

Rigid Amorphous Fraction in Polymer Nano-Composites

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Very often, semicrystalline polymers show significantly smaller relaxation strengths at glass transitions than expected from the non-crystalline fraction. This observation could be explained by the introduction of a rigid amorphous fraction (RAF) which contributes neither to the heat of fusion or X-ray crystallinity, nor to the relaxation strength at glass transition. The RAF is assumed to be in a glassy state above the common glass transition temperature. From the heat capacity, the temperature and time dependence of the RAF can be obtained. For semicrystalline polymers it is possible to study the creation and disappearance of the RAF in situ during isothermal crystallization and stepwise melting.

A RAF can be detected for polymer nano-composites as well. The RAF was calculated from the calorimetric relaxation strength, Δc_p , as $RAF = 1 - \text{filler fraction} - \Delta(c_p \text{ filled})/\Delta(c_p \text{ pure})$. For polystyrene (PS) calcium phosphate and polymethyl methacrylate (PMMA) silicon oxide nano-composites based on 10 nm filler RAF is on the same order as the filler content. Although the filler is thermally inert, no devitrification of the RAF is observed before degradation of the polymer, even for high heating rates. High heating rates were applied to avoid long residence times at high temperatures.

For the PMMA nano-composite, a RAF is observed for emulsion polymerized composites but not for melt blended nano-composites. Deagglomeration and the interaction of the polymer with the particle surface play an essential role for the occurrence of RAF.