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Si-O-Cx nano-composite negative electrodes for next generation lithium ion batteries formed by plasma spray PVD[1]

MAKOTO KAMBARA, The University of Tokyo

Silicon is a promising material for negative electrode of lithium ion batteries (LIB) owing to its high theoretical capacity. However, this material inevitably suffers from huge volume change as large as 400% during charge/discharge process, i.e. alloying and de-alloying with lithium ions, which causes pulverization of the electrode itself and thus the loss of the electric path within the electrode, only after several charge cycles. Approaches to overcome this difficulty have been reported from the structural control point of views, such as design of materials at nanometer length scale, including nano-porous structure, nanowire structure and composites with other elements. Practically speaking, such a nano-structuring has to be also done by the process that has the potential to be developed to meet the industrial throughputs in future. With this in mind, we have demonstrated plasma spraying for production of the Si-Cx nano-composite powders and showed their potential as negative electrode by the reasonably high capacity and cycle stability. Another advantage of this process is that cheap raw materials, such as metallurgical Si (mg-Si), can be used as Si source so that industry compatible low cost is also anticipated. With these as background, we attempted plasma spraying with SiO powders as another Si source and the fundamental battery properties were characterized in comparison with the case with mg-Si powders. In brief, aggregate powders with 0.1-5μm in size having 20-50 nm a-SiO as the primary particle was produced by plasma spraying from 15μm feedstock SiO powders at the feeding rate of 2.4 g/min. Upon addition of CH4 gas, reduction and disproportionation reaction of SiO is promoted, leading to the formation of nano-composite a-SiOx particle with $<$10 nm Si at its core. The half coin cells with these powders as electrode have shown 1000 mAh/g after 50 cycles with reasonable retention efficiency of $>$99.7%.

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