Rheology and Microstructure of Battery Electrode Slurries

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Abstract

Understanding the rheological behavior of lithium-ion battery slurries is essential for optimizing electrode manufacturing, as rheology governs processability, microstructure uniformity, and defect formation. Across different electrode chemistries, battery slurries commonly exhibit complex flow behavior arising from multi-scale particle networks and their evolving interactions under shear.

In anode slurries based on graphite, carbon black, and CMC, we identified distinct rheological regimes determined by the applied shear rate: yielding at low shear ($<0.1\ s^{-1}$), thixo-viscoelastic behavior at intermediate shear (0.1–$10\ s^{-1}$), and shear thinning at high shear ($>10\ s^{-1}$). When formulated at high solid content, these suspensions additionally exhibit apparent shear thickening and anti-thixotropy, driven by the growth of frictional contact networks under sustained shear. Flow reversal measurements reveal that contact stress increases with both shear rate and shearing time, reflecting a frictional contact mechanism. The resulting contact network partially persists after flow cessation, as indicated by slow relaxation governed by hydrophobic interactions. These findings establish how frictional contacts dictate shear-thickening behavior in concentrated anode slurries. Beyond graphite-based anodes, this particle interaction framework can be extended to other battery material systems with different particle sizes and surface chemistries. In addition, coating edge quality in slot-die coating processes is discussed as another critical aspect of electrode manufacturing.