Theory of SEI Formation in Rechargeable Batteries: Capacity Fade, Accelerated Aging and Lifetime Prediction

Matthew B. Pinson[1] and Martin Z. Bazant[2,3]
Departments of [1]Physics, [2]Chemical Engineering and [3]Mathematics, Massachusetts Institute of Technology
77 Massachusetts Ave, Cambridge MA 02139, USA

An understanding of cycle is critical to the successful application of rechargeable batteries. We present a single particle model quantifying capacity fade due to formation of a solid-electrolyte interphase (SEI) on the negative electrode, a primary source of capacity fade in lithium ion batteries.[1]

This model accounts for the reaction rate forming SEI on the surface of active material particles, and the transport of reacting molecules through the existing SEI layer. Published experimental data[2, 3] show that the reaction and transport rates depend on temperature, and this dependence can be quantified using an Arrhenius equation: see figure 1.

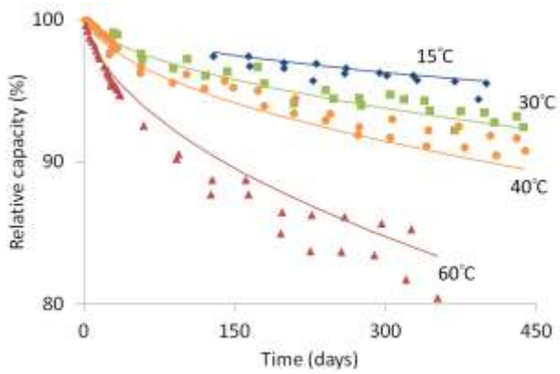

Figure 1: The capacity fade model explains observed fade at a range of temperatures, with parameters obeying an Arrhenius equation. Experimental data from (3).

The combination of the single particle model with the Arrhenius equation can be used to produce a quantitative prediction for long term capacity fade using short time 'accelerated aging' results at higher temperature. Such a prediction is superior to that obtained using the simpler assumption that capacity loss is simply proportional to the square root of elapsed time: see figure 2.

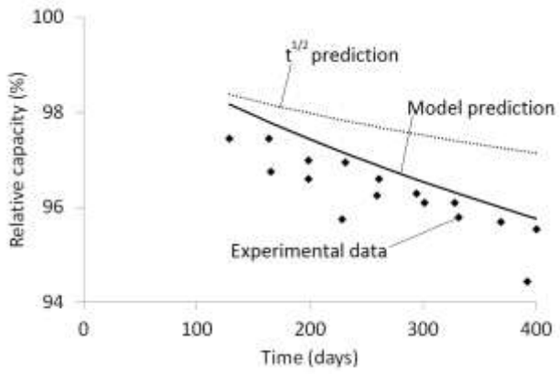

Figure 2: The capacity fade model can be used to provide quantitative predictions using accelerated aging data. Experimental data from (3).

We also extend the model to silicon electrodes, where large volume expansion on lithiation leads to rapid capacity fade. This application of the model predicts that capacity fade is linear, with the capacity loss per cycle proportional to the square root of the time per cycle. This prediction agrees with published experimental data:[4] see figure 3.

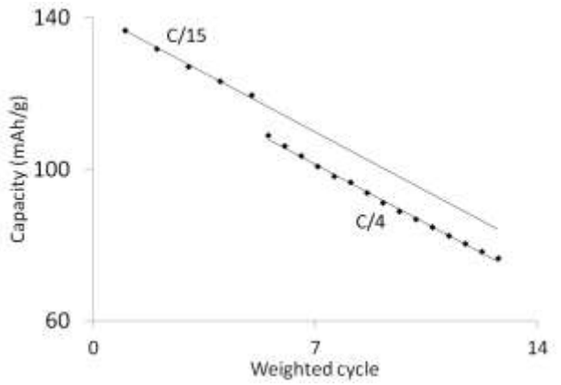

Figure 3: The capacity fade model applied to silicon accurately predicts the effect of cycling rate on fade. Experimental data from (4).

In addition to these results, the capacity fade model predicts a normal distribution of battery lifetimes, as observed in experimental results.[5] Furthermore, computational results indicate that there is essentially no spatial variation in SEI formation through the electrode, justifying the use of the single particle model.

1. M. B. Pinson and M. Z. Bazant, submitted to *Journal of The Electrochemical Society* (2012).

2. P. Liu, J. Wang, J. Hicks-Garner, E. Sherman, S. Soukiazian, M. Verbrugge, H. Tataria, J. Musser and P. Finamore, *Journal of The Electrochemical Society*, **157**, A499 (2010).

3. A. J. Smith, J. C. Burns, D. Xiong and J. R. Dahn, *Journal of The Electrochemical Society*, **158**, A1136 (2011).

4. L. Ji, H. Zheng, A. Ismach, Z. Tan, S. Xun, E. Lin, V. Battaglia, V. Srinivasan and Y. Zhang, *Nano Energy*, **1**, 164 (2012).

5. J. I. Park, J. W. Park, M. Jung, Y. H. Huh and S. J. Bae, *Journal of the Society of Korea Industrial and Systems Engineering*, **31**, 153 (2008).

This work was supported by Samsung via the Samsung-MIT Program for Materials Design in Energy Applications.