

TWO-STEP YIELDING BEHAVIOR OF CONCENTRATED LITHIUM-ION BATTERY ANODE SLURRY

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ABSTRACT

The nonlinear rheology of a concentrated lithium-ion battery anode slurry was investigated under large amplitude oscillatory shear and interpreted using sequence physical process (SPP) analysis. A complex interaction between three anode slurry components—graphite (Gr) as an active material, carbon black (CB) as a conductive additive, and carboxymethyl cellulose (CMC) as a binder—results in a two-step yielding behavior, which is represented by the secondary plateau in dynamic strain and stress sweep tests. Through the examination of intra-cycle rheological transition under oscillatory shear flow, we demonstrate that a two-step yielding behavior is represented by double deltoids in SPP analysis. Two-step yielding is observed in Gr-CMC slurries but not in CB-CMC slurries, indicating that Gr and CMC are the principal causes of two-step yielding in an anode slurry. A sedimentation test performed on a dilute Gr-CMC solution resulted in phase separation between graphite particles, with CMC adsorbed on their surface and graphite particles aggregated by hydrophobic attraction. This suggests that there are two different forms of interactions in a concentrated slurry: a hydrophobic interaction between graphite particles and a physicochemical interaction resulting from CMC adsorption on graphite particles. The initial yielding stage is associated with the hydrophobic interaction between graphite particles, resulting in a network structure that is expected to be brittle and break under a small strain. The second yielding stage is attributable to the interaction between concentrated CMC, as evidenced by the overlap between the secondary deltoid of the anode slurry and the single deltoid of the concentrated CMC solution in SPP analysis.

REFERENCES

1. Kim, Y., Kim, S., Kim, B. S., Park, J. H., Ahn, K. H., & Park, J. D. (2022). Yielding behavior of concentrated lithium-ion battery anode slurry. *Physics of Fluids*, 34(12), 123112, 2022. <https://doi.org/10.1063/5.0128872>