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NAVAL SURFACE WARFARE CENTER
DAHLGREN DIVISION • WHITE OAK DETACHMENT
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FOREWORD

The Naval Mine Warfare community has a need for improved availability, reliability, and performance of batteries required to power mines and mine countermeasures. A secondary issue in battery development is to reduce the number of variables in these batteries. To accomplish these goals, a standard family of cells for mine warfare applications has evolved. The current program effort requires the development of a AA-size cell which can be used in several mine batteries.

This report describes the availability and chemistries of various AA cell technologies and presents a test plan for their evaluation for mine battery development.

We wish to thank Mr. Clyde W. Bowers (NSWCMWEA-Code 7000) and Mr. Frank Visk (NSWCDDWODET-Code G94) for providing system requirements used in the test plan preparation and Mr. Clinton Winchester of the Electrochemistry Branch (Code R33) for beneficial editorial comments.

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Approved by:

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ABSTRACT

As part of an effort to reduce battery procurement problems in the Navy, a program has been developed to standardize battery chemistries and cell sizes. Mine batteries are being developed from a standardized family of cells. Currently, several mercury-based cells are being used in mine batteries; however, these have limited energy and power densities and present uncertain long-term availability and disposal issues. The lithium/thionyl chloride electrochemical technology is being considered as a long-term solution to these problems.

This report describes the surveillance effort that gave rise to selection of AA-size lithium/thionyl chloride cells for inclusion in the standard family of cells for mine battery development. A test plan to verify this choice and to identify potential cell or battery production and performance problems is also provided.

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CHAPTER 1

INTRODUCTION

The Navy has experienced some difficulty in the procurement of batteries for mine warfare systems. Because of unique mine system requirements, mine battery development has led to a proliferation of designs, and virtually every system has its own unique battery. In turn, each battery may use a unique cell because of the host of different chemistries available. This leads to low procurement quantities with a subsequent decrease of interest by available suppliers and increased qualification and production costs. Changing environmental policies, which are severely restricting the availability of mercury-based batteries in current use, were also an important consideration in the selection process.

A program has been developed to resolve problems of procuring batteries for mine warfare systems. This program relies on a standard set of cells which can be combined to meet the wide range of Navy mine applications. A family of three cell sizes has been developed to meet the long-term needs of a series of Navy mines. An A-size cell has been designed to meet long, low-rate discharge requirements. C-size and #6-size cells have been designed to provide low, moderate and pulse power for mine batteries. This standard family of cells is to be extended to include a AA-size cell for use in mines and mine countermeasures.

The stringent power requirements of future weapons systems necessitate replacing aging technologies with the higher energy lithium battery technology. An evaluation of commercial lithium AA-size cells was undertaken to assess the feasibility of incorporating them into the standard family of primary cells. To initiate the program, potential battery manufacturers were requested to provide technical product information with an emphasis on the ability to meet military requirements, including long term storage and low temperature operation. From a list of 31 possible suppliers, approximately a dozen viable candidates emerged. Two companies which are foreign suppliers without representatives available in the U.S., i.e., Great Northern (Hellesens) and G. N. Alkaline, were not contacted. Other foreign companies reputed to manufacture AA lithium cells, namely, Sony, Hoppecke, and Toshiba, were nonresponsive. The following companies do not manufacture a AA size lithium cell at this time: Alexander Battery Co., Ballard, BST, Catalyst Research, Duracell, Gates, Moli Energy, Panasonic, Promeon, Ray-O-Vac, Sanyo and Ultra Technologies. The Electrochem Industry Division of Wilson Greatbach is reputed to market a $Li\text{-}SOCl_2$ AA cell made by Maxell. Several of the companies providing AA cells manufactured in the U.S. are foreign owned. These include Battery Engineering Inc. (Hitachi), Power Conversion Inc. (British - Burmingham Tire & Rubber), and SAFT America (French - Alcatel Alsthom). Foreign companies with U.S. distributors include: Hoppecke, Maxell, Tadiran, and Varta. American manufacturers of AA size lithium cells include: Alliant Tech Systems, Eagle-Picher Industries Inc., Energy Conversion, EIC Laboratories Inc., Eveready, and Yardney Technical Products. Only Eveready and Eagle-Picher provide an off-the-shelf AA lithium cell. Eveready has a 1.5 volt lithium/iron disulfide (Li-FeS_2) cell marketed in Canada under the name LithEon. Eagle-Picher has a high capacity, 3.6 volt, "square AA" cell marketed under the Keeper 2 trademark.

The desire for greater energy density, improved storage life, wider availability, and minimum maintainance resulted in the consideration of only lithium-based cell chemistries. Many of the older, non-lithium battery technologies were precluded from consideration for mine warfare applications because of increasing environmental concerns. In the chapters that follow, the available AA-size lithium cell technologies and their potential use in mine applications are discussed.

CHAPTER 2

LITHIUM PRIMARY CELL TECHNOLOGY

Eight different chemistries produced in the AA-size were considered as potential candidates for mine or mine countermeasure battery applications. The nominal working voltage, capacity, and energy density for each AA-size cell technology are summarized in Table 1. LeClanche, alkaline, and mercury cells have been included for comparison.

A brief summary of available AA-size lithium electrochemical technology follows. Much of the information presented was obtained from manufacturer's literature or via telephone conversations with technical representatives.

LITHIUM/IRON DISULFIDE (Li/FeS_2)

Eveready's LithEon 1.5V No. L91 cell is reported to operate between -40°F and 160°F. The cell typically operates for 150 hours at 1.25V at 100 ohms (12.5 mA) under ambient conditions. The advantages are: the cell is mass produced for the commercial market, operates 3 hours at 0.5A at 1.0V, contains a vent, and has a positive temperature coefficient (PTC) switch that reverts to operation after cooling. The disadvantages are low operating voltage and low energy density relative to other lithium systems.

LITHIUM/COPPER OXIDE (Li/CuO)

The copper oxide series LC 6 cell made by SAFT France employs a solid cathode, bobbin design. It has an open circuit voltage (OCV) of 2.4V and discharges at 1.5V for over 200 hours at 10 mA. The primary advantage of this technology is good retention of capacity after high temperature storage. It can retain 85 percent of cell capacity after 53 months ambient storage which includes cycling at 50°C (122°F) for six hours a day, four days a week. The cell has good voltage regulation even at low temperatures; a constant 1.1 volts can be maintained for 1600 hours at a 1000 ohm load. The capacity decreases rapidly with operating temperature. The cell performs poorly under high rate (>40 mA) discharge.

TABLE 1. COMPARISON OF AA SIZE CELL DATA

CELL CHEMISTRY	NOMINAL WORKING VOLTAGE (Volts)	CAPACITY (Ah)	ENERGY (Wh)	WEIGHT (g)	ENERGY DENSITY (Wh/kg)	(Wh/l)
Zn/C	1.2	1.0	1.2	14.7	80	170
Alkaline Zn/MnO_2	1.2	1.7	2.0	23	86	250
Zn/HgO	1.35	1.85	2.5	30	83	329
Hg/Cd	0.8	2.4	1.9	30	63	238
Li/SO_2	2.8	1.0	2.8	14	200	360
Li/CuO	1.5	3.4	5.0	17.4	275	650
Li/FeS_2	1.2	1.9	2.3	15	152	238
Li/ $CuO_4(PO_4)_2$	2.5	2.3	5.8	17	341	746
$Li/SOCl_2$ standard	3.3	1.8	6.0	17	353	789
$Li/SOCl_2$ square AA	3.5	2.6	9.1	25	364	1022
$Li/SOCl_2$ optimum	3.3	2.2	7.3	17	429	878
Li/ $SOCl_2$-BrCl	3.4	2.0	6.8	16	425	915
Li/ SO_2Cl_2-X	3.4	2.0	6.8	17	400	732
Li/MnO_2	2.8	2.0	5.6	21.5	260	651

LITHIUM/COPPER OXYPHOSPHATE ($Li/Cu_4O(PO_4)_2$)

The copper oxyphosphate series LCP 6, AA cell is manufactured by SAFT France. The cell employs a solid cathode, bobbin construction with a nominal operating voltage of 2.5V. This cell contains no vent and can expell its internal parts upon abuse. It is capable of operating between -40°C and 70°C (158°F). It delivers 2.3 Ah at 1 mA at 20°C. At higher rates, e.g. 75 ohm loads, the voltage regulation becomes less stable and the operating voltage falls off rapidly with decreasing temperatures, decreasing from 2.3V at 70°C to 1.6V at -20°C.

LITHIUM/SULFUR DIOXIDE (Li/SO_2)

This technology is available from Power Conversion Inc. as a AA-size cell, model GO6/1. The major advantages are that it offers good performance over a wide range of temperatures from -54°C to 71°C (-65°F to 160°F), has power density greater than most of the primary cells previously cited, and has a demonstrated five-year shelf life with a projected shelf life of 10 years at room temperature and more than one year at 71°C. This cell chemistry is generally used for high-rate applications.

LITHIUM/MANGANESE DIOXIDE (Li/MnO_2)

There are two suppliers of AA-size Li/MnO_2 cells: Varta Batteries Inc. and Dowty Batteries. There are notable differences in design and performance.

Varta claims cell type CR AA has 2.0 Ah capacity (4.6 mA at 2.8V for 430 hours) at 20°C. The cell construction is a bobbin design. Safety and vibration test data are available for these cells. The cell is designed for good low-rate performance.

The Dowty cell type LIM 145H is a spirally wound cell design with a vent. This cell provides a higher rate capability than the Varta AA cell. The maximum continuous discharge is 275 mA. It has a nominal capacity of 1.1 Ah at 26 mA at 20°C to a 1.5V cutoff voltage.

The Li/MnO_2 solid cathode design offers good storage life with capacity losses of approximately 0.5 percent a year at room temperature. For each 10°C increase in temperature, the self-discharge rate approximately doubles. The manufacturers claim that the Li/MnO_2 system has instant startup with minimal voltage delay after long storage (in comparison to liquid cathodes). The system releases no toxic gases upon venting. However, discharge performance at temperatures below -20°C is poor.

LITHIUM/SULFURYL CHLORIDE (Li/SO_2Cl_2 - X)

Typical performance for an unoptimized Li/SO_2Cl_2 AA-cell containing ~ 0.5g lithium is shown in Fig. 1 (courtesy of EIC laboratories). Note that there are DOT requirements which regulate shipping cells containing more than 0.5g lithium. An optimized cell, i.e., a cell with the same mass of electrodes but containing thinner electrodes to maximize efficiency at higher current densities, might be expected to provide 20 percent more capacity.

The sulfuryl chloride system requires an additive (X) to provide suitable storage without corrosive degradation. It can be expected to lose about 4 percent capacity at 25°C during the first year and 2.5 percent each year thereafter. This system is designed for intermittent or pulsed profile applications and generally has superior high temperature performance.

Wilson Greatbatch (E•I) also employs a Cl_2 additive, referred to as CSC, which is rated to perform between -32°C and 92°C. The CSC electrochemical system offers several advantages: an OCV of 3.9V with a nominal operating voltage of 3.4V, high energy density, and the ability to provide moderate to high power at continuous discharge rates of 150 mA. The cell is rated at 2 Ah at a rated discharge current of 50 mA. The cell is a spiral wound design with an internal fuse to prevent short circuit abuse. This technology is not as well understood nor does it perform as well at low temperatures as the other liquid cathode systems. Wilson Greatbach has a specially designed PMX series that features excellent thermal cycling capabilities and is designed for extreme shock and vibration. The cell is rated at 1.6 Ah at 20 mA. It operates between 0°C and 150°C.

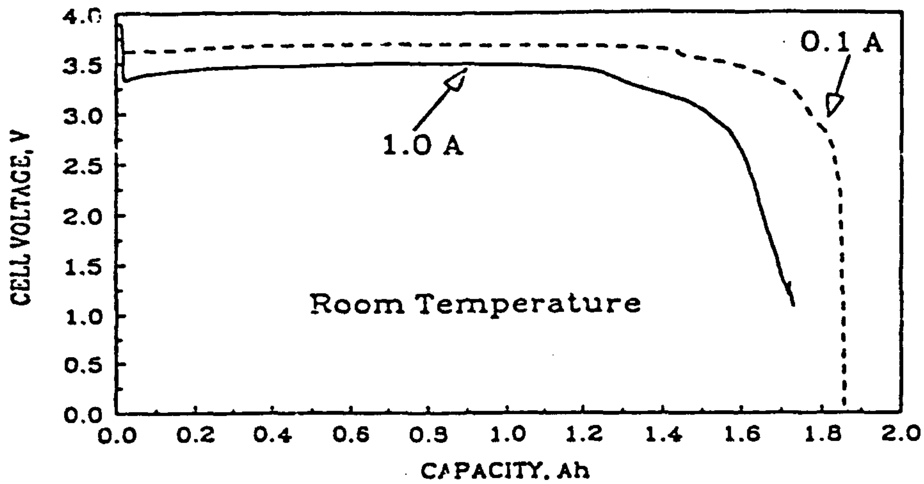
EIC Laboratories, GTE (now Yardney Technical Products) and NSWCDD have enhanced the performance of the SO_2Cl_2 technology by employing a mixed electrolyte in which the SO_2Cl_2 also contains $SOCl_2$ and/or SO_2.

LITHIUM/BROMINE COMPLEXED THIONYL CHLORIDE ($Li/SOCl_2$-BrCl)

Electrochem Industries has two spirally wound cell designs - an unoptimized (0.5g Li), series 3B27, which provides 1.6 Ah capacity and an optimized series 3B64 cell, which is capable of delivering ~2.0 Ah capacity. This technology, referred to as BCX, is composed of thionyl chloride and a bromine-chloride complex additive to enhance low temperature performance. It is intended for low to moderate rate discharge and typically delivers 2.0 Ah at 20°C under a 20 mA continuous drain. The cells operate between -40°C and 72°C. These cells are not vented.

LITHIUM/THIONYL CHLORIDE ($Li/SOCl_2$)

The thionyl chloride system is produced in both a bobbin design for maximum efficiency at low discharge rates, and a spirally wound construction for high rate or pulse applications. A prismatic design, manufactured by Eagle-Picher Industries under the "Keeper" trademark, is intended for extended capacity at very low discharge rates.



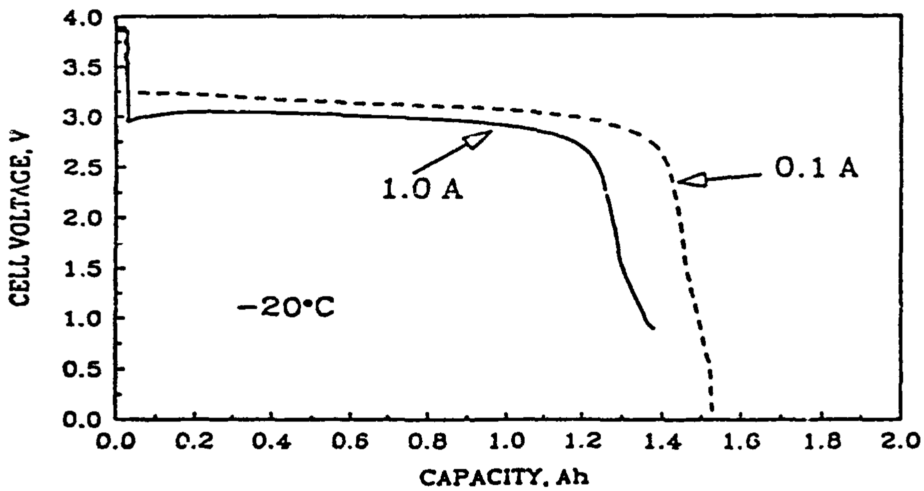


FIGURE 1. DISCHARGE CURVES FOR AA-SIZE LITHIUM SULFURYL CHOLORIDE CELLS AT (A) ROOM TEMPERATURE AND (B) -20°C.

The operating temperature range of the Li/$SOCl_2$ chemistry is between -55°C and 150°C. Cell discharge at higher temperatures can be achieved by employing a magnesium-lithium alloy for the anode. The latter design is often employed for oil well logging and drilling operations. The performance of a bobbin cell should deliver a nominal 3.4V at a current density of 1 mA/cm^2. A capacity loss of less than 2 percent per year during ambient room temperature storage can be expected. Capacity losses on the order of 5 percent per year have been observed for spirally wound cells with a thick electrode design. The theoretical capacity of a cell containing a maximum DOT allowable amount of 0.5g Li per cell is 1.93 Ah.

CHAPTER 3

AVAILABILITY OF COMMERCIAL AA-SIZE CELLS

The most mature AA-size cell technologies available are the Li/MnO_2, the Li/SO_2, and the Li/$SOCl_2$ electrochemical systems. Of these, the most accessable is the Li/$SOCl_2$ technology, which has a relatively large number of manufacturers of AA-size cells and is not limited to foreign suppliers.

Some companies are relatively small, such as Energy Conversion (ECO) and Battery Engineering Inc. (BEI), which employ about two dozen people. However, BEI is owned by a large foreign company, Hitachi. Small companies such as ECO and EIC Laboratories Inc. may be willing to supply cells in limited numbers that meet specific requirements, e.g. cells/batteries with a low magnetic signature.

Many of the larger battery manufacturers are foreign owned, including SAFT, Hitachi, Power Conversion Inc., Varta, Dowty, Hoppecke and Tadiran. Several suppliers of $SOCl_2$ batteries have either sold or liquidated the business (Gould and GTE), discontinued military portions of their lithium technology (Duracell), or have been forced into bankruptcy (Altus).

Variations in the technical performance data reported by suppliers will depend on the design, size and cell components. Consequently, the attributes of each supplier should be considered on their own merits.

Hitachi has two AA cells; one is a bobbin cell marketed under the Maxcell ER6C series that is rated at 1.9 Ah at 100 uA (only 1.1 Ah at 1 mA); the other, BEI cell type 14-48, is also a bobbin design which is rated at 1.8 Ah at 10 mA.

Electrochem Industries provides a series 3B940TC bobbin cell that has performance characteristics similar to the Maxcell ER6C.

Yardney Technical Products does not offer an off-the-shelf product but has considerable cell development experience. An older AA cell developed and tested approximately 10 years ago by the former GTE battery group (now part of Yardney), provided 1.6 Ah at a nominal current drain of 10 mA. At a current density of 1 mA/cm^2, capacities of 1.2, 2.0 and 1.6 Ah were measured at -20, 25, and 80°C, respectively.

SAFT's bobbin design LS6 BA series nominally provides 1.8 Ah and 3.5V at 5 mA at 20°C. A summary of performance data excerpted from a SAFT brochure (with permission) for an unoptimized AA-size Li/$SOCl_2$ cell is shown in Figure 2 as a function of temperature and discharge rates. The maximum recommended drain rate for this design is 100 mA at 20°C which still provides 50 percent of the nominal capacity. The next generation AA cell, series LS1450, will contain a vent.

Tadiran makes three types of AA bobbin cells: (1) model TL-2100 (Li Xtra) rated at 1.9 Ah at 1.5 mA and 1.3 Ah at 30 mA; (2) model TL-5903 (high capacity Xtra) rated at 2.4 Ah at 1.0 mA and 1.3 Ah at 30 mA; (3) model TL-5104 designed for memory back-up has a rated capacity of 1.75 Ah at 1.5 mA. The model TL-5903 has comparable capacity to the model TL-2100 when discharged at drain rates higher than 5 mA. The Xtra models are reported to have excellent voltage-response time, recovering to 3.3V within 10 sec after one year storage at room temperature under a 200 ohm load.

Eagle-Picher has a Keeper series of "square AA" low-rate optimized, prismatic cells referred to as model LTC-30P; these cells have unusually high capacities of 2.6 Ah and 2.4 Ah at about 5 mA and 2 mA, respectively. The higher cost per cell than is typical of other Li/$SOCl_2$ reflects the extra cost associated with meeting DOT shipping regulations for cells containing more than 0.5g Li. This cell is safe on shorting but will rupture if charged. It has no vent. The unoptimized version (<0.5g Li), model LTC-20, conservatively provides 1.6 Ah. Similar performance can be accomplished in a smaller cell, model LTC-16P, using a 2/3A size cell which also meets the 0.5g Li DOT requirements. Although the 13.5 mm side width meets the AA requirements of cell widths between 13.5 and 14.5 mm, the 19 mm diagonal is more characteristic of an A-size cell diameter. Eagle-Picher reports a reliability of 0.998 from last year's supply. A program to develop a modified chemistry to enhance storage and to reduce voltage delay is currently in progress. In early 1993, a standard bobbin AA non-magnetic cell is expected to be available.

Power Conversion Inc. (PCI) has developed an optimized (>0.5g Li) carbon-limited bobbin cell, with a vent, capable of delivering 2.2 Ah at a C/1000 rate. The cell, model T06/4, is currently approved for use in the Expendable Mobile Accoustic Training Target (EMATT). Model TO6/5 is the same cell design except that it contains only 0.5g Li to meet DOT shipping regulations for hazardous materials.

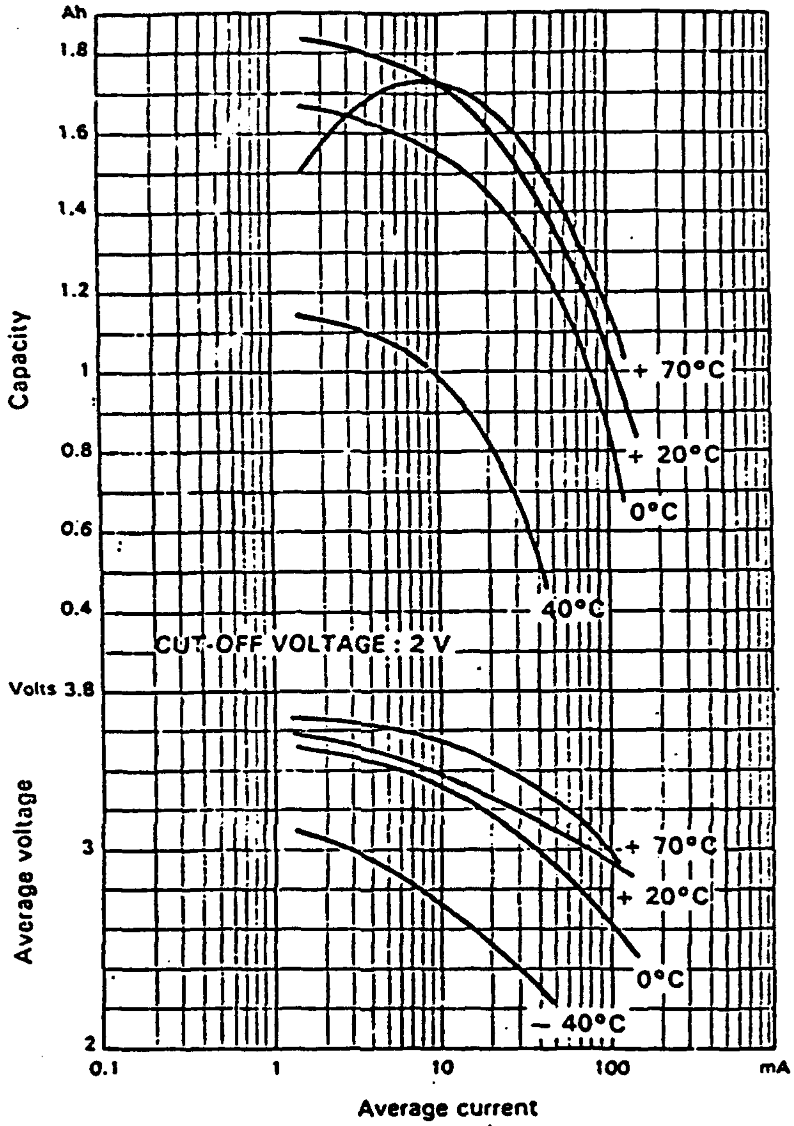


FIGURE 2. PERFORMANCE DATA FOR AA-SIZE (SAFT LS 6) $LI/SOCL_2$ CELLS AS A FUNCTION OF TEMPERATURE AND DISCHARGE RATES

CHAPTER 4

AA CELLS FOR NAVAL MINES

APPLICATIONS

As a starting point, R33 personnel discussed the potential application of AA size cells for mine batteries with the Naval Surface Warfare Center Mine Warfare Engineering Activity (NSWCMWEA). The mine batteries identified as most suitable for AA size lithium cells were the MK 117, 141, 142 and EX143 series. We examined the weapon specifications for these batteries and defined the most suitable broad spectrum test program for evaluation of the best chemistry and design configuration.

The immediate test objective was to select and purchase AA cells from various commercial vendors and to characterize their performance relative to Navy mine requirements. The cell test plan, shown as a flow chart in Figure 3, was developed to cover these broad spectrum needs for the preliminary evaluation. Of special interest are low temperature performance (typically -2°C), moderate rate discharge at ~3 mA, pulse behavior, capacity, start-up and performance after storage. The evaluation process is expected to provide criteria by which optimum AA cell performance can be measured.

Some applications require a low-magnetic signature for batteries. Magnetic signature is screened using magnetometer readings measured 4.5 inches from the cell. Cell casing, caps, tabs, current collector screens, and fill tubes all contribute to the cell's magnetic signature. Careful selection of these materials is required to reduce the magnetic signature. Preliminary studies performed at NSWCDD and NSWCMWEA indicate that many commercial cells that are designated non-magnetic have magnetic signatures on the order of 30 gamma per cell. One small company, however, provided a $Li/SOCl_2$ AA cell with a magnetic signature of 8 gamma. However, the magnetic signature is a secondary characteristic of a desirable cell technology. The primary emphasis of the current program is performance of the cell under discharge conditions typical of mine warfare applications.

RECOMMENDATIONS

From the available AA-size cell chemistries listed in Table 1 (Li/CF_x is not available in a AA cell), the $SOCl_2$ technology was selected as the most suitable choice to meet the power requirements of current and future mine applications. Several of the cell technologies either performed poorly at low temperature or had inferior voltage or energy relative to the $Li/SOCl_2$ system. Although the Li/SO_2 system possessed superior low temperature

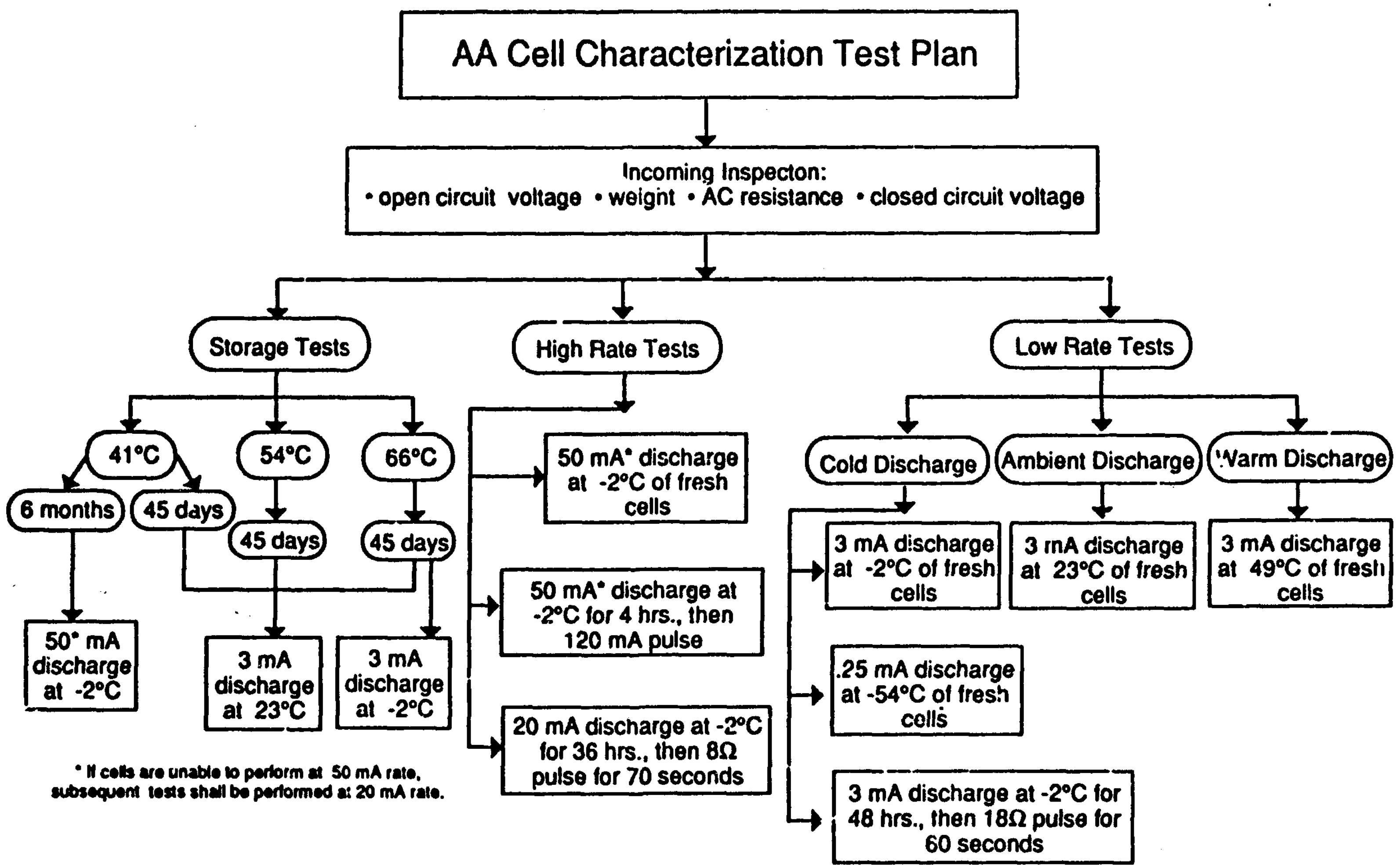


FIGURE 3. TEST PLAN FOR EVALUATION OF AA-SIZE LITHIUM CELLS FOR NAVY MINE BATTERIES

capabilities and would probably meet all the mine battery requirements, the $SOCl_2$ system offered about twice the energy density. Additionally, there was only one vendor of commercial AA-size SO_2 cells whereas several sources of the $SOCl_2$ technology were available. The Li/MnO_2 technology is the most mature commercial lithium system but was not available in a AA-size from U.S. based producers. It offers high energy density and high voltage and is reported to perform well at the low temperature (minimum -2°C) required for sea water operation. This technology offers the added benefit of producing less toxic and corrosive gases than $SOCl_2$ in the event of abusive venting. Thus, the Li/MnO_2 cell is an ideal candidate for comparison testing with the Li/$SOCl_2$ cell.

The Li/MnO_2 cell from Varta was selected from the two foreign suppliers because of its lower rate, long life design. Preliminary examination of the voltage, current, and capacity requirements indicates the MK 117 and the MK 135 (non-AA) are best matched by the use of a 3.0V system such as the Li/MnO_2 technology.

The present MK 141, MK 142, and MK 143 would benefit most from the Li/$SOCl_2$ technology. The primary concern with the Li/$SOCl_2$ technology is that exposure to high temperature during uncontrolled storage leads to capacity losses and possible severe voltage delay, especially under high rate or low temperature discharge.

Among the $SOCl_2$ suppliers, SAFT was recommended for evaluation due to the perceived superior storage/start-up performance of larger SAFT cells previously evaluated by the Navy. Power Conversion Inc. (PCI) was chosen as a potential supplier because a cell with low magnetic signature was indicated to be available. Wilson Greatbatch was selected because they market the widely available commercial Maxcell AA cell from Hitachi. Eagle-Picher was considered as a vendor because their Keeper square AA-type cell offered greater capacity in about the same size package as a cylindrical cell. Tadiran offered three types of Li/$SOCl_2$ AA-size cells. These cells were not considered for evaluation because of a limited program budget for performing cell tests.

Improvements in Li/$SOCl_2$ technology have evolved over the past years. These include new vents, separators, design modifications, the use of carbon blends, and anode coatings. Catalysts and new electrolyte salts have improved high rate performance, especially at low temperatures. A second report will be issued upon completion of the characterization program for commercial AA cells.

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As part of an effort to reduce Navy battery procurement problems, a program has been developed to standardize battery chemistries and cell sizes. Currently, several mercury-based cells are being used in mine batteries; however, thee have limited energy and power densities and present uncertain long-term availability and disposal issues. The lithium/thionyl chloride electrochemical technology is being considered as a long-term solution to these problems. This report describes the surveillance effort that gave rise to selection of AA-size lithium thionyl chloride cells for mine battery development. A test plan to verify this choice and to identify potential cell or battery production and performance problems is also provided.

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