Cold sintering process for development of all solid-state batteries

Joo-Hwan Seo[a], Wonho Lee[b], Yongjun Leng[c], Chao-Yang Wang[c], Enrique D. Gomez[b], Thomas E. Mallouk[d], Clive A. Randall[a]

[a] *Department of Materials Science and Engineering, The Pennsylvania State University, University Park, PA 16802, USA*

[b] *Department of Chemical Engineering, The Pennsylvania State University, University Park, PA 16802, USA*

[c] *Department of Mechanical and Nuclear Engineering, Pennsylvania State University, University Park, PA 16802, USA*

[d] *Department of Chemistry, The Pennsylvania State University, University Park, PA 16802, USA*

E-mail: jks5678@psu.edu

All solid-state batteries have been extensively studied to replace the conventional Li-ion batteries with their advantages such as high safety, stability and energy density. Fabrication processes of the all solid-state usually require very high temperature over 800 °C for sintering of the solid electrolyte layer and composite electrode layers.

Recently, we have developed a low temperature ceramic process, named "cold sintering process", for sintering of ceramics and ceramic-based composites[1-2]. This process is a very promising process not only to save energy but also to avoid any material losses and side reactions during the sintering process. Using CSP, highly densified structures can be obtained through a mediate dissolution-precipitation process in transient aqueous conditions at low temperature. We successfully demonstrated that a highly densified Li-ion cathode can be fabricated by the cold sintering process at 180 ~ 240 °C[3-4]. The $LiFePO_4$-based binder-free thin composite cathode cold sintered showed very high volumetric capacity density, rate performance and cyclability.

The cold sintering process can be employed to fabricate highly densified electrode and electrolyte layers for all solid-state batteries. It is expected that the conductivity, gravimetric and volumetric energy density of the solid cell can be improved by densifying the microstructures of each layers and improving the contacts between them. The cold sintering process enables to improve the intimate contacts between active materials, conductive carbon and solid electrolyte particles as well as sinter the solid electrolyte layer at a low temperature.

In this presentation, we introduce how to apply the cold sintering process in the fabrication of all solid-state Li batteries. Using CSP, we have prepared all-solid-state Li-ion batteries with NASICON-type electrolytes (e.g., $Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3$) and various electrode materials such as $LiFePO_4$, graphite, $Li_4Ti_5O_{12}$ and so on. Their microstructures, properties and electrochemical performances will be discussed.

References:

[1] J. Guo. H. Guo, A. L.Baker, M. T. Lanagan, E. R. Kupp, G. L. Messing, C. A. Randall, Angew. Chem. Int. Ed. 55 (2016) 11457-11461.

[2] J. Guo, S. S. Berbano, H. Guo, A. L. Baker, M. T. Lanagan, C. A. Randall, Adv. Funct. Mater. 26 (2016) 7115-7121.

[3] J.-H. Seo, J. Guo, H. Guo, K. Verlinde, J. Guo, D. S. B. Heidary, R. Rajagopalan, C. A. Randall, Ceramics International 34 (2017) 15370-15374.

[4] J.-H. Seo, K. Verlinde, J. Guo, D. S. B. Heidary, R. Rajagopalan, T. E. Mallouk, C. A. Randall, Scripta Materialia 146 (2018) 267-271.