Ultrasonic Spray Pyrolysis of Hierarchically Structured Cathode Materials

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Aerosol processing is widely used in industry to manufacture advanced materials such as pigments and oxide ceramics.[1] Simple and low cost continuous processing methods such as ultrasonic spray pyrolysis generate fine particles by decomposing atomized droplets containing metal precursors, and can be easily scaled up. Product particles with high purity are readily separated from the gas stream, without any liquid by-products unlike wet chemistry processes. In addition, multicomponent materials are also easily prepared by this route.

We have recently used a solution combustion technique to prepare layered mixed-metal oxide nanoparticles,[2] and are now exploring the possibility of making related battery materials using ultrasonic spray pyrolysis. Our initial work has successfully demonstrated that spherical $LiFePO_4/C$[3] and $LiCoPO_4/C$[4] particles can be prepared with good electrochemical performance. The particles are approximately 10 μm in diameter, but consist of nano-scale primary particles coated with carbon and have an interconnected network of nanopores that allow rapid transport of ions and electrons. The spherical morphology and relatively large size of the secondary particles allow easy processing of the materials into composite electrodes, and alleviate the energy density penalty often associated with nanostructuring. This illustrates the advantages of hierarchical structuring for battery materials.

The ultrasonic spray pyrolysis technique is now being extended to the preparation of other oxides and polyanionic based cathode materials with high voltage or capacity characteristics. By modifying spray pyrolysis conditions, the sizes of the secondary particles, made up of nanometric primary particles, can be varied from 1 μm to ~10 μm (Figure 1). Figure 2 shows an SEM image of spray-pyrolyzed $LiFe_{1/3}Mn_{1/3}Co_{1/3}PO_4/C$ particles,[5] indicating that multicomponent materials are easily prepared using this method. For this talk, we will discuss the microstructure of these materials and their relevance to the electrochemical properties.

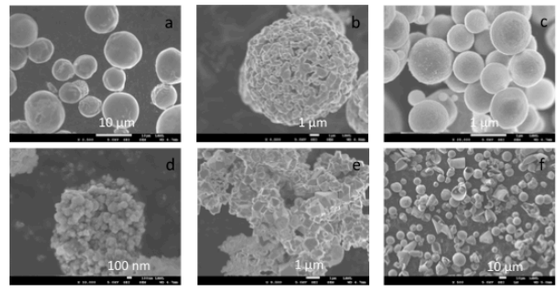

Figure 1. Scanning electron micrographs (SEMs) of $LiNi_{0.5}Mn_{1.5}O_4$ particles made by spray pyrolysis: a) as-made from 1M metal nitrate solution using a 120 kHz nozzle, b) particle from a, heated at 900° for 15 minutes c) as-made from 1M metal nitrate solution using a 2.4 MHz nozzle, d) as-made from 1M nitrate solution with glycine added, 120 kHz, e) sample d heated to 900°C for one hour, f) as-made 3M metal nitrate solution and 120 kHz.

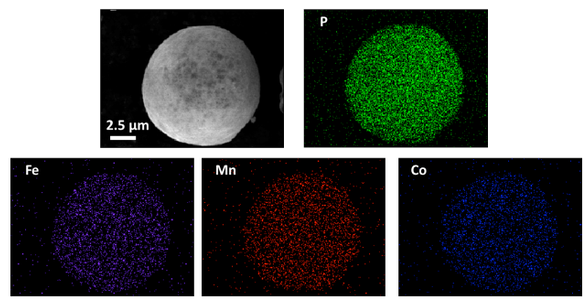

Figure 2. SEM image of $LiFe_{1/3}Mn_{1/3}Co_{1/3}PO_4/C$ composite particle and EDX elemental mapping for P, Mn, Fe, and Co.

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