Aqueous Processing of Graphite Anodes for Lithium Ion Batteries

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Recently, as electronic devices, in particular, portable electronic devices such as PDAs, cellular phones, and notebook computers are widely used in various areas, batteries for driving these electronic devices have been actively developed in the direction of small size, thin thickness, lightweight, and high performance. Rechargeable lithium batteries have been used as main driving sources for portable electronic devices due to advantages such as lightweight and high energy density.

Graphite anodes for lithium ion batteries were fabricated in an aqueous system using sodium carboxymethyl cellulose (CMC) and emulsified styrene butadiene rubber (SBR). The transition from a nonaqueous to an aqueous mixing/coating process may involve unexpected difficulties in preserving the electrical properties for battery applications. These problems mainly originate from the distinct physicochemical properties between graphite particulates and water and graphite particulates and organic liquids. In addition, slurry formulations that are compliant with the suspended medium, including the binder and other functional additives, have to be newly determined. Experimental results on the preparation of a stable slurry are presented to demonstrate a successful aqueous mixing and coating process for an electrode composed of natural graphite. This was confirmed by the measurements of the electrokinetic behavior, the flow behavior, microstructural observations, and measurement of the pore size. It was found that the dispersion stability of the graphite suspension was improved when both the SBR and CMC were incorporated into the graphite suspension. An unstable graphite suspension resulted form the use of CMC alone. Electrochemical experiments using a Li/organic electrolyte/as-cast natural graphite anode halfcell and cylindrical lithium ion cells exhibited an initial discharge capacity above 340 mAh/g, improved chargedischarge efficiency and an excellent rate capability. Furthermore, from the charge-discharge cycling test of 2400mAh-class Li-ion battery, the cycle life was discussed in relation with the adhesion strength of the electrode.

REFERENCES

- 1. U. Paik, V.A. Hackley, and H.W. Lee, J. Am. Ceram. Soc., **82**(4) (1999) 833-840.
- 2. Y.-M. Choi, U. Paik, and K.-H. Kim, Korean Patent Application No. P2003-0040085 (2003).
- 3. R. Barbucci, A. Magnani, and M. Consumi, Macromolecules, **33** (2000) 7475-7480.



Fig. 1. Electrokinetic behavior of graphite suspension prepared at a mass fraction of 5% solids with both CMC and SBR as a function of CMC concentration. The suspensions measured for electrokinetic analysis were prepared with a 1.5% SBR mass fraction based on the solids contents.



Fig. 2. Flow diagram versus solids concentration for suspensions containing graphite, 1.5% SBR, and 1.5% CMC at a mass fraction on solids. CMC has two different degrees of substitution; DS of 0.70 (squares, broken line) and DS of 1.28 (circles, dotted line). The as-drawn lines are best-fit results of viscosity data measured at the constant shear rate of 400 sec⁻¹.



Fig. 3. . First charge-discharge curves of the natural and artificial graphite anodes prepared using our aqueous method and a conventional non-aqueous method, respectively, in a 1.1 M LiPF6 EC/EMC/DMC(30/60/10 vol.%) solution.