Controlled Surface Oxidation of LK-702 Graphite Anode Material Using CO_2

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Graphites have attracted considerable attention as anode materials for lithium ion cells due to their high reversible capacity and relatively low irreversible capacity during the first few formation cycles. There have been several studies aimed at further decreasing the irreversible capacity of graphite anodes; one approach toward this goal has been to modify the surface of the graphite by oxidation. Mild air oxidation of graphite has been demonstrated by Peled and coworkers to lower the irreversible capacity of Li-ion anodes.[1] In contrast, Rubino and Takeuchi found an increase in irreversible capacity following air oxidation of synthetic graphite LK-702.[2] Since differences in graphite materials and the choice of electrolyte will influence the amount of irreversible capacity in a lithium ion cell, a careful study was undertaken using LK-702, with CO_2 used as the oxidizing atmosphere. The oxidation of graphite by CO_2 has been reported by Winter and coworkers to produce a mild burn-off of carbon compared to the more aggressive reaction of O_2 with graphite at high temperature.[3] Thus, heat treatment of LK-702 under an atmosphere of CO_2 was investigated here as a means to carefully study the results of surface oxidation on the electrochemical properties of the graphite.

Oxidized LK-702 samples were prepared using a variety of reaction times and temperatures and then characterized by percent mass loss, XRD, SEM, and BET surface area measurements. Notably, the specific surface areas of the oxidized graphite samples did not linearly correlate with oxidation time or mass loss. Between 0% and 2% mass loss the graphite samples displayed a 50% increase in surface area, while between 2% and 6% mass loss surface area measurements were nearly constant at 5.0 m^2/g, and above 6% mass loss the surface area decreased to 4.2 m^2/g. SEM analysis was used to identify particle morphology and size following CO_2-oxidation. The oxidized graphites were used as anode materials for Li-ion cells, and the ability of these materials to intercalate lithium was studied. The reversible capacity and capacity fade over 100 cycles for the graphite materials were relatively unaffected by the treatment, but the irreversible capacity did vary as a function of the percent reaction, displaying general trends related to the specific surface area of the samples.

References

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2) R. S. Rubino, E. S. Takeuchi, *J. Power Sources*, 81-82, (1999) 373-377.
3) J.P. Olivier, M. Winter, *J. Power Sources*, 97-98 (2001) 151-155.