Thixotropy, Shear Rheology and Electron Transport in Model Carbon Black Suspensions Under Flow

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Electron transport in complex fluids, biology, and soft matter is a valuable characteristic in many natural and industrially relevant processes, including electrochemical energy storage. These processes often employ mixed conductor-insulator systems in which electron transport properties are fundamentally linked to the microstructure and dynamics of the conductive phase. While microstructure and dynamics are well-recognized as key determinants of the electrical properties, a unified description of their effect has yet to be determined, especially under flowing conditions. The conductivity and shear viscosity are measured for model conductive colloidal suspensions to build a unified description by exploiting both recent quantification of the effect of flow-induced dynamics on charge transport and well-established relationships between electrical properties, microstructure, and flow. In this work, direct structural measurements over a hierarchy of length scales spanning from nanometers to tens of micrometers are used to determine the microstructural origin of the suspension's thixotropy, viscosity and electrical conductivity under flow. These experiments were performed on a series of dense suspensions consisting of high-structured carbon blacks The shear-induced microstructure was measured at a range of applied shear rates using Rheo-VSANS (very small angle neutron scattering) and Rheo-USANS (ultra-small angle neutron scattering) techniques. New instrumentation combining rheology simultaneous with dielectric spectroscopy and neutron scattering interrogation of microstructure is presented. A shear-thinning viscosity is found to arise due to the self-similar break up of micrometer-sized agglomerates with increasing shear intensity. This self-similarity yields a master curve for the shear-dependent agglomerate size when plotted against the Mason number, which compares the shear force acting to break particle-particle bonds to the cohesive force holding bonds together. The relevance of the Mason number is demonstrated to unify both the rheology and conductivity of concentrated suspensions such as those relevant to the processing of carbon black suspensions for applications including battery slurries. A new rheological constitutive equation derived from population balance and particle-level physics is introduced to model the thixotropic rheology. Finally, the use of this applied rheology to engineer the process of coating battery slurries is discussed and illustrated with a classroom laboratory exercise.