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Review

A review of electrochemical cells and liquid metal battery (LMB) parameter development

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Abstract: Liquid Metal Battery (LMB) technology is a new research area born from a different economic and political climate that has the ability to address the deficiencies of a society where electrical energy storage alternative are lacking. The United States government has begun to fund scholarly research work at its top industrial and national laboratories. This was to develop liquid metal battery cells for energy storage solutions. This research was encouraged during the Cold War battle for scientific superiority. Intensive research then drifted towards high energy rechargeable batteries, which work better for automobiles and other applications. Intensive research has been carried out on the development of electrochemical rechargeable all-liquid energy storage batteries. The recent request for green energy transfer and storage for various applications, ranging from small-scale to large-scale power storage, has increased energy storage advancements and explorations. The criteria of high energy density, low cost, and extensive energy storage provision have been met through lithium-ion batteries, sodium-ion batteries, and Liquid Metal Battery development. The objective of this research is to establish that liquid metal battery technology could provide research concepts that give projections of the probable electrode metals that could be harnessed for LMB development. Thus, at the end of this research, it was discovered that the parameter estimation of the Li//Cd-Sb combination is most viable for LMB production when compared with Li//Cd-Bi, Li-Bi, and Li-Cd constituents. This unique constituent of the LMB parameter estimation would yield a better outcome for LMB development.

Keywords: LMB; SOC; terminal voltage; resistance; capacitance; time; charge; open circuit voltage

1. Introduction

Liquid metal batteries (LMBs) have the ability to store energy coming from generating, transmitting, and distribution stations. The storage capability dynamics of the LMBs would greatly enhance their efficiency and reliability, particularly when intermittent renewable energy technologies (such as wind and solar) are incorporated [1]. For a long time, batteries have been considered a strong alternative remedy owing to their advantages of easy handling, simplicity in maintenance, and flexibility in sitting [2]. The twenty-first century technological advancement has shown the feasibility of developing high-capacity energy storage devices out of many liquid combinations of alkali or alkaline-earth metals as well as metalloids [2]. Though the major challenge to widespread acceptance of battery technology is its high cost of production. Hence, the description of a Lithium–Antimony-Cadmium (Li//Sb-Cd) liquid metal battery could meet the performance requirements for stationary energy storage [2,3]. The LMB comprises liquid lithium as the negative electrode, a molten salt electrolyte, Lithium Chloride-Lithium Fluoride-Lithium Iodide (LiCl-LiF-LiI) fusion, and an antimony-cadmium alloy as the positive electrode, which would separate on its own accord by density into three distinct layers. This is due to the immiscibility property of the electrolyte and the electrodes inter-phase. The liquid characteristics of the constituents of the LMB bring to mind the merits of high current density and prolong cycle life or life span over conventional battery technology [2,4].

2. Brief review on liquid metal battery (LMB)

The advancement of LMB innovation has been re-visited by a group of research pathfinders since 2006 at the Massachusetts Institute of Technology (MIT), United States of America. The recent MIT investigations were conducted as a result of the work carried out at Argonne National Laboratory in the 1960s. The study began with Sodium-Bismuth (Na-Bi), but after a short while, attention quickly shifted towards Magnesium-Antimony (Mg-Sb) and Lithium-Lead-Antimony (Li-Pb-Sb) candidates [2]. For alkali and alkaline-earth liquid metal electrodes, their solubility in their halides increases with the halide atomic number in the order of Fluoride < Chloride < Bromide < Iodide (F < Cl < Br < I). Hence, the alkali and alkaline-earth liquid metals and sodium-based systems have the main challenge of their high solubility in liquid electrolytes [5]. The electronic conductivity of the sodium system is high, and it experiences a high self-discharge current. The improvement on sodium-based systems would necessarily require the development of an electrolyte that would minimize sodium's solubility challenge [5]. The United States government began to fund scholarly research work at its top industrial and national laboratories. This was to develop an all-liquid cell for energy storage solutions. This research was encouraged during the Cold War battle for scientific superiority. Intensive research then drifted towards high energy rechargeable batteries, which are better suited for automobiles and other applications [7]. The motivation was due to the incessant power outages around the globe at that time. For liquid metal battery applications, an elevated operating temperature is detrimental because of its tendency to corrode, mostly for alkaline or alkaline earth metals. The major drawback of the Mg-Sb candidate is that it has a low cell voltage of 0.21 V. This made the Mg-Sb system impracticable. In Mg-Sb collaborations, NaCl-KCl-MgCl2 electrolyte was examined at a very high temperature, producing a high charge efficiency, a deceased voltage efficiency, and a fairly low electrolyte conductivity of 0.8 S cm^{-1} [4]. The cell performance characteristics of the Li-Sb LMB was 1.9 Ah theoretical capacity cell, built-in with a Li electrode, a Sb-Pb electrode and a LiF-LiCl-LiI electrolyte [7]. A potential of 1.2 V was measured for Li-Sb LMB, which is relatively low, and a self-discharge current of 0.6 $mAcm^{-2}$ was significantly lower than 20 $mAcm^{-2}$ [8]. Intensive research has been conducted on the development of electrochemical rechargeable all-liquid energy storage battery. The recent request for green energy transfer and storage for various applications, ranging from small-scale to large-scale power station has increased energy storage advancements and explorations. The criteria of high-energy densities, low cost, and extensive energy storage provision have been met through lithium-ion batteries, sodium-ion batteries, and LMBs [6].

3. Description of a liquid metal battery (LMB)

An LMB cell is made up of two metal electrodes that are in their liquid states splitted by an electrolyte that separate it into three layers based upon their density and immiscibility property is shown in **Figure 1**:

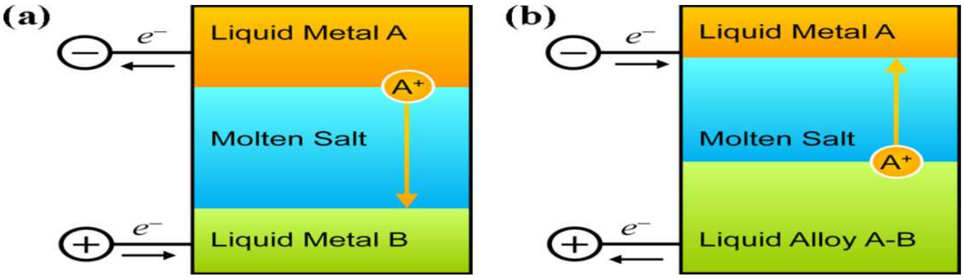


Figure 1. Schematic diagram of a Liquid Metal Battery (L.M.B.) upon **(a)** discharging and **(b)** charging [2].

Table 1. Proposed developmental parameters of the LMB cell.

Design parameters	Li//Cd-Sb	Li//Cd-Bi	Li-Bi	Li-Cd
Electrode composition in moles	47-36-17 48-36-16 49-35-16 50-34-16	42-38-20 47-33-20 45-35-20 44-32-24	45-55 40-60 50-50 48-52	70-30 60-40 50-50 65-35
Operating temperature in O °C	450–500	400–500	350–400	300–400
Electrode area in cm^2	1.30–2.50	1.00–1.50	1.00–2.00	1.50–2.00
Inter-electrode distance in cm	1.00–1.50	1.00–2.00	1.50–2.00	1.50–2.00
Charge–discharge current, A	0.34–0.55	0.34–0.55	0.34–0.55	0.34–0.55
Coulombic efficiency in %	80–99	80–99	80–99	80–99
Voltage efficiency in %	65–80	65–80	65–80	65–80
Average discharge voltage in volts	0.65–0.75	0.65–0.75	0.65–0.75	0.65–0.75
Theoretical capacity in Ah	0.75–0.90	0.75–0.90	0.75–0.90	0.75–0.90
Discharge capacity in Ah	0.55–0.80	0.55–0.80	0.55–0.80	0.55–0.80
Voltage input in volts	12.0 12.1 12.3 11.9	12.5 12.6 11.8 12.0	11.6 11.7 11.9 12.1	11.5 11.6 11.8 11.7
Voltage output in volts	220 215 225 235	225 220 230 230	230 225 235 225	235 230 240 220

4. Material preparation and procedure

1) All samples would be carried out in a controlled glove box environment through the melting of Lithium (Li) and alloys of Antimony (Sb) and Cadmium (Cd). When the alloys are completely fluid, a tungsten wire would be inserted and the alloys would be allowed to cool, ensuring intimate contact between the electrode material and the tungsten wire.
2) The electrolyte would be prepared in the controlled atmosphere by the combination of Lithium Chloride (LiCl), Lithium Iodide (LiI), Lithium Bromide

(LiBr) and Lithium Flouride (LiF) in a vessel. The vessel would be positioned in the test vessel and dried under vacuum some hours and then the temperature could be increased to a higher temperature in the heater.

3) The container containing the electrolyte would be passed through argon gas, before the temperature would be increased to a higher temperature for some hours in order to remove traces of residues from it. On cooling, the electrolyte would be removed from the vessel in the glove box and stored until use.
4) Electrochemical measurements could be carried out in a test vessel sealed against the outer environment.
5) The samples of the induction-melted electrodes prepared initially in No 1 above would be located in an alumina vessel with the pre-melted electrolyte. The assembling of the LMB would be done in the controlled glove box.
6) The charge tester would be used to measure the charge and discharge characteristics of the battery.

5. Battery model with variable parameter equations

Table 2 presents an estimation of the variable parameters in terms of efficiency, standard error, statistics and probability. The different values in terms of efficiency, standard error, statistics and probability gives a mathematical value that defines the variable parameters used and their outputs for LMB estimation.

Table 2. A table of the battery model with variable parameters in terms of efficiency, standard error, statistics and probability of estimation.

Variable parameters	Efficiency	Standard error	Statistics	Probability
OCV	0.000347	0.007368	0.047076	0.9626
R	0.067178	0.054790	1.226096	0.2233
I	–0.003409	0.002345	–1.453833	0.1494
CP	–0.000164	0.000293	–0.560209	0.5767
T	0.000396	0.000356	1.113499	0.2684
TV	–0.001823	0.008424	–0.216355	0.8292
CHG	0.000597	0.001011	0.590636	0.5562
C	0.647579	0.128301	5.047355	0.0000

Estimation command:

$$\mathrm{LSM, SOC, OCV, R, I, CP, T,TV, CHG} \tag{1}$$

Estimated model equation:

$$\mathrm{SOC} = \mathrm{C}(1) \times \mathrm{OCV} + \mathrm{C}(2) \times \mathrm{R} + \mathrm{C}(3) \times \mathrm{I} + \mathrm{C}(4) \times \mathrm{CP} + \mathrm{C}(5) \times \mathrm{T} + \mathrm{C}(6) \times \mathrm{TV} + \mathrm{C}(7) \times \mathrm{CHG} + \mathrm{C}(8) \tag{2}$$

Substituted coefficients:

$$\begin{gathered}\mathrm{SOC} = 0.000346863777113 \times \mathrm{OCV} + 0.0671776941118 \times \mathrm{R} - 0.00340942723159 \times \mathrm{I} - 1.63965521212 \times 10^{-6} \times \\ \mathrm{CP} + 0.000395913781329 \times \mathrm{T} - 0.00182250310982 \times \mathrm{TV} + 0.000597393328656 \times \mathrm{CHG} + 0.6475785799\end{gathered} \tag{3}$$

Estimation command abbreviations:

Least Square Method (LSM), State of Charge (SOC), Open Circuit Voltage (OCV), Resistance (R), Current (I), (4) Capacitance (CP), Temperature (T). Terminal Voltage (TV) Charge (CHG).

6. Discussion of results of the simulated parameters for LMB development

The parameters of all sections of the LMB are based on the initial selection of the structural elements combinations previously stated above. All the relevant parameters were simulated at varying range of values for all input and output parameters.

Figure 2 explains that at the various levels where the SOC parameters are dynamic or vigorous. At the various SOC terminals at the horizontal side, the SOC values are fairly vigorous but at some certain values the SOC parameter values begin to display a high dynamic, particularly at 33, 45, 54, 57, 79 and 85. Hence, to conclude, the highest SOC for the LMB battery would be set at 85%, which is absolutely a good SOC value fit for the battery production.

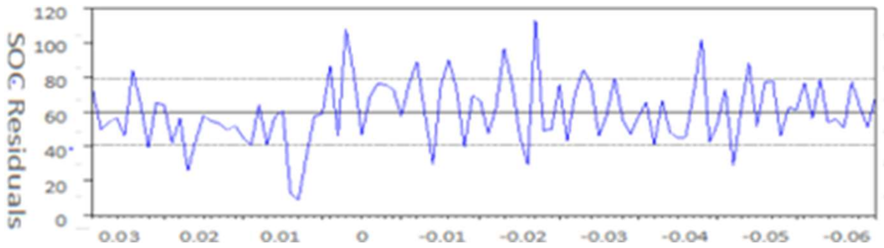


Figure 2. State of charge (SOC) residual parameters.

From **Figure 3**, an analysis was examined between the Residual, Actual and the fitted battery SOC parameters. It was observed that the graph of the Residual and Actual values of the battery SOC value looks the same. It is only that the fitted display has better steady-state values of SOC.

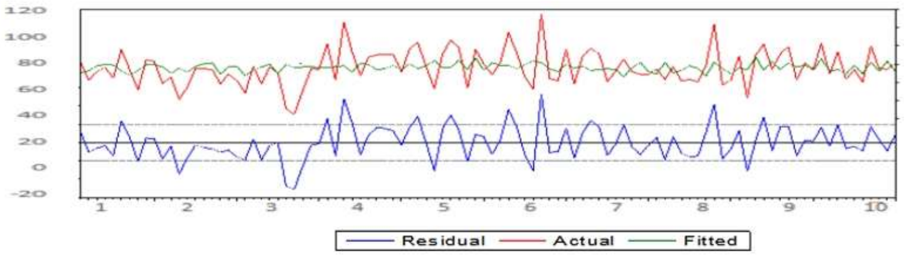


Figure 3. Comparison of the residual, actual and the fitted battery state of charge (SOC).

From **Figure 4**, the battery's standardized residuals for both the positive and the negative value are stable. At the positive side of the graph, the residuals are in regular patterns showing that the State of Charge is implementable.

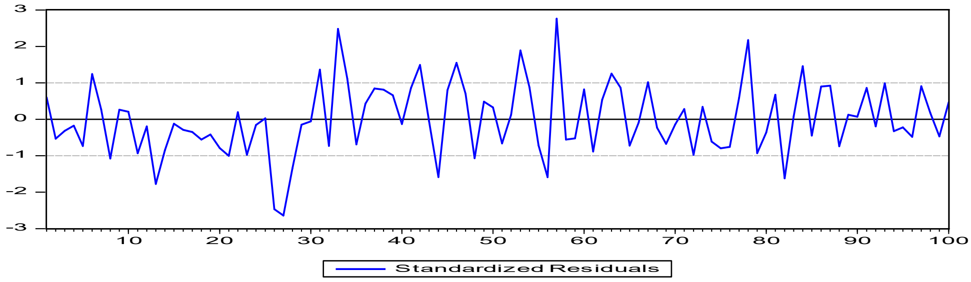


Figure 4. Simulation of standardized residuals for the battery's state of charge.

From **Figure 5**, the Open Circuit Voltage (OCV) must be within the permissible operational voltage of a battery. The Open Circuit Voltage (OCV) for the LMB as stated in the graph is 12.19 V.

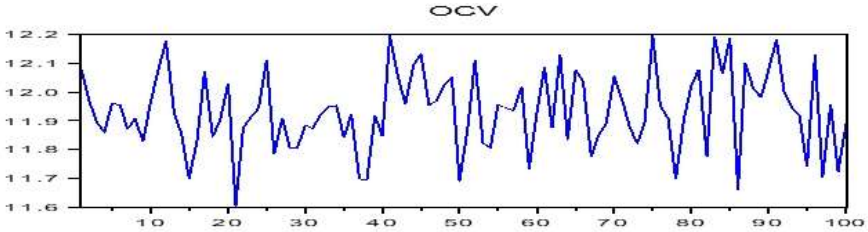
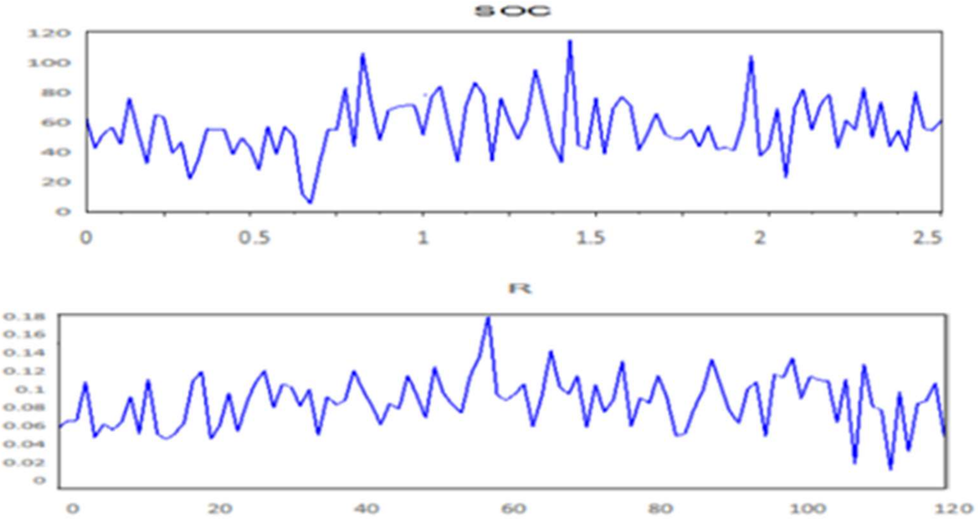


Figure 5. Simulation of Open Circuit Voltage (OCV) for the LMB.

From **Figure 6**, the SOC and Resistance parameters were plotted. The graphical signal output of the SOC parameter shows that the SOC signal is at various points is stable until the peak point where the value is highest at 0.876, which depicts 87.6%.



Figures 6. Simulation of actual State of Charge (SOC) and Resistance (R) respectively.

For **Figure 7**, the resistance parameter, there was unsteady state display of the resistance value at different points until the point where the steady state peak value for output resistance at 0.09 Ω was measured at exactly 50.

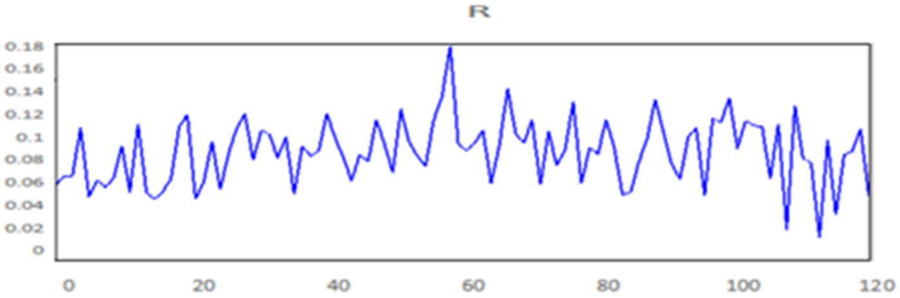


Figure 7. Simulation of output resistance (R) of the battery.

From **Figure 8**, the current at the peak point was 4.0 A and other values for current fluctuation span between 3.0 A and 3.8 A. Meaning that the current is within a pre-determined range. Taking the average of the different values of current at 3 A and at 3.8 A is equal to 3.4 A which is the estimated actual current of the LMB.

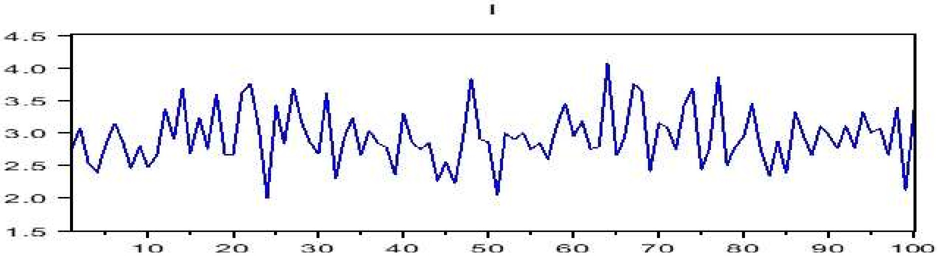


Figure 8. Simulation of the input current (I) of the battery.

The actual time in **Figure 9** as stated in the above graphical representation is 34 minutes.

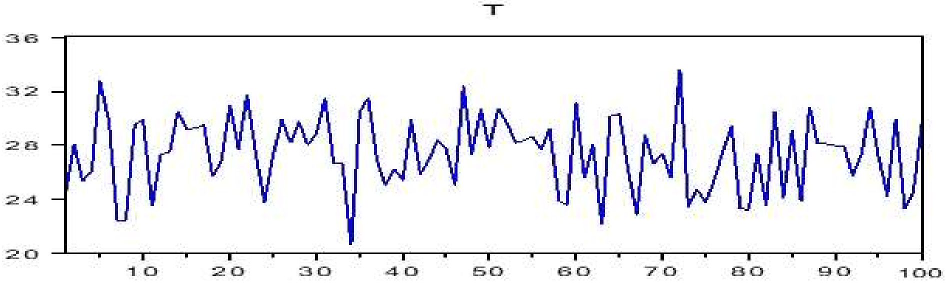


Figure 9. Simulation of the actual time (T) of the battery.

From **Figure 10**, the charge has varying values between 0 and 100. But the actual charge that is displayed at the graphical representation is at 98 coulombs (C).

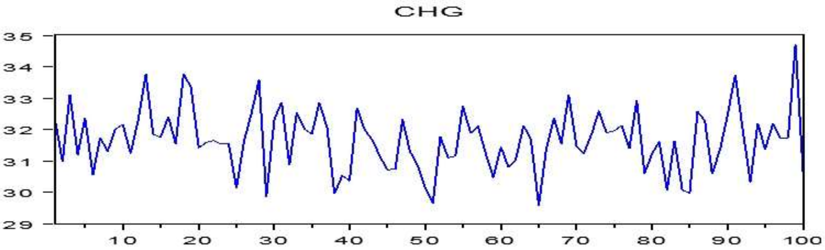


Figure 10. Simulation of the charge (CHG) of the battery.

The simulated graph at **Figure 11** for the terminal voltage (TV) of the battery is captured at point 12.1 V.

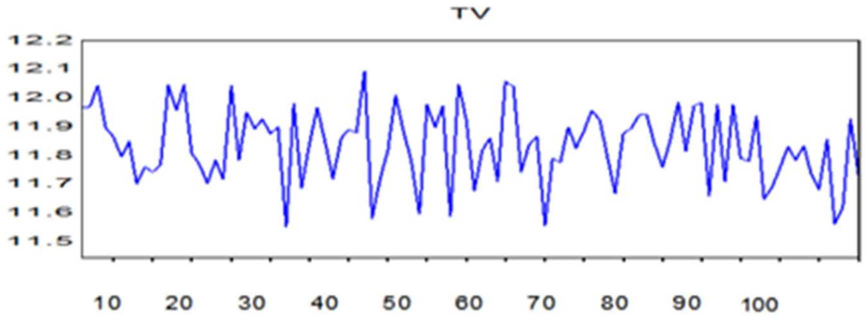


Figure 11. Simulation of the terminal voltage (TV) of the battery.

From **Figure 12**, the capacitance displayed at the graphical representation is at 3.6 × 103 Farads (3.6 KF).

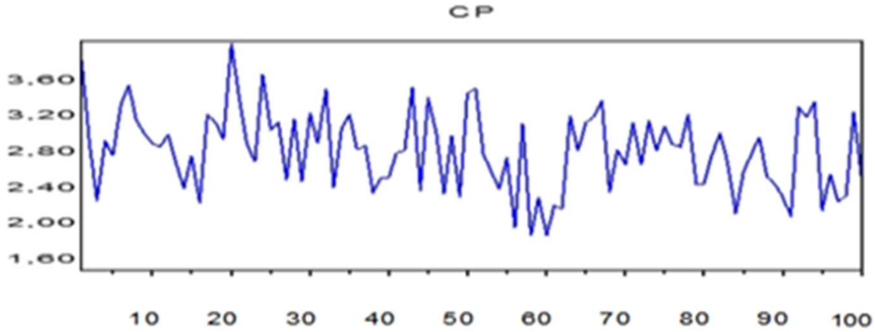


Figure 12. Simulation of the capacitance (CP) of the battery.

7. Conclusion

Liquid Metal Batteries (LMBs) have some dynamic characteristics in parameter estimation during various charge and discharge periods. Due to parameter estimation through the comparative analysis of the simulated chemical constituents of varying metals and electrolytes, the Li//Cd-Sb combination is most viable for LMB production when compared with Li//Cd-Bi, Li-Bi, and Li-Cd constituents. The values for Open Circuit Voltage (OCV), the Terminal Voltage (TV), Charge (CHG), Time (T), Current (I), Resistance (R), Capacitance (CP), and State of Charge (SOC) are determined for the preferred LMB combination for battery development. This would make the operation of the LMB highly practicable for both domestic and small industrial uses.

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