ROLE OF PLASTICIZING MOLECULES ON THE AMORPHOUS PHASE DYNAMICS OF POLYLACTIDE

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ABSTRACT

Many works have shown the possibilities offered by the use of Poly(lactic acid) (PLA) in the packaging field. In this context, the study of mechanical and barrier properties is crucial and it is obvious that the microstructure and the amorphous phase properties have to be considered. Unfortunately, the inherent brittleness and low strain properties impose manufacturing constraints for a large amount of applications. Plasticization could be a way to improve the mechanical properties. In this work, we studied the influence of the addition of a plasticizer, Acetyl Tributyl Citrate (ATBC), on the molecular mobility and the amorphous phase dynamics of a wholly amorphous PLA. The understanding of the influence of plasticization on polymer dynamics is of fundamental importance for a deeper comprehension of the glass transition phenomenon and of plasticized PLA properties.

The amorphous phase mobility is investigated by Broadband Dielectric Spectroscopy BDS and Temperature Modulated Differential Scanning Calorimetry TMDSC to determine the impact of the ATBC incorporation (0 to 18 % w/w) in PLA. Since the theory proposed by Adam and Gibbs, it is well accepted by the scientific community that the main relaxation process in the supercooled liquid state, called α relaxation process, is cooperative approaching the dynamic glass transition region: a structural unit can move only if a certain number of neighboring structural units are also moving. This concept implied the notion of Cooperative Rearranging Region (CRR), which can be estimated in terms of cooperativity volume according to different models.

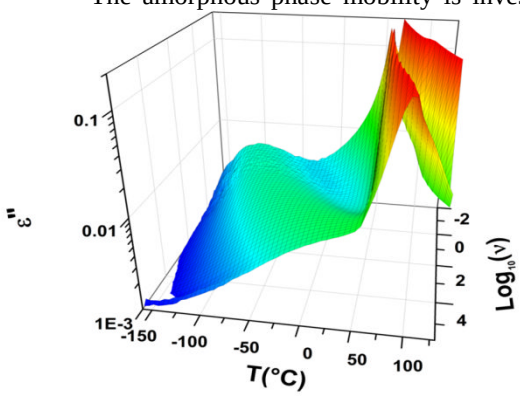

Fig. 1: Imaginary part of the complex dielectric permittivity vs. temperature and frequency.

It is shown that cooperativity volume decreases drastically when the plasticizer content increases. Moreover, presence of ATBC leads to a modification of the amorphous phase dynamics. Indeed, additivation leads to huge decrease of the fragility index from a PET-like to a PDMS-like behavior. At a given temperature, the kinetics of the molecular motions are accelerated by plasticizing molecules. Investigation of physical aging of plasticized materials shows a noticeable increase of relaxation speed even in the glassy state.

So, It has been shown that the modifications of interchain interactions due to plasticization could be quantified by fragility index, CRR volume and recovery function.

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