COUPLING BETWEEN AMORPHOUS AND CRYSTALLINE PHASES REVEALS FROM MOLECULAR MOBILITY STUDIES

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ABSTRACT

The amorphous phase geometrically confined at the nanometric length scale in semi-crystalline polymers, exhibits different conformational dynamics with respect to the pure amorphous situation. The purpose of this work is to use the molecular mobility dependence of the amorphous phase on the microstructure to obtain information on the coupling between phases.

Poly(lactic acid) (PLA) is used in this work because different microstructures can be obtained: strain induced crystallization from mono or bi axial drawing, spherulitic crystallization from the melt or from the glassy state. Moreover PLA exhibits at least four different crystalline forms (α, β, γ, δ) depending on the crystallization conditions. X ray diffraction and thermal analyses were performed on PLA with crystallinity degrees varying from 0% to 73%.

We show that different types of amorphous phases are obtained. By example for incomplete spherulitic crystallization three fractions of amorphous phases exist: the first fraction is the amorphous phase which composes the matrix. The second and third fractions are trapped in the spherulites. Due to coupling between the crystalline lamellas and the amorphous parts, the second fraction (the rigid amorphous fraction RAF) cannot develop cooperative motion at the glass transition. The third fraction, called the mobile amorphous phase has restricted dynamics due to confinement and coupling between phases. We examine also the crystallographic influence: δ crystallization yields to a high RAF quantity while α crystallization generates only a small RAF content.

Comparing the Cooperative Rearranging Regions (CRR) size and the amorphous domain size, we evidence a confining effect, causing an amorphous phase thickness decrease during crystallization, and inducing a drastic CRR size reduction.