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LIMITING FACTORS TO ADVANCING THERMAL BATTERY TECHNOLOGY FOR NAVAL APPLICATIONS

BY PATRICK B. DAVIS AND CLINTON S. WINCHESTER

RESEARCH AND TECHNOLOGY DEPARTMENT

OCTOBER 1991



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FOREWORD

Increasing power requirements for evolving naval systems have provided a strong incentive to improve the present lithium thermal battery technology. Improvements benefit missile, small vehicle, and sonobuoy capabilities. The Electrochemistry Branch (Code R33) of the Naval Surface Warfare Center (NAVSWC) has continued to study developments over the last 10 years and examine promising avenues for further advancement of the technology. This report summarizes these avenues for advancement.

The authors would like to acknowledge the many Navy, Army, Air Force, and Department of Energy program offices, laboratories, and centers that have supported thermal battery efforts in the past, without whom the current technology would not be possible. We would also like to thank the two U.S. thermal battery manufacturers for their willingness to share their knowledge so readily: Eagle-Picher of Joplin, Missouri, and Saft America of Cockeysville, Maryland.

Approved by:

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ABSTRACT

Thermal batteries are primary reserve electrochemical power sources using molten salt electrolytes which experience little effective aging while in storage or dormant deployment. Thermal batteries are primarily used in military applications, and are currently used in a wide variety of Navy devices such as missiles, torpedoes, decoys, and training targets, usually as power supplies in guidance, propulsion, and Safe/Arm applications.

Technology developments have increased the available energy and power density ratings by an order of magnitude in the last ten years. Present thermal batteries, using lithium anodes and metal sulfide cathodes, are capable of performing applications where only less rugged and more expensive silver oxide/zinc or silver/magnesium chloride seawater batteries could serve previously. Additionally, these batteries are capable of supplanting lithium/thionyl chloride reserve batteries in a variety of specifically optimized designs.

Increases in thermal battery energy and power density capabilities are not projected to continue with the current available technology. Several battery designs are now at the edge of feasibility and safety. Since future naval systems are likely to require continued growth of battery energy and power densities, there must be significant advances in battery technology. Specifically, anode alloy composition and new cathode materials must be investigated to allow for safe development and deployment of these high power, higher energy density batteries.

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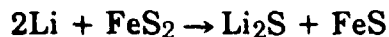
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INTRODUCTION

The Electrochemistry Branch (Code R33) of the Naval Surface Warfare Center is continually evaluating thermal battery technology for advanced naval systems. A thermal battery is a primary battery whose electrolyte is a solid and nonconducting salt at room temperature. The electrolyte is rendered molten by a pyrotechnic heat source which is an integral part of the battery cell stack. Battery activation and internal thermal equilibration to its operational temperature of approximately 500°C normally occur within 0.5 second of initiation. It is primarily the high conductivity of the electrolyte at this elevated temperature that allows the battery to be discharged at high rates. This class of battery technology is considered a reserve type battery since it is inactive until the pyrotechnic is initiated (normally accomplished by an electric pulse to a squib). Once activated in this manner, the battery will only remain active for its particular design life and cannot later be "reactivated"; as such, it is a one-shot device. A flow chart of the activation sequence is given in Figure 1. Figure 2 is a thermal battery cross section. Although the term "thermal battery" refers to a family of batteries which include a variety of specific chemistries (see Table 1), this discussion will focus on the lithium/iron disulfide chemistry because of its higher energy and power densities. The overall cell reaction between anode and cathode material for the lithium/iron disulfide system is given below:



Thermal batteries are very application specific; they are designed and hand assembled to meet very specific electrical and environmental parameters. Before a battery can be accurately designed, requirements such as electrical performance (current, voltage regulation, activation delay, electrical noise, operating life, etc.), battery activation, environmental conditions, and mounting provisions must be completely defined.

Thermal batteries are primarily used where one of the following conditions occurs: (1) an application demands a very long shelf life (up to 20 years) with no battery maintenance, (2) a high power density is required, or (3) an application which requires a very rugged design. Some advantages and disadvantages of the thermal battery system are given in Table 2.

In the early 1970's, lithium thermal batteries began being retrofitted into existing applications (such as those filled by other thermal batteries or reserve, silver oxide/zinc batteries). Because of an inherently superior power density, requirements were usually met with relative ease. However, recent battery development efforts have served to highlight design areas of concern when pushing the technology to its limits. These limits include maximizing power density, energy density, and overall energy content. This report will examine these limits as they relate to present lithium thermal battery technology, and discuss routes to improve these systems.

DISCUSSION

Figure 3 is a Ragone plot of a variety of power sources indicating power density versus energy density. As can be seen, the Li-FeS₂ system has very little competition for high power applications (>500 W/kg). However, emerging naval applications are requiring even higher power densities. For example, one projected sonobuoy application requires >2000 W/kg. The problem of attaining a high power density is made worse by specifications that simultaneously require a high energy density. (The same sonobuoy application requires an energy density in excess of 13 Wh/kg.) Presently, there are definite limits to what power densities can be achieved and even more restrictions on what energy densities can be maintained. Table 3 lists performance limits that have been demonstrated in actual batteries, along with the most common performance values found in production batteries. It should be noted that the high end of almost any of the demonstrated limits listed in Table 3 can be reached only at the expense of other characteristics. The factors that limit energy and power density are discussed below.

Energy density is limited by the very nature of the battery; most of the battery consists of support structures such as the can, header, connector, insulation, activation mechanism, pyrotechnic heat source, binder materials, and electrolyte. In addition, because elemental lithium melts far below the operational temperature of the battery, anodes are made either as an alloy with aluminum or silicon, or held in a matrix of fine iron particles. Consequently, only 20 to 40 percent of the anode is lithium (the remainder being inactive). Also, to provide acceptable voltage regulation, the active cathode material (FeS₂) is normally proportioned at a 10:1 ratio of the active lithium available. It is estimated that a completed battery consists of less than 35 percent active electrochemical constituents. Therefore, for the entire battery to realize significant gains in energy density, large increases in the active electrochemical constituents must be made. Increased cell thicknesses tend to result in an increased overall energy density, but as Figure 4 indicates, this benefit quickly reaches a plateau. Increased cell mass also slows the activation process and limits the practical current density due to increasing resistivity during discharge. But more importantly, cell mass is dictated by the electrical requirements placed on the battery and cannot arbitrarily be changed in an attempt to increase energy density. Therefore, the only way to improve energy density is to make some very major changes in the present thermal battery system. Such changes might include alternate cathode materials, an improved FeS₂ cathode material, or an alternate lithium alloy that contains a higher percentage of lithium allowing discharge at a higher potential. These alternatives will be addressed below.

Power density as determined at the battery level is a function of the current density at the electrode, cell thickness (capacity), cell size, and cell duty cycle. Although very high power pulses can be taken of short duration (thus, yielding a seemingly extraordinary power density figure), we will limit our examination to batteries capable of extended high power discharge. In addition, we will not consider cell thickness in detail, as it is defined by the total energy requirement; as previously discussed, energy density cannot be improved easily. Discharges at an extremely high current density pose several problems. First, as shown in Figure 5, voltage will be depressed linearly with increases in current density. This is often intolerable due to voltage regulation constraints on the battery. High current densities reduce delivered energy density through inefficient electrochemical utilization. Figure 6 indicates useful power density gains can be had by using current densities of up to

60 A/in². However, current densities of this magnitude can only be sustained for short periods of time (0.1 to 2.0 seconds), after which either a severe drop off in current or voltage will occur. Capacity (energy) loss is also a major concern. Since we have already established that the overall energy density of thermal batteries is strongly tied to the small portion of the battery that will actually be discharged, it is obvious that as much of the active materials as possible must be used. But, discharging a thermal battery at rates above approximately 20 A/in² results in severe capacity losses. Figure 7 is an idealized graph of energy density delivered versus current density. When designing batteries for actual naval applications, these capacity losses almost always dictate a current density ceiling of 7 to 20 A/in² for extended discharges.

Other issues that limit volumetric power and energy densities as measured at the system level are linked to form factor restrictions. Because the electrochemical components start out as powders, pellet formation requires uniaxial compression under high pressure (16 to 40 tons/in²). This processing step places two restrictions on a completed battery: (1) due to the problems associated with pressing noncircular shapes, this process dictates for ease, cost, and reliability of manufacture that most of the designs will be right circular cylinders (because the cell parts are circular) and (2) because a cell size of 6 inches in diameter requires a press of 300 to 1000 tons (depending on cell thickness), battery diameters are presently limited to about that level. Volumetric power and energy densities are lost due to poor packing efficiency of the right circular cylinder. Presently, several designs use rectangular parts to increase volumetric power and energy densities (one example being a 77 Wh/kg, 275 W/kg battery), but do so at a cost of manufacturability. In addition, this type of design usually suffers a penalty in gravimetric energy density (a non-cylindrical design has to be more robust to withstand the internal pressure developed during discharge, translating into a heavier case weight).

Routes to higher specific energy and/or power include the following:

- (1) Use of lower melting point or lower resistivity electrolytes.
- (2) Use of more thermally stable cathodes.
- (3) Use of more energetic pyrotechnic sources.
- (4) Use of advanced structural alloys or composites.
- (5) Development of higher voltage cathodes.
- (6) Development of better insulations.

Each of these will be discussed in detail below.

Use of lower melting point electrolytes can help improve energy density. This is accomplished primarily by decreasing the amount of pyrotechnic heat source required in raising the battery to its operating temperature. Table 4 lists the various electrolytes commonly used along with their melting points and resistivity.¹ Although use of these electrolytes can be effective for certain applications, lower melting point electrolytes will not normally improve power density since the conductivity of these electrolytes is usually lower (with corresponding ohmic voltage drop). While lower resistivity electrolytes (such as the LiF-LiCl-LiBr system) are superior for use in high power applications, their relatively high conductivity at room temperature may cause problems that include shortened shelf life.

Use of more thermally stable cathodes (>600°C) would offer several benefits. One such benefit would be a higher allowable battery operating temperature, thus, a corresponding gain in electrolyte conductivity and power. Another benefit stems

from safety and reliability concerns. Batteries designed for higher current densities can suffer from various failure modes that include localized heating within the battery. Localized increases in temperature can serve to increase the rate in which battery components thermally decompose. Thermal decomposition occurs with a corresponding release of additional heat. This cycle can lead to localized or total battery thermal runaway that might include a battery venting or more severe failure. A higher decomposition temperature cathode would allow either higher current densities or a higher operating temperature without these events occurring. One cathode that has been examined is synthetic FeS_2 . Commonly used FeS_2 cathode material is derived from naturally occurring pyrite deposits or as a by-product from the processing of base or noble metal ores. Thus, the commonly used material may vary in composition, purity, and particle size greatly depending upon source and lot. The synthetic material has highly uniform physical and chemical properties. In addition, it has six times the surface area (offering an improved rate capability), and a higher thermal decomposition temperature.² Figure 8 is a comparison of synthetic and natural FeS_2 cathodes at two different current densities. Cost of the synthetic material is higher, but would not increase battery cost significantly.

Use of more energetic pyrotechnic materials would of course reduce the amount of material required to raise the battery to operating temperature. One class of higher performance heat source is a nickel-titanium based fuel mix. Advantages are rapid burn rate propagation (> 10 inch/sec) and gasless combustion with high flame temperatures and energy release per volume and mass. Problems associated with these materials are cost, toxicity, and sensitivity to ignition during battery manufacture. An additional problem associated with advanced exothermic sources is the limited ability of the battery anode and cathode materials to tolerate exceptionally energetic thermal sources without cascading into thermal runaway.

Use of advanced structural alloys or high temperature composites for the building of battery cases and header would offer weight advantages. Although little volume could be saved, overall gravimetric energy and power densities would improve slightly (5 to 15 percent demonstrated). Titanium based materials are normally thought of as the chief replacement materials due to superior strength and melting point. However, due to the higher cost and difficulties in perfecting the hermetic seal technology for battery terminals, the moderate energy and power density gains that can be achieved have not been realized to date.

Development of higher voltage cathodes have been pursued for some time. Candidate cathode materials are listed in Table 5. Promising materials are those with either an increased thermal decomposition temperature or a higher theoretical energy density. However, none of the systems offers trouble-free gains. The CuF_2 and the CuCl_2 systems have low practical energy densities, and present electrolytes are incompatible with the CuF_2 system due to the high voltage. The MnO_2 system has a low decomposition temperature, and the $(\text{CF})_x$ material has both a low decomposition temperature and a low volumetric energy density. The VO_x material has a slightly higher resistance than is desired and is very difficult to manufacture. The CoO_x is difficult to manufacture and is suspected of being carcinogenic.

Development of improved insulations includes vacuum based technologies and aerogel ("foamed" ceramic materials). However, these materials offer only small gains (present insulators are quite good) compared with many problems their use poses. These problems include high cost, handling issues, and shelf life (for vacuum based insulators). It is not expected that these alternatives will come into wide use within the foreseeable future.

SUMMARY AND CONCLUSIONS

Advanced materials and engineering can help improve present thermal battery technology. Gains would be seen in energy and power densities and/or safety of high power batteries. However, most routes to significant increases in energy and power density involve breakthroughs in present technology. Of the possible improvements discussed above, the examples that could most easily be incorporated are the synthetic FeS_2 and advanced structural materials.

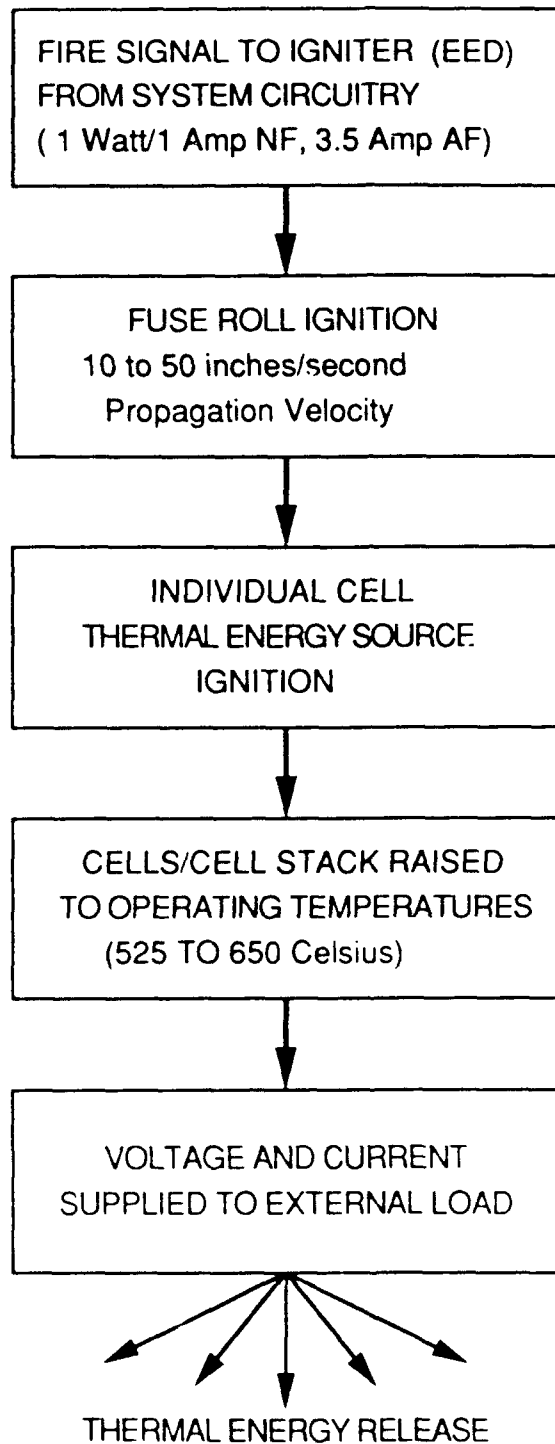


FIGURE 1. THERMAL BATTERY FUNCTIONAL BLOCK DIAGRAM

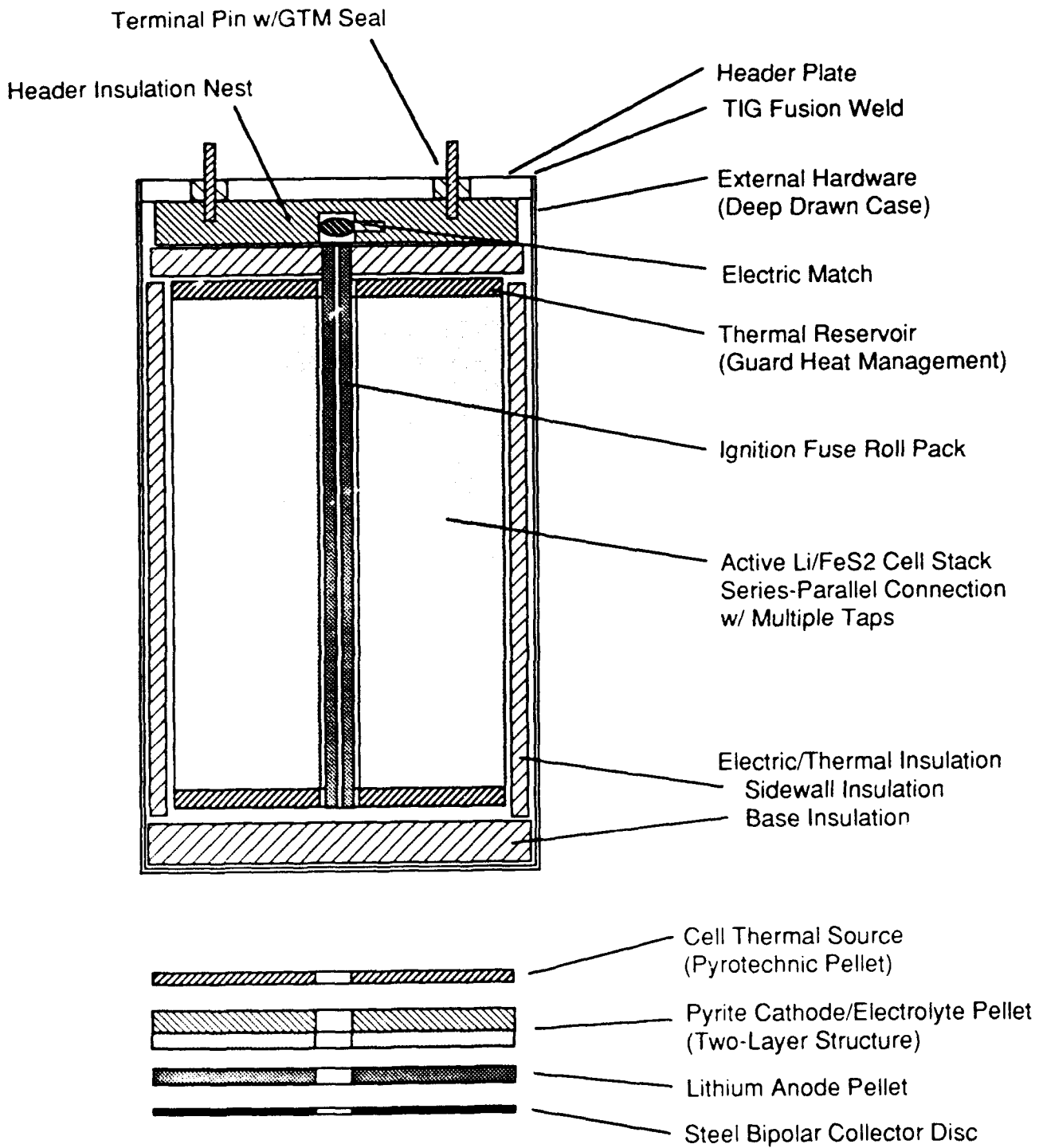


FIGURE 2. LITHIUM/IRON DISULFIDE THERMAL BATTERY FUNCTIONAL ELEMENT CROSS-SECTION

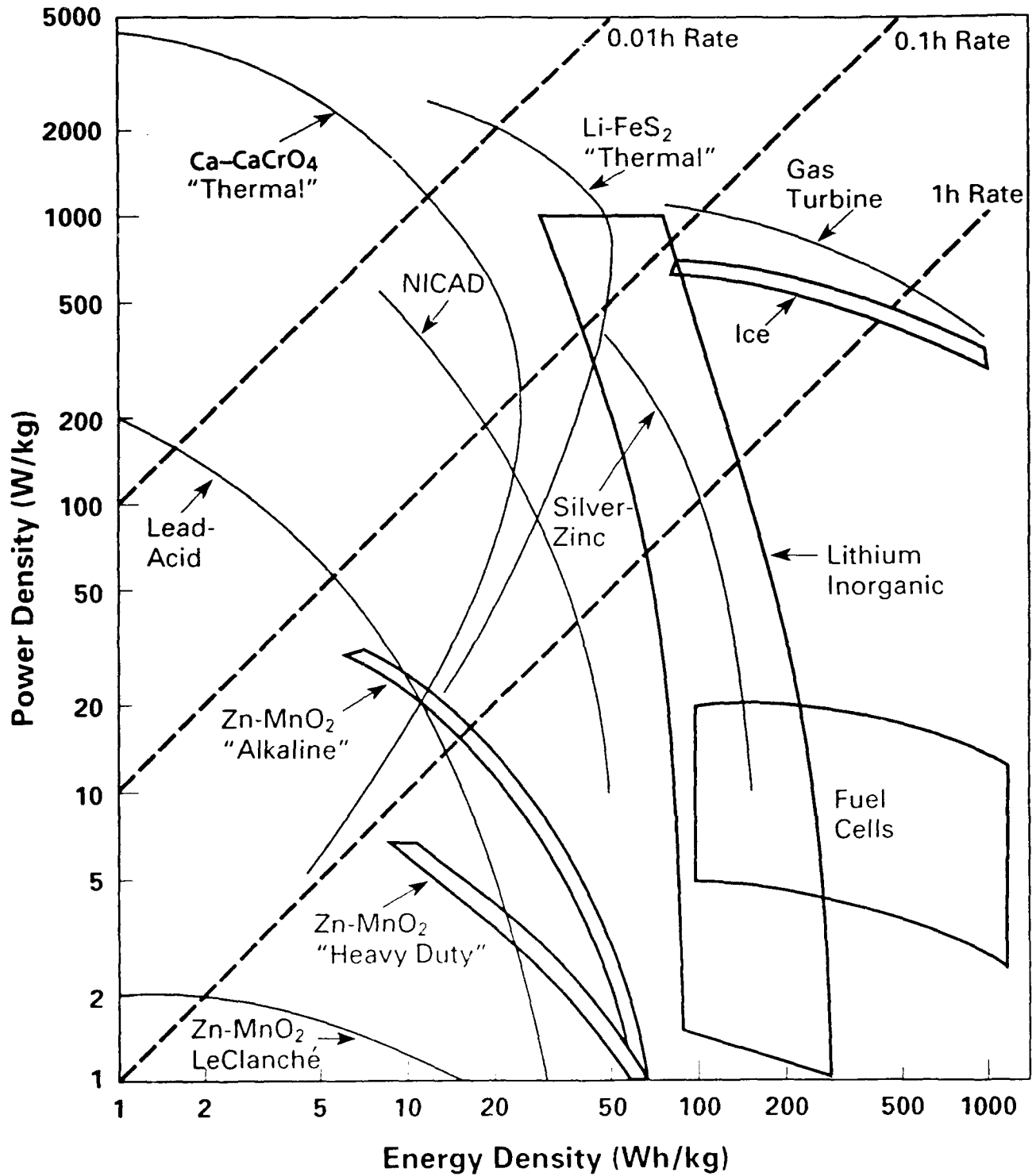


FIGURE 3. RAGONE PLOT

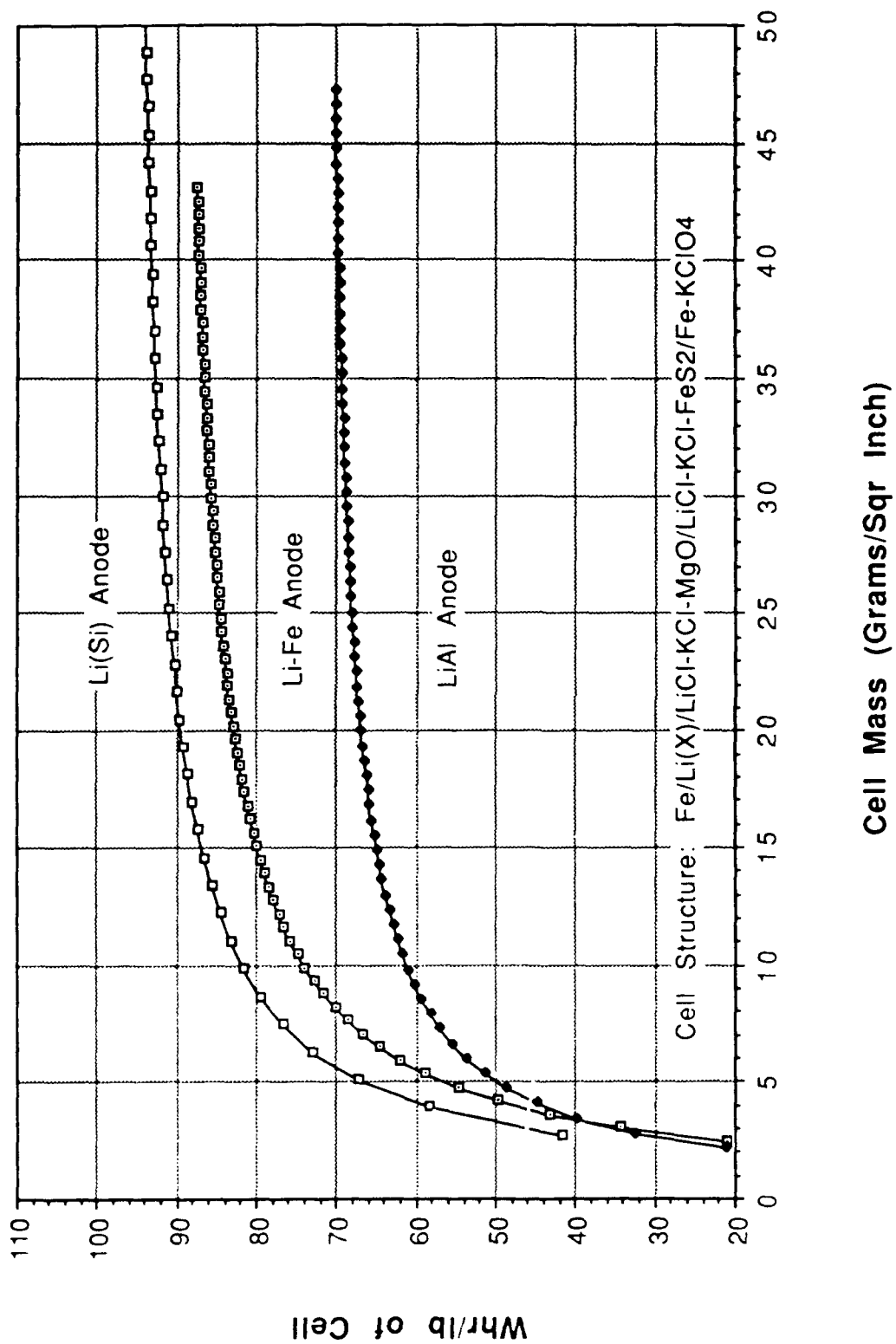


FIGURE 4. POTENTIAL SPECIFIC ENERGY VERSUS ANODE TYPE

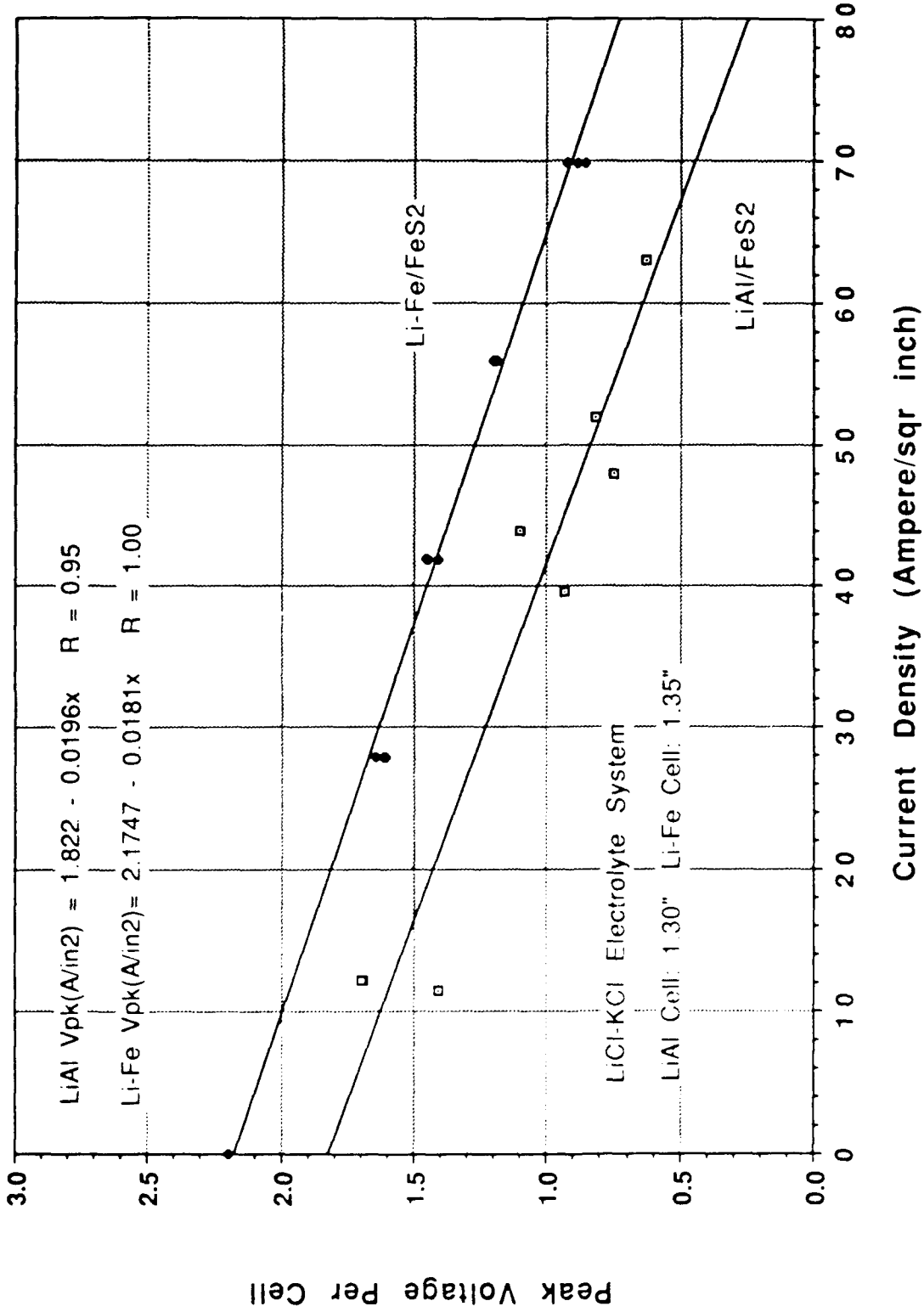


FIGURE 5. COMPARISON OF PEAK VOLTAGE VERSUS CURRENT DENSITY FOR LiAl AND Li-Fe/IRON DISULFIDE CELLS

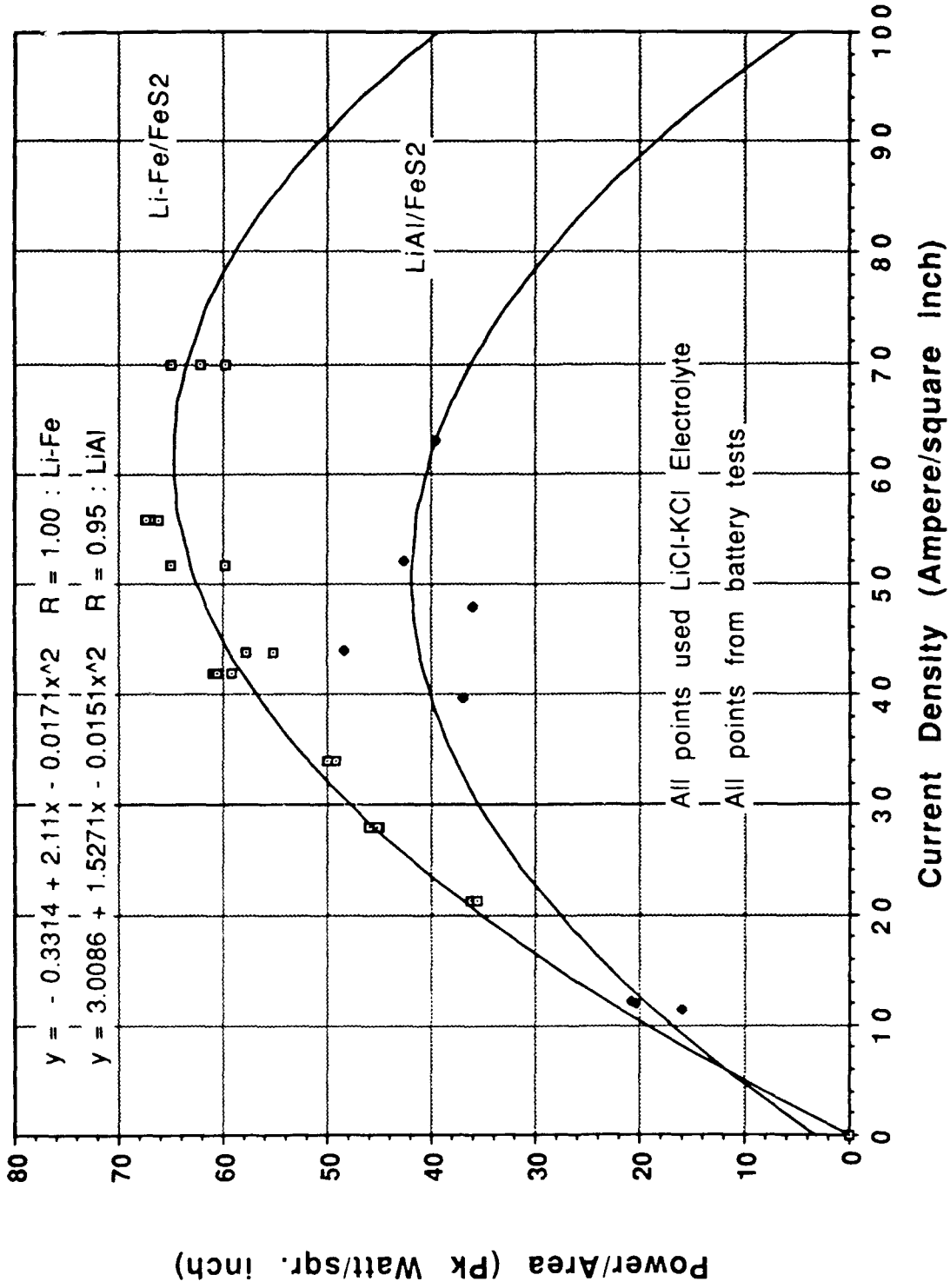


FIGURE 6. PEAK POWER VERSUS CURRENT DENSITY FOR LiAl AND Li-Fe/IRON DISULFIDE CELLS

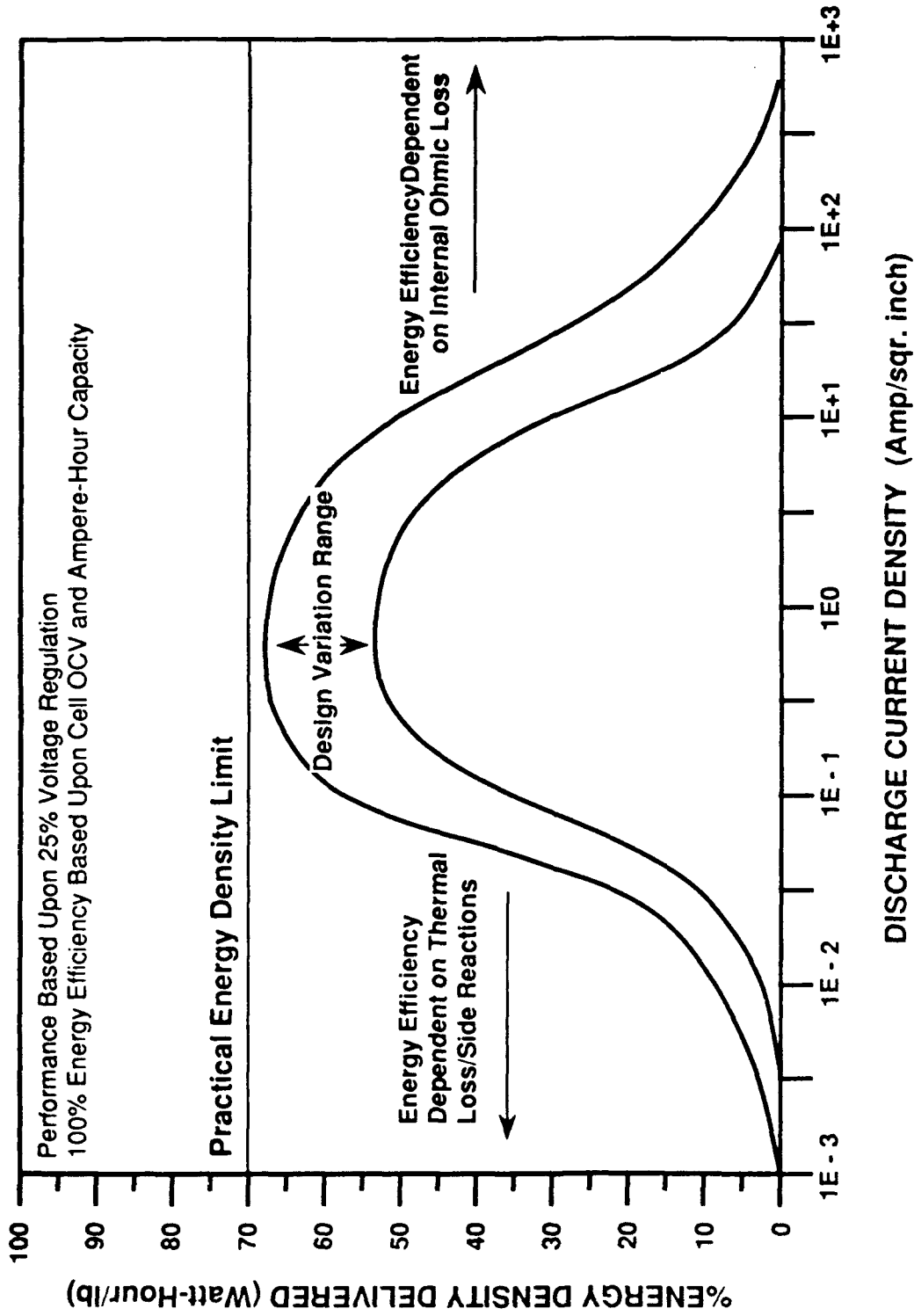


FIGURE 7. AVAILABLE CELL ENERGY DENSITY VERSUS CURRENT DENSITY

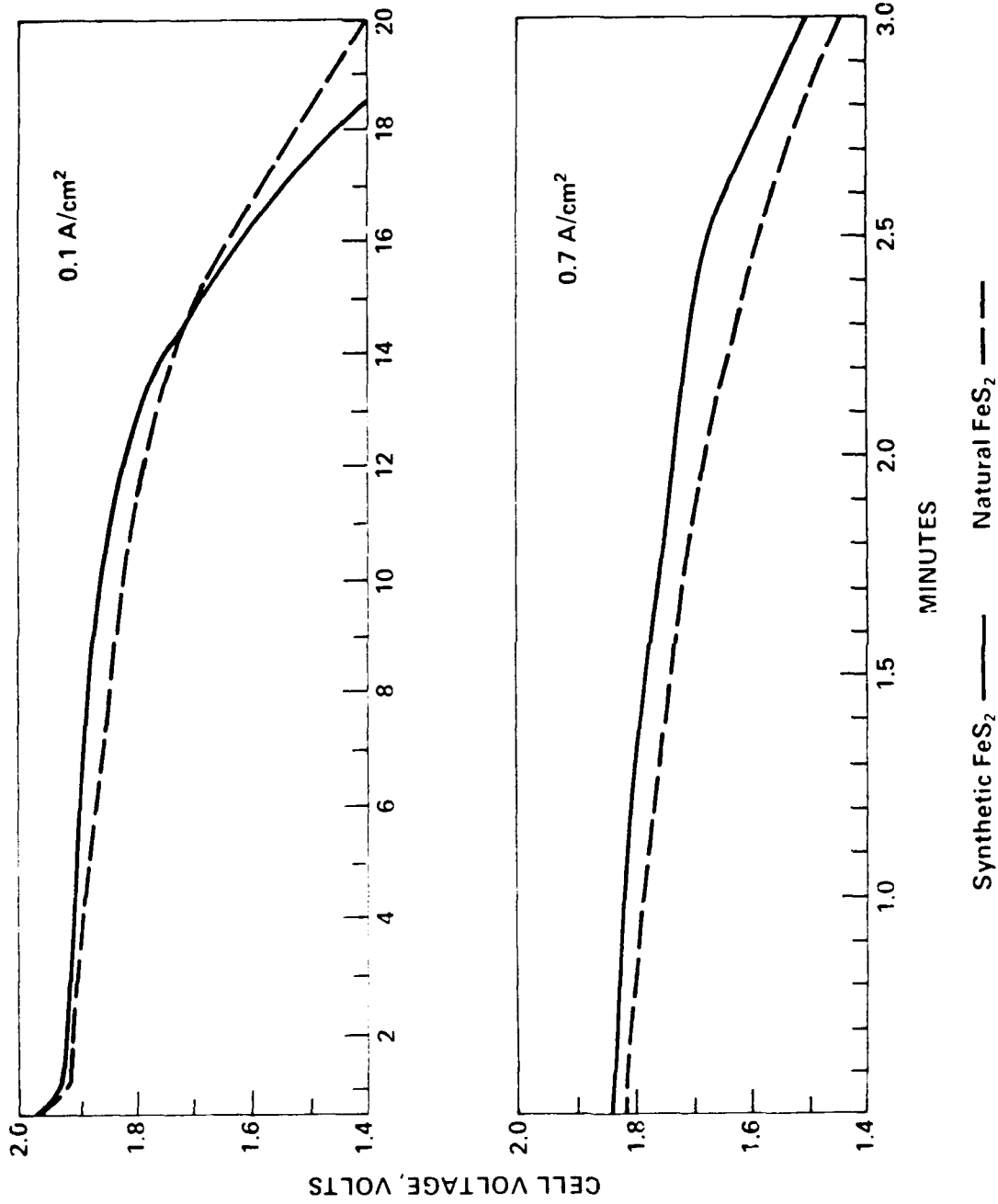


FIGURE 8. TYPICAL SINGLE-CELL DISCHARGE CURVES

TABLE 1. TYPES OF THERMAL BATTERIES

Electrochemical System (Anode/Electrolyte/Cathode)	Typical Cell Voltage	Cell Characteristics and Applications
<u>Systems Used in New Designs:</u>		
Li/LiCl-KCl/FeS ₂	1.6 to 2.2	Highest capacity cell, low electrical noise, long service life.
Ca/LiCl-KCl/K ₂ Cr ₂ O ₇	3.3	Short activation time.
<u>Old Systems Still in Use:</u>		
Ca/LiCl-KCl/WO ₃	2.4 to 2.6	Used for fuse applications with low shock and vibration.
Ca/LiCl-KCl/CaCrO ₄	2.2 to 2.6	Used for high shock and vibration and short operating life.
Mg/LiCl-KCl/V ₂ O ₅	2.2 to 2.7	Used for high shock and vibration and short operating life.

TABLE 2. ADVANTAGES AND DISADVANTAGES OF THERMAL BATTERIES

Advantages	Disadvantages
Long storage life	Short activated life
No maintenance device	High Surface Temperature
No self discharge	Nonlinear Output Voltage
High power density	One shot device
Wide operating temperature	Hand assembled, high unit cost
Extremely rugged	
No external heating required	
No outgassing	
High reliability	
Low life cycle cost	
Fast activation	

TABLE 3. THERMAL BATTERY PERFORMANCE LIMITS

	Limits Demonstrated	Most Common
Discharge Voltage	1.5 to 2000V	10 to 40V
Current Density	0.0 to 12 Amp/cm ²	.01 to .1 Amp/cm ²
Discharge Life	0.05 to 7200 seconds	5 to 180 seconds
Activation Time	0.005 to 30 seconds	0.3 to 1.0 second
Voltage Regulation	5 to 70 percent of V _{max}	20 to 30 percent of V _{max}
Cell Diameter	0.25 to 16.0 inches	1.0 to 3.0 inches
Battery Length	0.20 to 48.0 inches	1.5 to 8.0 inches
Battery Mass	15 to 62,000 grams	100 to 7000 grams
Specific Energy	0.2 to 75.0 Wh/kg	11.0 to 50 Wh/kg
Specific Power	0.0 to 46 KW/kg	138 to 1900 W/kg

TABLE 4. AVAILABLE ELECTROLYTES

	Melting Point	Resistivity Ohm-cm at 500°C 35 wt% MgO Binder
LiCl-KCl	350°C	0.85
LiBr-KBr-LiCl	330°C	1.15
LiBr-KBr-LiF	280°C	0.95
LiF-LiCl-LiBr	440°C	0.55
LiF-LiCl-LiBr-LiI	340°C	0.45

TABLE 5. ALTERNATE CATHODE MATERIALS

Cathode	Theoretical OCV with Lithium Anode	Decomposition/ Melting Point	Theoretical Specific Energy
CuF_2	3.54V	950°C	1650 Wh/kg
MnO_2	3.5V	535°C	1005 Wh/kg
CuCl_2	3.1V	620°C	1125 Wh/kg
$(\text{CF})_x$	3.1V	580°C to 640°C	2180 Wh/kg
VO_x	2.7V	1200°C	1100 Wh/kg
CoO_x	3.9V	900°C	550 Wh/kg

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13. ABSTRACT (Maximum 200 words) <p>Thermal batteries are primary reserve electrochemical power sources using molten salt electrolytes which experience little effective aging while in storage or dormant deployment. Thermal batteries are primarily used in military applications, and are currently used in a wide variety of Navy devices such as missiles, torpedoes, decoys, and training targets, usually as power supplies in guidance, propulsion, and Safe/Arm applications.</p> <p>Technology developments have increased the available energy and power density ratings by an order of magnitude in the last ten years. Present thermal batteries, using lithium anodes and metal sulfide cathodes, are capable of performing applications where only less rugged and more expensive silver oxide/zinc or silver/magnesium chloride seawater batteries could serve previously. Additionally, these batteries are capable of supplanting lithium/thionyl chloride reserve batteries in a variety of specifically optimized designs.</p> <p>Increases in thermal battery energy and power density capabilities are not projected to continue with the current available technology. Several battery designs are now at the edge of feasibility and safety. Since future naval systems are likely to require continued growth of battery energy and power densities, there must be significant advances in battery technology. Specifically, anode alloy composition and new cathode materials must be investigated to allow for safe development and deployment of these high power, higher energy density batteries.</p>			
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