QUANTIFYING MULTI-SCALE NETWORK PROCESS-STRUCTURE-PROPERTY RELATIONSHIPS IN POLYMER MATERIALS THROUGH SCATTERING DATA DECOMPOSITION

By

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ABSTRACT

In this work, we present an optimization-based methodology to identify network process-structure-property relationships in multiphase polymeric materials. Control over enhanced properties of polymer materials requires knowledge of the local (nanoscale) and global (microscale) structure of the crystalline and amorphous domains present in the material. A quantitative description of the structure of these domains can be provided by x-ray and neutron scattering techniques, which probe ordered density fluctuations in a material. An observed scattered intensity signature is then modeled as the weighted sum of linearly independent components, where the profile of a component depends on the shape, aggregation, and extent of fundamental structural units present in the material. We therefore seek to simultaneously extract the shape of these components and their contribution to the observed intensity, through decomposition of process-dependent scattering datasets, without assumptions on polymer structural geometry. This decomposition methodology is based on network component analysis (NCA), a nonlinear program (NLP) used to generate a unique, globally optimal matrix decomposition. This program is then reformulated as a mixed-integer nonlinear program (MINLP) to overcome the limitation that NCA requires a priori knowledge of the connectivity between the experimental and underlying component signatures. The methodology is then applied in the study of ethylene/alpha-olefin copolymer crystallization from the melt using wide- and small-angle x-ray scattering (WAXS/SAXS) data, where the variables of interest are time, side chain branch length and isothermal crystallization temperature. The validity of the decomposition is first confirmed through extraction of well-known amorphous and crystalline component signatures present in WAXS signatures. It is then applied in the decomposition of SAXS data to quantify the structural evolution of crystalline and amorphous domains at the nanometer scale. Finally, a model study of amphiphilic triblock copolymers in solution is carried out using simulated small-angle neutron scattering (SANS) data in order to extract components based solely on knowledge of where density contrast matching occurs.