Contribution ID: **10** Type: **invited talk**

Shapes of the free volume holes in amorphous polymers as estimated by positron annihilation lifetime spectroscopy

Friday, 28 June 2019 08:30 (25 minutes)

Intermolecular spaces in polymer chains form the so called free volume, a useful concept to understand mechanical and transport properties of polymers. Quantification of the free volume can be obtained theoretically, using appropriate lattice models, as well as experimentally, through suitable probes. Among these, positronium (Ps) has become widespread due to the non-destructive character of the technique, the capability of Ps to preferentially localize inside the free volume holes and the correlation between the Ps lifetime and the size of the holes. In most of the investigations the cavity is approximated to a sphere. However, this may bias the evaluation of the free volume fraction. We show that by coupling results from Ps lifetime and specific volume measurements for amorphous polymers at equilibrium and the predictions of the Simha-Somcynsky equation of state it is possible to shed light on the dimension of the holes, on their morphology as well as on their expansion with temperature. In fact, in spite of their irregular shape, non-spherical cavities are generally found to give a better agreement between the theoretical free volume fraction and the experimental results.

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Session Classification: Friday