Preparation of lithium iodide-doped Fe-containing Li_2S-based positive electrode materials applicable for Li-S battery

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Lithium sulfide (Li_2S) is one of the promising cathode active materials for high-energy rechargeable lithium batteries because of its high theoretical capacity (*ca.* 1170 mAh・g^{-1}) [1]. However, this material is both electronically and ionically resistive, which gives rise to relatively low electrochemical performance in the cells [2]. Recently, we have tried to prepare Fe-containing Li_2S-based positive electrode material (Li_xFeS_y, typically Li_8FeS_5) and found that the cells showed the discharge capacity of more than 700 mAh・g^{-1} [3]. However, this material showed relatively rapid capacity degradation with cycling; typical capacity retention after 30 cycles was *ca.* 36%. In the present work, we have tried to incorporate lithium iodide (LiI) into Li_8FeS_5 for improving the cycle capability of the cells, after the previous report on the LiX(X=Cl, Br, I)-doped Li_2S applicable in all-solid-state batteries [4]

Li_2S and FeS with a molar ratio of 4 : 1 were blended thoroughly, and the mixture was then treated by the SPS process at 600^{o}C [3]. The resulting pellet (Li_8FeS_5) was ground and mixed with LiI (molar ratio of 1 : 0.1-1.0) and acetylene black (weight ratio of 9 : 1), and then it was mechanically milled for 8 h to yield the Li_8FeS_5-*x*LiI sample. Electrochemical lithium extraction / insertion reactions were carried out using lithium coin-type cells with 1M $LiPF_6$ / (EC + DMC) electrolyte at a current density of 46.7 mA・g^{-1} (corresponding to 0.04C).

The obtained Li_8FeS_5-*x*LiI samples were black in color, and the XRD patterns showed that they consisted of low-crystalline Li_2S. The electrochemical tests for the Li_8FeS_5-*x*LiI sample cells showed that the initial discharge capacity decreased with the incorporated LiI content (*x*); typically *ca.* 630 mAh・g^{-1} for the Li_8FeS_5-0.5LiI, which is lower than that of non-doped Li_8FeS_5 (*ca.* 810 mAh・g^{-1}). However, the Li_8FeS_5-0.5LiI sample cell showed much improved cycle capability; the capacity retention after 30 cycles was *ca.* 72%, which was nearly double of the Li_8FeS_5. The improved mechanism was examined using the X-ray absorption and scattering techniques, and the results will be presented in the Conference.

Acknowledgements

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