Development of a quasi-solid composite electrolyte for 3D-structured batteries

Hirokazu Munakata, Yoshiyuki Shibata, Kiyoshi Kanamura

Department of Applied Chemistry, Graduate School of Urban Environmental Sciences
1-1 Minami-Osawa, Hachioji, 192-0397 Tokyo, Japan

E-mail: *munakata@tmu.ac.jp*

So far, the development of lithium-ion batteries has been carried out in a layer-by-layer configuration, in which an electrolyte layer is sandwiched with anode and cathode layers. However, it is impossible to realize both high energy density and high power density in this configuration since the diffusion of Li^+-ions in the electrode becomes difficult as increasing the electrode thickness. As a result, the practical thickness of electrode is limited up to ~ 100 μm in current lithium-ion batteries. In order to overcome this limitation, we have focused on 3D-structured electrode configurations[1)]. For example, an interdigitated electrode configuration is available (Fig. 1). This comfiguration can provide a constant distance (Li^+-ion diffusion length) between cathode and anode even if the electrode heights (the amounts of anode and cathode materials) are increased. Therefore, both high energy density and power density can be realized at the same time. However, it is difficult to use a conventional separator to avoid an internal short circuit between anode and cathode in this electrode configuration. Here, we developed a composite electrolyte composed of Li^+-ion conducting solid electrolyte powder ($Li_{0.35}La_{0.55}TiO_3$, LLT) and a nonvolatile liquid electrolyte (N,N-diethyl-N-methyl-N-(2-methoxyethyl)ammonium bis(trifluoromethanesulfonyl)imide including 0.32 mol kg^{-1} of lithium bis(fluorosulfonyl)imide) to solve the short circuit problem. When the volume content of LLT was 30%, an ionic conductivity of 1.7×10^{-3} S cm^{-1} was obtained at 30 °C, and the composite electrolyte had fluidity suitable to introduce into the gap beween anode and cathode and was then solidified after leaving to stand (Fig. 2). This quasi-solid-state property (thixotropy) improved not only the safety but also the cycle performance of 3D-structure battery. For further improvement, we also examined the composite electrolyte including methyl methacrylate as a gelling agent.

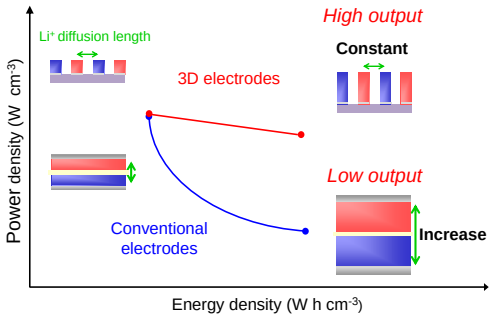

Fig. 1 Schematic relationships between energy density and power density in conventional and 3D-structured electrode configurations

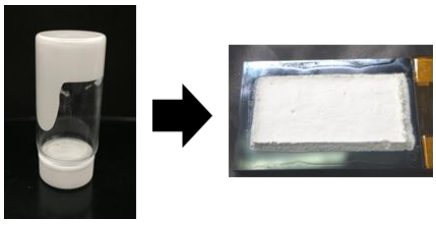

Fig. 2 The quasi-solid-state property of the prepared composite electrolyte

[1] K. Yoshima, H. Munakata, K. Kanamura, *J. Power Sources*, **208** (2012) 404