Nanostructured particle-polymer composites: filler dispersion and chain conformation from well-defined model to "real" systems, correlation with mechanical properties

Jacques Jestin
Accrediation to Supervise Research – Pierre et Marie Curie University Paris

Laboratoire Léon Brillouin UMR12 CEA-CNRS, CEA Saclay 91191 Gif/Yvette Cedex France
jacques.jestin@cea.fr

Nanocomposite materials have raised a great interest over the last twenty years. This deep attraction comes from the outstanding potential of applications of these systems in a broad range of industrial fields. Putting together "soft" polymer chains and "hard" particles of similar nanometer size opens also the way to new questions of fundamental physics associated to polymer chains conformation and dynamic, dispersion mechanisms and confinement effects. However, the large variety of possible assemblies between different kind of polymers and particles makes difficult to dispose of a general trend to describe the correlations between the local structure of the materials and their macroscopic properties. Despite of unusual performances of mechanical reinforcement, adhesion or deformability, the general viscoelastic behaviors in nanocomposites remain still not fully described due to specific interactions, difficult to identify and to complex organizations covering a large range of typical sizes from the nanometer to the micrometer.

The present work exposes the correlations that can be established between particle dispersion and polymer chain conformation with the viscoelastic behavior of the materials at rest and under deformation across different examples of nanocomposites. We first discuss different strategies (chemical grafting, magnetic field, specific additives or controlled sample processing) to control the filler dispersion inside the polymer matrix with different particles (colloidal or industrial silica, magnetic nanoparticles) dispersed in polymers (Polystyrene, Poly (methyl methacrylate)) or elastomers (Styrene-Butadiene-Rubber). We go progressively from well-defined model systems toward more complex "real" systems. We obtain a broad range of various morphologies from individual particles, to compact aggregates, oriented chains of particles to hierarchical networks described quantitatively by Small Angle X-ray Scattering (SAXS) and microscopy (TEM) at the different length scales (particle and aggregate shapes, sizes and inter-object correlations). With a combination of several methods, the viscoelastic behavior has been determined and analyzed quantitatively with hydrodynamic and percolation models: the reinforcement is mostly driven by the aggregate morphologies and by the network strength (depending on inter-particle and inter-aggregate interactions). The flow behavior is mostly driven by the polymer chain contribution whose physical origin is related to specific polymer-particle interactions. This origin is discussed according to the different existing models. With a combination of SANS with different contrast variation methods, we demonstrate that the polymer chain conformation is not modified by the filler suggesting that the number of chains involved in the viscoelastic processes is low. By following the scattering of both filler and chain under deformation, we confirm that the non-linear mechanical behaviors are mostly dominated by the filler reorganizations. We finally propose some strategies to progress in the understanding of the specific polymer-chain interactions with nanocomposites using grafted nanoparticles. We suggest focusing on the chain dynamic with inelastic neutron scattering and some synthesis developments to manage the interface chain mobility or the polymer-chain interactions without changing the filler dispersion. In a more extended view, we discuss the necessity to go further toward industrial systems with the development of new "real-time" experiments associated with the design of the future SANS spectrometers.

Keywords: Nanocomposites, filler dispersion, polymer chain conformation, Small Angle Neutron and X-ray Scattering, viscoelastic behavior, reinforcement, deformation.