

### **LiMn<sub>x</sub>Ni<sub>1-x</sub>O<sub>2</sub> Cathodes Revisited**

S. B. Schougaard and J. B. Goodenough  
University of Texas at Austin  
Mechanical Engineering, ETC 9.102  
1 University Station, C2200  
Austin, TX, 78712

The layered LiMO<sub>2</sub> compounds with M = Co or Ni<sub>1-x</sub>Co<sub>x</sub> are used commercially when high rates and small volume are needed; but cobalt is expensive, and operation on the Co<sup>4+</sup>/Co<sup>3+</sup> and/or the Ni<sup>4+</sup>/Ni<sup>3+</sup> couples leads to oxygen evolution when more than half of the Li is removed. In LiNi<sup>2+</sup><sub>1-x</sub>Mn<sup>4+</sup><sub>x</sub>O<sub>2</sub> compounds, on the other hand, both the Ni<sup>3+</sup>/Ni<sup>2+</sup> and the Ni<sup>4+</sup>/Ni<sup>3+</sup> redox couples are present due to the introduction of tetravalent Mn in the Ni sites and both redox couples are accessible in the 4V region. This occurs because of strong pinning at the O<sup>2-</sup>:2p<sup>6</sup> band, which is why we find no step in the voltage profile when transferring from one couple to the other. In our initial study of this system (Cushing BL, Goodenough JB, *solid state sciences*, 4(11-12):1487), it was shown that carbon coating the particles increased the performance at higher current densities. Unfortunately, the capacity for both the uncoated and coated materials was only about two-thirds of what had previously been reported for the LiMn<sub>x</sub>Ni<sub>1-x</sub>O<sub>2</sub> system (Ohzuku T, Makimura Y, *Chem Lett.* (8): 744). The latter report showed that in Li<sub>1-x</sub>Ni<sub>0.5</sub>Mn<sub>0.5</sub>O<sub>2</sub> a capacity of ~150 mAh/g can be attained while experiencing only a modest fade.

We present here data that allow us to understand the difference between our material and that of Ohzuku *et al.* i.e. we attempt to explain why materials with nominally the same composition have so very different electrochemical performances. The study is based on a combination of synthetic approaches and X-ray diffraction analysis. Using an improved synthesis, we are able to make cathode material, that at a 0.1 mA/cm<sup>2</sup> rate, has an average voltage of 4 V vs. Li, has a capacity of 160-170 mAh/g if operated over the range 4.3-2.75 V, has little or no capacity fade after 50 cycles, and does not contain any Cobalt.