Development and Analysis of a Lithium Carbon Monofluoride Battery-Lithium Ion Capacitor Hybrid System for High Pulse-Power Applications

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Abstract

Although Li/CF_x and Li/CF_xMnO₂ have two of the highest energy densities of all commercial lithium primary batteries known to date, they are typically current-limited and therefore are not used in high-power applications. In this work, a Li/CF_xMnO₂ battery (BA-5790) was hybridized with a 1,000 F lithium ion capacitor to allow its use for portable electronic devices requiring 100 W one-minute pulses. An intelligent, power-management board was developed for managing the energy flow between the components. The hybrid architecture was shown to maintain the battery current to a level that minimized energy loss and thermal stress. The performance of the Li/CF_xMnO₂ hybrid was compared to the standard Li/SO₂ battery (BA-5590). The hybrid was shown to deliver the same number of 100 W pulse cycles as two BA-5590 batteries, resulting in a weight savings of 30% and a volumetric reduction of 20%. For devices requiring 8 hours of operational time or less, a 5-cell Li/CF_xMnO₂ hybrid was found to be a lighter (55%) and smaller (45%) power source than the existing two BA-5590 battery option, and a lighter (42%) and smaller (27%) option than 1½ BA-5790 batteries alone. At higher power requirements (>100 W), further weight and size improvements can be expected.

1. Introduction

Portable electronic devices often have load profiles that consist of intermittent pulses of high power separated by a much longer, more modest baseline-power demand. One of the mainstay batteries used by the military for such applications has been the lithium/sulfur dioxide (Li/SO₂)-based BA-5590 primary battery. With an energy density of $\sim 200 \text{ Whkg}^{-1}$ (230 WhL^{-1})[1,2], the BA-5590 is noted for its ability to meet high-power requirements and to operate at low temperatures (-40°C). However, major limitations of the BA-5590 for defense applications continue to be its weight and volume. The total weight of batteries required to complete a mission can be a logistics problem, thus reducing the fighting capability of the soldier. Additionally the military, similar to the consumer, desires miniaturized, less cumbersome devices. Reducing the weight and volume of the power source promotes user agility and would be of great value to the Marine Corps and the Army.

Alternative technologies with higher energy densities have been developed, such as lithium/carbon monofluoride (Li/CF_x) which have approximately twice the energy density of Li/SO₂ cells (Table I). However to date, Li/CF_x technologies have not replaced the Li/SO₂ in military dominance due to their inability to meet the power demands required for numerous electronic devices. The limited high-rate performance of the Li/CF_x battery is thought to be the result of kinetic limitations associated with breaking C-F bonds and the poor electrical conductivity of the CF_x material creating high internal impedance during discharge [3,4].

Over the years, numerous attempts to increase the rate capability of the Li/CF_x electrochemistry and thereby increase operational life have had limited success. CF_x is an intercalation compound formed by the reaction between carbon powder and fluorine gas. This process forms strong covalent C-F bonds with sp³ hybridization that induces charge localization and results in a significant reduction in electrical conductivity as the value of x increases [5]. Approaches to solving the issue of low electrical conductivity to improve rate capability while maintaining a high energy density include: partial carbon

fluorination so that any unreacted carbon can serve as a conductor between the CF_x particles [6,7], carbothermal treatment of the CF_x that results in the formation of sub-fluorinated CF_x [8], and pretreatment of CF_x using a "solvated electron" reduction method to obtain a thin layer of graphitic carbon coating on the CF_x particle surfaces [9]. Additional material approaches to improving poor-rate performance have included blending CF_x with more conductive cathode materials such as manganese dioxide (MnO_2), with the expectation that an optimized cathode mixture ratio would provide enhanced cell rate capability while maintaining the high energy density advantage of CF_x [10]. These studies demonstrated promising improvements in cathode rate capability, though none were at a level required for future high-power portable devices. Thus batteries composed of CF_x cathode materials still cannot deliver their full capacity under high-power conditions, making them an unsuitable replacement for the low energy density Li/SO_2 -based batteries in military applications.

A hybrid battery-capacitor architecture containing both a high-energy device (battery) and a high-power device (capacitor) is a potential solution to address the unmet military need for light, miniaturized devices that can provide a series of intermittent high-power pulses superimposed over a steady, low-power baseline. Combining a battery with a capacitor allows the capacitor to support the high-power transients, thus lowering the battery discharge current and improving energy utilization. In a CF_x battery-capacitor hybrid, each component would operate under optimal discharge rate conditions: short-duration, high-power pulses are supplied by the capacitor, and the CF_x battery operating at low to moderate discharge rates fulfills the baseline power demand. During periods of low-power events, the CF_x battery would recharge the capacitor.

Several studies have indicated success in utilizing hybrids with capacitors to improve the usable capacity of primary lithium and non-lithium batteries in applications requiring high-power pulses. However, few have focused on providing such augmentation while minimizing weight and volume – key for military applications. Most previous investigations have used electrochemical double layer capacitors (EDLCs) for

the high-power component. Atwater et al. observed a significant improvement in a primary zinc-air battery using a simplistic hybrid design consisting of the battery and an EDLC in parallel with no interface electronics [11]. The hybrid zinc-air achieved double the number of high-power pulses compared to the battery alone. Penella et al. [12] observed a 33% improvement in runtime with a primary, silver oxide-zinc battery when hybridized with an EDLC and more recently, Cain et al. [13] noted a 46% increase in retrievable energy from a primary Li/MnO₂ battery. A larger number of studies have also focused on hybridizing secondary (rechargeable) batteries with EDLCs. For example, Cain et al. observed a 40% increase in capacity with a lithium ion rechargeable battery in the above study [13]. Others have also investigated hybridization of lithium ion batteries with EDLCs as a method to increase cycle life and improve operating temperature performance, in addition to extending battery run time [14-19]. These enhanced properties are particularly relevant for electric, hybrid electric and plug-in vehicles where high-power surges can lead to excessive cell heating, battery degradation and safety concerns. To determine the viability of a hybrid system for these and other applications, significant experimental work, mathematical simulations, and investigations to improve power/energy management have been conducted [e.g., 20-21]. Though these studies have successfully demonstrated improved battery performance, a thorough analysis of the weight and size of the devices has often been neglected.

The benefits of a hybrid battery-EDLC power source, unfortunately, are not without consequences. Enhanced battery performance can be at the expense of the overall system's weight, volume and complexity [22-24]. These repercussions are particularly damaging for portable applications when long (> 30 seconds) high-power pulses are required. EDLC's have low energy density compared to batteries. A capacitor bank would therefore need to be oversized to meet a long-duration pulse requirement, resulting in poor realization for portable military applications. However, recent developments in EDLCs have made the development of hybrid systems requiring long pulse times significantly more feasible.

Traditional EDLCs have a symmetric design comprised of two identical carbon electrodes, with a total cell capacitance equal to one half of the electrode capacitance. Recent efforts to develop higher energy density capacitors have led to an asymmetric design, where one of the double-layer carbon electrodes is replaced with a battery material that undergoes facile, reversible, Faradaic redox reactions [25]. Here, the capacitance of the battery electrode is more than 10 times that of the capacitive electrode ($C_1 \gg C_2$). As a result, the total capacitance of an asymmetric cell is doubled that of the symmetric design. In addition, asymmetric capacitors typically operate at voltages 50% higher than that of the symmetric capacitors. Therefore, the theoretical energy ($E = \frac{1}{2}CV^2$) of an asymmetric capacitor is approximately four times greater than the symmetric design. One promising asymmetric capacitor that has demonstrated high energy density utilizes a carbon negative electrode which reversibly intercalates lithium. Referred to as LICs (Lithium Ion Capacitors), the cells have demonstrated energy densities 3-5 times greater than that of traditional EDLCs and exhibit a cycle life of 1,000,000. Unlike traditional EDLCs, LICs can hold charge for extensive periods of time (~3-5% voltage loss after 3 months)[26,27] allowing energy to be available for immediate use. They have also been found to be more abuse tolerant than lithium ion rechargeable batteries due to the lack of a Faradaic electrode [28].

To the best of our knowledge, there are no reports of hybridizing a primary lithium battery with a lithium ion capacitor. Furthermore, studies with EDLC-containing hybrids have rarely included an in-depth analysis on the size of such devices and their potential for portable applications. The present study reports our efforts to develop a novel hybrid power source that exploits the high energy density benefits of CF_x batteries and the high power capability of a higher energy density capacitor. Our focus was to determine if, and under what conditions, a hybrid power source consisting of a $Li/CF_x/MnO_2$ battery and a LIC would be more advantageous than the conventional, Li/SO_2 BA-5590 battery for portable devices requiring one-minute 100 W pulses.

2. Test Procedures and Hybrid Design

2.1 Materials

Li/CF_xMnO₂ D-cells and batteries were manufactured at Eagle Picher Technologies (Joplin, MO). The cell, with a nominal capacity of 16 Ah, utilized an aluminum case and weighed 81 g. The percentage of the active cathode materials was not known and is proprietary. The electrolyte consisted of a blend of dimethoxyethane, propylene carbonate, tetrahydrofuran and lithium perchlorate. The 10-cell Li/CF_xMnO₂ battery, identified as the BA-5790 (Figure 1a), had a rated capacity of 15 Ah when operated at the 24 V mode and 30 Ah when operated at the 12 V mode. The BA-5790 (61.0 mm x 111.8 mm x 127.0 mm) weighed 1030 g. The half BA-5790 (Figure 1b), composed of five D-cells, weighed 531 g and had a rated capacity of 15 Ah at 12 V.

Li/SO₂ D-cells (LO 26 SHX) and ten-cell batteries (BA-5590 B/U, Figure 1c) were manufactured at SAFT America (Valdese, NC). Unlike Li/CF_xMnO₂ that contained a solid cathode material, Li/SO₂ cells consisted of a gaseous positive electrode material, SO₂, dissolved in a solution of acetonitrile and lithium bromide. The nominal capacity of the Li/SO₂ cell was 7.5 Ah. The BA-5590 battery (62.2 mm x 111.8 mm x 127.0 mm) had a rated capacity of 15 Ah when operated at the 12 V mode and 7.5 Ah when operated at the 24 V mode. The weights of the cell and BA-5590 B/U battery were 85 g and 1,006 g, respectively.

JM Energy (Yamanashi, Japan) supplied the prototype 1,000 F LIC (Figure 1d). The LIC was selected based on its availability, previous performance/safety analysis [28] and robust cell design. The design did not necessarily reflect improvements which may be incorporated in future, manufactured cells. The cells (11 cm x 7 cm x 1.1 cm) weighed 163 g. The electrolyte consisted of a blend of ethyl methyl carbonate, ethylene carbonate, dimethyl carbonate and lithium hexafluorophosphate. An undisclosed ingredient was added as a flame retardant.

Fig. 1- Photograph of military batteries used in this investigation: (a) Eagle Picher Li/CF_xMnO₂ BA-5790 battery containing ten Li/CF_xMnO₂ D-cells, (b) Eagle Picher half BA-5790 battery containing five Li/CF_xMnO₂ D-cells, (c) Saft BA-5590 battery consisting of ten Li/SO₂ D-cells, and (d) 1,000 F LIC.

2.2 Hybrid Design

The hybrid system consisted of three main parts: the battery, the capacitor, and the power management board. The power management board controlled the flow of energy between the battery and the capacitor in response to the load demand. It contained all the necessary sensors, microprocessor based controls, and switching power electronics to allow interconnection of the battery and the capacitor to realize hybrid operation. The board was sized (12 cm x 10.5 cm x 0.16 cm, 71.5 g) to fit within the footprint of a BA-5590 package, making it suitable for portable military applications.

The power management board differed from a typical battery management system which is often responsible for balancing charge on a series string of rechargeable batteries. For this application the focus was on primary batteries and a single capacitor which do not require cell balancing. As our work progresses and rechargeable and series cells are employed within the hybrid, the power management function will expand to include battery management and cell balancing as well. A block diagram of the power management board is provided in Figure 2a. The power management algorithm which transitions the device between capacitor charge and discharge modes is diagramed in Figure 2b, and the electrical model of the hybrid system is depicted in Figure 2c.

The architecture of the hybrid system can be seen by examining Figure 2a. The battery is connected to the power management board, which routes the high power bus in parallel with a bidirectional DC/DC converter. Although prior investigations have used a direct connection between a battery and a bank of series connected EDLCs, that architecture limits the voltage range of the capacitors to that of the battery and therefore restricts the amount of energy that can be extracted from the capacitors. Other designs effectively connect a series bank of capacitors directly to the load in order to eliminate having a DC/DC converter (with its efficiency loss) directly in series with the capacitors [20]. However, that approach requires voltage cell balancing circuitry to ensure that charge is evenly distributed in the capacitors over a

Fig.2-Hybrid system. (a) Block diagram of Intelligent Power Management Board. Main components include: a bi-directional DC/DC converter, a controller to manage the power distribution, a microprocessor, a serial link enabling direct communication with the power management board, a temperature sensor, and an output DC/DC converter, (b) power management algorithm flow, and (c). electrical model of the hybrid system.

number of charge/discharge cycles. A direct connection to the load also means that the energy available from the capacitors is constrained by the allowable voltage variation of the load. In this study, a single LIC cell was used, thereby minimizing the size, cost, and complexity of the hybrid design. The bidirectional DC/DC converter in series with the LIC extracts the maximum energy from the LIC regardless of the battery or load voltage. The parallel connection of the battery pack and capacitor (via bidirectional DC/DC converter) was passed through an output DC/DC converter that allowed the user to generate a regulated voltage output to the load. For this study, the output DC/DC converter was not used.

The power management board (Figure 2a) consisted of several design elements including communications hardware and software, sleep circuitry, microprocessor/controller, and power electronics. A detailed description of each is provided below:

The communications hardware included serial receive/transmit circuitry necessary for enabling the user to directly communicate with the power management board.

The communications software allowed the user to vary the control configuration in order to conform to a particular experiment being conducted. For example, the battery current limits could be modified through serial communications based upon the particular load power to be supported. The board also allowed the user to access instantaneous readings of the system measurements.

The sleep circuitry reduced the system power consumption and thus improved overall system efficiency. It monitored the system to determine if any discernable load was present. If no load was detected, then a count-down was initiated before the system was put into a low-power mode. The low-power mode shut down all circuitry that was not required to maintain the system in a sleep state or to detect the presence of a load in the future.

The microprocessor/controller executed the power management algorithm (Figure 2b), which allowed the device to operate in one of three distinct modes in response to an applied load. Monitored parameters used as inputs into this algorithm included battery current and voltage, load power and capacitor voltage. The three modes included: Hybrid, Recharge, and Battery. The Hybrid Mode was selected when the load requirements exceeded the defined battery control current set point and the LIC was not fully depleted. In this situation, the system functioned as a Hybrid by providing power to the load using contributions from both the LIC and battery (or the LIC only if the battery were depleted). The controller operated a bidirectional power converter to step up (boost) the voltage of the LIC from its nominal 3.0 V to the nominal 12.0 V of the main bus to assist in high load-power circumstances (Figure 2c). The Recharge Mode was selected when the load requirements were less than the defined control set point of the battery and the LIC voltage was measured to be less than the maximum defined. In this state, the controller operated the bidirectional power converter to step down (buck) the voltage from the battery to the LIC delivering power for the recharge. The Battery Mode was selected when the required load power was less than the defined battery control set point or when the load was greater than the battery set point but the LIC was depleted. The battery provided sole support of the load with all converter electronics disabled.

The power electronics circuitry incorporated a bidirectional DC/DC converter (Figure 2c) that enabled charging and discharging of the power dense LIC component in the hybrid architecture. The DC/DC converter bridged the voltage gap between the battery and LIC, allowing both to operate in the same system (Li/CF_xMnO₂: ~14-10 V, LIC: ~3.8-2.2 V). The efficiency of the converter was typically ~90% and was a function of the duty cycle resulting from the particular power level in which the hybrid system was operating. The current out of the battery was regulated to keep the battery operating efficiently via a proportional-integral current controller which varied the duty cycle on the MOSFET switches. In addition, the circuitry had safety

features that monitored the LIC to ensure operation within its recommended voltage range, ensured the maximum current rating of the MOSFETs was not exceeded, and monitored the battery voltage and temperature. The power electronics were shut down if any preset fault conditions were exceeded to protect the system.

Two unique hybrid designs were examined. The first was a "single-unit" (Figure 3a) with the same footprint as a BA-5X90 military battery (6.2 cm x 11.2 cm x 12.7 cm) and consisted of five Li/CF_xMnO₂ D-cells, a 1,000F LIC, and the power management board. The board was programmed so that the operating range of the LIC was 4.0 V - 2.2 V. The second design, referred to as the "Power Augmentor" (12.5 cm x 11.0 cm x 3.8 cm, 380g, Figure 3b), was an attachable device containing only the power management board (Figure 3c) and capacitor. The Power Augmentor had an ITT Cannon P/N CA 110821-6 mating connector so that it could easily attach to a BA-5X90 battery (Figure 3c) when high power pulses were required, but removed for lower power demands. The Power Augmentor was programmed so that the LIC operated between 3.8 V and 2.2 V. This voltage range was selected to ensure maximum rechargeability (1,000,000) by minimizing chemical reactions between the electrode and the electrolyte. Hybridization of both the BA-5X90 batteries was evaluated with the Power Augmentor.

2.3 Cell Characterization

Li/CF_xMnO₂ and Li/SO₂D-cells underwent galvanostatic testing at currents ranging from 1 A to 3 A to determine the effect of discharge rate on cell capacity and operating temperature. All tests were conducted at ambient conditions. Two Type-K thermocouples (Omega Chromel/Alumel), held in place by electrical tape, were positioned on the outer circumference of the cell to monitor temperature every 3 minutes. Cells were placed in an expanded 2 inch polystyrene insulating container with an R factor of approximately four. A MACCOR Series 4000 Automated Test System was used to discharge cells to an endpoint of 2.0 V. At least two cells were evaluated per test condition. Voltages were measured at least every 2 seconds.

Fig. 3- Photograph of (a) the single unit hybrid having the same format of the BA-5X90 and consisting of five lithium D-cells, 1,000 F LIC, and power management board, (b) the attachable Power Augmentor consisting of the 1,000 F LIC and the power management board, (c) the power management board within the Power Augmentor, and (d).the Power Augmentor (with two pair of alligator clips used for voltage sensing in the laboratory test environment) attached to the BA-5790 battery.

The 1,000 F LIC was cycled at a constant power of 100 W between 3.8 V and 2.2 V at ambient temperature using a MACCOR Series 4000 Automated Test System. Self-discharge was monitored by charging the cell and then measuring the voltage daily with a voltmeter.

2.4. Battery and Hybrid System Evaluation

Both the Li/CF_xMnO₂ BA-5790 and Li/SO₂ BA-5590 batteries and hybrid systems were evaluated using either a MACCOR Series 4000 Automated Test System or a BK Precision 8500 DC Electronic Loader. The test procedure [100 W for 1 minute, followed by 12 W for 9 minutes, repeat until the battery (or hybrid system) voltage fell to 10 V] was designed to provide harsh conditions that would clearly demonstrate the effect of high-rate pulse load applications on battery/hybrid service life and temperature. Type-K thermocouples were taped onto the surface of batteries (ten positioned on the BA5790, six positioned on the BA5590) to monitor temperature during the test. (Note that although the hybrid's power management board had the capability for measuring battery temperature, external thermocouples were used for data collection in these experiments.) An Agilent 34970 Data Acquisition unit was used to monitor the cell/battery temperatures (at least every 3 minutes) and voltages (at least every 2 seconds). As a safety precaution, tests were conducted in a chemical fume hood with an average air flow of 92 feet per minute.

3. Results and Discussion

3.1 Cell Characterizations

We evaluated Li/CF_xMnO₂ and LIC cells to determine the operational limits of the fundamental units of our hybrid design. Tests were conducted on Li/SO₂D-cells for comparison purposes.

The voltage and temperature profiles of Li/CF_xMnO₂ D-cells discharged at various constant currents under ambient temperature conditions (~20 °C) are shown in Figure 4a,c. Unlike Li/SO₂ cells that

Fig. 4- Voltage and temperature profiles of (a, c) Li/CF_xMnO₂ and (b, d) Li/SO₂ D-cells discharged at a constant current of 1 A, 2 A, 2.5 A and 3 A.

displayed flat working voltages (~ 2.8 to 2.7 V, Figure 4b), Li/CF_xMnO₂ D-cells exhibited discharge profiles with two distinct plateaus attributed to the reduction of MnO₂ (3.3 V to ~ 2.5 V) and CF_x (~ 2.5 V to ~ 2.0 V). Cell temperature increased sharply when the cell voltage entered the second plateau. At 3.0 A, only $\sim 60\%$ of the rated capacity was delivered before the cell skin-temperature rose to 85 °C and the test was terminated for safety reasons. An increase in cell temperature with discharge rate has been previously reported for both Li/CF_x cells[29,30] and Li/CF_xMnO₂ batteries[31]. One possible explanation for the generated heat is the large activation energy associated with breaking C-F bonds. As noted by Read et al., a large activation energy would lead to poor kinetics during discharge and significant heat generation[32]. Increased cell temperature has also been reported to result from the decomposition of an intermediate discharge product (i.e. CFLi_x:S_y), where S is a solvent molecule that is co-intercalated with the lithium upon discharge to form LiF and C[33,34]. At currents ≤ 2.5 A, high cell skin-temperatures did not limit performance and the high rated cell capacity(15 Ah) was achieved(Table II).

The corresponding voltage and skin-temperature profiles of Li/SO₂ D-cells discharged under identical conditions are shown in Figure 4b,d. At 3 A, the highest skin cell temperature observed was ~ 50 °C and cells delivered 7.0 Ah. There is therefore no benefit, in terms of delivered capacity, to use Li/CF_xMnO₂ in a hybrid system if cells are required to operate at 3 A or higher. At currents ≤ 2.5 A, Li/CF_xMnO₂ cells provide twice the capacity as Li/SO₂ cells and there is a clear advantage for their use. It should also be noted however that Li/SO₂ cells discharged at currents ≥ 2.0 A exhibited a substantial voltage drop and subsequent voltage delay (Figure 4b) when the load was first applied. Voltage delay results from a passivation layer that forms on the surface of the lithium anode when exposed to electrolyte, solvent, or impurities [35]. Under normal operating conditions, the passivation layer does not affect cell performance. However, when a high-power load is placed on the cell, the resistance of the film can cause the cell's voltage to drop significantly. The cell voltage eventually recovers as the discharge progresses and the film is disrupted, but for certain applications, the voltage delay cannot be tolerated.

The 1,000 F capacitor delivered 45 seconds at 100 W when it was discharged between 3.8 V and 2.2 V, as shown in Figure 5. When the cell was charged to 3.8 V, the self-discharge rate was 4% over 2 months. When charged to 3.0 V, the self-discharge rate dropped to ~ 0%.

The cell investigations confirmed the following:

- A primary, lithium D-cell consisting of a CF_x cathode material (CF_xMnO_2), delivered approximately twice the capacity as a cell utilizing a SO_2 cathode when discharged at ≤ 2.5 A. There is a significant energy density advantage (both gravimetric and volumetric) in using $\text{Li}/\text{CF}_x\text{MnO}_2$ cells as opposed to Li/SO_2 cells for portable electronic devices.
- The energy density advantage of a $\text{Li}/\text{CF}_x\text{MnO}_2$ cell may be over shadowed by safety concerns. Significant heat is generated when CF_x is discharged at high rates. Cells discharge at 3 A can reach temperatures exceeding 85 °C. Safety concerns may be exacerbated when cells are closely stacked together, as in a military BA-5X90 battery.
- The 1,000 F LIC cannot sustain a 100 W load for one minute. In a hybrid system, the battery would be required to power-share for applications requiring 100 W pulses for greater than 45 seconds.
- Unlike EDLCs, a LIC has a relatively low self-discharge rate. This should increase energy efficiency in prolonged hybrid use situations.

3.2 Battery Characterizations

In order to fully understand the benefit of a $\text{Li}/\text{CF}_x\text{MnO}_2$ -LIC hybrid system, the $\text{Li}/\text{CF}_x\text{MnO}_2$ BA-5790 and Li/SO_2 BA-5590 batteries were assessed using the same test protocol as the hybrid. The Li/SO_2 BA-5590 was evaluated as a benchmark, as it is commonly used in military applications.

The BA-5590 is constructed from two parallel strings of five Li/SO_2 D-cells connected in series (5s2p). Each nominally 12 V section has control electronics consisting of a current fuse, a thermal fuse and a

Fig. 5- Voltage profile of a 1,000 F LIC discharged at 100 W.

diode to prevent accidental charge. The plug connection from the battery allows the two strings to be discharged either in parallel (12 V) or in series (24 V), depending on the configuration of the mating electronics. When configured to deliver 12 V, each string of cells has a limited current of 3 A as a safety precaution against overheating. As a result, the maximum power that can be drawn from a BA-5590 battery is approximately 72 W.

Experiments confirmed that a single Li/SO₂ BA-5590 (5s2p) cannot deliver 100 W pulses (Table III). The 3 A fuse limit was immediately exceeded, and the voltage fell below the 10 V termination limit. Restarting the experiment proved ineffective. Only when two BA-5590s were connected in parallel (5s4p) was the test able to proceed and the battery deliver 100 complete cycles (16.7 h, Figure 6a). It should be noted however, that two attempts were required before the battery was able to discharge. During the first attempt, the initial 100 W pulse resulted in an immediate voltage drop below the cut-off voltage (9.38 V) and the test terminated. The second attempt was able to proceed because the initial battery voltage was 0.6 V above the termination limit. The voltage drops observed in both tests are attributed mainly to a resistive lithium dithionite (Li₂S₂O₄) anode film resulting from the reaction of the soluble cathode, SO₂, with lithium [36,37]. Once the experiment was underway, the Li/SO₂ battery displayed a flat cycling profile. An expansion of the discharge plot (Figure 6b) shows that during the 9-minute 12 W segments, the battery voltage was approximately 2 V higher than during the 100 W, 1-minute pulse. The highest temperature observed on the outer surface of the BA-5590 case during the test was 35°C.

The BA-5790, composed of ten, Li/CF_xMnO₂ D-cells (5s2p) and a 3 A fuse, was unable to sustain the 100 W load, similar to its Li/SO₂ counterpart (Table III). When the 100 W pulse commenced, the fuse limit was immediately reached and the voltage dropped below 10 V. It was only when two Li/CF_xMnO₂ BA-5790 batteries were connected in parallel (5s4p) that the unit was able to support the 100 W load and deliver 208 cycles (34.7 h). The potential versus time graph (Figure 6c-d) showed the characteristic two-

Fig. 6- Voltage profiles of two, BA-5X90 batteries connected in parallel (5s4p) and cycled according to the test protocol (1 minute at 100 W, 9 minutes at 12 W, repeat): (a, b) Li/SO₂ BA-5590 batteries [It should be noted that during the initial 100 W pulse, the battery voltage fell to 9.38 V (below the cut-off voltage). Only after restarting the experiment could the test continue], and (c, d) Li/CF_xMnO₂ BA-5790 batteries. Figures b and d are expanded versions of plots a and c, for clarity.

voltage plateau observed with the D-cells. The highest temperature recorded on the outer surface of the battery case was (50°C).

The battery investigations confirmed the following:

- Single Li/SO₂BA-5590 and Li/CF_xMnO₂BA-5790 batteries (5s2p) cannot deliver 100 W pulses. The 3A fuse limit is exceeded. Thus battery-capacitor hybrids must maintain the battery current < 3A.
- Two Li/CF_xMnO₂BA-5790 batteries in parallel (5s4p) can deliver twice the capacity as two Li/SO₂BA-5590s in parallel (5s4p).
- The temperature of the Li/SO₂BA-5590 and Li/CF_xMnO₂BA-5790 batteries increases during high-power discharge.
- The Li/SO₂BA-5590 can exhibit significant voltage drop during initial high-power discharge.

3.3 Hybrid Investigations

Given the power limitations of the high energy density Li/CF_xMnO₂BA-5790, we developed a hybrid power system that would give the military the option of using Li/CF_xMnO₂ batteries for both low and high power applications. Our goal was a power source that was lighter and smaller than the present Li/SO₂BA-5590. To accomplish this objective, an intelligent power management board was developed to manage the energy flow between the battery and the capacitor (Section 2.2).

Under optimal conditions, the LIC capacitance would be sized to meet the entire 1-minute, 100 W pulse-load, eliminating battery stress during the pulse and prolonging battery life. During the course of this investigation however, limited cell sizes (capacities) were available due to the newly emerging LIC technology. Of those that were available, the 1,000 F (0.44 Ah) cell was selected for use based on its small profile. However, the 1,000 F capacitor could only operate for 45 seconds when discharged at 100 W between 3.8 V and 2.2 V (see Section 3.1) and thus, the full advantage of the hybrid could not be realized. Accordingly, the board was programmed to allow the battery to work in tandem with the

capacitor to meet the high-power load. Given safety concerns regarding the temperature rise of Li/CF_xMnO₂ D-cells at high currents (Figure 4c), and the additional limitations of heat transfer when cells are assembled within the BA-5790 battery case, the power management board was configured to limit cells to the minimum current required to meet the load demands. For hybrid systems incorporating 5s2p and 5s4p batteries, the board was programmed so that the cell current never exceeded 2.5 A and 1.5 A during the 100 W and 12 W load pulses, respectively. For hybrid systems with 5s1p batteries, the limit was increased to 2.9 A (100 W) and 2.5 A (12 W).

Despite the 45-second LIC limitation, the hybrid designs in this study demonstrated an improved ability to deliver high-power pulses with reduced operating temperatures compared to non-hybridized BA-5790 batteries. As shown in Figure 7a, the single-unit Li/CF_xMnO₂ hybrid design (Figure 3a) successfully delivered fifty 100 W pulses, whereas a single, Li/SO₂ BA-5590 battery did not deliver any (Section 3.2). The voltage profiles of both the hybrid and the battery component displayed two distinct plateaus that were similarly observed for the Li/CF_xMnO₂ D-cell (Figure 4a). As expected, an expansion of the voltage profiles clearly demonstrated that both the LIC and the battery contributed to the 100 W load demands, while the battery provided sole power during the 12 W loads (Figure 7b). The battery and hybrid voltage immediately fell at the initiation of the high-power pulse. Both voltages remained low as the two components supported the 100 W load. Upon completion of the 1-minute pulse, the hybrid voltage increased while the battery voltage continued to decline as it accommodated the 12 W load and charged the LIC. Once the capacitor was fully charged, the battery voltage rapidly increased and then remained relatively constant until the next high-power load pulse.

During this test, the battery temperature appeared to plateau around 62 °C (Figure 7a). In contrast, an un-hybridized five-cell Li/CF_xMnO₂ battery (5s1p, 3 A fuse removed) that was subjected to identical testing demonstrated cell temperatures exceeding 70 °C (Figure 7c) and the experiment was terminated due to safety concerns.

Fig. 7- Voltage profiles of (a-b) a five D-cell Li/CF_xMnO₂ battery - LIC hybrid, and (c) a five D-cell Li/CF_xMnO₂ non-hybridized battery without a 3 A fuse, cycled according to the test protocol (1 minute at 100 W, 9 minutes at 12 W, repeat).

(Although the manufacture states that cells can operation to 85°C, an upper limit of 70°C was selected for experiment termination based on military specification MIL-STD-1472F.) In addition to safety concerns, high operating battery temperatures accelerate chemical reactions and accelerate aging. Therefore, hybrid architectures are not only a technique to reduce cell temperatures and improve battery safety, but can also extend battery life.

After the successful demonstration of the power management board in the single-unit configuration, the board was then coupled to the 1,000 F LIC and placed in a durable housing as a detachable, reusable unit (Power Augmentor) to provide boost power as needed. When the Power Augmentor was coupled to a half-size Li/CF_xMnO₂ BA-5790 (5s1p), the unit delivered 58 cycles (Table III), achieving similar results as the single-unit design described above. Connecting one full-size Li/CF_xMnO₂ BA-5790 (5s2p) with the Power Augmentor resulted in 96 cycles (Figure 8a) - approximately the same number of cycles achieved with two, Li/SO₂ BA-5590 batteries (5s4p, Figure 6a). This equates to a 49% reduction in disposable battery weight (Table III) and a 30% reduction in total weight (battery and Power Augmentor). A low disposal battery weight is desirable, as it is a reflection of the burden to supply batteries to the field.

During testing of the BA-5790 (5s2p)-Power Augmentor hybrid, the temperature of one Li/CF_xMnO₂ cell rose to 65 °C (Figure 8b). The hottest cells were located in the center of the battery. This is in agreement with thermal conductivity modeling and measurements previously studied by Listerud et al. [31]. In comparison, the highest battery temperature observed with two Li/SO₂ BA-5590s connected in parallel (5s4p) was 33 °C. However the actual Li/SO₂ cell temperature may have been higher. Unlike the conformal Li/CF_xMnO₂ BA-5790 design (Figure 1a) that allowed thermocouple positioning directly over the individual cells, the Li/SO₂ BA-5590 design is cuboid (Figure 1c) and has stagnant air pockets around the cells. For the Li/SO₂ BA-5590, thermocouples were placed on the center of each face of the battery case and were not located directly over each cell. It should be noted that the 1 minute, 100 W pulse

Fig. 8- Voltage and temperature profiles of one BA-5X90 battery (5s2p) - LIC hybrid system cycled according to the test protocol (1 minute at 100 W, 9 minutes at 12 W, repeat): (a, b) hybrid incorporating BA-5790 (10 Li/CF_xMnO₂ D-cells; thermocouples placed on case directly opposite individual cells), and (c, d) hybrid incorporating BA-5590 (10 Li/SO₂ D-cells; thermocouples placed on the center of each face of the battery case).

experiments conducted during this investigation represents extreme conditions and is not representative of field use. A considerable reduction in cell temperature can be achieved by reducing the pulse duration to 30 seconds. In the case of the hybridized Li/CF_xMnO₂ half-BA-5790 (5s1p), limiting the pulse duration to 30 seconds while maintaining the previous current settings on the power management board reduced the highest cell temperature by 15°C.

The hybrid architecture was shown to also benefit the performance of the standard military Li/SO₂BA-5590 battery. Attachment of the Power Augmentor to a single BA-5590 allowed 47 cycles (Figure 8c) compared to 0 cycles with the non-hybridized battery (Section 3.2). Although the LIC was not observed to lower the battery operating temperature (Figure 8d), it did minimize voltage delay. The initial 100 W pulse remained above 10 V (Figure 8c), compared to the non-hybridized system which resulted in a voltage drop to 9.38V for two, BA-5590s (5s4p).

3.4 Comparative Analysis

To more clearly understand the potential benefit of a hybrid Li/CF_xMnO₂-LIC design, a weight and volume analysis comparing hybrid systems to the individual Li/SO₂(BA-5590) or Li/CF_xMnO₂(BA-5790) batteries was conducted. Table IV summarizes the battery/hybrid weight and volume required for a 1-minute 100 W pulse/9-minute 12 W load profile over various mission durations (8 h, 16 h, 24 h). Two BA-5590 Li/SO₂ batteries are presently needed to meet this requirement and they have a weight and volume of 2060 g (2 x 1030 g) and 1766 cm³ (2 x 883 cm³) respectively. Thus, only those hybrid systems not exceeding 2060 g in weight or 1766 cm³ in volume were analyzed. Hybrid calculations were based on the Power Augmentor design unless otherwise noted.

The comparative analysis demonstrated that hybridization of a Li/CF_xMnO₂ BA-5790 battery (5s1p configuration) with a 1,000 F LIC led to a significant savings in weight and volume for missions 8 hours or less. The Power Augmentor was found to reduce the weight and volume of the power source by

55%(wt.) and 45%(vol.) with respect to BA-5590 (Li/SO₂, 5s4p) battery and by 42% (wt.) and 27% (vol.) with respect to BA-5790 (Li/CF_xMnO₂, 5s3p) battery. It is important to note that these benefits were observed despite the LIC, power management board, and their assembly being far from optimized. Additional benefits in weight and volume would likely be realized with further refinement. Within the context of the current design, the advantage for durations between 8 – 16 hours is less clear. The hybrid (5s2p) is superior (30% lighter, 20% smaller) to the existing BA-5590 (Li/SO₂, 5s4p). However, although the hybrid (5s2p) is 10% lighter than the BA-5790 (5s3p) it is 6% larger. This trade-off between weight and volume may be advantageous with respect to the Li/CF_xMnO₂ electrochemistry as hybridization would reduce the battery operating current and thus lower the operating temperature.

It should be further noted that the results presented in this study suggest that the benefit of the hybrid becomes more pronounced as the power level is increased. This is a logical conclusion, as the LIC is a power-dense device implying significant benefit will be realized in the area of increased load-power requirements. For example, increasing the power level to 140 W from 100 W prohibits the use of the 1½ BA-5790 (5s3p) since the 3 A fuse limit per 5s1p battery string is exceeded and an additional ½ BA-5790 (5s1p) is required. However, a hybrid configuration implementing the 1000F LIC with 1 BA-5790 (5s2p) is capable of supporting a 1-minute, profile with a high-power pulse of 140W. In regards to the comparative analysis during 1-minute 140 W challenges, the hybrid provides ~30% improvement in weight and ~20% improvement in volume. Substantially greater benefit can be realized using the hybrid technology as power levels are further increased. This is a noteworthy finding as the results suggest that advanced portable electronics requiring greater power may be developed since they can now be supported by this technology without substantial penalty of weight.

4. Conclusions

To improve the high-power performance of lithium batteries containing aCF_x cathode material, we investigated the use of a hybrid system utilizing a LIC. An intelligent power management board was

developed to manage the energy flow between the battery and the capacitor. We showed that a detachable hybrid device, consisting of a 1,000 F LIC and a power management board, allows the military to utilize high energy density $\text{Li/CF}_x\text{MnO}_2$ (BA-5790) batteries for applications requiring both low and high power. The capacitor was able to buffer the high-power demands of the load, thus reducing the stress on the battery, lowering cell operating temperature and achieving greater energy utilization. One $\text{Li/CF}_x\text{MnO}_2$ BA-5790 battery hybridized with a LIC delivered approximately the same capacity as two, Li/SO_2 BA-5590 batteries when cycled under a simulated communications testing protocol (100 W for 1 minute, 12

W for 9 minutes). A hybrid design could reduce the weight and volume of batteries required for high pulse-power devices. In theory, this would reduce the number of battery types used by the military and thus improve logistics and affordability. It should be noted that the benefits of a hybrid architecture observed in this study were achieved using an un-optimized LIC due to limited availability of a newly emerging technology. Increasing the capacitance/energy density of the LIC, or accepting a 45-second pulse instead of a 60-second pulse, would allow the full benefit of the hybrid to be realized. Future efforts will focus on using higher energy density LICs as they become available. This would allow the LIC to meet the entire 100 W one-minute pulse requirement, thus lowering the $\text{Li/CF}_x\text{MnO}_2$ cell current ($2.5 \text{ A} \rightarrow 1.2 \text{ A}$) and reducing BA-5790 battery operating temperatures.

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Table I. Comparison of Lithium Primary D-Cell Technologies

	Li/SO ₂	Li/MnO ₂	Li/CF _x	Li/CF _x MnO ₂
Capacity at 2 A (Ah)	7.5	11	19	16
Open Circuit Voltage (V)	3.0	3.3	3.0	3.3
Nominal Voltage (V)	2.8	3.0	2.6	2.6
Operating Temperature (°C)	-60 to +70	-40 to +72	-20 to +90	-40 to +85
Weight (g)	85	115	69	81
Specific Energy (Wh/Kg) @12 V	255	287	716 (Al case)	514 (Al case)
Manufacturer, Number	SAFT, LO 26SX*	Ultralife, U10013**	Spectrum Brands, BR-20 ⁺	Eagle Picher, LCF-133 ⁺⁺

* Saft specifications document 31033-2-1005 (October 2005). ** Ultralife specifications document UBI-5099, Rev F20 (October 2010).⁺Rayovac marketing data sheet 123-BA02A (March 2016).⁺⁺Eagle Picher preliminary technical data sheet for Li-CF_x/MnO₂ hybrid.

Table II. Performance of D-Cells Discharged at Various Constant Currentsto a 2.0V Cutoff

Current	Li/SO ₂		Li/CF _x MnO ₂		
	Capacity	Highest Temp.	Capacity		Highest Temp.
(A)	(Ah)	(°C)	to 87°C (Ah)	to 70°C (Ah)	(°C)
1	7.5	37	15.0	15.0	49
2	7.1	45	15.2	15.2	69
2.5	7.1	47	15.2	7.0	81
3	7.0	52	10.1	7.1	87

Table III. Results of Hybrid Testing

Energy Storage Device	Cycles 1 min @ 100W, 9 min @ 12W	Disposable Unit		Reusable Unit		Total Weight/Volume (g/cm ³)
		Weight (g)	Volume (cm ³)	Weight (g)	Volume (cm ³)	
1 Li/SO ₂ BA5590 (5s2p)	0	1006	883	0	0	1006 /883
1 Li/CF _x MnO ₂ BA5790 (5s2p)	0	1030	883	0	0	1030/883
1½Li/CF _x MnO ₂ BA5790 (5s3p)	158	1561	1325	0	0	1561/1325
2 Li/SO ₂ BA5590 (5s4p)	100	2012	1766	0	0	2012/1766
2 Li/CF _x MnO ₂ BA5790 (5s4p)	208	2060	1766	0	0	2060/1766
1 Li/SO ₂ BA5590 (5s2p) + LIC	47	1006	883	380	523	1386/1406
½ Li/ CF _x MnO ₂ BA5790 (5s1p) + LIC	58	531	442	380	523	911/ 965
1 Li/CF _x MnO ₂ BA5790 (5s2p) + LIC	96	1030	883	380	523	1410/1406
1½ Li/CF _x MnO ₂ BA5790 (5s3p) + LIC	155	1561	1325	380	523	1941/1728

Table IV. Weight and Volume Comparison of Battery/Hybrid Systems
(Power Source Cavity:~6.2 cm x ~25.4 cm x ~11.2 cm)

Power Source	8 Hour Duration		16 Hour Duration		24 Hour Duration	
	Total Weight (g)	Total Volume (cm ³)	Total Weight (g)	Total Volume (cm ³)	Total Weight (g)	Total Volume (cm ³)
LIC + ½ BA-5790, 5s1p	911	965	-	-	-	-
LIC + 1 BA-5790, 5s2p	1410	1406	1410	1406	-	-
LIC + 1½ BA-5790, 5s3p	1872*	1766*	1872*	1766*	1872*	1766*
LIC + 1 BA-5590, 5s2p	1386	1406	-	-	-	-
1 BA-5590, 5s2p	-	-	-	-	-	-
2 BA-5590, 5s4p	2012	1766	2012	1766	-	-
1 BA-5790, 5s2p	-	-	-	-	-	-
2 BA-5790, 5s4p	2060	1766	2060	1766	2060	1766
1½ BA-5790,5s3p	1561	1325	1561	1325	1561	1325

* Uses Design 1 (Single-Unit).