Supporting information

Contact resistance stability and cation mixing in Vulcan-based LiNi_{1/3}Co_{1/3}Mn_{1/3}O₂ slurry

for semi-solid flow batteries

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The flow cell used to characterize Vulcan-based NCM slurry is presented in figure 1S. It consists of several components that can be assembled either in a 2 or 3 – electrode configuration with minor adjustments due to its modular set-up. Two thick Teflon plates sandwich the components: current collectors, separator(s) and Teflon channel(s) or gaskets, and strengthen the battery. Fluidic slurries for electrochemical characterization are pumped into the cell through an inlet and flow along the channel as defined by gasket(s). In this manuscript, only one Teflon gasket was used since the counter electrode was metallic lithium. For electrochemical and impedance data measured in a 3-electrode configuration, the flow cell was assembled with a Li metal strip sandwiched between separators.



Figure 1S Flow cell used for electrochemical testing either in a 2- or 3- electrode

configuration

The viscosity of Vulcan-based NCM slurry was measured using a rotational viscometer with spindle size SC-18 and results are presented in figure 2S. Its viscosity changes as a function of shear rate (pseudo plastic behavior) i.e. between 100 to 1000 Cp, in good agreement with viscosities reported for concentrated slurries [1–3].



Figure 2S Viscosity values as a function of shear rate for the Vulcan-based NCM slurry.

The electrochemical performance of a KB-based NCM catholyte was evaluated in flow mode in order to demonstrate its limited cycling stability compared to Vulcan-based NCM slurry. The KB-based slurry was formulated and prepared following recipe described in experimental section; a mixture of 4.6 % of NCM, 7.4 % of KB carbon and 88 % in weight of electrolyte stirred overnight at 500 rpms. GCD results for the KB-based NCM slurry in flow mode 2 mlmin⁻¹ and 2-electrode configuration are presented in figure 3S. Current used was 0.33 mAcm⁻². The slurry delivers capacity 20 mAh but it becomes non-fowable before reaching cut-off voltage 4.6 V leading to unstable voltage readings > 20 mAh.



Figure 3S GCD profile of KB-based NCM slurry measured in flow mode 2 mlmin⁻¹ and 2electrode configuration as a function of electrochemical capacity at 0.33 mAcm⁻².