Photoelectron Spectroscopy (PES) studies of intermetallic anodes for Li-ion batteries

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Intermetallic anodes are today exploited as possible anode materials in Li-ion batteries. Lithium insertion mechanisms and reversible capacities have been described for a range of materials such as: Cu_6Sn_5 [1], InSb [2], Cu_2Sb [3], SnSb [4], MnSb and Mn_2Sb [5], etc. All these materials exhibit a capacity loss during the first discharge (during lithium insertion). This loss can be explained partly in terms of lithium ions trapped in oxide impurities (formed during synthesis of the crystalline intermetallic) and partly by electrolyte reduction and the accompanying formation of a Solid Electrolyte Interphase (SEI) on the electrode surfaces [6].

Cu_2Sb and SnSb were selected for this study because of their stable electrochemical cycling performance. We have here chosen to investigate both systems for an electrolyte consisting of 1M $LiPF_6$ in EC/DEC (ethylene carbonate and dimethyl carbonate). Our goal is to describe the surface chemistry of these materials before and after electrochemical cycling by Photoelectron Spectroscopy (PES) using synchrotron radiation (SR) and AlK_α radiation. The advantages of using SR are the high intensity of the excitation radiation compared to monochromatised light, and the possibility for varying the photon energy. The latter enables a depth profile to be obtained in a non-destructive way and also the detection of photoelectrons with the same kinetic energy for different elements. The SR-PES measurements were performed on the I411 at the Swedish National Synchrotron Radiation Laboratory MAX.

The electrode/electrolyte interface influences the electrochemical performance and also the thermal stability of the battery [7]. However, very little is known about the thermal stability of the intermetallic materials. The thermal stability of electrochemically cycled intermetallic anodes has here been studied by Differential Scanning Calorimetry (DCS). The results will be discussed in the light of surface chemistry results from PES measurements.

All results will be related to the case of the well-characterised SEI layer formed on graphite and lithium metal.

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References

[1] L. Fransson, E. Nordström, K. Edström, L. Häggström, J. T. Vaughey and M. M. Thackeray, *J. Electrochem. Soc.,* **149** (2002) A736.
[2] C.S. Johnsson, J.T. Vaughey, M.M. Thackeray, T. Sarakonsri, S.A. Hackney, L. Fransson, K. Edstrom and J.O. Thomas, *Electrochem. Comm.*, **2** (2000) 595.
[3] L.M.L. Fransson, J.T. Vaughey, R. Benedek, K. Edstrom, J.O. Thomas and M.M. Thackeray, *Electrochem. Comm.*, **3** (2001) 317.
[4] J. Yang, M. Winter and J.O. Besenhard, *Solid State Ionics*, **90** (1996) 281.
[5] L. Fransson, J.T. Vaughey, K. Edström and M.M. Thackeray, *J. Electrochem. Soc.* 2002. *In press*
[6] A. Ulus, Y. Rosenberg, L. Burstein and E. Peled, *J. Electrochem. Soc.*, **149** (1998) 4195.
[7] A.M. Andersson, M. Herstedt, A.G. Bishop, K. Edström, *Electrochim. Acta,* **47**, (2002) 1885.