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The Mechanism Study on the Difference between High and Low Voltage Performance of Li3V2(PO4)3

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Abstract : As one of most popular polyanionic compounds in lithium-ion cathode materials, Li3V2(PO4)3 has always suffered from the low rate capability especially during 3~4.8V, which is considered to be related with the ion diffusion resistance and structural transformation during the Li+ de/intercalation. Here, as the change of cut-off voltages, cycling numbers and current densities, the process of SEI interfacial film's formation-growing- destruction-repair on the surface of the cathode, the structural transformation during the charge and discharge, the de/intercalation kinetics reflected by the electrochemical impedance and the diffusion coefficient, have been investigated in detail. Current density, cycle numbers and cut-off voltage impacting on interfacial film and structure was studied specifically. Firstly, the matching between electrolyte and material was investigated, it turned out that the batteries with high voltage electrolyte showed the best electrochemical performance and high voltage electrolyte would be the best electrolyte. Secondly, AC impedance technology was used to study the changes of interface impedance and lithium ion diffusion coefficient, the results showed that current density, cycle numbers and cut-off voltage influenced the interfacial film together and the one who changed the interfacial properties most was the key factor. Scanning electron microscopy (SEM) analysis confirmed that the attenuation of discharge specific capacity was associated with the destruction and repair process of the SEI film. Thirdly, the X-ray diffraction was used to study the changes of structure, which was also impacted by current density, cycle numbers and cut-off voltage. The results indicated that the cell volume of Li3V2 (PO4)3 increased as the current density increased; cycle numbers merely influenced the structure of material; the cell volume decreased first and moved back gradually after two Li-ion had been deintercalated as the charging cut-off voltage increased, and increased as the intercalation number of Li-ion increased during the discharging process. Then, the results which studied the changes of interface impedance and lithium ion diffusion coefficient turned out that the interface impedance and lithium ion diffusion coefficient increased when the cut-off voltage passed the voltage platforms and decreased when the cut-off voltage was between voltage platforms. Finally, three-electrode system was first adopted to test the activation energy of the system, the results indicated that the activation energy of the three-electrode system (22.385 KJ /mol) was much smaller than that of two-electrode system (40.064 KJ /mol).

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