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INTERFACIAL ELECTROCHEMISTRY REGULATION ON CARBON ANODES BY AN ETHER-BASED ELECTROLYTE FOR SODIUM-ION BATTERIES

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Introduction

Carbon materials are the most promising anode for sodium-ion batteries (SIBs), which have already been practically applied in large-scale energy storage market. However, as the commercialized anode in lithium-ion batteries, graphite can only deliver limited capacity in SIBs. Researchers had to turn to more amorphous and defective carbon materials. Unfortunately, the initial coulombic efficiency (ICE) of these carbon materials are typically very low (below 40 %) and far from the practical demand. In order to address this critical low-ICE bottleneck, we propose two fundamental approaches from both electrode and electrolyte sides to regulate the interfacial electrochemistry on carbon anodes and improve the ICE without sacrificing the capacity.[1,2]

Materials and Methods

Graphite oxide was prepared from graphite powder using a modified Hummers method. Reduced graphene oxide (rGO) was obtained by a two-step thermal treatment of graphite oxide. Porous graphene monolith (PGM) was obtained by hydrothermal treatment of graphene oxide colloidal suspension. The Al_2O_3/PGM was obtained through soaking and stirring PGM in $AlCl_3$ solution, followed by filtration, drying and post annealing.

Results and Discussion

Here, rGO was investigated as a model anode with high specific surface area and massive defects. When modifying the conventional ester-based electrolytes to ether-based electrolytes, a dramatic improvement in ICE from 39 % to 74.6 % is obtained. Besides, the reversible charge capacity is almost twice higher in ether-based electrolytes after 100 cycles at 0.1 A/g. The improvement of electrochemical performance resulted from the modified SEI microstructure and composition. Ether-derived SEI has a denser organic layer at the exterior and a relatively uniform mixture of organic/inorganics at the interior, which can facilitate the sodium ions diffusion and improve the efficiency of sodium storage.[1] Moreover, we illustrated that the energy barrier to charge transfer at the electrolyte/electrode interface is the factor dominating the interfacial electrochemical characteristics and it is significantly reduced (a factor of 2.7) in ether-based electrolytes.[4]

In addition, we proposed an effective but simple remedy for deactivating defects in the rGO with Al_2O_3 nanocluster coverage, in order to further regulate the SEI. It was shown that these Al_2O_3 nanoclusters suppressed the decomposition of conductive sodium salt in the electrolyte, resulting in the formation of a thinner and more homogenous SEI. Meanwhile, Al_2O_3 nanoclusters could reduce the detrimental etching of the SEI by hydrogen fluoride (HF). As a result, both

efficiency and stability of rGO-based electrode can be further enhanced.[3]

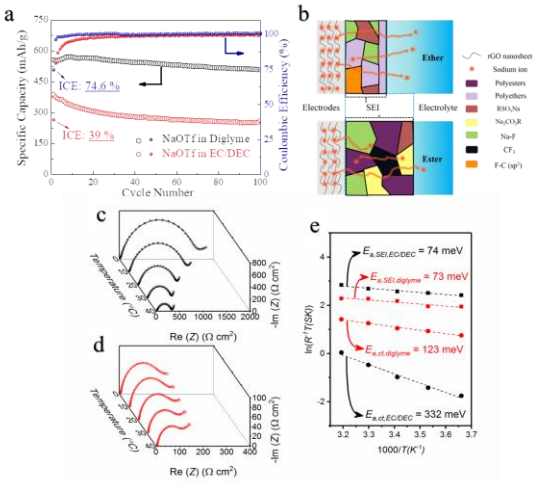

Figure 1. a) the comparison of electrochemical performance, b) the scheme for the different composition of SEI and c) temperature-dependent EIS of rGO electrodes in different electrolytes.

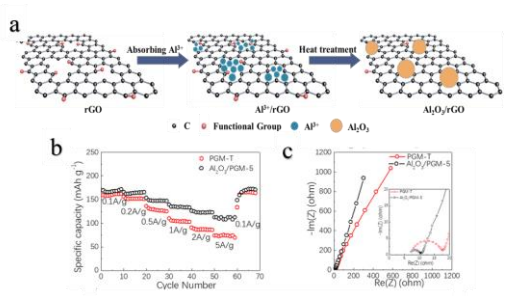

Figure 2. a) Scheme of the formation of Al_2O_3 nanoclusters on defective rGO, b) Rate performance and c) EIS comparison of electrodes with or without Al_2O_3 nanoclusters shielding.

Conclusions

Ether-based electrolytes can effectively regulate the interfacial electrochemistry on carbon anodes and improve the ICE without sacrificing the reversible capacity. Besides, the well-designed Al_2O_3 nanocluster shielding can further enhance the efficiency and cycling stability.

Acknowledgment

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