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**Polymer Structure and Dynamics in Polymer / Layered-Silicate Nanocomposites<sup>1</sup>**

SPIROS H. ANASTASIADIS<sup>2</sup>, Foundation for Research and Technology-Hellas and Univ. of Crete, Greece

Polymer/layered silicate nanocomposites are of particular interest among different nanohybrids because of their anticipated superior properties. Mixing polymers with layered inorganic materials can lead to three different types of structure, depending on the interactions between the constituents: phase separated, intercalated and exfoliated. Intercalated hybrids, where the polymer is confined within the inorganic galleries, can serve as model systems for the study of the static and dynamic properties of macromolecules in nano-confinement. We describe our recent efforts to elucidate the effects of severe confinement utilizing hydrophilic nanohybrids of PEO or hyperbranched polymers mixed with Na<sup>+</sup>-MMT. Intercalated hybrids with mono-, bi- and tri-layers of chains are obtained for all compositions covering the complete range from pure polymer to pure clay. Severe confinement influences significantly the structure of the polymer: the PEO chains intercalated within the inorganic galleries as well as those in close proximity to the outside walls are purely amorphous; it is only when there is significant excess polymer outside the completely filled galleries that the bulk polymer crystallinity is abruptly recovered. In contrast, when the inorganic is incorporated as silica nanoparticles, the crystallinity varies smoothly with composition whereas a population with a lower melting temperature near the inorganic surfaces is observed under strong confinement. The dynamics of the polymers confined within the galleries is probed by quasi-elastic neutron scattering and dielectric spectroscopy. The very local dynamics of the confined chains show similarities with those in bulk, whereas the segmental dynamics depend very strongly on the polymer/inorganic interactions varying from much faster to much slower or even frozen dynamics as the strength of the interactions increases.

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