Carbon-based catholytes for semi-solid Li/O_2 flow battery

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Li/O_2 batteries can ensure the requirement of specific energy > 500 Wh kg^{-1} [1]. Although Li/O_2 system has an intrinsically low efficiency, the use of a carbonaceous semisolid catholyte in a flow battery can be the keystone to improve its cycling performance. Indeed, it permits to: i) decouple energy and power; ii) increase cell areal capacity and energy by alleviating current collector passivation; iii) improve current rate response and decrease recharge overvoltage by the carbon percolating network.

We have already demonstrated a semi-solid lithium redox flow air battery (SLRFAB), based on O_2-saturated catholyte (conductive carbon particles added to a non-aqueous electrolyte). In SLRFAB, the passivation by the Li_2O_2 discharge product at the solid carbon-based current collector is alleviated, while the redox sites are multiplied because the oxygen redox reactions (ORR) mainly occurs at the dispersed carbon particles [2]. The development of slurries that match high electrochemical performance and rheological features suitable for an efficient flow in a battery is challenging. The effect of changing catholyte composition on SLRFAB has been deeply discussed. By a cell design evaluation, the specific energy can be increased changing the carbon content from 2% to 10%, reaching the outstanding value of ca. 900 Wh kg^{-1} [3].

Increasing the carbon content in the SLRFAB catholyte is an arduous approach because the carbon particles affect the viscosity of the catholyte. Thus, the best compromise between the energy of the SLRFAB and the energy required by pump for the catholyte flow, the latter being essential to the battery operation, should be found.

For the first time we address the challenging study of semi-solid catholyte formulation for Li/O_2 batteries. A range of catholytes for a SLRFAB is investigated, using different carbon and carbon contents. Specifically, catholytes based on Super-P® and Pureblack® in O_2-saturated TEGDME LiTFSI electrolyte have been studied with a focus on cyclability and rate response. The electrochemical results are presented and discussed and related to the morphology, rheology and electrical conductivity of the semi-solid catholytes.

The work demonstrates that up to hundreds of cycles with areal capacity of 4 mAh cm^{-2} and currents higher than 0.5 mA cm^{-2} are achievable using 10 wt.% Pureblack® catholyte.

These results open new paths towards the development of post-Li-ion and new redox flow batteries [4].

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References:
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