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Development of Li/$(CF_x)_n$ Battery at Sandia National Laboratories For Long-Lived Power Sources Applications

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Executive Summary

- Prepared cathode with uniform coating
  - Coated on 12 μm Al with 3 μm carbon coating
  - Carbon coating was found to be necessary for improved adhesion of $(CF_x)_n$.
- Deposited Li anode vapor on 10 μm Cu
- Achieved 3.6 Ahrs in 18650 cells
- Cell capacity proportional to cathode length
  - 36” long cathode gave 3.6 Ahrs
  - 29” long cathode gave 2.9 Ahrs
- Cell impedance remained nearly constant for voltages >2.2 V and increased below 2.2 V
- We will leverage our understanding and experience gained in this work to prepare Li/$(CF_x)_n$ battery for Long-Lived Power Sources Applications

Objectives

- Develop in-house facility to:
  - Fabricate $(CF_x)_n$ electrodes
  - Wind electrode rolls
  - Prepare 18650 cells
- Optimize capacity in 18650 cells
  - Perform electrochemical tests for
    - capacity at different temperatures
    - Impedance at different cell voltages

Outline

- Brief description of $(CF_x)_n$ chemistry
  - Highest theoretical energy/capacity compared to other Li primary cathode/catholyte
  - Very low self-discharge (0.5%/year near RT)
  - Very little voltage delay compared to other Li-primary cells
- Preparation of $(CF_x)_n$ electrodes
  - Partial list of in-house facilities
- Fabrication of 18650 cells with SNL coated electrodes
  - Evaluate performance in SNL-E with $LiBF_4$
- Summary

Description of the Cathode

- This chemistry has solid cathode as opposed to $SOCl_2$ which has liquid catholyte
- The equivalent weight for the $(CF_x)_n$ = 31 g
  - $(CF_x)_n$ has one of the lowest equivalent weights and hence the highest specific capacity of primary battery chemistry.
- Capacity = 865, mAhr/g

Equipment for Preparing Slurry

BYK Gardner Slurry Maker

Brookfield Viscosity Meter

Equipment for Coating and Rolling

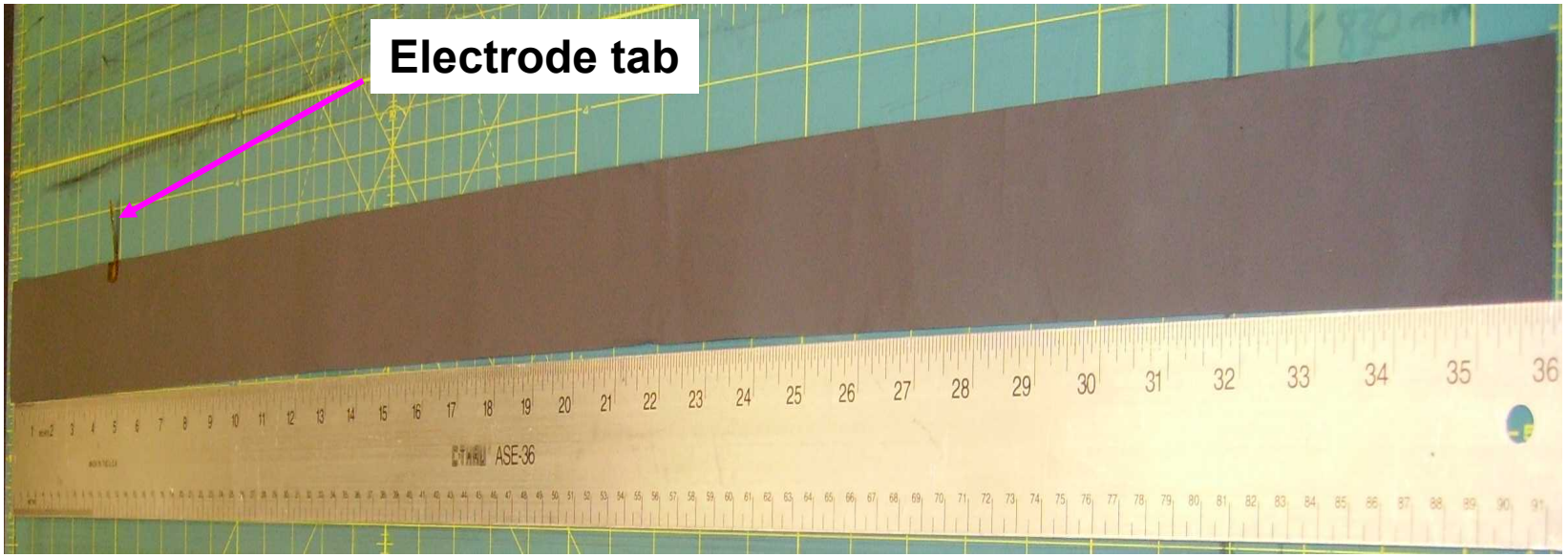
Electrode Coater Hohsen Model # HSCS-200

Hohsen Manual winder

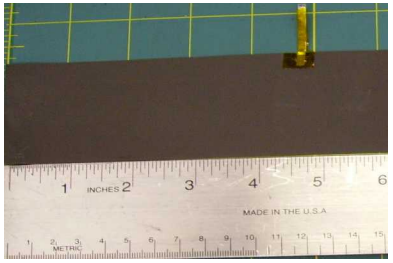
Cathode Slurry

- Composition
  - 90 w% active material
  - 5 w% acetylene black
  - 5 w% PVDF binder
- Slurry viscosity 230-240 cP
- Aggregate particle size < 5 micron

Photo of Cathode

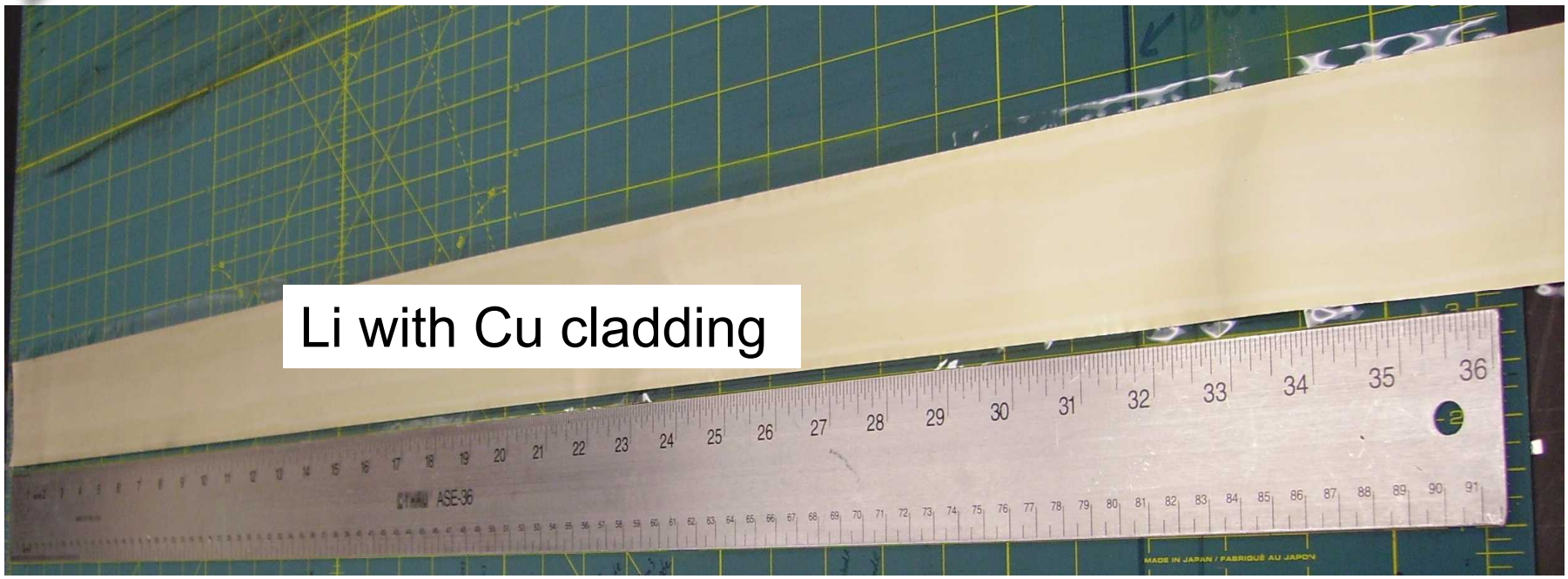


SNL coated $(CF_x)_n$ electrode with tab welded ultrasonically



Cathode material on Al current collector with 3 micron carbon for improving adhesion. Electrode thickness~4.2 mil.
The tab indicated above is ~ 4.75 inches from end.

Photo of Anode



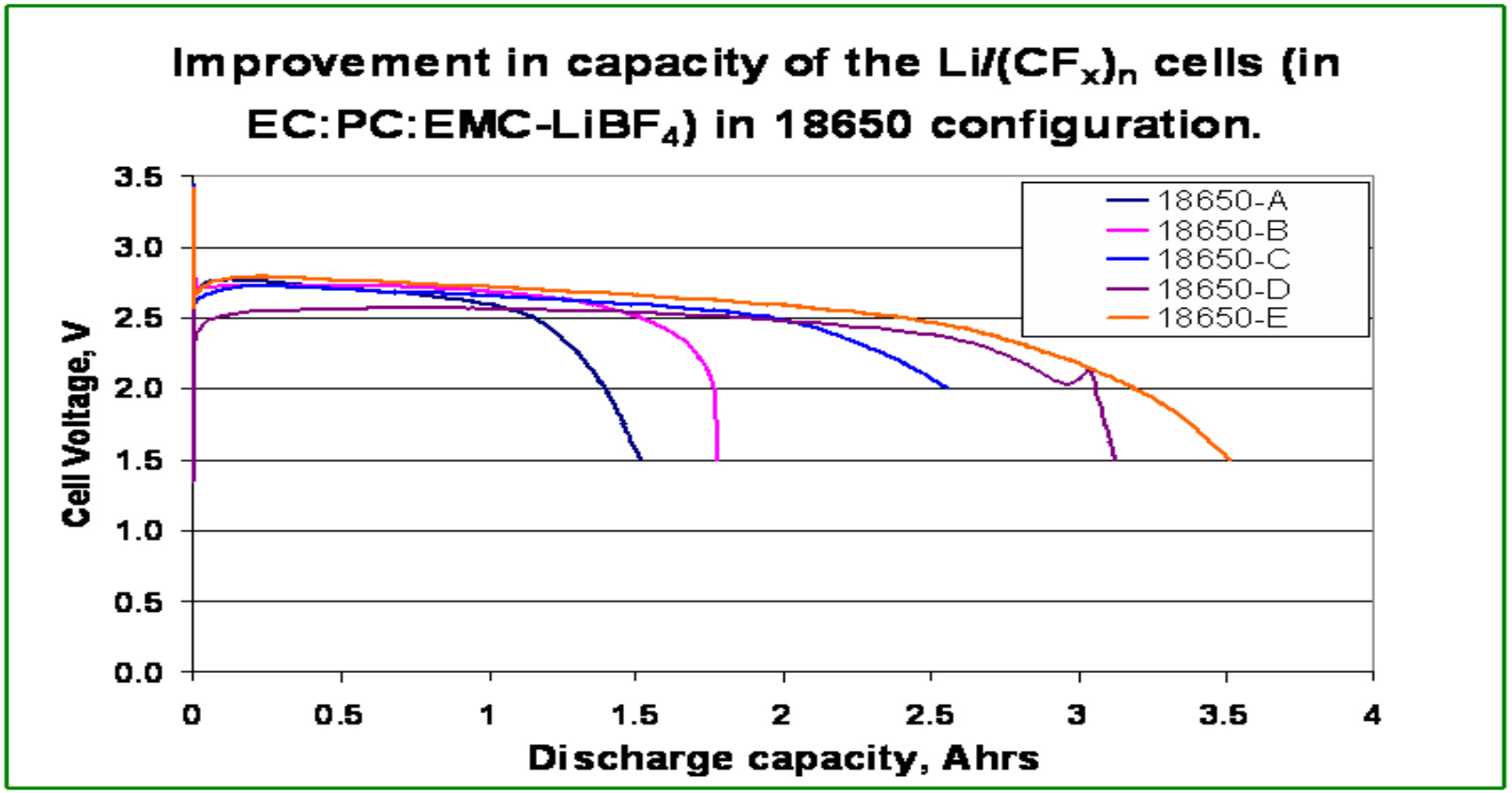
Li electrode with Cu cladding.

Total electrode thickness:	**50 µm**
Cu substrate:	**10 µm**
Li ~ 20 µm on each side:	**40 µm**

Photo of a Finished Cell

Sandia-Built 18650-Cell

Increasing Capacity Output in 18650 Cell

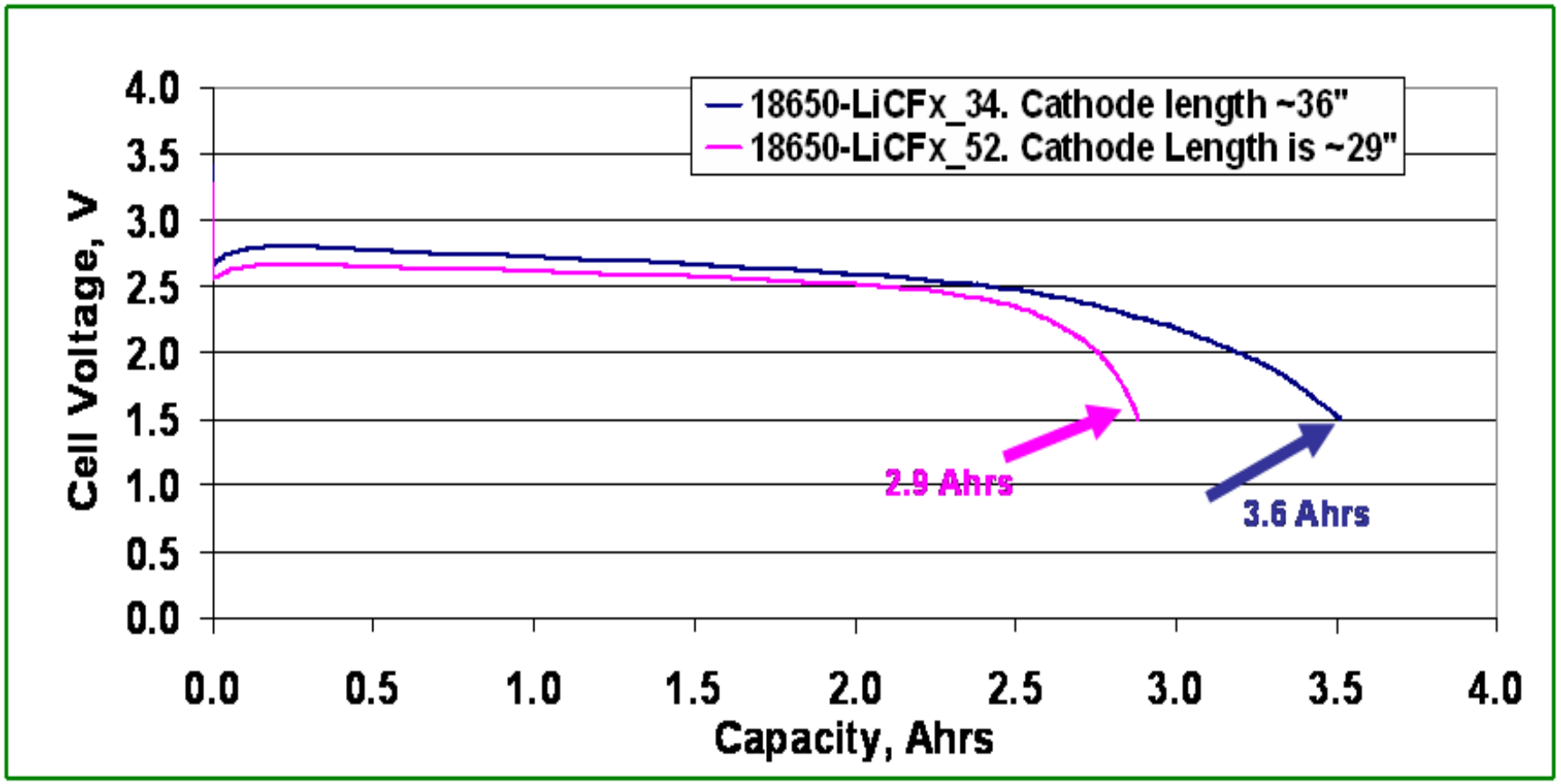


By constantly optimizing electrode quality, we increased delivered capacity from 1.5 Ahrs to 3.6 Ahrs

Increasing Capacity Output in 18650 Cell Cont'd

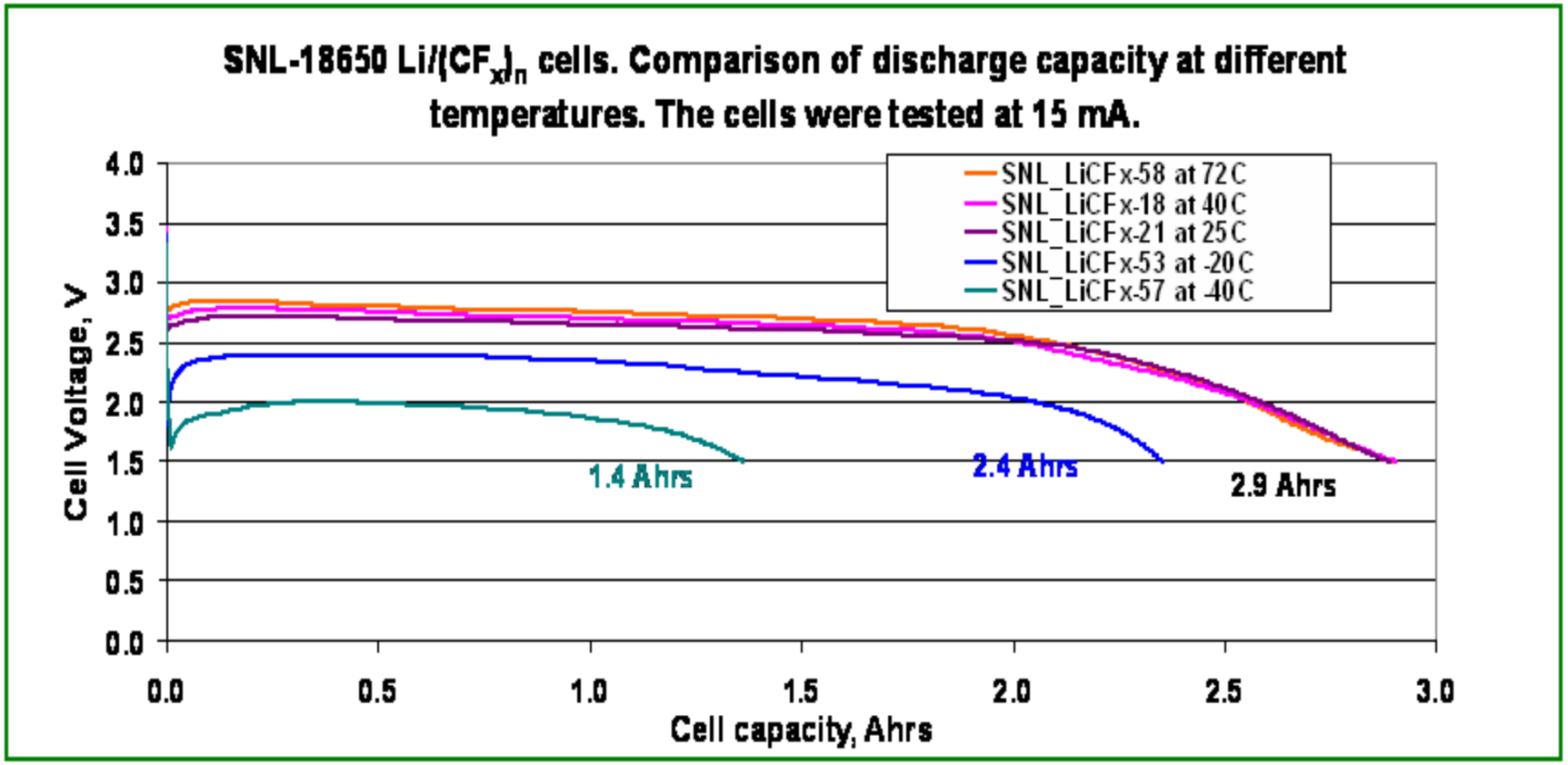
- Enhancement in capacity was obtained by fine tuning the slurry properties
  - Aggregate particle size and the viscosity of the slurry influence the electrode performance
- Anode thickness and roll tightness influence the cell performance

Discharge Capacity of Li/$(CF_x)_n$ 18650 Cells is Proportional to Cathode Length



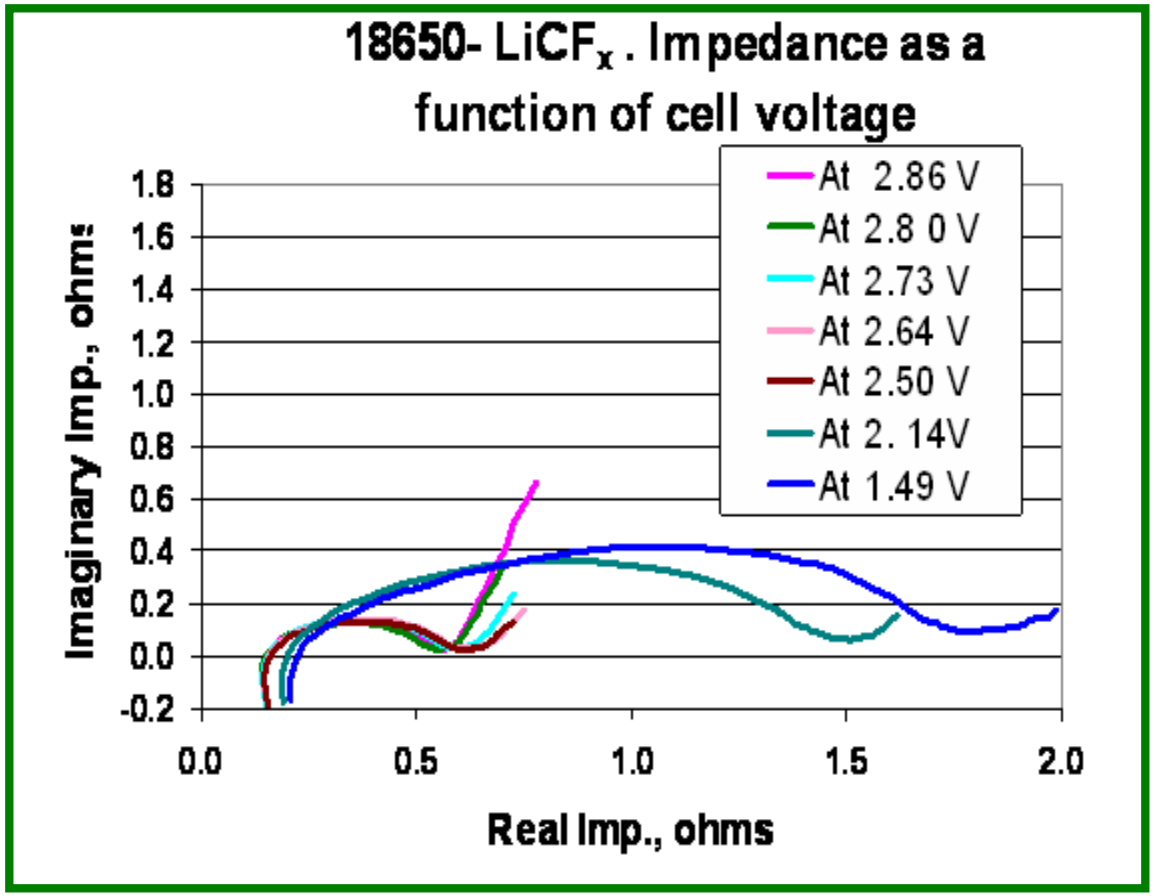
Lower operating voltage for the shorter cathode since the cells were discharged at the same current (15 mA).

Discharge Capacity of 18650 Cells at Different Temperatures



Delivered capacity at -20°C is >80% and at -40°C >40% of the 25°C capacity

Cell Impedance with Voltage

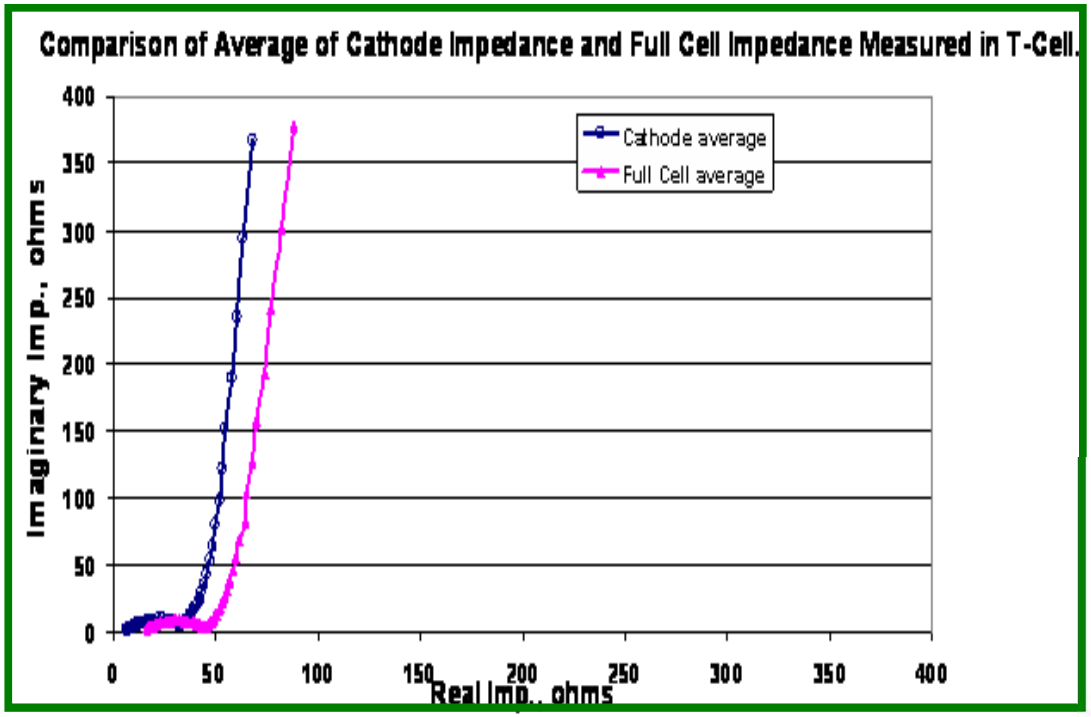


The cell impedance was collected between 50kHz and 50 mHz

Small increase in impedance with cell voltage.
However, at lower cell voltages the impedance increased dramatically.

Where is the impedance loop coming from. Cathode? Anode? or both.

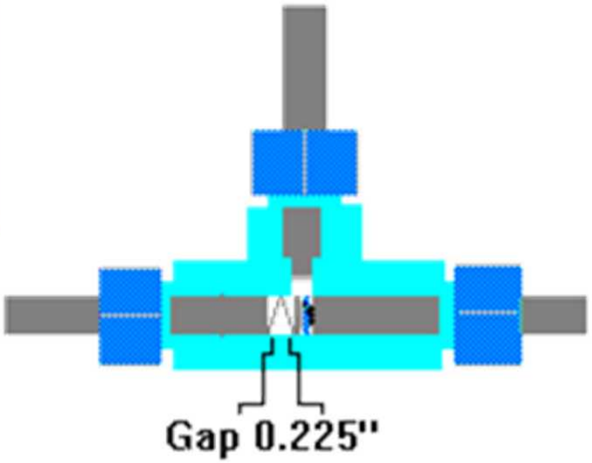
Cathode Contribution to Cell Impedance is Significant



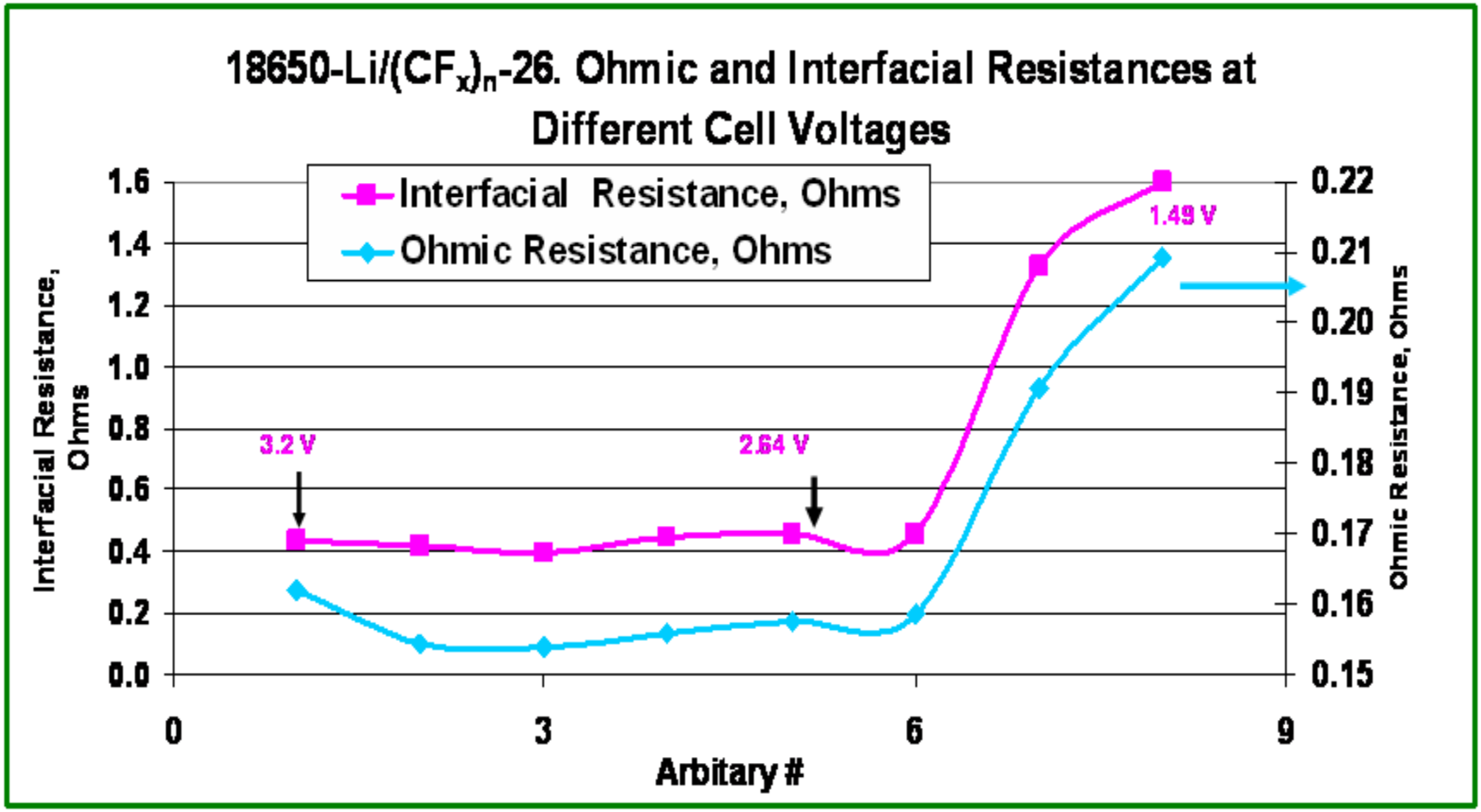
Impedance was collected in 3-Electrode Cell.

Cathode impedance makes up most of the cell impedance

T-Cell



Interfacial and Ohmic Resistances vs. Cell Voltage



Summary

- Prepared $(CF_x)_n$ electrodes with PVDF binder.
- Designed special Li anode with very thin Cu cladding.
- Built several 18650 cells.
  - Evaluated for capacity
  - Measured cell impedance at different voltages
- Increased cell capacity to 3.6 Ahrs by optimizing the slurry properties.
  - Both the aggregate particle size and the viscosity of the slurry are important
  - Tightness of the cell roll is also found to be important
- In the temperature range 25-72^{o}C the discharge capacity is practically the same
- At -20^{o}C, the capacity is 81% of the higher temperature capacity
- At -40^{o}C, the delivered capacity is ~ 47% of the higher temperature capacity
- Cell impedance was nearly constant at voltages above 2.2 V but increased significantly below 2.2 V may be due to accumulation of LiF.
- We will draw up on the experience gained in this effort and fine tune our skills to prepare Long-Life lithium batteries for SNL applications.