Temperature Controlled Pressure Characterization (TPC) of Lithium-Ion Batteries with Silicon based Anodes.

M. Sternad [(1)*], E. Lanzer [(1)], N. Hochgatterer [(1)];
S. Koller [(1)], P. Raimann [(1)], M. Schmuck [(1)],
M. Schweiger [(1)], M. Takehara [(2)], M. Ue [(2)], M. Winter [(1)].

[(1)] Graz University of Technology, Stremayrgasse 16, A-8010 Graz, Austria.

[(2)] Mitsubishi Chemical Group Science and Technology Research Center, Inc., Battery Materials Laboratory, 8-3-1, Chuo, Ami, Inashiki, Ibaraki 300-0332, Japan.

*michael@sternad.com

During their cycle life, the active electrode material inside lithium-ion batteries perambulates several stadiums of transformation and degradation, whereas especially the film forming on the anode surface during the first charge results in electrolyte decomposition, leading to an increase of gas pressure in the cell's headspace. The attempt to create "role assigned electrolytes" e.g. by adding small amounts of film forming agents, in addition to common components like EC or DEC, normally results in an enhanced trend to gas production, either from the anodic and / or from the cathodic electrode surface during cycling.

To provide an industry-near test method for the evaluation of assembled anode - electrolyte - cathode combinations in respect of their future use as a commercial product, the Temperature Controlled Pressure Characterization (TPC) examination method has been developed. This test procedure enables the *in situ* measurement of the gas pressure and the electrode temperature with high accuracy and resolution in a temperature stabilized, swagelok based examination environment (typical temperature fluctuation +/- 0.1°C, Figure 1). Temperature changes during the electrochemical experiment, for example caused by resulting reaction and current heat are the base for a mathematic temperature compensation, that enhances the quality of the yielded pressure data.

The cell's electrode set-up was designed as an amorphous silicon layer anode on copper carrier, while the cathode consisted from $LiCoO_2$ / aluminum grid, in combination with a polypropylene based separator.

Using the fore described test conditions, several experiments using an electrolyte containing EC / DEC 3:7, 1M $LiPF_6$ with and without an additive amount of 5 % wt. vinylene carbonate (VC) have been performed (e.g. Figure 2). Subsequent, the anode surfaces have been studied using SEM (Figure 3).

Figure 1: Swagelok based TPC Pressure Test Cell.

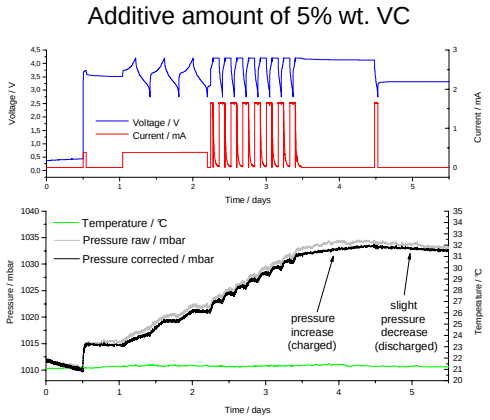

Figure 2: TPC experiment of EC / DEC 3:7, 1M $LiPF_6$ + 5 % wt. VC.

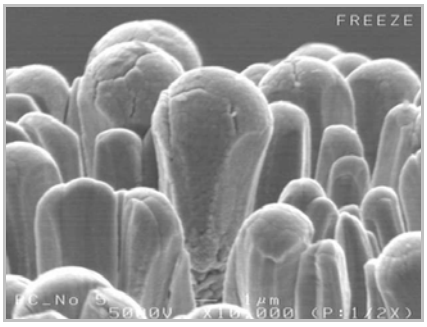

Figure 3: Lateral SEM image of anode surface after 10 cycles in EC / DEC 3:7, 1M $LiPF_6$ + 5 % wt. VC.

References

1. D. Goers, M. Holzapfel, W Scheifele, E. Lehmann, P. Vontobel, P. Novak., *J. Power Sources* 2004, *130*, 221-226.
2. M. Holzapfel, H. Buqa, L. Hardwick, M. Hahn, A. Würsig, W. Scheifele, P. Novak, R. Kötz, C. Veit, F.-P. Petrat, *Elektrochim. Acta.* 2006, *52*, 973-978.
3. W. Kong, H. Li, X. Huang, L. Chen, *J. Power Sources* 2005, *142*, 285-291.
4. J.-S. Shin, C.-H. Han, U.-H. Jung, S.-I. Lee, H.-J. Kim, K. Kim, *J. Power Sources* 2002, *109*, 47-52.