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Optimized nanoparticle formation during plasma spray for enhanced storage capabilities with transfer entropy evaluation

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Silicon nanoparticle is the promising candidate for anode of high density lithium ion batteries (LiB) as it possesses high theoretical capacity and attains high cyclability. Its nanostructure, however, conversely contains high oxygen content per unit mass due to its high specific surface area and hence the irreversible phase amount increases, which deteriorates the initial coulombic efficiency and the cycle capacity eventually. Furthermore, the nanoparticles in general tend to aggregate and hinder the homogeneous particle dispersion more in a complex organic-inorganic mixture slurry containing the less-polar organic solvent which is necessary for stable mixture with sulfuric solid electrolyte. The resultant inhomogeneous electrode structure is considered as the main cause for the decrease in the battery cyclability. Therefore, these silicon nanoparticles are to be produced with suppressed surface oxidation during synthesis while maintaining the nanosizing of the particles. Even so, from the production standpoint, to meet the quantity demands from the ever-growing EV market, nanoparticles have to be produced by high throughput method also using affordable raw materials. With these requirements in mind, we have successfully produced silicon nanoparticles with suppressed surface oxidation by plasma spray starting with low-cost powder feedstock at the industry-compatible high throughputs [2]. In particular, based on our understanding that the nanoparticles form via the cocondensation of high temperature along the gas streamline within the plasma jet, we have also attempted to introduce in-reactor axial cyclone to make the plasma gas streamline uniform to homogenize the nanoparticle formation path. Our fluid dynamics simulation shown in Fig.1 have revealed that the gas streamline was appreciably aligned by the cyclone and the gas cooling path until the powder collection filter was significantly shortened with less temperature and spatial fluctuation. These suggests that the condensation path of silicon nanoparticles are to be more uniform and the particles are effectively cooled down. In fact, the nanoparticles so produced clearly exhibited that the average particle size was reduced and the size variation was also appreciably suppressed. The battery cell using these nanoparticles have exhibited the increased capacity and cycle stability. Therefore this obviously demonstrated that the homogenization of gas streamline in the condensation-associated temperature range directly leads to the uniform particle formation and also thus the uniform battery electrode structure, which contributes to higher battery cyclabilities. In order to quantify the degree of the gas streamline homogenization, we have employed the transfer entropy for the gas stream characteristics [3]. As a result, the degree of temperature and the gas streamline position variations was quantitatively evaluated as the potential guideline for particle formation. Statistical correlation with the nanoparticles so produced and the battery performance will be discussed in the presentation.

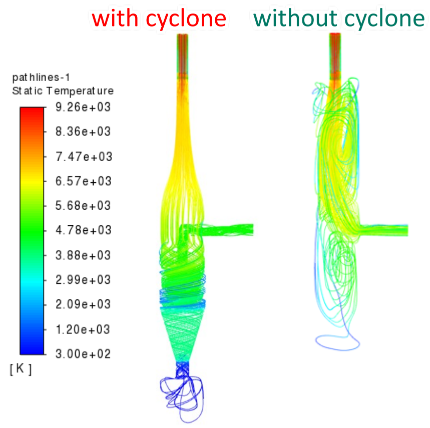

Fig.1 Simulated streamlines and temperature distribution of the plasma jet with/without axial-cyclone. Plasma was generated by ICP at the top of the reactor and the gas was taken out through the horizontal tube via the filter. The gas streamlines are seen to align appreciably, suppressing the circulating flows and leading to uniform spatiotemporal condensation.

References

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