報告番号	甲　第14732号

主　論　文　の　要　旨

論文題目　**Analysis and design of Li ion flow in the materials for batteries**
(電池材料中におけるリチウムイオンの流れに関する設計と分析)

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論　文　内　容　の　要　旨

As environmental issues are attracted much more, attention on "carbon zero" has been focused much more. Upon this attention, the demand for electric vehicles, not conventional vehicles using fossil fuels, has been increasing. However, on the current electric vehicles with lithium (Li) ion battery (LIB), once the battery is damaged by an accident, a fire may occur due to unusual reactions with Li ions caused by unexpected contact between electrodes and its exposure to air, which is a huge problem on instability. Furthermore, the current electric vehicles have disadvantages on short mileage and low charging speed of the battery. To solve these problems, two developments on the battery materials are required. One is the development of a solid-state electrolyte, which has been focused as an alternative of a liquid electrolyte for the stability, and the other is the improvement of the electrode of the battery. Therefore, active research for developing both a solid-state electrolyte and the electrode has been continued to improve mileage and charging speed with the stability. However, there have been two problems that the conductivity of Li ion in solid-state electrolytes is generally lower than that in liquid electrolytes, and the flow of Li ion in electrodes has not been known well because it has a discrepancy that it requires higher crystallinity with smaller Li ion flow. Therefore, these are the barrier to achieve the long mileage and fast charging with its stability of a battery.

From the viewpoint of the flow of Li ions in materials, the development of materials for solid-state electrolytes and electrodes would help improve the performance and stability of batteries.

Therefore, this research developed battery materials to improve the Li ion conductivity of stable solid-state electrolyte and the capacity of anode. For the development of a solid-state electrolyte, a novel synthesis method, which can fabricate it easily and quickly, was proposed. Furthermore, it was found that the conductivity of Li ion was improved. On the other hand, for the improvement of the performance in the anode, the parameter sets related to the characteristics were constructed and with these sets, the correlation function that can predict specific capacity and electrical conductivity for carbon materials was successfully proposed.

In Chapter 1, a general introduction to the batteries, lithium-ion batteries, and solid-state batteries that have been used so far. In addition, lithium ions move when charging and discharging with lithium-ion batteries and solid-state batteries currently in use and under R&D. The reasons and difficulties of looking at this flow of lithium ions were explained. The structure of the material used for the solid-state electrolyte and the electrode and a method for solving the above difficulties were presented. Finally, we explained the purpose and concept of this study.

In Chapter 2, the analysis device and analysis method used in this study were introduced; XRD: for measuring the crystal structure of the material, Raman: for investigating at the crystal state of the surface, SEM: for observing the particle state on micro scale, EDX: for detecting the distribution of elements, and BET: for calculating the surface area of the material. In addition, a method for measuring Li ion conductivity and capacity, which are important for results in this study, was also introduced. Finally, as a result of the method to verify whether it is statistically meaningful, we conclude this chapter with an explanation of the p-value.

In Chapter 3, Zr was introduced as a substrate adding to LiF due to its stability, and LiZrF was synthesized with a simple method to improve lithium-ion conductivity. quickly adding Zr into LiF through liquid methanol as a medium. The interfacial contact with the electrode, which was one of the essential factors related to the battery performance in the solid-state electrolyte, was examined with SEM by mixing it with the cathode material. Compared to LiF with the cube structure, LiZrF showed rock-salt structure, therefore it was found the contact would lead to be smoother movement of Li ions. As a result, it was found that Li ion conductivity was improved with the addition of Zr-by changing from the FCC structure of LiF to the P31m structure of LiZrF.

In Chapter 4, the characteristics and predict equation of carbon materials used as conductive agents and anode materials in batteries were presented. On various carbon materials, such as graphite, SWCNT, MWCNT and carbon black, etc., data for the crystallinity was collected and it was examined with correlation analysis from nano to micro scales. Among these characteristics, capacity was largely influenced by the crystallinity along the c-axis, which was important factor in battery performance. In the aspect of the Li ion flow, when Li ion were intercalated between two graphene layers, the gap between the graphene layers increased, and during its intercalation, the increased area in the gap then decreased after Li ion passed away along its basal plane. Based on these results, a predict equation as well as the major factors and the phenomena for predicting capacity was proposed. As a result of validation process of the proposed function with the known results of previous research and statistical method, it was found that the function showed high predictivity.

In Chapter 5, a summary of the doctoral thesis and future directions were presented. In Chapter 1, introduction of the necessity and direction of research on a battery for electric vehicles was described. In Chapter 2, Introduction of the way to measure Li ion conductivity and capacity, including how to analyze the structure and properties of materials, and of the way to statistically verify the results was presented. In Chapter 3, the novel synthesis method of LiZrF material for improving low lithium-ion conductivity was presented with a fast and easy fabrication process, which lead to improve the Li ion conductivity as a problem of solid-state electrolyte of the battery. In Chapter 4, the high-predictable function as well as the major factors and the phenomena, which were related to the capacity of carbon materials, used as anodes and conductive agents in batteries, were presented.

This Ph.D. thesis designed and analyzed materials that will improve the stability and performance of the batteries for electric vehicles from the point of view of the Li ion flow. With considering these results, this research could provide novel and possible motivations to scientists and engineers who are dedicated to the research of battery materials and other energy devices for further improvement of its stability and performance.