All-Solid-State Lithium-Ion Batteries Using Oxide Solid Electrolyte Assembled by Spark Plasma Sintering

Toyoki Okumura, Tomonari Takeuchi and Hironori Kobayashi

Research Institute of Electrochemical Energy, National Institute of Advanced Industrial Science and Technology (AIST), 1-8-31 Midorigaoka, Ikeda, Osaka 563-8577, Japan.

To develop an all-solid-state lithium-ion battery (ASS-LIB) using oxide solid electrolyte for applications such as electric vehicles, the thicknesses of stacked electrode and electrolyte layers need to be curtailed to several ten to hundred μm order. This can be achieved through powder technology. In the ASS-LIB, the good contact interfaces should be prepared by a simple powder sintering process, and also produces little ion-blocking impurities. Well-known high lithium-ion conductive oxides possess of over 10^{-4} S cm^{-1}; however, these required sintering at high temperature beyond 1000 °C to facilitate good contact with the electrodes. In addition, most electrode materials produce impurities after sintering. To address these issues, the ASS-LIBs were assembled by the use of oxide solid electrolyte with low melting point ($Li_{2+x}C_{1-x}B_xO_3$)[1] and/or the use of spark plasma sintering (SPS)[2], which could be processed at low temperature.[3]

For assembling ASS-LIB, composite electrode powder was firstly prepared from a mixture of 70 wt% $LiCoO_2$ and 30 wt% $Li_{2.2}C_{0.8}B_{0.2}O_3$ electrolytes. Then, Au/composite electrode powder/ $Li_{2.2}C_{0.8}B_{0.2}O_3$ powder was assembled by SPS process at 450 °C for 1 min under 30 MPa of pressure. Lithium foil was used as a reference/counter electrode. A poly(ethylene oxide) -based polymer electrolyte film was inserted between the lithium foil and the $Li_{2.2}C_{0.8}B_{0.2}O_3$ electrolyte separator to reduce the interfacial resistance with adhesion as possible. Electrochemical charge-discharge test was performed at a constant current of 10 μA cm^{-2} at 60 °C. The ASS-LIB shows an initial charge-discharge profile which is similar to the liquid electrolyte case, and the discharge capacity is 118 mAh g^{-1}. No impurity peak was observed in the powder XRD pattern of the $LiCoO_2$-$Li_{2.2}C_{0.8}B_{0.2}O_3$ composite electrode after SPS process. Therefore, the reversible capacity of ASS-LIB could be measured in this study, thanks to the use of $Li_{2.2}C_{0.8}B_{0.2}O_3$ with low melting point and SPS process.

References

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