Rheology-Guided Optimization of Aqueous Graphene Slurries for Scalable Supercapacitor Electrodes

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Abstract

The conventional way of electrode fabrication for batteries and supercapacitors relies on organic solvents such as N-Methyl-2-pyrrolidone (NMP) and Dimethylformamide (DMF). Both of these solvents are costly, highly regulated, toxic, volatile, and environmentally hazardous [1]. On the contrary, aqueous-based slurry formulations offer a safer, more sustainable, and cost-effective route for graphene electrode fabrication. Despite these advantages, water-based processing introduces distinct challenges arising from its high surface tension and evaporation-driven drying behaviour. Aqueous processing can lead to binder migration, density gradients across electrode thickness, and residual moisture retention, all of which adversely affect electronic conductivity, electrode-current collector contact, and long-term stability. Furthermore, electrode microstructure, porosity, and batch-to-batch reproducibility are strongly governed by the interactions between graphene and the binder system. In particular, excessive binder content can disrupt conductive pathways and increase equivalent series resistance (ESR), underscoring the need for systematic slurry optimization [2]. In this study, graphene slurries were formulated using an aqueous CMC-SBR binder system, with total binder content varied between 5 and 7.5 wt% and solids loading between 24 and 30 wt%, while maintaining a fixed conductive additive fraction. Rheological characterization, including viscosity–shear rate behaviour, yield stress, and thixotropic recovery, combined with castability assessments, was employed as a gating criterion to identify processable slurry compositions. The results establish a reproducible rheological window for uniform tape casting and defect-free electrode fabrication. This work provides a scalable framework for translating aqueous graphene slurries from laboratory formulation to practical supercapacitor electrodes, supporting low-ESR, high-performance devices through environmentally sustainable processing routes.

References

[1] Sonia Reenu, L Phor, A Kumar, S Chahal, J. Energy Storage, 84 (2024), 110698-110734
[2] P Luís, S Martin-Fuentes, M Arnaiz, J Ajuria, J Appl Electrochem, 55 (2025), 2841-2852

Figures

Figure 1: Schematic illustrating rheology-guided optimization of aqueous graphene slurries.