PNNL-SA-27904

UC-900

RECEIVED

FFR 18 1997

OSTI

Final Project Report

Pacific Northwest National Laboratory Collaboration with Moltech Corporation to Manufacture Lithium Polymer Batteries (C/PNL/061)

John D. Affinito

August 1996

**Prepared for U.S. Department of Energy
under Contract DE-AC06-76RLO**

**Pacific Northwest National Laboratory
Operated for the U.S. Department of Energy
by Battelle**

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor Battelle Memorial Institute, nor any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, produce, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or Battelle Memorial Institute. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PACIFIC NORTHWEST NATIONAL LABORATORY

operated by

BATTELLE

for the

UNITED STATES DEPARTMENT OF ENERGY

under Contract DE-AC06-76RLO 1830

Printed in the United States of America

Available to DOE and DOE contractors from the

Office of Scientific and Technical Information, P.O. Box 62, Oak Ridge, TN 37831;

prices available from (615) 576-8401, FTS 626-8401

Available to the public from the National Technical Information Service,

U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Final Project Report

Pacific Northwest National Laboratory Collaboration with Moltech Corporation to Manufacture Lithium Polymer Batteries (C/PNL/061)

John D. Affinito

August 1996

**Prepared for U.S. Department of Energy
under Contract DE-AC06-76RLO**

**Pacific Northwest National Laboratory
Operated for the U.S. Department of Energy
by Battelle**

Pacific Northwest National Laboratory Collaboration with Moltech Corporation to Manufacture Lithium Polymer Batteries

PURPOSE/OBJECTIVE

Initially: MOLTECH intended to start process scale up to manufacture lithium polymer batteries for various market segments beginning with batteries for consumer electronic applications. MOLTECH intended to use proprietary materials of their own combined with several PNNL technologies in their manufacturing process. The ultimate intent of the PNNL/MOLTECH collaboration was to fabricate a complete lithium polymer battery, consisting of seven basic layers, entirely under vacuum in an integrated, in-line, manufacturing process. MOLTECH was to: license PNNL patent 5,260,095 for the vacuum extrusion of polymer electrolyte and cathode layers; enlist PNNL to develop the processes for the vacuum deposition of the battery layers; commission PNNL to help design, build, install, and debug it's initial production equipment and, in the work outlined here, have PNNL determine the deposition rate to be expected for the lithium anode layer.

As the deposition rate of lithium was expected to be the rate limiting step in the manufacture of the vacuum deposited multilayer battery. The work to be performed under this CRADA was to attempt to determine the expected lithium deposition rate by utilizing the PNNL vacuum roll coater and rod-fed electron beam evaporation system. Once the lithium rate was determined the type, and number, of lithium deposition sources required in the production system could be specified. This information was to be used to complete the design phase, which will be proceeding separately and in parallel with the lithium rate determination work of this CRADA. Using the information obtained from this CRADA, PNNL would also assist in the design, construction, installation, and shake down of the MOLTECH lithium polymer battery production facilities.

Expansion Of Objectives: As the project developed successfully, the scope expanded to encompass all of the layers of the battery structure. To develop all of the vacuum deposition processes necessary for complete vacuum fabrication of a seven layer, thin film, lithium polymer battery on continuously moving, flexible substrate, utilizing materials used/supplied by Moltech corporation and patented PNNL processes. A schematic, typical of the battery layers, appears to the right as figure 1.

FIGURE 1. Layer Structure For Vacuum Deposited Battery Using: PML, LML, e-beam, and Thermal Deposition Sources On Flexible, Web Substrate

Encapsulation layer: <u>PML</u> evaporated polymer	2µm
Copper current collector: e-beam evaporated	0.5µm
Lithium anode: evaporated from thermal source	10-50µm
Anode stabilizer: evaporated from thermal source	0.1-0.5µm
Polymer electrolyte: extruded by <u>LML</u> process	10-50µm
Polymer composite cathode: extruded by <u>LML</u> process	25-100µm
Nickel current collector: e-beam evaporated	0.5µm
Polyester Substrate	5-15µm

SUMMARY OF ACTIVITIES PERFORMED

Technical Tasks: We tested various polymer and metal foil substrates for suitability to the task along with the deposition process development for the various layers of the battery proper.

We demonstrated that sputtered, or evaporated current collectors of Al, Cu, or Ni could be deposited sufficiently thick ($0.5\ \mu\text{m}$), in a single pass, to work without the heat load destroying a polymer substrate.

We designed, had Moltech build, and installed an enclosure in the web coater to contain the lithium evaporation system and extraneous lithium deposits. We installed the electron beam evaporation system to be used for the lithium deposition in the enclosure in the PML vacuum web coater system (see figure 2 below). We then e-beam evaporated Li at thicknesses up to $75\ \mu\text{m}$ in a single pass at 1 meter per minute line speeds. Again, without the heat load destroying a polymer substrate. The optimal thickness is from $15\ \mu\text{m}$ to $30\ \mu\text{m}$. We installed, and made operational, a chemical glove box to act as a staging area for preparation of lithium evaporation crucibles and storage of lithium coated substrate materials. We also built a thermal evaporation source for the Li layer which has not been tested yet.

FIGURE 2. LAYOUT OF THE PML WEB COATER

We vacuum extruded polymer electrolyte and composite polymer cathode layers. The vacuum extruded electrolyte layers demonstrated better thickness uniformity, surface smoothness, and adhesion to the current collecting layer than Moltech has been able to achieve in air. Comparison with electrolyte films fabricated in bench top operations showed our vacuum deposited films to have identical ionic conductivity. The vacuum extruded cathode layers demonstrated thickness uniformity and surface smoothness similar to what Moltech has been able to achieve in air. However, the adhesion was much better than what Moltech has been able to achieve in air.

We began the design of Moltech's pilot fabrication facility which incorporates the technology of the present project. We then selected a vendor to build the hardware and the hardware is now being built.

Papers and Presentations: Following are a list of technical publications and presentations that deal in part or exclusively with the work of this CRADA.

J.D. Affinito, "Polymer Multilayer Technology Integrates Thin Film Deposition", in B-TIP Technology Spectrum, October 1994, No 58, pg. 1-5.

J.D. Affinito, P.M. Martin, M.E. Gross, and W.D. Bennett, 1994. "Vacuum Deposited Polymer Films: Past, Present, and Future Applications." In: Proceedings of the Eighth International Vacuum Web Coating Conference, November 6-8, 1994, Las Vegas, Nevada.
TALK GIVEN AS WELL

J.D. Affinito, P.M. Martin, M.E. Gross, C.A. Coronado, and E.N. Greenwell. Paper titled "Vacuum Deposited Polymer/Metal Films for Optical Applications", was presented at the International Conference of Metallurgical Coatings and Thin Films in San Diego, Ca., in April 1995. The paper is to appear in an upcoming issue of Thin Solid Films - which issue has not yet been disclosed to the authors. **TALK GIVEN AS WELL**

P.M. Martin, J.D. Affinito, M.E. Gross, C.A. Coronado, W.D. Bennett, and D.C. Stewart, "Multilayer Coatings on Flexible Substrates", Proceedings of the 38th Annual Technical Conference of the Society of Vacuum Coaters, Chicago, Illinois, April 3-8, 1995 **TALK GIVEN AS WELL**

J.D. Affinito, M.E. Gross, P.M. Martin, C.A. Coronado, and G.C. Dunham, "High Rate Vacuum Deposition of Polymer Electrolytes", to appear in the Proceedings of 42nd Annual National Symposium of the American Vacuum Society, 1995. **TALK GIVEN AS WELL**

J.D. Affinito, M.E. Gross, C.A. Coronado, and P.M. Martin, "Vacuum Deposition Of Polymer Electrolytes On Flexible Substrates." Invited to give submit paper and give Plenary talk at the Ninth International Conference on Vacuum Web Coating on 11/13/95 in Tucson, Arizona. **PLENARY TALK GIVEN AS WELL**

As well, through several interviews, and an exchange of literature, we were able to get the March 1995 issue of *FUTURETECH* devoted our technology. *FUTURETECH* is a monthly publication that devotes each issue to one *promising* new technology that is ready for commercialization.

FUTURETECH No. 183, March 1995. Technical Insights, Inc.

Pre-prints of some of the publications are included in the appendix.

SIGNIFICANT ACCOMPLISHMENTS

Technical Accomplishments: We have demonstrated that all 7 of the layers of Moltech's lithium polymer battery are compatible with simultaneous, in-line, vacuum deposition onto a flexible plastic substrate via PNNL's PML and LML technology (see figure 1 above). This includes the first ever demonstration of vacuum deposited thin film polymer electrolyte and polymer cathode layers. We also demonstrated that all of these materials, including lithium, could be deposited in a single pass without melting the substrate (detailed in pre-prints of papers included in appendix). The technology developed in the CRADA received an R&D-100 Award recognizing it as one of the 100 most significant new product developments in 1995.

Business Accomplishments: The technical accomplishments were impressive enough that Moltech renegotiated their license arrangement from a non-exclusive license to an exclusive license. We are also assisting in building the prototype production hardware, a 9-chamber in-line vacuum web coater, for initial battery production at Moltech's Tucson facility.

Follow-on funding to support further development of the battery and its production system has been obtained from various sources external to the Department of Energy.

SIGNIFICANT PROBLEMS

There were only two problems of note. One was the extreme sensitivity of the lithium films to oxidation during deposition and removal from the chamber. This required careful attention to chamber base pressure, protection of the lithium layer in vacuum, chamber venting with inert gas, and immediate removal of the film from the chamber to an argon dry box. The second problem was the extremely high enthalpy of condensation of lithium causing the polyester substrate to burn as the lithium film was deposited. This required cooling the drum to -5 C and adjusting the web speed and deposition power to optimize rate and minimize substrate heating.

DOE/LABORATORY BENEFITS REALIZED

DOE/PNNL have benefited through the development of two additional capabilities (e-beam evaporation and slot die extrusion in the vacuum roll coater) that will supplement the ongoing PNNL vacuum roll coating program. Past ER work at PNNL has developed the capability needed for this work to be undertaken. In turn, this work has benefited our ongoing ER and

CRADA work by thoroughly examining the PML flash evaporation process in a manufacturing application. PNNL has also licensed PNNL patent 5,260,095 for the vacuum extrusion of polymer electrolyte and cathode layers. The success of this work has also generated numerous requests for invited (one plenary) talks at conferences. *We received a Federal Laboratory Consortium (FLC) Award for Technology Transfer for this work and the R&D-100 Award mentioned previously.*

INDUSTRY BENEFITS REALIZED

From this program MOLTECH should realize benefits in at least four very crucial areas: a higher performance product; a more reliable product; a less expensive manufacturing process; and a manufacturing process that needs less initial investment in capital facilities. In particular, it is expected that the PNNL processing technologies will provide better inter-layer adhesion and a more active lithium layer. These improvements should provide improved current carrying and cycle life capability while making the cell much less susceptible to lithium dendrite formation/shorting. As well, since the vacuum deposited layers do not require constant pressure to ensure electrical contact, the battery casing can be much lighter and simpler in design - further reducing cost and increasing power density.

RECOMMENDED FOLLOW-ON WORK

The vacuum extrusion of polymer electrolyte layers should be pursued for the fabrication of electrochromic, switchable, windows and mirrors. PNNL has already developed, in another chamber, the ability to deposit the electrochromic (EC) and transparent conducting (TC) oxides necessary to this task. They were used in a process that vacuum deposited EC/TC/GLASS, then the glass was removed and a proprietary polymer electrolyte (PE) was spread in air on the layered materials. Then the PE/EC/TC/GLASS was returned to the vacuum chamber where we deposited another EC and TC pair to yield TC/EC/PE/EC/TC/GLASS.

We fabricated these devices up to 12 inches by 12 inches in area. The company that participated in these tests abandoned the technology because they did not believe the 3-step (vacuum-air-vacuum) process could make the devices for less than about \$8-\$10 per square foot. We think that, with our vacuum extrusion process, we can get the price to \$2 per square foot. To perform this task we would need to transfer our TC and EC oxide technology into our web coater (mainly buying sputtering targets and evaporation material) and deposit the oxide in-line with our extruded polymer electrolyte. On a flexible substrate, the material could be used in retrofit applications as well as in new construction.

POTENTIAL BENEFITS FROM PURSUING FOLLOW-ON WORK

If inexpensive electrochromic windows and mirrors can be developed they will offer new alternatives to energy conservation and daylighting initiatives for buildings and vehicles. Currently, the only commercial electrochromic devices are switchable mirrors for automotive rear view mirrors. These cost 10's of dollars per square foot for the EC mirror alone. The high price is the reason other applications have not yet taken off. Even in the auto industry, the auto companies would like electrochromic sun roofs but the price-to-performance ratio is too high.

APPENDICES

reprints
and preprints
removed
dsg

To supply more details on the technical aspects of the process technology, the following four pre-prints or publications are included:

J.D. Affinito, "Polymer Multilayer Technology Integrates Thin Film Deposition", in B-TIP Technology Spectrum, October 1994, No. 58, pg. 1-5.

J.D. Affinito, P.M. Martin, M.E. Gross, C.A. Coronado, and E.N. Greenwell. Paper titled "Vacuum Deposited Polymer/Metal Films for Optical Applications."

J.D. Affinito, M.E. Gross, C.A. Coronado, and P.M. Martin, "Vacuum Deposition of Polymer Electrolytes On Flexible Substrates."

Distribution

No. of
Copies

Offsite

Ted Vojnovich
Technical Program Manager
Office of Computational and
Technology Research
Laboratory Technology
Research Division
U.S. Department of Energy
Room G434 (ER-32)
19901 Germantown Road
Germantown, Maryland 20874

No. of
Copies

Onsite

DOE/Richland Operations Office

NL Hieb K8-50

12 Pacific Northwest National
Laboratory

JD Affinito (3) K3-59
BJ Harrer (2) K9-21
Information Release Office (7)