Optimizing the performance of organic Li-battery electrode materials: the example of dilithium trans-trans benzenediacrylate

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Organic electrodes represent a promising alternative as new electrode materials for Li-ion batteries (LiBs). Among the different existing categories, especially conjugated carbonyl materials have gained interest due to their abundance, cheapness and recyclability. However, they are associated with three major drawbacks which prevent their commercial usage: solubility in liquid electrolytes, poor volumetric density and poor conductivity [1,2].

Recently, dilithium trans-trans benzenediacrylate ($BDALi_2$) performances have been improved by a simple method such as in situ carbon-coating in the liquid state [3], but the large particle size obtained after drying restricts the performance at elevated current rates. Combining the benefits of in situ carbon-coating with a high surface area, would therefore be beneficial. Here freeze-drying has shown to be a useful method for reducing the size of water-soluble particles and to be compatible with in-situ carbon-coating [4].

A series of aqueous solutions have been prepared with different concentrations of $BDALi_2$: 2, 5, 6, and 7 wt%. The samples were submitted to a freeze-drying process, where after electrodes were prepared by mixing the resulting powders with carbon SP (33 wt%).

Galvanostatic cycling performed between 0.9 and 3 V at a rate of 1 Li^+ per 10 h in 1 M LiTFSI/DMC electrolyte, showed that the best observed performance is seen for the 6 wt% material (Fig. 1). An improvement in particle size can also be observed (Fig. 2). The combination of freeze drying with in-situ carbon coating was expected to give even better results but, this was not the case, which might be explained by a high cell resistance resulting from a poorly connected carbon matrix in the electrode.

By increasing the carbon SP content to 50 % of the total weight (using a sample of 6 wt% of dispersed $BDALi_2$ in solution and there after freeze-dried), a clear improvement could be made, indicating that this problem can be overcome.

Galvanostatic cycling at a comparatively high C-rate showed promising results which cannot be explained merely by the additional amount of carbon additive: good capacities and moderate capacity fading are achieved when cycled at C/2 and 2C (corresponding to one lithium every hour or 15 minutes for the di-lithiated compound, respectively, Fig. 3).

A simple and potentially cost-effective method for optimizing the electrode formulation of water-soluble organic electrode material for LiBs will thus here be presented, using a combination of in situ carbon coating and freeze-drying.

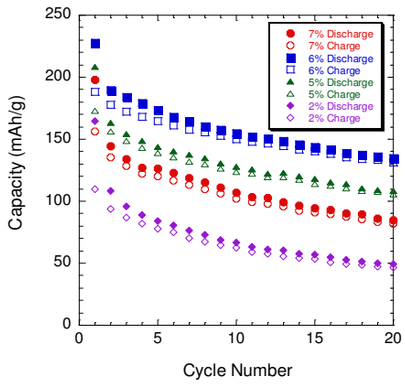

Fig. 1 Capacity retention curve of a Li half cell using dilithium benzenediacrylate prepared by freeze-drying of an aqueous solution with different concentrations and cycled galvanostatically between 0.9 and 3 V at a rate of 1 Li^+/10 h (C/20) in 1 M LiTFSI/DMC.

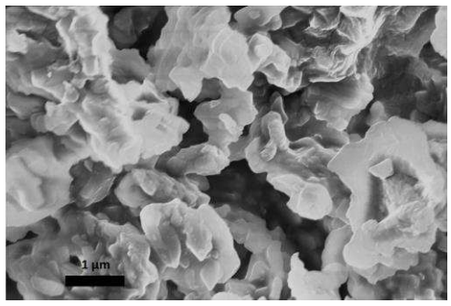

Fig. 2 SEM of $BDALi_2$ prepared by freeze-drying from a 6 wt% aqueous solution.

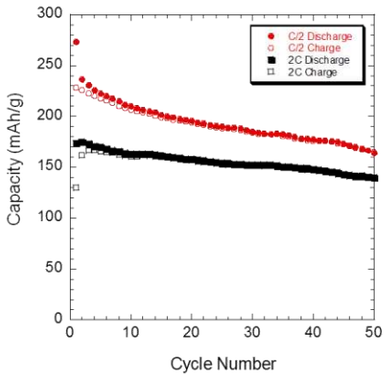

Fig. 3 Capacity retention curve of a Li||$BDALi_2$ half-cell prepared by freeze-drying a 6 wt% aqueous solution of $BDALi_2$ with in situ carbon-coating (50% in total weight) and cycled galvanostatically between 0.9 and 3 V at a rate of C/2 or 2C in 1 M LiTFSI/DMC.

References

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