Silicon/Carbon Composite Negative Electrode Materials

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With a theoretical capacity of 4200 mAh/g, silicon is an appealing negative electrode material for rechargeable lithium batteries. However, silicon electrodes are plagued by large volume changes during cycling and poor room-temperature kinetics.[1] Recent efforts have focused on improving silicon's capacity retention by using silicon/carbon composites. Umeno et al. achieved stable cycling with reversible capacities over 550 mAh/g with carbon-coated silicon.[2] Wang et al. reported large capacities (~600 mAh/g) for silicon and graphite milled for 150 hr.[3] Li et al. reported capacities as large as 2100 mAh/g (on a silicon-only basis instead of a total composite mass basis) for electrodes with a composition of 40 wt.% Si nanopowder, 40 wt.% carbon black, and 20 wt.% PVDF.[4] The reversible capacities dropped to approximately half of the original capacity over 20 cycles for the highest capacity electrode.

We have prepared silicon/carbon composites by milling commercial silicon and natural graphite powders. The cycling performance is shown in Fig. 1 for a few electrode compositions for identical cycling conditions. The reversible capacities are based on the total composite powder masses (i.e., the masses of both silicon and carbon are included). In all cases, the composites had large reversible capacities well over graphite's theoretical capacity of 372 mAh/g. As expected, increasing the silicon content in the composite increased the maximum reversible capacity while also increasing the capacity fade rate. With other cycling conditions, we have observed capacities over 600 mAh/g for more than 50 cycles. These electrode materials have been characterized by cyclic voltammetry and *in situ* and *ex situ* x-ray diffraction.

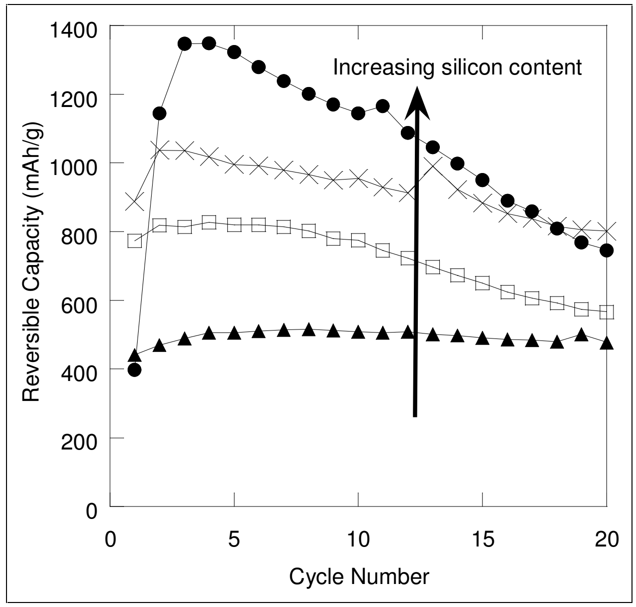

Figure 1

References

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