Preliminary evaluation of rechargeable lithium cells for a totally-implantable ventricular assist device

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Abstract

A preliminary evaluation of the performance characteristics for three types of rechargeable, 'AA' size, lithium cell chemistries, namely Li/TiS_2 (two different manufacturers), Li/MoS_2 and Li/MnO_2, was carried out in order to determine their potential usefulness in the internal (implanted) battery for the electrohydraulic ventricular assist device (EVAD) being developed. The major parameters studied at 37 °C were discharge rate capability, self-discharge and cycle life. The cycle life of the lithium cells above the minimum 30 min discharge time specified for EVAD were short, with the Li/MoS_2, Li/MnO_2 and two Li/TiS_2 cells giving 80, ~11, 37 and 101 cycles, respectively, under pulsed discharge conditions. The 24 h, self-discharge study of all the cells at 37 °C showed <1.2% decrease in capacity. Discharge rate studies showed that the Li/TiS_2 cells from both manufacturers offered higher observed specific energies (85 and 133 W h/kg) and energy densities (203 and 273 W h/l), lower internal resistances (155 and 84 mΩ) and larger observed capacities (0.83 and 1.00 A h) when compared to the Li/MoS_2 (49 W h/kg, 126 W h/l, 153 mΩ and 0.58 A h, respectively) and Li/MnO_2 (56 W h/kg, 131 W h/l, 350 mΩ and 0.39 A h, respectively) cells operating under average EVAD load conditions. The cycle life and operating times of cells that were pulse discharged to mimic actual EVAD operating conditions were shorter than those that underwent cycling with an average EVAD load. When compared to other energy sources and the EVAD design specification, it was concluded that none of these prototype lithium cells were currently suitable for use in the EVAD due to their low cycle life.

Introduction

One of the common disorders causing suffering and premature death in our society is heart disease for which a mechanical pump, such as a total artificial heart (TAH) or a ventricular assist device (VAD) would provide a clinically viable solution. Researchers have been developing such devices [1–3] and it has been proven clinically that these pumps could be used to sustain blood circulation until the recovery of the natural heart [4–7] or as a bridge to transplantation of a donor heart [8, 9].

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A totally-implantable electrohydraulic ventricular assist device (EVAD) is under development in this laboratory and has been previously described [10, 11]. A key component in the development of these devices is the need for a rechargeable, high-energy, reliable, safe, implantable battery. The performance characteristics at 37 °C of three types of nickel/cadmium (Ni/Cd) cells for possible use in the EVAD's internal battery have been previously described [11, 12]. The use of Ni/Cd cells yields an internal battery that is heavy and has limited operating capacity. Rechargeable Li cells have higher specific energy (W h/kg) and energy density (W h/l) than rechargeable Ni/Cd cells, due in part to their lower anode weight and higher electrochemical cell potential. These properties provide advantages over the Ni/Cd cells, as a smaller size and lighter weight battery may be implanted while still providing the same, or longer, periods of operation. Therefore, the investigation of rechargeable Li cells for their potential use in the EVAD is of great interest.

Untereker [13] has described several primary Li chemistries that have been successfully used in the pacemaker industry. These cells have a very long shelf life and are ideally suited for long-term implants with a low energy demand (i.e., in the order of μW). However, the power requirement for the EVAD (estimated to be 10–10.5 W, with an average operating voltage of 12 V) is several orders of magnitude greater than that of a pacemaker or most implanted devices. An estimated average discharge current of 0.8–0.9 A will be demanded of the internal battery during the EVAD's operation. Based on the size of one manufacturer's defibrillator [14], a maximum volume of 120 ml and thickness of 2.0 cm was specified for EVAD's internal battery, which precludes the use of cells larger than the 'AA' size. These power demands on the cells could result in discharge rates as high as 1.5 C, depending on the cell chemistry. It has also been demonstrated that pulse discharge conditions more accurately mimic the power requirements of the implanted EVAD [12]. However, few studies have been reported in the literature dealing with the 37 °C operating performance of rechargeable Li cells at high discharge rates. For example, one recent study involved investigating the capacity and cycle life characteristics of Li/Li_xCoO_2 cells while operating at 37 °C under an average pulsatile load of 1.0 A [15].

In this paper, a preliminary study of performance characteristics, such as discharge rate capability, self-discharge and cycle life under constant current and pulse current discharge, were determined for three rechargeable Li chemistries. The cell chemistries studied here included lithium/manganese dioxide (Li/MnO_2), lithium/molybdenum disulfide (Li/MoS_2) and lithium/titanium disulfide (Li/TiS_2, two different manufacturers). Only a limited number of cells of each type were utilized in this study. The feasibility of using rechargeable Li cells for implantation in the EVAD application is discussed in terms of these preliminary results. Although nonrechargeable Li cells are already used in pacemakers and defibrillators, the use of rechargeable Li cells in implantable devices does raise some safety concerns. However, for the purposes of this study, the safety and reliability of rechargeable Li cells have not been evaluated.

Experimental

The rechargeable Li/TiS_2 cells from manufacturers #1 and #2 were rated at 0.9 and 1.0 A h, respectively. The Li/MnO_2 cells were rated at 0.6 A h and the Li/MoS_2 cells were rated at 0.8 A h. All of the cells used in this study were 'AA' size, precommercial or research prototypes. Therefore, the product uniformity and quality

TABLE 1

Number of cells examined in each study

Cell type	Discharge rate study	Self-discharge study	Capacity fade study	
			Constant current	Pulsed current
Li/MoS_2	2	2	3	2
Li/MnO_2	2	2	1	2
Li/TiS_2 #1	2	2	2	4
Li/TiS_2 #2	3	2	1	2

control of these cells may be lower than that found in the final commercially-available product.

The cells were cycled using an automatic battery cycler (Techware Systems, Model ABC). The battery cycler was controlled by a Stride computer (Model-430, Stride Micro) and supported by battery cycling software developed by Techware Systems. Unless otherwise stated, all experiments were carried out at 37 ± 0.2 °C in an environmental chamber (Tenney Engineering, Inc., Model TH-Jr.).

In order to break up the protective Li/electrolyte reaction film on the Li anode and to standardize the cells for use in the subsequent experiments, all cells were precycled prior to their use in the study. This precycling consisted of ten cycles with a charge rate of 0.1 C and a discharge rate of 0.3–0.5 C at 37 °C. Table 1 shows the number of cells tested in each of the following studies. As this Table shows, the interpretation of the results from this preliminary report is limited by the small number of cells examined in each category.

Discharge rate

The cells used in this study were charged at a 0.1 C rate and discharged at various rates ranging from 0.3–2.5 C. The cells were discharged several times throughout the study at a 0.3 C rate in order to correct the observed capacities for natural capacity fade due to cycling. The average discharge currents (0.8–0.9 A) expected for the EVAD translate into discharge rates of 0.8–1.0 C for the two types of Li/TiS_2 cells, 1.3–1.5 C for the Li/MnO_2 and 1.0–1.1 C for the Li/MoS_2 cells.

Self-discharge

The cells used in this study were charged at a 0.1 C rate and were then discharged at 0.875 C for the Li/TiS_2 #1 cells, 0.3 C for the Li/TiS_2 #2 cells and 0.5 C for the Li/MnO_2 and Li/MoS_2 cells. A series of open-circuit periods ranging from 1–24 h were imposed in between the charge and discharge steps. In addition, cycles containing no open-circuit period were imposed in between the cycles that contained an open-circuit period in order to correct the observed capacities for natural capacity fade due to cycling.

Capacity fade

In this paper, the cycle life (CL_{50}) of a cell is defined as the number of charge/discharge cycles that a cell undergoes while retaining its capacity above one half of its original or first cycle value. The charge and discharge steps were terminated at

TABLE 2

Charge and discharge termination voltages, cycle life (CL_{50}) and initial operation times (0.833 A average discharge) for the rechargeable lithium 'AA' size cells at 37 °C

Cell type	End of charge (V)	End of discharge (V)	Discharge conditions	CL_{50} (cycles)	Initial operating time (min)
Li/MoS_2	2.3	1.3	Pulse current	188	38
			constant current	468	41
Li/MnO_2	3.4	2.4	Pulse current	169	28
			constant current	182	35
Li/TiS_2 #1	2.6	1.6	Pulse current	44	52
			constant current	53	60
Li/TiS_2 #2	2.7	1.7	Pulse current	97	67
			constant current	92	73

the manufacturer's recommended voltages limits listed in Table 2 for the four types of Li cells used in this study. The charge current was a 0.1 C rate for all four types of cells. Two types of discharge regimes were employed, one using a constant current of 0.833 A, representing an average EVAD load of 10 W for a 12 V battery, and the other using a pulse current regime, which also averaged 0.833 A. The pulse regime was designed to mimic the diastolic and systolic load demands of the pumping heart as required by the EVAD and consisted of 0.4 s at 0.416 A followed by 0.2 s at 1.667 A. This pulse discharge cycle was repeated until the cell reached its discharge termination voltage. The pulse current was supplied by a power level simulator (PLS) developed in our laboratory.

Results and discussion

Discharge rate

There were three main reasons for evaluating discharge rate capability of the different Li cell technologies. The discharge rate study is designed to determine if a cell can provide sufficient capacity under a particular discharge current, to determine its operating time at these load conditions, and to observe the cell's discharge voltage-time profile under various load conditions.

Figure 1 shows the normalized capacity, as a function of discharge rate, for each type of cell. The observed cell capacities were first corrected for their natural capacity fade due to cycling. Capacity normalization was then carried out by dividing the measured cell capacity at a particular discharge rate by the measured capacity at the 0.3 C discharge rate. The discharge rate was obtained by dividing the selected discharge current used by the nominal, or manufacturer's rated, ampere-hour capacity for each type of cell. It was found that the pair of cells of each type behaved similarly and all of the cell types showed an approximate linear relationship between the capacity and discharge rate at low rates. However, at higher discharge rates the capacity of the cells dropped off at a quicker rate due to increased polarization within the cells. The Li/MnO_2 cells showed the largest decrease in capacity with increasing discharge rate, whereas the Li/TiS_2 #1 and #2 cells gave the smallest decrease in capacity with increasing discharge rate.

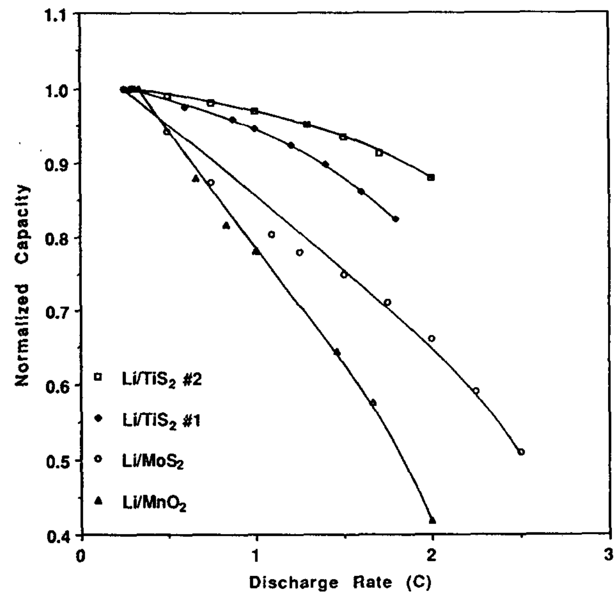

Fig. 1. Normalized capacity as a function of discharge rate for the four lithium 'AA' size cells at 37 °C.

Cell voltages are typically depressed with increasing discharge currents. The midpoint voltage (MPV) is defined as the cell voltage at one half of the cell's discharge capacity and can be determined from the cell's discharge voltage-time profile. Figure 2 shows the MPV, as a function of discharge rate, for the Li cells studied. All four types of Li cells show a linear decrease in MPV with increasing discharge rate. This linear relationship reflects the fact that ohmic polarization within the cells in the factor resulting in the observed 'IR' or voltage drop. The effect of the discharge rates on the discharge voltages in the Li/TiS_2 #2 cells having the smallest slope and the Li/MnO_2 cells having the largest slope. If Ohm's law is assumed, the effective internal d.c. resistance of the cells at their 50% capacity point (R_{50}) can be obtained from the slopes of the lines in a similar graph to that of Fig. 2 plotting MPV versus discharge current (A). Based on these slopes, Table 2 shows that the Li/TiS_2 #2 and Li/MnO_2 cells have the lowest and the highest internal d.c. cell resistances, respectively. The internal d.c. resistance observed for the Li/MoS_2 cells in this study is similar, albeit lower, to the literature [16] value of 220 mΩ. It should be noted that, a comparison between the internal cell resistances measured by the d.c. and a.c. methods was not carried out for these Li cells. However, other studies in our laboratory on Ni/Cd cells have shown that internal cell resistances measured by this d.c. method are 2–3 times higher than those measured by the a.c. method. As shown by the data in Figs. 1 and 2, the higher the internal d.c. cell resistance, the greater the voltage is depressed at a particular discharge rate, and the greater the reduction in a cell's capacity. This reduced cell capacity is the result of an increasingly-depressed cell voltage reaching the constant termination voltage more rapidly as the discharge rate is increased.

Table 3 also shows wide variations in the performance of the four types of Li cells under EVAD load conditions. The Li/TiS_2 #2 cells have the best discharge rate capability, as demonstrated by their high discharge capacity and long operating time.

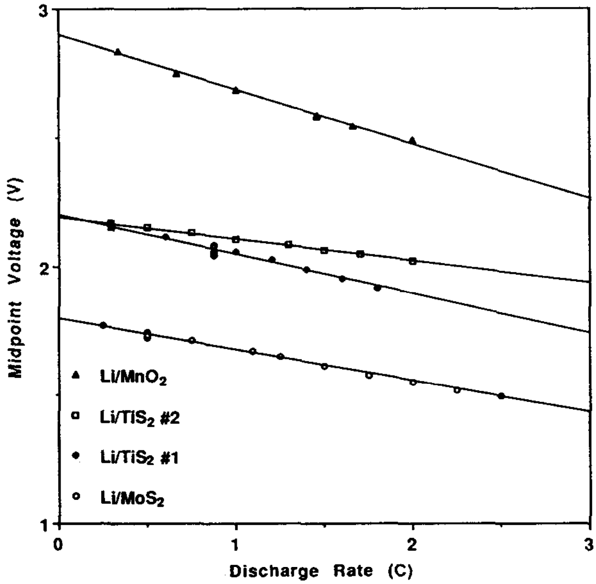

Fig. 2. Midpoint voltage (MPV) as a function of discharge rate for the four lithium 'AA' size cells at 37 °C.

TABLE 3

Performance characteristics of the rechargeable lithium 'AA' size cells (0.875 A discharge) at 37 °C

Characteristics	Li/MoS_2	Li/MnO_2	Li/TiS_2 #1	Li/TiS_2 #2
Manufacture's rated capacity (A h)	0.8	0.6	0.9	1.0
Midpoint voltage (V)	1.67	2.59	2.07	2.10
Capacity (A h)	0.58	0.39	0.83	1.00
Percent of rated capacity (%)	72	65	92	100
24 h capacity loss (%)	1.2	0.4	0.1	0.1
Initial operating time (min)	40	27	57	69
Internal resistance (R_{50}, mΩ)	153	350	155	84
Cell weight (g)	19.92	18.16	20.20	15.75
Cell volume (ml)	7.70	7.70	8.47	7.70
Specific energy (W h/kg)	49	56	85	133
Energy density (W h/l)	126	131	203	273

In addition, these cells have the highest observed specific energy and energy density. The Li/TiS_2 #1 cells are very similar to the Li/TiS_2 #2 cells and, not surprisingly, are rated a close second. The Li/MoS_2 and Li/MnO_2 cells show moderate discharge rate capability. However, the lower cell voltage and capacity of Li/MoS_2 cells, as well as their higher cell weight (see Table 3), resulted in a lower specific energy and energy density and make them less attractive for the EVAD application. The lower cell capacity of Li/MnO_2 cells also resulted in lower specific energy and energy density despite their higher cell voltage. The specific energies and energy densities were calculated using the average weights and volumes, respectively, of the cells examined in this study. These average cell weights and volumes are listed in Table 3.

Self-discharge

Most rechargeable cells lose some of their capacity on standby after they have been charged and the rate of capacity loss is different for different cell chemistries. Such capacity loss is defined as self-discharge. Temperature is also an important factor in the rate of self-discharge. Most Li cells in the EVAD application will require between 5–10 h to be fully charged, assuming an operating time of one half hour per day at the approximate *C* rate discharge demanded by the EVAD. Therefore, the cells will be on open circuit for the remaining 13.5–18.5 h of the day. As a result, a cell chemistry having a low self-discharge rate is desirable for the EVAD application.

Figure 3 shows the normalized capacity, as a function of open-circuit period, for the Li cells studied. These capacities have been corrected for natural capacity fade due to cycling and have been normalized in order to better compare the four types of cells. The normalization was carried out by dividing each discharge capacity by the capacity value first observed for that cell. Table 3 lists the percentage of capacity lost with respect to the initial capacity for each type of cell after a open-circuit period of 24 h at 37 °C. This maximum period of 24 h was selected as a typical amount of time between deep discharge uses of the internal battery by the patient. As is shown in Fig. 3 and Table 3, all of the cells examined showed low self-discharge rates with

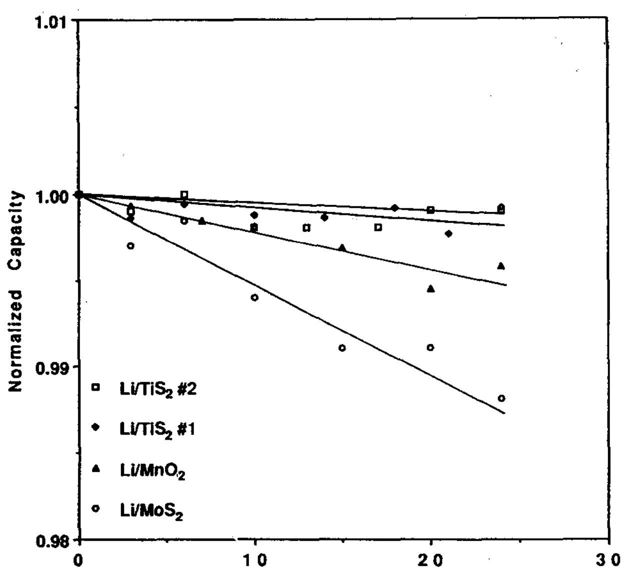

Fig. 3. Normalized capacity as a function of open-circuit time for the four lithium 'AA' size cells at 37 °C.

normalized capacities <1.2% of their initial capacity after 24 h. In a similar study at 37 °C [11], Ni/Cd cells showed a 10–15% reduction in capacity over the same time period. Loss of this much capacity or operating time for the Ni/Cd cells may be a potential safety problem. It was also found that the pair of cells of each type behaved similarly with respect to the amount of capacity loss during a particular open-circuit period.

Capacity fade

Rechargeable Li cells have been known to have a lower cycle life than rechargeable Ni/Cd cells because of the highly-reactive nature of the Li metal anode toward the electrolyte [17–19]. The irreversible reaction of Li with the electrolyte results in a loss of active electrode material and, hence, a loss of cell capacity during cycling. Many studies have been carried out to investigate the stability of Li metal or its alloys toward various electrolytes and combinations of electrolytes in order to improve the rechargeability of the Li electrode [20–25]. Operating temperature also affects the performance of rechargeable Li cells. It has been found in our laboratory, as well as others [26], that the cycle life and capacities of Li/MnO_2 cells cycled with moderate discharge rates (0.3–0.7 C) at 37 °C were greater than those observed for the same cells at room temperature. This increased discharge capacity is most likely due to higher electrolyte conductivity at 37 °C and, hence, a lower internal cell resistance. We speculate that the increased cycle life at 37 °C for these cells may be due to an increased Li cycling efficiency, which may result from the quicker formation of a denser than usual protective Li/electrolyte reaction film that reduces any further interaction and reaction between the Li metal anode and the electrolyte.

The determination of the cycle life for the rechargeable Li cells under the EVAD operating conditions is important for several reasons. The physician or medical engineer must know the amount of service life that can be expected from the internal battery and when it should be replaced. Important diagnostic information on the internal battery is also obtained by knowing trends, such as those observed in the discharge capacity fade during battery cycling, that indicate a normal battery as opposed to those trends that identify an abnormal battery which needs to be replaced.

It was found in our study that several cells of the same type, undergoing the same cycling regime, yielded varying results. These differences were possibly due to the fact that most cells were hand-made research prototypes and that product uniformity available in commercial production was therefore absent to some degree. Therefore, we have only presented the best results obtained in our studies. We feel this is justified, as one would expect improved cell performance from a commercial-ready cell that has had more stringent quality assurance and control measures applied in its manufacturing process.

Figure 4 shows the operating time, as a function of cycle number, for the four types of lithium cells. These were tested under both pulse and constant current discharge conditions (0.833 A average) that represent of an average EVAD load of 10 W for a 12 V battery. All of the cells were cycled at a 100% depth-of-discharge in this study, with the Li/TiS_2 #1, Li/TiS_2 #2, Li/MoS_2 and Li/MnO_2 cells experiencing relative discharge rates of 0.93 C, 0.83 C, 1.04 C and 1.39 C, respectively. The capacity fade trend for the four types of Li cells under pulse and constant current discharge are quite similar. However, there were a number of discontinuities in the capacity fade trend for the Li/MnO_2 and Li/MoS_2 cells. This may have been the result of temporary electrical isolation and later reconnection of cathode and/or anode materials to their respective current collectors.

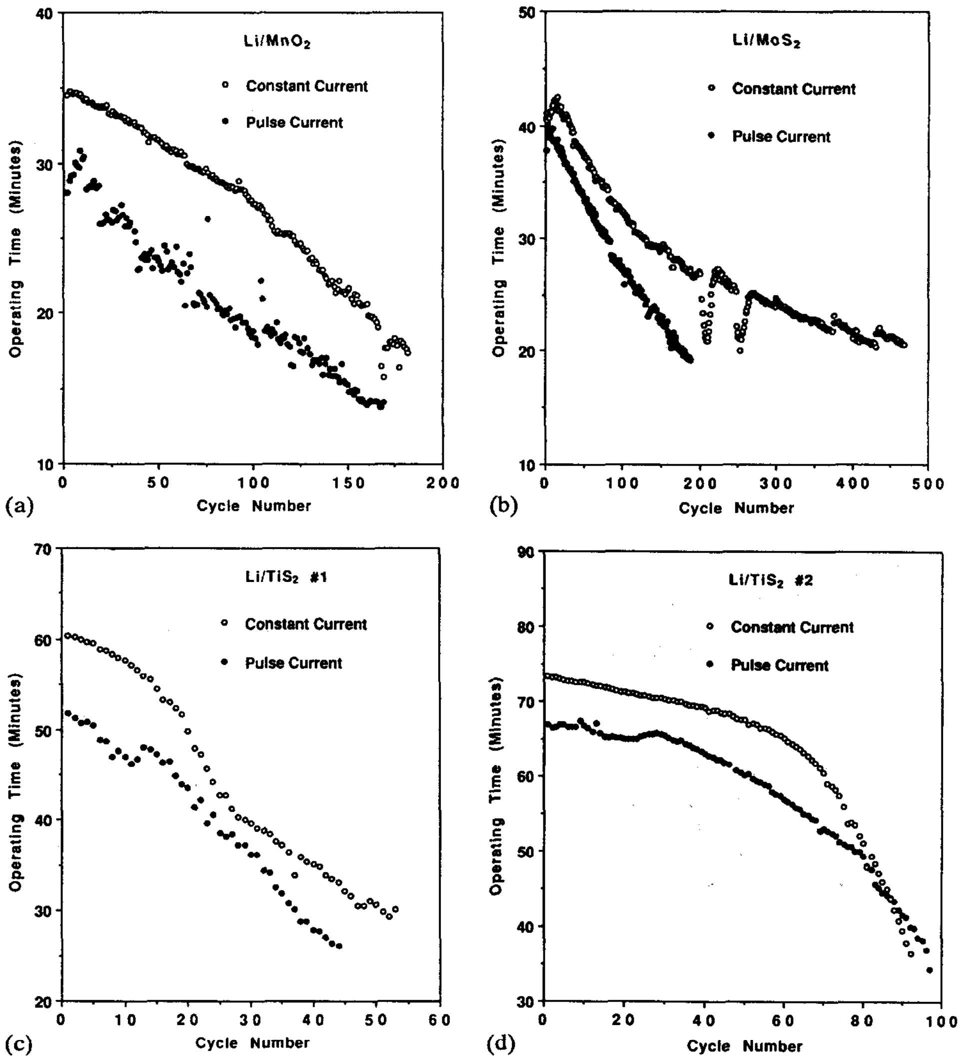

Fig. 4. Operating time as a function of cycle number for the four lithium 'AA' size cells operating under constant current or pulse current discharge (0.833 A average discharge) at 37 °C.

The Li/MoS$_2$ and Li/MnO$_2$ cells behaved similarly and exhibited relatively low capacity fade rates. On the other hand, the Li/TiS$_2$ #1 cells exhibited the fastest capacity fade of the four types of cells studied. Table 2 lists the cycle life (CL_{50}) and initial operating times for the cells under their specific cycling conditions. Although having shorter operating times than the Li/TiS$_2$ cells, the Li/MoS$_2$ and Li/MnO$_2$ cells gave longer cycle life even at higher relative discharge rates. Figure 4 and Table 2 also show that the cycle life of the cells that were pulse discharged were shorter than those that underwent constant current discharge cycling. The Li/TiS$_2$ #2 cells were the only exception to this observation. As is also shown in Fig. 4 and Table 2, the

TABLE 4

Comparison of cycle life to 50% of initial capacity (CL_{50}) and above 30 min discharge time (0.833 A average discharge) for the rechargeable lithium 'AA' size cells at 37 °C

Cell type	Discharge conditions	CL_{50} (cycles)	Above 30 min (cycles)
Li/MoS_2	Pulsed current	188	80
	constant current	468	127
Li/MnO_2	Pulsed current	169	~11
	constant current	182	64
Li/TiS_2 #1	Pulsed current	44	37
	constant current	53	50
Li/TiS_2 #2	Pulsed current	97	101
	constant current	92	96

operating times for those cells discharged under the pulse current were shorter than those discharged under constant current for all four types of Li cells studied. This observation is most likely due to the cell voltages being depressed during the higher pulse current discharge, which will lead to the cells reaching their discharge end-point voltage more rapidly and yielding shorter operating times.

It is envisioned that the EVAD patient would utilize their internal battery only once per day for a 30 min period. Therefore, the initial depth-of-discharge would be as little as 40–60% for the Li/TiS_2 cells, 85–100% for the Li/MoS_2 cells and 70–80% for the $LiMnO_2$ cells, which would extend their cycle life. If cycle life for EVAD is defined as the number of cycles yielding more than 30 min of discharge time then, as seen in Table 4, the Li/MoS_2 cell gave slightly more cycles under constant current discharge conditions than did the Li/TiS_2 #2 cell. However, this relationship is reversed when pulsed current conditions were used, with the Li/TiS_2 #2 cell yielding slightly more cycles than did the Li/MoS_2 cell. The cycle life above 30 min discharge time for the Li/TiS_2 #1 and Li/MnO_2 cells were relatively similar and much shorter than the other cells.

Conclusions

The performance of prototype Li/TiS_2, Li/MoS_2 and Li/MnO_2 cells were studied at 37 °C in this preliminary evaluation. Their performance, in terms of their discharge rate capability, self-discharge and cycle life, under the EVAD operating conditions were evaluated and compared. The internal d.c. cell resistance for the Li/TiS_2 #1 and Li/MoS_2 cells were moderate, while the Li/MnO_2 cells showed the highest internal resistance and the Li/TiS_2 #2 cells showed the lowest. All four types of cells exhibited insignificant rates of self-discharge for the EVAD application during a 24 h open-circuit period at 37 °C. It was found that both of the Li/TiS_2 cells offered higher specific energies and energy densities than the other Li cells studied.

The Li/TiS_2 cells that were studied provided longer operating times than the other Li cells. The initial operating times of about 1 h for the Li/TiS_2 cells are suitable for the EVAD application. The Li/MoS_2 and Li/MnO_2 cells experienced higher relative discharge rates and, yet, exhibited a greater number of cycles to 50% of their initial capacity (CL_{50}) than did either of the Li/TiS_2 cells. However, if cycle life for EVAD is defined as these number of cycles yielding more than 30 min of discharge time then the cycle lives of the cells are quite short. Therefore, assuming a daily operating time of only 30 min during the required one year minimum implant time (i.e. 365 cycles), none of the Li cells studied here would be able to meet this EVAD design specification, although the Li/TiS_2 #2 cell may come close. Additional cycling studies using a 30 min depth-of-discharge under pulse discharge conditions at 37 °C will be required in order to confirm this.

This study also found that pulsatile current loads, such as that demanded by the EVAD, have an effect on the cycle life and capacities of rechargeable Li cells. The cycle life and operating times of the cells that were pulse discharged were shorter than those that underwent constant current discharge cycling. Additional studies are required in order to determine whether there are any safety problems associated with pulse discharging rechargeable Li metal-based cells under EVAD operating conditions.

In conclusion, none of the four types of Li cells studied in this paper are presently able to fulfil the performance requirements required for EVAD's internal battery. Significant improvements in cycle life and possibly higher cell capacity are required before these cells may be considered for use in an implantable device such as the EVAD. Other factors, such as safety and reliability of rechargeable Li metal-based cells, are also major concerns with respect to the implantable use of these cells. These issues must be addressed once the cells are able to meet all of their performance requirements. For these and other reasons, rechargeable Li metal-based cells will not be considered as a candidate for the EVAD implantable power source at this time.

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