shear-thinning behaviour of PLA. The K-BKZ constitutive equation was shown to represent the rheological properties of the resins accurately. Damping functions such as Marruci and Wagner successfully predicted the nonlinearity of the rheological properties beyond linear viscoelastic region (LVE). Also, the extensional viscosity data revealed the reduced viscosity values for the hybrid materials to levels below the average values lower than the volumetric average values, pointing to the changes in the molecular interactions of PLA in the presence of PHA. This study reflects on the effect of processing conditions on the rheological behaviour of PHAs as biobased, biodegradable polymers and their hybrid materials in emerging markets.