

A nonlinear framework for the rheological characterization of polymer nanocomposites

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Processing of polymer nanocomposites involves rheologically complex materials in complex flow configurations. The material is most often subjected to nonlinear deformations, i.e. stretching, orientation, disentanglement of polymer chains occurs, with the interactions between flow field, matrix and filler(s) dictating the overall flow dynamics and subsequent material properties/performance. In this framework, the nonlinear material response of various nanocomposites was probed in oscillatory shear. The shear stress output signal analysis was performed in the framework of Fourier-Transform (FT) analysis and Tchebyshev polynomial decomposition. A range of nanofillers, e.g. graphite nanoplatelets, graphene, hybrids, nanocellulose, and thermoplastic polymers, e.g. with varying molecular topologies, were tested. The rheological percolation threshold was determined with superior sensitivity using nonlinear material parameters. In linear viscoelastic frequency sweeps, the additional elastic contribution of the filler network buildup can be difficult to detect in the limit of low angular frequencies. In contrast, at the percolation threshold, the shear stress nonlinearities disclose a comparatively strong angular frequency dependent material response, readily available via the FT relative higher harmonics and the elastic and viscous relative Tchebyshev coefficients. Furthermore, nonlinear materials parameters show unique features related to the type of material compositions, especially related to orientation dynamics [1].

[1] Kádár et al., (2017) *Linear and Nonlinear Rheology Combined with Dielectric Spectroscopy of Hybrid Polymer Nanocomposites for Semiconductive Applications*, *Nanomaterials* 7, 23