

FINAL REPORT

BA00001-10 (Defense Logistics Agency)

**Advanced Manufacturing Process for Lower cost Rechargeable Lithium-ion
Batteries for DOD Including the BB2590**

Contract #SP4701-10-C-0032

Submitted by

LithChem Energy

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Executive Summary

LithChem Energy (LCE) successfully designed and built a partially automated lithium-ion bi-cell production machine to produce lower cost prismatic lithium-ion batteries for the DOD. This machine was completed and started up to produce a short run of lithium-ion bi-cells with lithium cobaltate as the cathode material. This system included the design and successful fabrication of two dip coaters, a brush tabber for the coated electrodes to form the weldable electrodes, and a coater for priming the electrode metal grids prior to dip coating. Work stations were constructed for quantitative cell filling, resistance welding of the cell pack electrodes, and for cell pack formation. The prismatic cell packs were designed to be 6 Ah with eight of these to fit into the space of a BB2590 to achieve 12 V and 24 V circuits. In a parallel effort, a new high energy capacity cathode material was optimized based on a lithium nickel cobaltate which achieved an energy density of 230 mAh/g in the voltage range of 2.6 V to 3.9 V at C/10. This cathode material also exhibited high power density up to 40 C discharge rates. This advanced manufacturing System and new cathode material have been designed to lower the cost of All-American made rechargeable lithium-ion cells and batteries for the DOD.

1.0 Goal of Program/Problem to Address

The goal of this Program is to provide an All-American Low Cost Manufacturing Process for the production of prismatic rechargeable cells and batteries for the DOD and especially for the War Fighter. Currently most of the DOD lithium-ion rechargeable cells/batteries are composed of combinations using Asian 18650 cells including the BB2590. This dependence is due to the much lower cost of the Asian and particularly the Chinese 18650 cells which are made on very large scale and also with lower labor costs. LithChem Energy (LCE) in its Final Report to DLA from its Phase I DLA SBIR identified the Asian 18650 rechargeable lithium-ion cell as the highest volume battery used by the DOD (DLA). The 18650 lithium-ion cylindrical battery (3.7 V) or cell is the

basic cell for manufacturing most DOD rechargeable lithium batteries such as the BB-2590 (The War Fighter's battery). Due to the low manufacturing cost (\$0.45/Wh), these are all imported from Asia and principally China and then assembled by the welding of connecting tabs into 12 V/24 V strings and inserted into the BB-2590 by assemblers such as Bren-Tronics and Ultralife. No American company has been able to begin approaching this low manufacturing cost/price of these 18650 cells which has resulted in a total dependence on the Asian market for DOD and consumer rechargeable lithium batteries. This particular 18650 battery/cell (3.7 V) is made in enormous volumes for laptop computer batteries and many other applications in a combination of labor intensive and semi-automated steps (e.g., BYD-China). The DOD percentage of sales is only a small fraction of the total consumer market and therefore is of no significant influence on demand or price. However, DLA and DOD desire to have a totally reliable All-American source for this vital manufacturing which currently means to reduce the cost of American manufacturing of these 18650 cells or a replacement cell for it. Further, CERDEC (Army Power Group) not only wishes to reduce the cost of these batteries, but has as its goal to reduce the weight of batteries that the War Fighter carries by 50% (double the energy density) since the battery load is already too heavy. These stated needs - to reduce American battery/cell manufacturing costs further, improve the energy density of these batteries, and to achieve All-American manufacture - cannot be accomplished in the present situation with Chinese and Asian manufacture focused on the enormous consumer market.

To achieve lower battery costs, LithChem Energy (LCE) has developed a different semi-automated cell manufacturing approach which is relatively low in capital investment (minimizing depreciation costs) with high flexibility to accommodate different lithium-ion cell sizes and enable the shorter production runs usually required by DOD to be cost effective. The labor content is reduced to the point that the cost of raw materials (which is essentially internationally equal) is the only significant component of the cost/price. In accomplishing this as an All-American company, LCE would provide longer term security of supply. In addition, the LCE semi-automated process would be used to make larger cells than the 18650, and the larger cell size lowers the fabrication cost per Wh in the fabrication process. For example, the fabrication of a 21 Wh cell/battery

which is readily done in a prismatic cell/battery form versus the current 7 Wh/18650 cylindrical cell/battery will achieve three times the Wh with approximately the same fabrication/manufacturing costs. In the BB-2590 for example, there are twenty-four 18650 cells which have to be connected versus the lower number of eight larger prismatic cells which would be required. In addition changing from a cylindrical cell to a prismatic cell will achieve a desirable increase in volumetric energy density by 21%.

The LCE automated cell/battery manufacturing process utilizes the same proven principles that continuous film processing of photographs used 20 years ago (Joe Kejha, TPOC, designed and built these machines). A roll of coated anode electrode and a roll of coated cathode electrode material along with a roll of separator film which acts as the carrier band are continuously fed into the assembly machine. The individual electrodes are cut and indexed and heat welded to the separator which is the continuous carrier through the machine. The resulting "string" of cells are Z-folded and cut to length to form the basic 3.7 V bi-cell, and then inserted into a hard prismatic aluminum case (case interior is plastic lined) with electrolyte and sealed. These cell packs are then run through formation, vacuum sealed, and then connected in parallel and series as required to form 12 V/24 V. In this completed program, LCE demonstrated this unique process and successfully tested the initial production run at the completion of the program. This rechargeable prismatic lithium-ion production line also utilized some production units already in place from another LCE production line for the initial demonstration in order to keep the program in budget.

Eight of the resulting prismatic 21 Wh, 3.7 V (nominal) cells/batteries can be inserted into each BB-2590 case to demonstrate that this low cost manufacturing approach will compete effectively with large scale low cost Chinese manufactured batteries.

However, now that the conformal battery approach is being pursued by DOD and especially the Army for the War Fighter, these production bi-cells packs can be configured to fit into a belt or vest and connected in series to achieve the desired battery voltage configuration. LCE can produce batteries on a limited run basis to demonstrate that the manufacturing labor cost per Wh for these new batteries produced by this new automated process is less than \$0.12/Wh and lower than the estimated labor cost for

Asian batteries when additional funding is available. The overall cost of these batteries with the reduction in labor cost and packaging costs (larger batteries) can provide an All-American battery which is also lower cost than the Asian sourced batteries used by DOD. These new cells/batteries will be incorporated into the new battery configurations proposed by and being developed by CERDEC and the DOD including the BB-2590 by working with the current assemblers of these batteries to change over to the eight lower cost LCE 3.7 V, 23 Wh prismatic cells. The replacement of the 24 cylindrical cells by the eight larger prismatic cells increases the volumetric energy density by 20% and offers approximately 10% gain in weight saving per Wh due to less packaging. (Note that higher capacity - more Wh per kg - will reduce the cost per Wh even further.)

The establishment of LCE as a low cost All-American manufacturer of lithium-ion cells/batteries based on this flexible automated manufacturing process for DOD will facilitate the introduction of new maximum energy density materials more readily. The large Chinese battery manufacturer is not interested in modifying its current production for the consumer to accommodate the interests of DOD/DLA/the US soldier. LCE has already developed a new cathode material which will achieve 40-50% improvement in energy density, or 1/3 reduction in the number of batteries required (reduction in battery weight). This can now be introduced when additional funding is available for the production runs for new cell designs to meet these new developments of DOD and CERDEC and the War Fighter (dismounted soldier). The goal of a manufacturing cost of less than \$0.50/Wh appears achievable with this new semi-automated cell/battery production process, the larger cell size and with the new higher capacity cathode material developed by LCE.

2.0 Technical Approach and Objectives

LCE combined three weight and cost reduction approaches/factors to make the lower cost All-American basic lithium-ion cell battery for the War Fighter (dismounted soldier). These factors were:

- 1) LCE designed and built an automated bi-cell production machine for prismatic lithium-ion cells which produces larger cells as bi-cells (eliminating the weight and cost of one carbon copper electrode). This reduces the labor cost per Wh through automation and by increasing the achievable cell size to >6Ah.
- 2) The prismatic cell design was changed to a bi-cell while also eliminating the cost and weight of half of the copper anodes per cell. The bi-cells are necessary to achieve high energy density by sharing the heavy copper current collectors of the anodes, in the middle of the bi-cells.
- 3) In parallel, a lithium nickel cobaltate mixed oxide cathode material was developed which achieved a capacity over 200mAh/g. This material can be made by LCE in house, leveraging off other production equipment already in place.

These factors in combination can achieve the goal of <\$ 0.50/Wh for an All-American made cell battery for the War Fighter (DOD) and DLA which can be made in limited production runs compared to the Asian commercial production.

To aid in the completion of this program within budget, LCE leveraged some of its production capability from existing low cost manufacturing machines from other programs (MDA missile battery) for prismatic primary cells manufacture, and to add new low cost machines to produce low cost lithium-ion rechargeable prismatic bi-cells.

Automated Bi-Cell Production

The LCE approach to produce low cost and high energy density cells and batteries was to use an automated continuous production process to form prismatic bi-cells. The larger the cell size the better, the elimination of half of the copper anodes is an advantage, as well as incorporating a maximum energy density cathode material, i.e., lithium nickelate.

Specifically the LCE production approach to making automated consistent and uniform thickness electrodes of any width, is to utilize the following basic production steps based on continuous rolls of metal grid electrodes cut to the desired width. **Figure 1** is a schematic of the overall production process for this low cost lithium-ion bi-cell program.

Process Steps:

Step 1) Precoat the metal (cathode or anode) grid continuously with a carbon based primer using a dip precoater and curing; **Figure 2a** shows the schematic of the primer coater which was designed and built. **Figure 2b** shows the actual functioning unit.

Step 2) Dip coat the primed coated cathode (aluminum) or anode (copper) grid metal electrode roll continuously through a cathode or carbon anode slurry mix (containing conductive carbon and binder), and then the acetone solvent is continuously heated and evaporated and the coated grid rewound to achieve a coated cathode or anode. By maintaining constant viscosity of the slurry by pump mixing, and constant but adjustable web speed, the uniform thickness is achieved on both sides of the web at once, and the dried coated continuous electrode is wound onto a hub or spool. See **Figure 3a** for the schematic of the coater. **Figure 3b** is the picture of the actual functioning unit (two of these for the program).

Step 3) The coated electrode grid (cathode or anode) is run through a brush tabbing machine, removing approximately 1" of the coating from the edge of the continuous coated metal grid to form a tabbed (bare metal grid) edge for cell tab welding. See **Figure 4a** for the tabber schematic. **Figure 4b** is the picture of the actual functioning unit (two of these for the program). All rolls of finished electrodes are vacuum dried with heat overnight and then brought into the dry room before being opened.

Step 4) A roll of the completed cathode and a roll of the completed anode are each fed simultaneously into the Single Cell Assembly Machine, cut into leaves (individual electrodes), adhesively (ethylene carbonate) laminated to the first continuous separator and indexed with exact spacing between the leaves. This results in a string of single cells with the cathodes and anodes all bonded to the separator which is between them. This string of cells is wound onto a spool with a slip clutch. **Figure 5a** shows the schematic of the Single Cell Assembly Machine. **Figure 5b** is the picture of the actual functioning unit.

Step 5) The Single Cells from the spool from the Single Cell Assembly Machine are then fed into the Bi-Cell Assembly Machine, where the second continuous separator roll

is fed and added on top of the anodes of the single cells, and the second cathode roll is fed and is cut into cathode leaves and added onto the second separator. Both are laminated using ethylene carbonate as the adhesive in the laminator to the single cells in an aligned and synchronized manner. This results in a string of bi-cells which are wound onto another spool. See **Figure 6a** for the schematic of the Bi-Cell Assembly Machine. **Figure 6b** is the picture of the actual functioning unit.

Step 6) For the automated bi-cell production system, LCE had to be certain that in the bi-cell machine which is fed by the single cell machine, the second cathodes of the bi-cells are properly cut and aligned with the anodes of the single cells, bonded (ethylene carbonate, an electrolyte component, is used as the adhesive) to the second separator, and the second separator bonded to the anodes. This precise automated process is controlled by a PLC unit and required considerable adjustment time and machine trials.

Step 7) The bi-cells are then unwound from the spool onto a table by a drive mechanism controlled by operator's foot switch.

Step 8) The desired quantity of bi-cells is then cut-off in the gap, and folded in the gaps, or flat wound into a cell pack, and the cathode and anode tabs, respectively, are welded together by a Resistance Welder. These two thicker gathered welded tabs are in turn welded to two thicker tabs which will later exit from a pouch. The cell packs are then inserted into aluminum pouch folders and filled with electrolyte with the Electrolyte Filling Machine. The cells are sealed temporarily and activated under vacuum, in the Activation Glove Box under argon atmosphere.

Step 9) The cell packs are then formatted by charge/discharge cycled on a Multi-station Battery Tester. These formatted cells are returned into the Re-Seal Glove Box, opened and vacuum degassed again and vacuum resealed on the Vacuum Packing Machine.

Step 10) The finished packs are removed from the Re-Seal Glove Box, stacked and connected in series for the desired voltage by the Resistance Seam Welder. The welded, full width terminal tabs of the packs are covered by insulating Kapton adhesive tapes. Each series pack stack is inserted into a hard aluminum sheet metal housing with

two final high voltage insulated, exiting battery terminal wires which are resistance welded to the first and last pack tabs.

Step 11) The last assembly step before closing the housing is the filling of the empty spaces in the housing around the tabs with a flexible silicone rubber resin to protect the battery from shocks and vibration. Two half shells of the housing in tight contact with the battery stack are then locked together by an adhesive or by a laser welding.

Each cell pack also has its thin voltage monitoring wires exiting from the housing to be connected to a BMS unit and a Charger.

The objective of the program is to use minimum touch labor, which is accomplished up to and including the unwinding of the bi-cells. From that point and up to the formation cycling, the cell packs handling can be done by a Robotic Station with one arm and holders, under argon in a Large Glove Box or dry room. This robotic step will be added later under a new program when available which is requested in the recommendations.

All of the previously described machines cost approximately 10% of the cost of competitive automated manufacturing processes for lithium-ion batteries.

LCE Very High Capacity Cathode Material for the Low Cost Rechargeable Battery

LCE already had been working on its own process and testing for lithium mixed metal oxide cathodes and focused on the lithium nickelate system, particularly due to the higher energy density of the lithium nickelate cathode materials. This was a successful development when a limited amount of cobalt (10%) was included to improve the conductivity of the lithium nickelate. This high capacity lithium cathode material achieves >200 mAh/g from 2.6 V to 3.9 V at C/5. **Figure 7** is a Ragone chart for an 18650 cell made with the LCE cathode material.

LCE took the following technical approach to produce high capacity cathode material based on its already developed lithium nickelate. A series of runs were made with varying amounts of cobalt oxide added to the lithium nickelate mixture. The unique preparation process which LCE uses is the reaction of lithium hydroxide with the nickel hydroxide and cobalt hydroxide in a stoichiometric ratio in a minimum amount of water.

The reaction mixture is stirred and heated to boiling and the mixture is fully reacted. The water is removed with heat and vacuum and the powder screened and then furnace, reground, and screened again. The resulting mixture lithium nickel cobaltate has a relatively higher surface area ($2.5 \text{ m}^2/\text{g}$) giving it a high power density along with high energy capacity with low impedance.

The objective for the high capacity cathode material was to increase the energy density of the battery, thus reducing the cost of the battery. For example, if the same weight of battery materials has twice the energy stored, cost per Wh is 50% less for the battery.

3.0 Program Accomplishments

The main accomplishment of this program was the successful design and fabrication of all the described unique, low cost automation machines, and to make them all work and function properly which resulted in lower cost pilot production of prismatic lithium-ion bi-cells. This proved that this new All-American manufacturing process is sound and can be used for lower cost production of high energy density prismatic rechargeable batteries for the military applications.

After the initial assemblies and tests, all machines had to be modified and improved. The most challenging was to make the Bi-Cell Assembly Machine function properly, where the second cathodes must be cut, aligned, and welded to the moving single cells through the laminator. This was successfully accomplished by installing a special sensor, and a cutter metering system with programming of the computer controller.

Another accomplishment was the development of high capacity lithium cobalt nickelate cathode which increased the energy density by 50% above the standard lithium cobaltate, and thus it also lowered the cost per Wh in the cells. This in combination with the described cell design and production methods, the American cells could be made at less than \$0.50/Wh cost which would make them competitive with Asian cells, and thus free our military from dependency on foreign suppliers.

4.0 Program Tasks

The Tasks for this Program are listed with respect to completion and accomplishment.

TASK 1: Design, fabricate components, assemble, and test (2) Anode Coaters and (1) Anode Tab Cleaner Machine to produce Anode Rolls having copper grid collector web with graphite coating for Automated Cell Assembly Machines

This was accomplished and photos of these completed units are shown in **Figures 2b, 3b, and 4b**. The corresponding similar cathode coating machines were already in place from another program and were used for the production of the cathode rolls.

TASK 2: Modify design, manufacture components, assemble, and test one Cell Assembly Machine for Single Cells and one Assembly Machine for Bi-Cells

Both of these machines were designed, fabricated and tested with the single cell machine feeding into the bi-cell machine for total automated prismatic lithium-ion bi-cell assembly and tested with lithium cobaltate cathode material. These are shown in the pictures of **Figures 5b and 6b**.

TASK 3: Acquire, install and prove out resistance welder for tab welding bi-cell packs

This was successfully accomplished with the resistance welding being successful for these bi-cell cell packs. The welder set up is shown in **Figure 8**.

TASK 4: Manufacture components, assemble and test (1) Bi-Cell Vacuum Dryer with Activation Glove Box

This was successfully accomplished. **Figure 9** shows this installation.

TASK 5: Manufacture components, assemble and test (1) Semiautomatic Cell Electrolyte Filling Machine

This was successfully accomplished. **Figure 10** shows this unit.

TASK 6: Purchase one Heat Sealer for sealing pouch cell packs after activation, and an Additional Vacuum Sealing Machine with Glove Box for reseal of the cell packs after degassing

This was successfully accomplished. **Figure 11** shows this installation. The seam welder was already in place from another program for welding full width bi-cell tabs to complete the cell pack. The LCE Maccor bi-cell testing equipment also was in place.

TASK 7: Test, debug, and improve individually all new machines for bi-cell production

This was successfully accomplished. This included improving the clutch and brake assemblies from the original design and installation to make them more robust. **Figure 12** shows an example of these new upgraded parts installations which were required.

TASK 8: Run Preproduction of bi-cell packs through all the steps, using standard cobaltate cathode and graphite anode materials

This was successfully accomplished. Test voltage and capacity diagrams for Initial Bi-Cells is shown in **Figure 13**.

TASK 9: In parallel with the above tasks, develop high capacity lithium cobalt nickelate cathode material with over 200 mAh/g capacity

This was successfully accomplished in-house at the 3 kg level using equipment already in place at LCE. **Figure 9** shows the Ragone of this material in an 18650 cell.

TASK 10: Run Production bi-cell packs and assemble conformal 12 V/24 V battery battery to fit into the volumetric space of a BB2590 or any other War Fighter battery using the LCE Lithium Cobalt Nickelate

This Task was not completed due to lack of additional funding.

Task 11: Submit the Final Report

This task has been completed with this submission.

5.0 Developments and Results

LCE leveraged its existing technology for prismatic primary cells to make rechargeable lithium-ion prismatic bi-cells and batteries by designing, fabricating and debugging through startup anode coaters and automated single and bi-cell assembly machines. This was in conjunction of utilizing a cathode coater and several other pieces of in-place equipment to actually demonstrate the continuous automated route to making prismatic lithium-ion bi-cells within the funding for this program.

LCE designed and built two different Cell Assembly Machines: 1) Single prismatic Cell Assembly Machine, and 2) Bi-Cell Assembly Machine. The rolls of coated anode carbon on copper grid, cathode coated on aluminum with separator rolls were continuously fed into the machines. The electrodes were cut and adhesively laminated with ethylene carbonate to continuous separator carriers in the laminators as is described in the Technical Approach section.

We have found that the separators (carriers) are not strong enough to be pulled and wound onto the take-up spool, so we added (2) PET films carriers with their feed and take-ups into both machines to take the tension loads. The exit of the completed bi-cells was then changed to winding onto the take-up spool. The continuous roll of bi-cells connected by the continuous separator was then later unwound onto a table by a specially designed un-winding drive mechanism controlled by the operator's foot switch.

The required length with a desired quantity of the cells is then un-wound, cut-off and flat rolled into a bi-cell cell pack and the bi-cell tabs are resistance welded together to (2) exiting main tabs.

The bi-cell cell packs are vacuum dried and then inserted into aluminum pouches in an argon glove box, then activated with electrolyte by the LCE designed and built filling machine, heat sealed and formation tested. The cell packs after formation and testing are degassed, resealed under vacuum and packaged in another glove box. The packs are then seam-welded by their terminal tabs in series for the desired voltage and stacked into hard aluminum cases. Two large custom glove boxes were also designed and built by LCE for vacuum drying the cells activation and reseal.

Combining the machinery from the MDA primary line with DLA Li-ion machines has created a full production, automated line for Li-ion cells, and demonstrated this manufacturing process. All of the described new machines in combination with the primary prismatic line machines are now working and functioning as intended and designed. First production bi-cell packs of “generic” bi-cell design for testing only have been produced by this new automation process.

LCE has made several improvements on the Single Cells Assembly Machine and the Bi-Cell Assembly Machine by adding three electric clutches with brakes in each, and also changed the programming. These changes make it possible to produce large format (long) cells from 12” to 20” long x 6” wide, and any cells with improved accuracy of alignment of the electrodes at higher speeds. These latest changes have been tested with paper “electrodes” first, and then with real electrodes, which had been cut and aligned and welded successfully the same way.

In parallel, we developed a new lithium cobalt nickelate cathode material with up to 220 mAh/g capacity using capacity test conditions from 2.6 V to 3.9 V at C/5. This material also has very high power density and will be used in this War Fighter battery. LCE production capability for this cathode material has been increased to approximately 20 mT/yr.

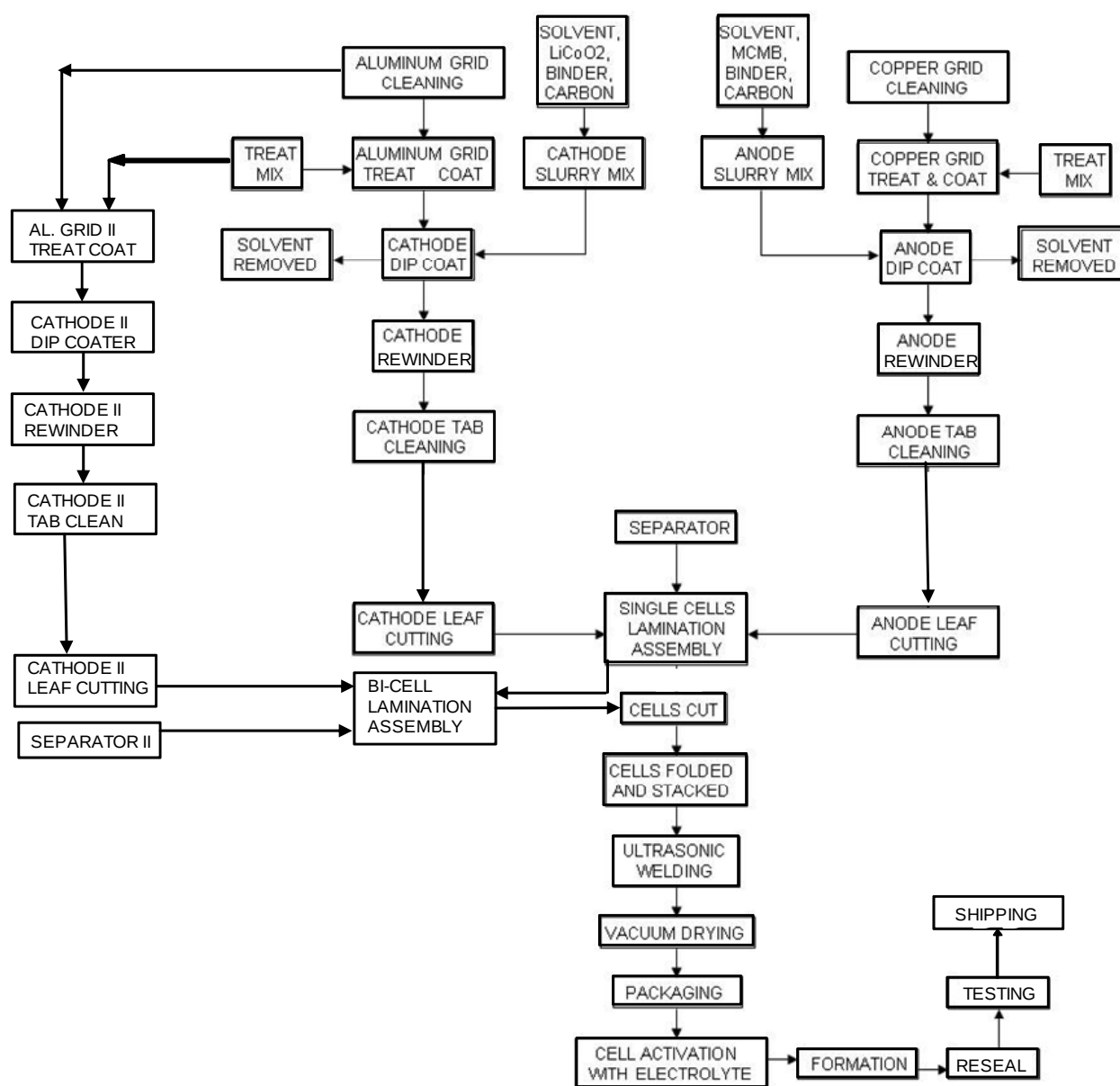
6.0 Conclusions and Recommendations

LCE has demonstrated its novel, low cost manufacturing process for mass production of prismatic, high energy density, rechargeable lithium-ion bi-cells/batteries. In addition, LCE has demonstrated a new high capacity cathode material for these batteries and related lithium batteries.

This bi-cell production process works and can make possible a substantial reduction in the cost of military batteries. However, more cells and batteries have to be produced to prove that it is a rugged and reliable system. Additional robotic automation of the cells packaging has to be built to increase the speed of the total through-put and additional equipment needed to make this line completely stand alone.

LCE will need more funding to accomplish this for these batteries and large format prismatic bi-cells and recommends additional funding to accomplish this.

The result will be low cost all-American manufactured military batteries and dual use batteries for DOD electric vehicles.



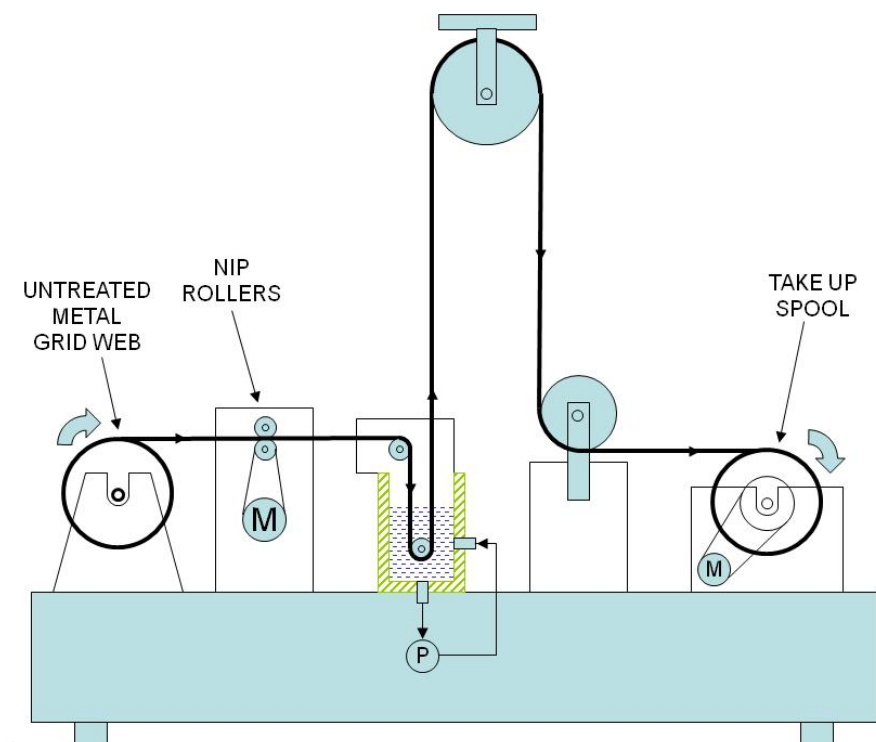


FIGURE 2A: SCHEMATIC OF PRIMER COATER



FIGURE 2B: PHOTO OF PRIMER COATER

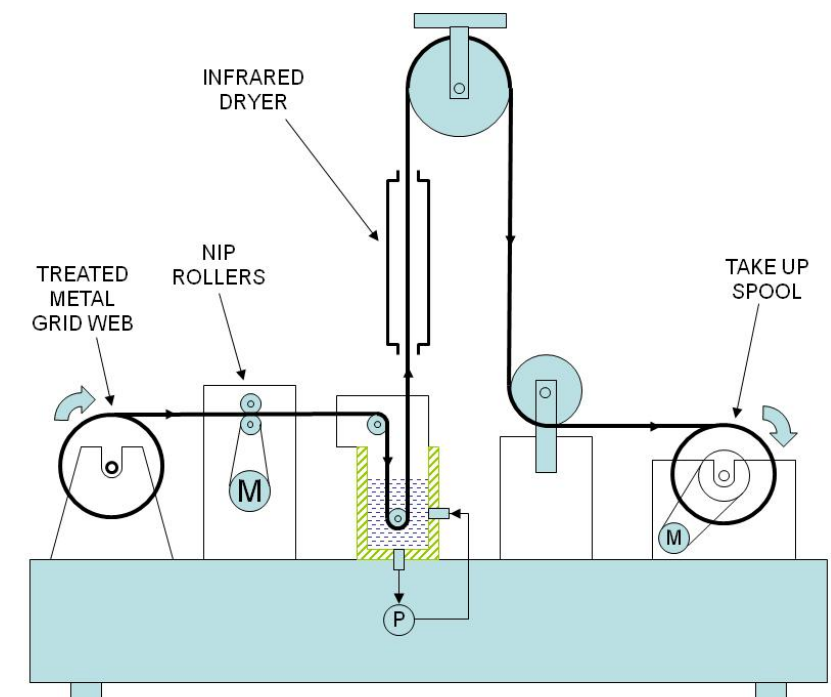


FIGURE 3A: SCHEMATIC OF ANODE COATER



FIGURE 3B: PHOTO OF ANODE COATER

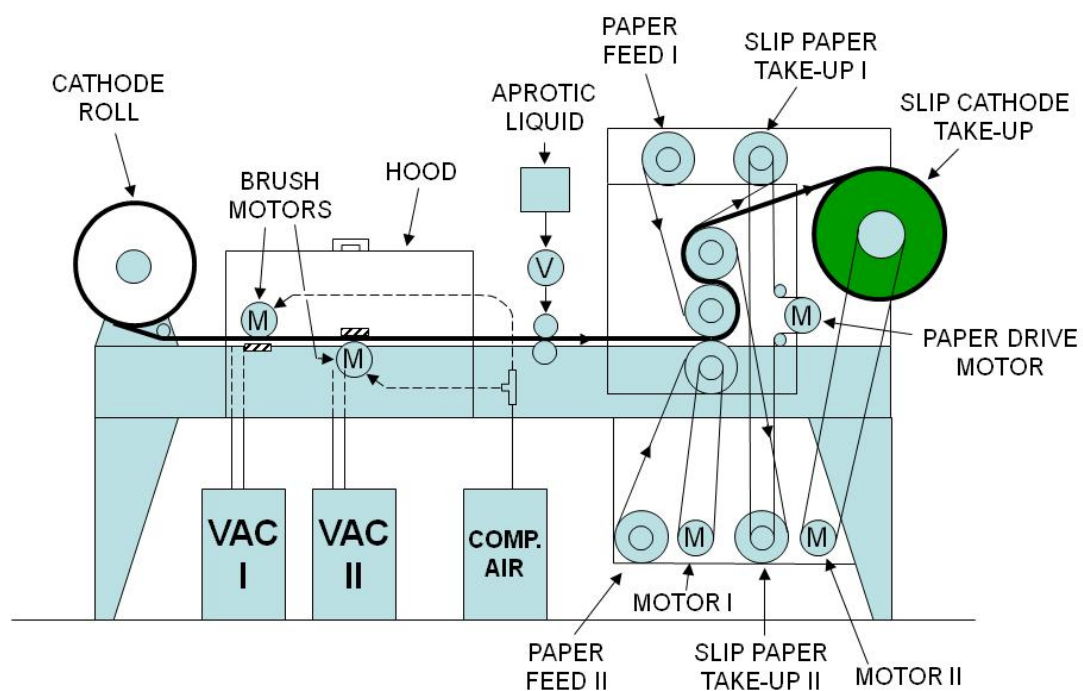


FIGURE 4A: SCHEMATIC OF TAB CLEANER



FIGURE 4B: PHOTO OF TAB CLEANER

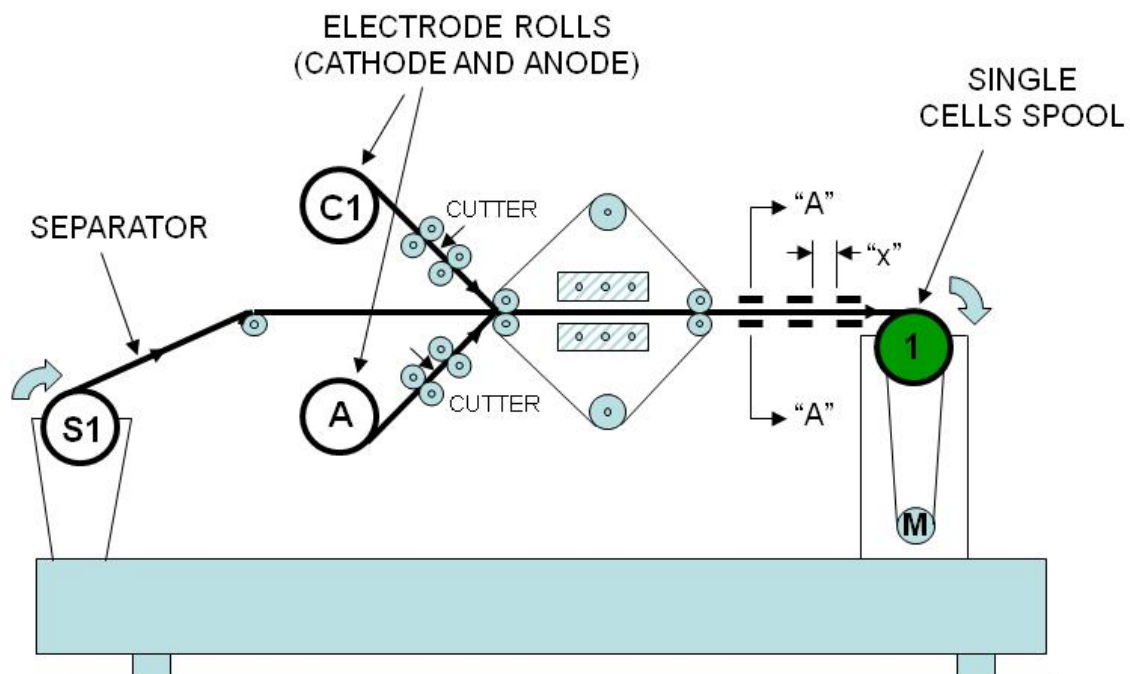


FIGURE 5A: SCHEMATIC OF SINGLE CELL ASSEMBLY MACHINE



FIGURE 5B: PHOTO OF SINGLE CELL ASSEMBLY MACHINE

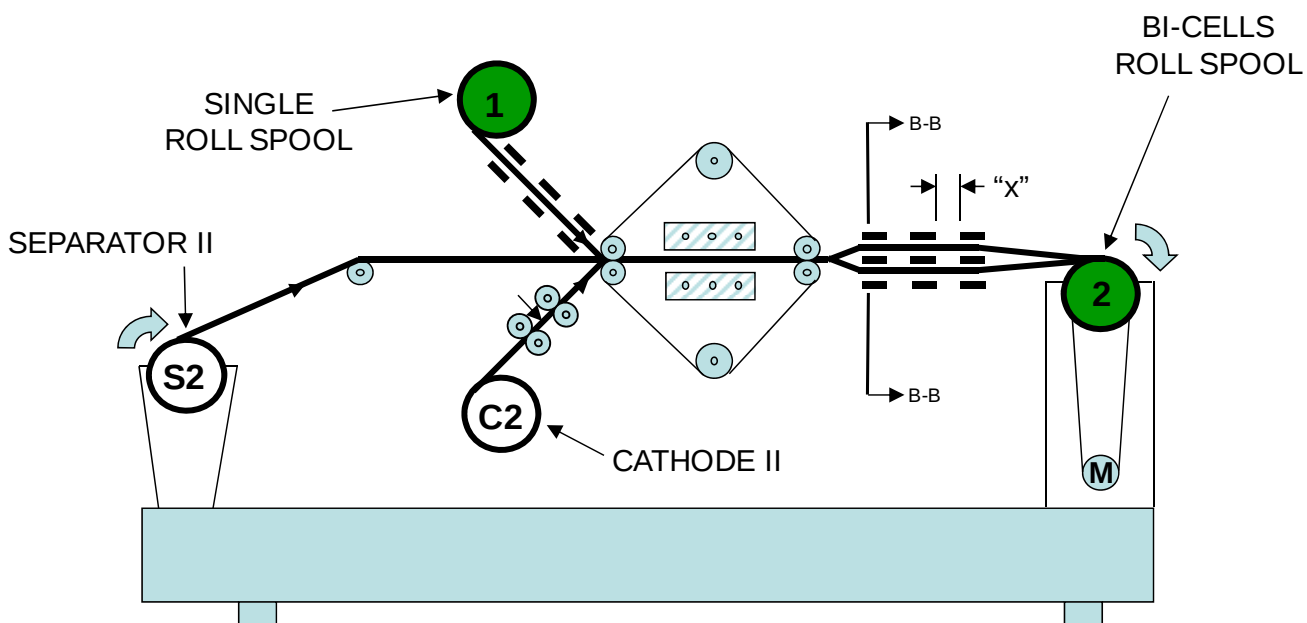


FIGURE 6A: SCHEMATIC OF BI-CELL ASSEMBLY MACHINE



FIGURE 6B: SCHEMATIC OF BI-CELL ASSEMBLY MACHINE

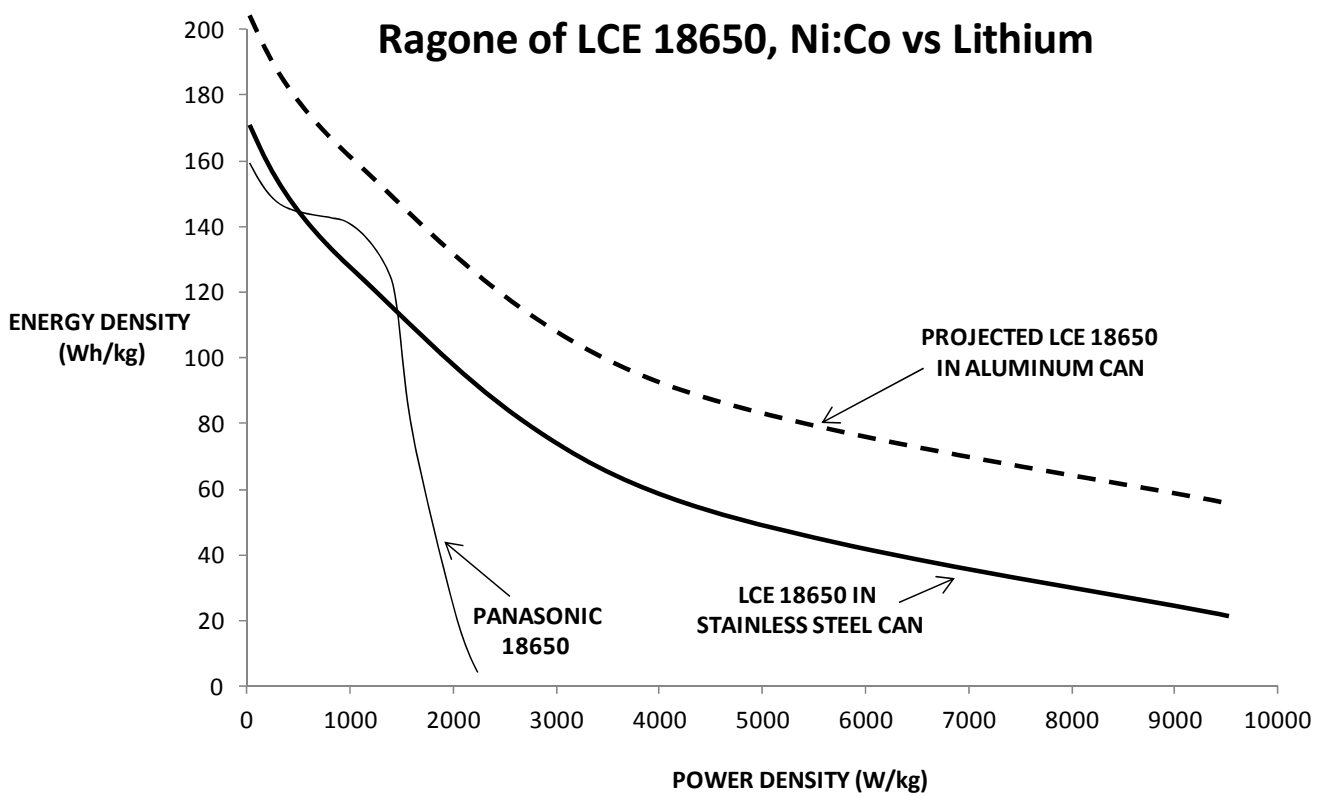


FIGURE 7: RAGONE OF LCE LITHIUM COBALT NICKELATE IN 18650 CELL

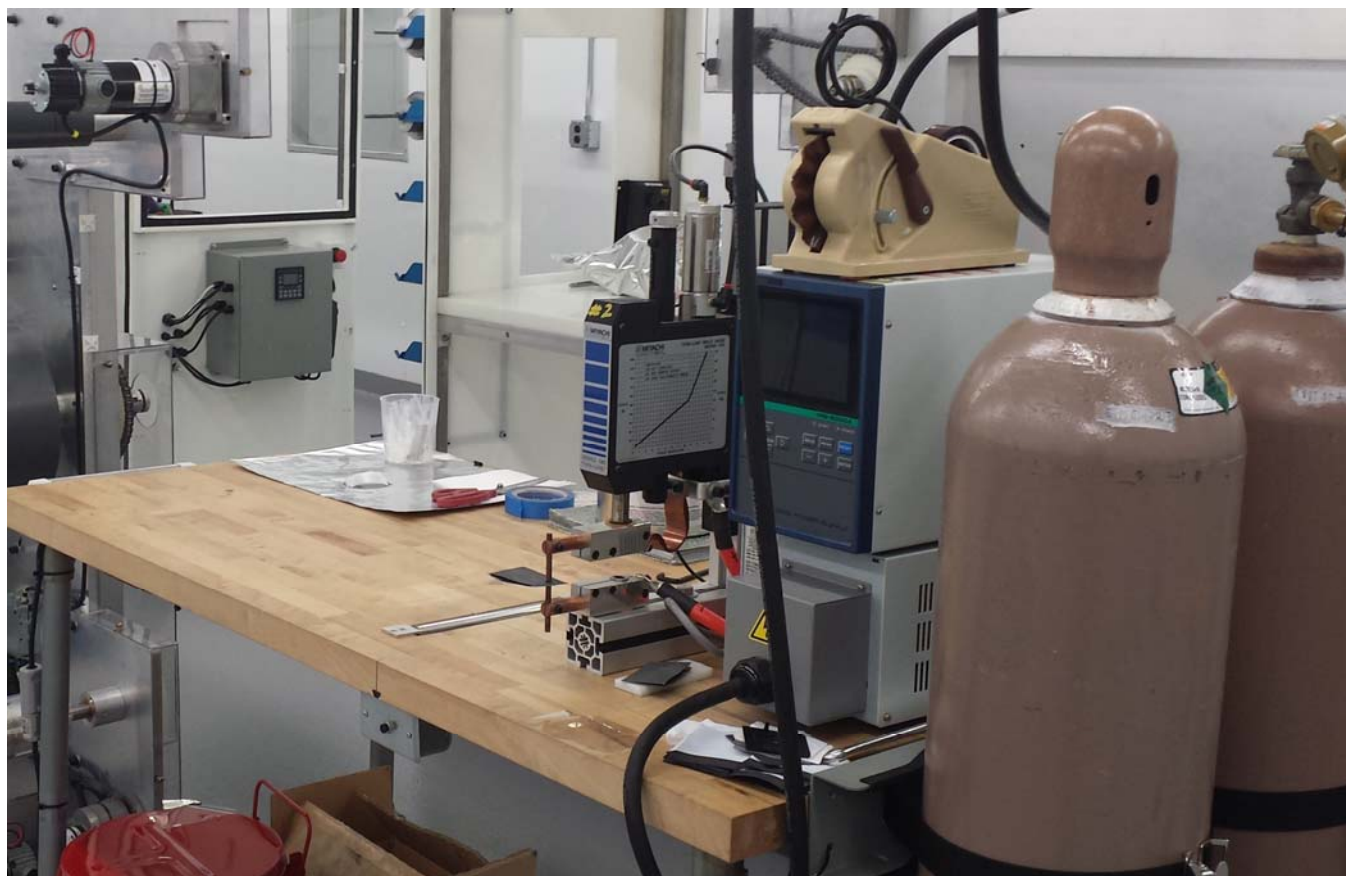


FIGURE 8: RESISTANCE WELDER

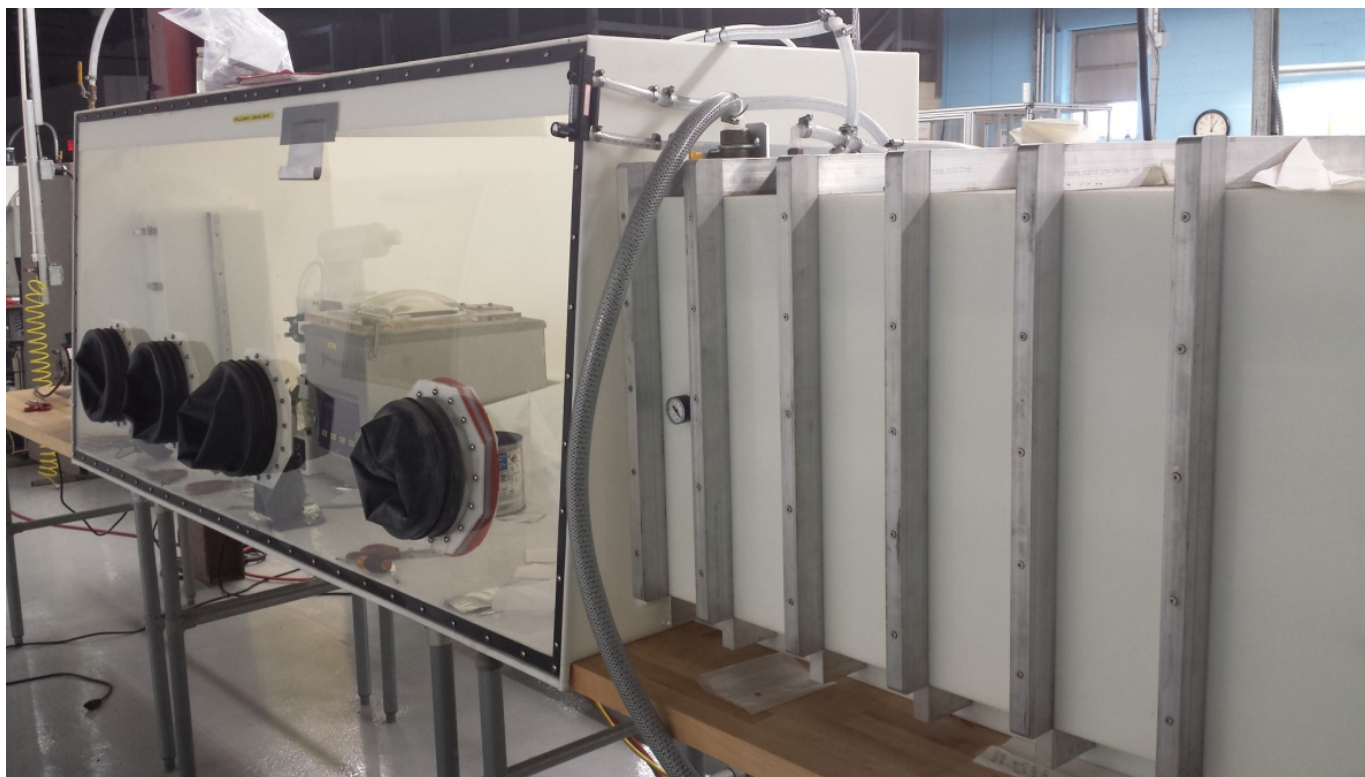


FIGURE 9: BI-CELL VACUUM DRYER WITH ACTIVATION BOX



FIGURE 10: SEMI-AUTOMATIC ELECTROLYTE FILLING MACHINE



FIGURE 11: VACUUM HEAT SEALER FOR VACUUM RESEALING OF POUCHES

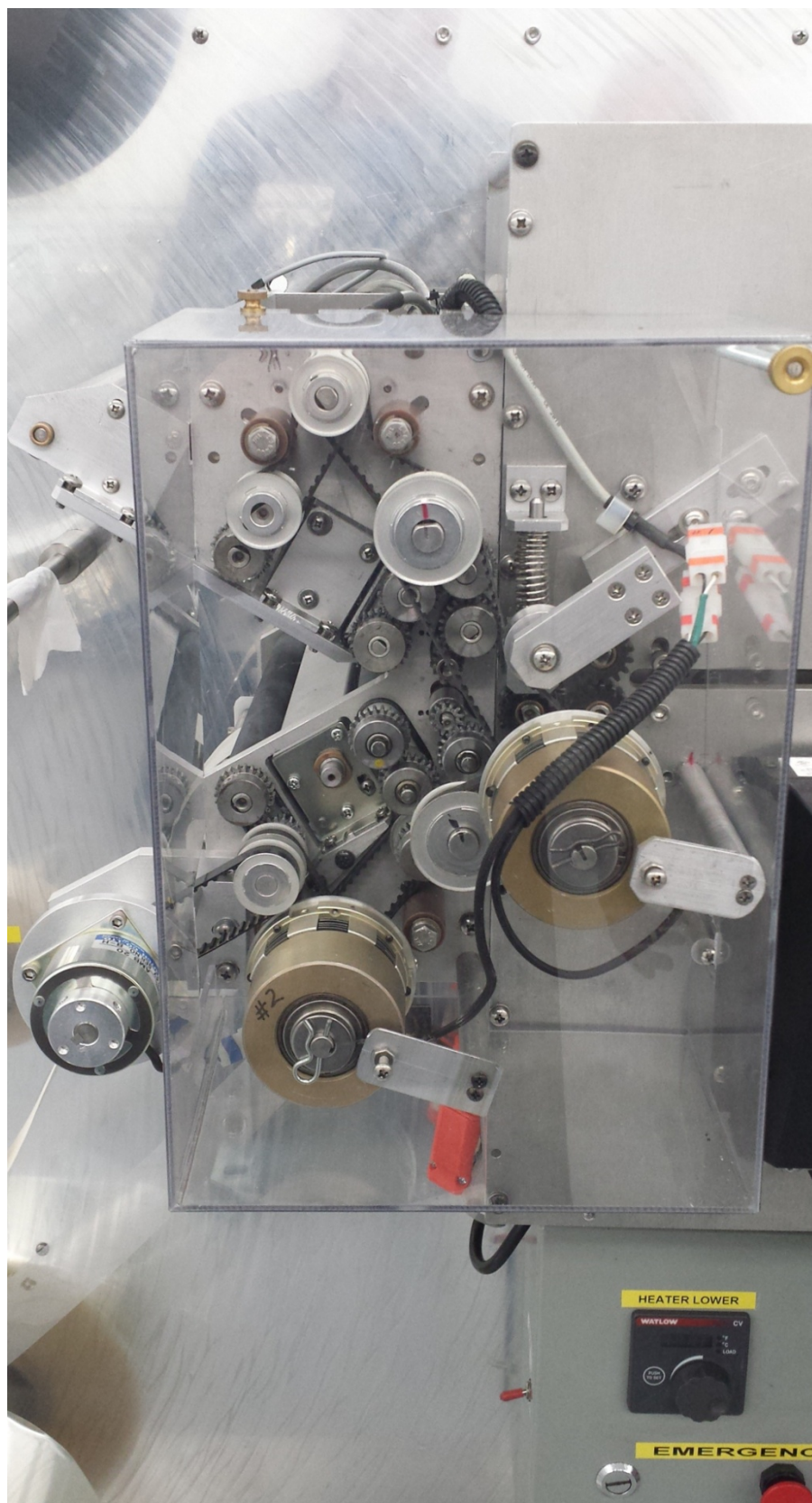


FIGURE 12: ADDITIONAL PARTS NEEDED TO UPGRADE THE BI-CELL ASSEMBLY

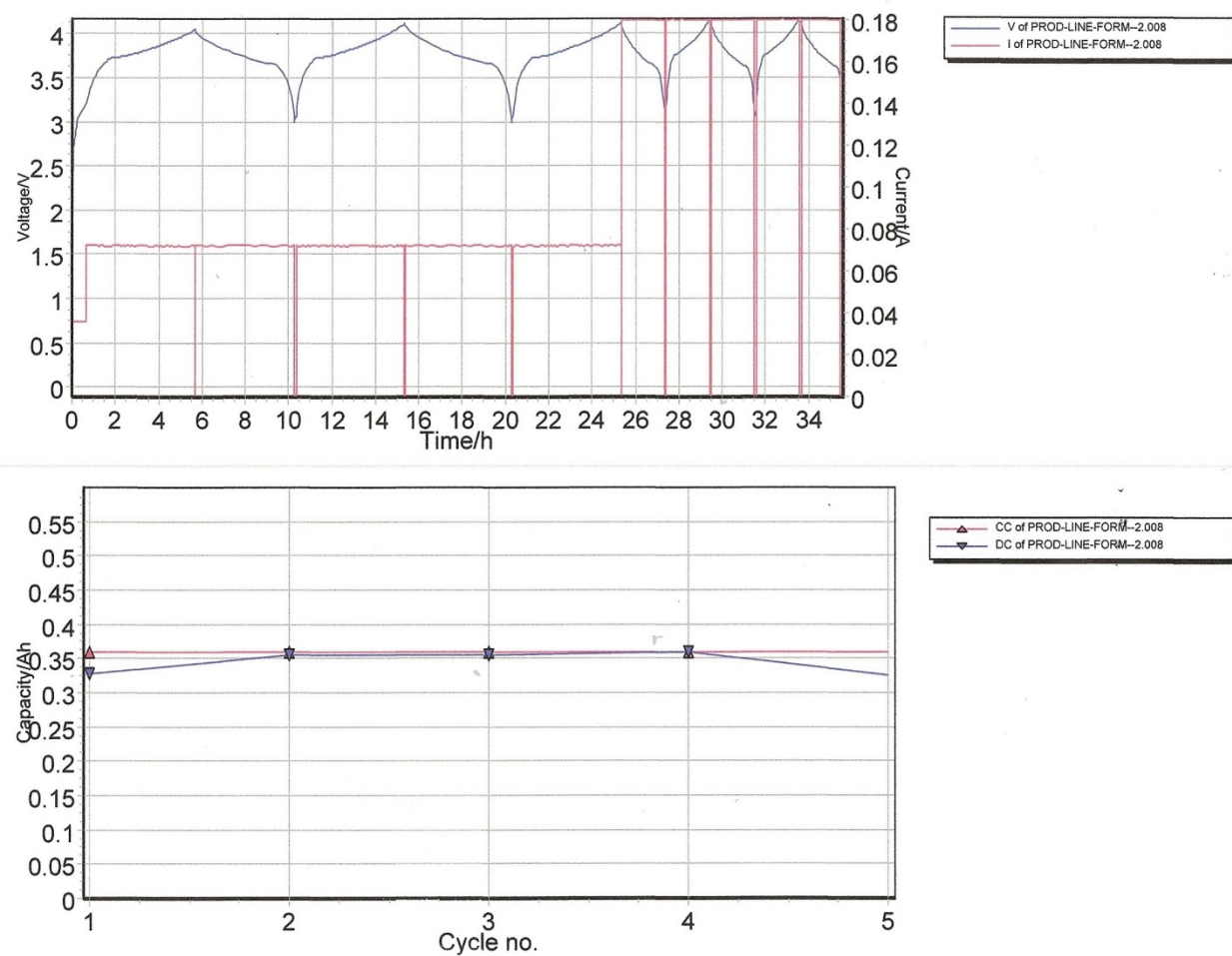


FIGURE 13: TEST RESULTS OF INITIAL TEST BI-CELLS ON PREPRODUCTION