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LITHIUM BATTERIES - AN OVERVIEW

by

G.D. Nagy

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ABSTRACT

In the search of high-energy density batteries, lithium systems have always been prime candidates because of their low equivalent weight and high potential. Problems have been concerned with handling and efficient utilization of the active components. The recent advent of promising commercial batteries has demonstrated the viability of lithium systems. This paper reviews some of the promising systems and their problems.

RÉSUMÉ

Parmi les piles électriques possédant une grande énergie massique, les systèmes à base de lithium sont populaires à cause de leur tension et faible poids équivalent. Cependant la fabrication et l'utilisation efficace des constituants actifs de ces cellules ont été des problèmes principaux. Récemment des piles fabriquées commercialement ont démontré la praticabilité des systèmes de lithium. Ce travail donne un compte rendu des systèmes encourageants et de leurs problèmes.

LITHIUM BATTERIES - AN OVERVIEW*

I wish to thank the organizers of this symposium for being able to talk to you today. One of the difficulties I had in preparing this talk was settling on a title. It is intended to be a review and should emphasize more promising systems. I don't wish to take anything away from the other speakers who will be talking later this morning about these more promising systems. The vagueness of the term overview permits me this latitude.

One of the things that impressed me about the literature was the rapid growth in this field. Work only really started in the late fifties and early sixties (1-6), yet in the last couple of years commercial versions have been manufactured. However, we must not become complacent about this fact as there is still much work to be done and many more changes will occur.

There have been rapid changes in other fields as well. I grew up only a few miles from this hotel, and I can remember tobogganing on a hill about a half mile from here. Now the natural hill has been replaced by an expressway interchange and an artificial tobogganing hill. In ten years these may also be replaced. I can also remember standing in-front of the old Toronto air terminal, at that time no larger than this hotel, and watching the maiden flight of the world's first commercial jet liner. It is not necessary to dwell on the subsequent expansion of the terminal and the jet aircraft industry and the need for changes in the near future.

I think that I have been reminiscing enough; I only wanted to emphasize the rapid growth and justify the prediction that Freud and Spindler made in 1971 in the book "The Primary Battery" (7). They stated: "It seems almost certain that within the next decade, nonaqueous batteries will outperform in energy density ... the lead/acid battery by a factor of 5 or 10 and they will have additional advantages in meeting a greater variety of environmental operating conditions as well".

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* Text of presentation at the Winter Symposia on Lithium Batteries and Electrothermics in Steelmaking, Ontario-Quebec Section of the Electrochemical Society, Toronto, February 15, 1974. Presented by Dr. G.D. Nagy.

TABLE 1

Energy Density of Primary Systems-"D" Cells

Low Current Drain, 50-100 mA

System	Wh/lb		Wh/cm^3	
	25°C	-40°C	25°C	-40°C
LeClanche	19.5	0	0.10	0
Alkaline-Manganese	26	4.8	0.14	0.02
Mercury	40	1.6	0.27	0.01
Magnesium	36	4.2	0.18	0.015
Lithium/SO_2	121	81.2	0.42	0.28

Table 1 shows a comparison of energy densities of various commercial "D" size primary batteries, discharged in our laboratories at currents of 50 to 100 mA (8). The magnesium cell refered to is magnesium/manganese dioxide. The energy density of 121 Wh/lb for the lithium/sulfur dioxide system at room temperature is impressive, but even more impressive is its energy density at -40°C. In fact we have discharged cells at -68°C and obtained 50% of the 25°C capacity. Considering that lead/acid batteries have an energy density of about 15 Wh/lb, the goal of Freud and Spindler is almost reached.

ELECTROLYTES

The electrolytes used for lithium batteries can be nonaqueous inorganic, nonaqueous organic and aqueous. For excellent reviews, I would refer you to "High Energy Batteries" by Jasinski (9), "Advances in Electrochemistry and Electrochemical Engineering", Vol. 8 (10) and "The Primary Battery" by

Hiese and Cahoon (11).

The nonaqueous inorganic electrolytes include fused salts, ammonia, hydrogen fluoride, sulfur dioxide, phosphorous oxychloride and thionyl chloride (9, 11-16). The fused salts, mainly alkali chlorides, sulfates, nitrates and carbonates are used at temperatures between 90 and 650°C (9, 17). Solutes for the other solvents include the alkali halides, perchlorates, nitrates and thiocyanates.

Nonaqueous organic electrolytes, at the present time are the most promising for useful batteries (10, 18). Propylene carbonate, dimethyl formamide, dimethyl sulfoxide, butyrolactone, acetonitrile, nitromethane, methyl formate, methyl acetate and ethylene carbonate are the most common. Dimethyl sulfoxide and ethylene carbonate cannot be used alone for low temperatures because of their high melting points. While the other solvents freeze in the vicinity of -25 to -50°C, mixtures can give lower temperatures. As mentioned earlier the lithium/sulfur dioxide system which uses propylene carbonate and acetonitrile with a bromide solute has operated efficiently at -68°C.

The solutes used with nonaqueous organic solvents are chlorides, bromides, perchlorates, thiocyanates, borofluorates and fluophosphates. The chlorides include double cation salts of lithium plus boron, aluminum, antimony, titanium and tin (12). Recently a cell using lithium hexafluoarsenate in propylene carbonate has been reported (12, 19, 20).

All operations with these electrolytes must be carried out in dry boxes or under controlled environments to minimize or eliminate impurities and traces of water (9-11, 19). Care must also be taken in the choice of solvent and solute as in many cases the electrodes can dissolve or self-discharge.

Recently, a lithium aqueous system has been described (21). The system has a theoretical energy density of 3880 Wh/lb if used as a seawater activated battery and 1687 Wh/lb if water must be supplied. The voltage of this high rate system is around 2.2V. At present, practical energy densities are much lower as parasitic corrosion limits the conversion efficiency to a maximum of 30%, but, there is encouragement that these problems can be solved.

LITHIUM ELECTRODES

Lithium anodes used in primary batteries usually consist of lithium ribbon pressed onto stainless steel, copper or nickel screens (19, 22-25). Pure lithium strips with stainless steel tabs (26) and extruded lithium anodes (27) have also been used. Utilizations between 90 and 100% can be obtained at current densities up to 4 mA/cm^2. Nickel screen dipped into molten lithium produce poorly adherent electrodes (22), while plated lithium does not offer advantages over the pressed kind (25).

For secondary batteries, plated electrodes work well, but can have lower surface areas (4, 25, 28) than pasted types (29, 30). Coulometric efficiencies of 75 to 100% have been achieved in propylene carbonate electrolytes (31, 32). During cycling and electrodeposition, alloying with other metals to form intermetallic components can occur (33, 34).

For molten salt batteries, lithium alloys with aluminum and magnesium (35, 36, 37) have been used. The most common contain magnesium with 13-15% lithium and 1.0 to 1.5% aluminum, or aluminum with 7 to 47% lithium. It is claimed that these alloys do not react with water below 200°C. Foam matrix electrodes made by the vapour deposition of lithium onto 95% porous nickel-chromium alloys have given short-lived operation in fused salt electrolytes (36, 37).

In general, the lithium electrode is well behaved in organic electrolytes as long as impurities are removed. Decomposition need not involve gassing except in the case of the presence of water (38, 39), in which a passivation layer of hydroxide or oxide is formed (40). The effect of this film is more pronounced at low temperature and accounts for some of the loss in energy density at -40°C shown in Table 1. At low temperature the film also increases the activation time. However, in the aqueous battery the film helps to slow down the corrosion reaction on open circuit.

LITHIUM COUPLES

The following 5 Tables show reactions, theoretical or thermodynamic cell potentials and theoretical energy densities of various lithium couples. These systems vary in development from laboratory curiosities to commercial products. I include the large list mainly for comparison purposes. Little mention will be made of some of the more promising systems in deference to those who are better able to speak on the topic and will be speaking later this morning.

For ease of comparison, I have chosen 4 subgroups: elemental, halide, oxide and sulfide couples.

ELEMENTAL COUPLES

Table 2 deals with elemental couples; the halogens, sulfur and oxygen. The halogens suffer mostly from the elaborate methods needed for containment and their toxicity. Although combined iodine has been used with lithium in an experimental solid state battery (41), the cells can only be discharged at microamps/cm^2 and suffer from diffusion of the iodine into the electrolyte. Energy densities up to 135 Wh/lb have been reported. This is a vast improvement over the 1 to 5 Wh/lb obtained for a similar system using AgI (42).

TABLE 2

Lithium/Elemental Couples

Reaction	Theoretical Cell Voltage	Theoretical Energy Density Wh/lb
$2Li + F_2 \rightarrow 2LiF$	6.05	2840
$4Li + O_2 \rightarrow 2Li_2O$	2.90	2362
$2Li + S \rightarrow Li_2S$	2.52	1335
$2Li + Cl_2 \rightarrow 2LiCl$	3.96	1137
$2Li + Br_2 \rightarrow 2LiBr$	3.52	493
$2Li + I_2 \rightarrow 2LiI$	2.78	252

The two systems of most importance are sulfur and chlorine. Both can be used as primary or secondary batteries. In nonaqueous and aqueous electrolytes (6), less than 20% of the sulfur is reduced (12, 15, 23). High temperature cells using a molten lithium chloride-potassium chloride electrolyte at 400°C can give up to 75% utilization (36, 37).

Lithium/chlorine, lithium/bromine and lithium/iodine cells with phosphorous oxychloride or thionyl chloride electrolytes have been investigated (15, 43). Like sulfur in the same electrolytes, the open circuit voltages are about 0.4 to 0.6 volts above the theoretical values reported here. Lithium/chlorine secondary batteries in lithium chloride at 600°C have been investigated for vehicle propulsion (44, 45). While 250 Wh/lb of the active ingredients have been obtained, the storage, heating and insulating systems will lower this significantly, possibly to the 20 Wh/lb of a similar primary thermal battery (46).

HALIDE COUPLES

Table 3 is the first of two on lithium/halide couples. These Tables do not contain all possible couples, but only representative ones.

Cobalt fluorides have been made into electrodes. However, utilization is low and the electrodes decompose during manufacture (47).

TABLE 3

Lithium/Halide Couples

Reaction	Theoretical Cell Voltage	Theoretical Energy Density Wh/lb
$3Li + CoF_3 \rightarrow 3LiF + Co$	3.63	969
$2Li + CoF_2 \rightarrow 2LiF + Co$	2.88	623
$2Li + CdF_2 \rightarrow 2LiF + Cd$	2.70	880
$2Li + HgF_2 \rightarrow 2LiF + Hg$	3.40	328
$Li + AgCl \rightarrow LiCl + Ag$	2.84	230
$2Li + PbCl_2 \rightarrow 2LiCl + Pb$	2.35	196

Experimental lithium/cadmium fluoride cells have given utilization of 75% (48, 49). The low operating voltage, of the order of 1.5 volts is due to poor conductivity of the cadmium fluoride. As secondary electrodes, utilizations of 15 to 42% have been obtained (50). Although not shown here a lithium/silver fluoride cell which has a high theoretical potential (4.78V) and high energy density (678 Wh/lb) gave over 3.0V with 70 to 100% utilization (51, 52). Poor charge retention on wet stand was a major short coming (53).

Lithium/silver chloride prototype secondary cells have been built (31, 54, 55). Utilization is very poor and they are really not suited for practical high energy density applications.

The more promising halide couples are shown on Table 4. I will be dealing with the carbon fluoride and copper halides later. Nickel fluoride and nickel chloride secondary cells have been built (56-58). Average run-out voltages were of the order of 1.75 volts. High conversion efficiencies can be obtained, but on cycling there is evidence of interaction between the halides and the electrolyte (29, 30).

TABLE 4

Lithium/Halide Couples (continued)

Reaction	Theoretical Cell Voltage	Theoretical Energy Density Wh/lb
$n\mathrm{Li} + (\mathrm{CF})_n \rightarrow n\mathrm{LiF} + n\mathrm{C}$	3.20	1026
$2\mathrm{Li} + \mathrm{CuF_2} \rightarrow 2\mathrm{LiF} + \mathrm{Cu}$	3.54	746
$2\mathrm{Li} + \mathrm{NiF_2} \rightarrow 2\mathrm{LiF} + \mathrm{Ni}$	2.83	623
$\mathrm{Li} + \mathrm{C_4F} \rightarrow \mathrm{LiF} + 4\mathrm{C}$	3.35	550
$2\mathrm{Li} + \mathrm{CuCl_2} \rightarrow 2\mathrm{LiCl} + \mathrm{Cu}$	3.07	504
$\mathrm{Li} + \mathrm{CuCl} \rightarrow \mathrm{LiCl} + \mathrm{Cu}$	2.75	316
$2\mathrm{Li} + \mathrm{NiCl_2} \rightarrow 2\mathrm{LiCl} + \mathrm{Ni}$	2.57	436

Carbon fluoride cells can use the monofluoride, tetracarbon fluoride and fluorinated carbon. These cells give energy densities in the order of 100 to 250 Wh/lb depending upon the electrolyte used (13, 15, 16, 20). Figure 1 shows run-outs for cells equivalent to the "D" size, for the monofluoride at various temperatures. Flat voltage curves are obtained and the low temperature performance is good. Utilization at room temperature is between 50 to 75%. However, in phosphorous oxychloride and thionyl chloride the equivalent of 400% has been obtained because of solvent reduction (13,16). Incidently lithium/carbon cells in the same electrolytes have given up to

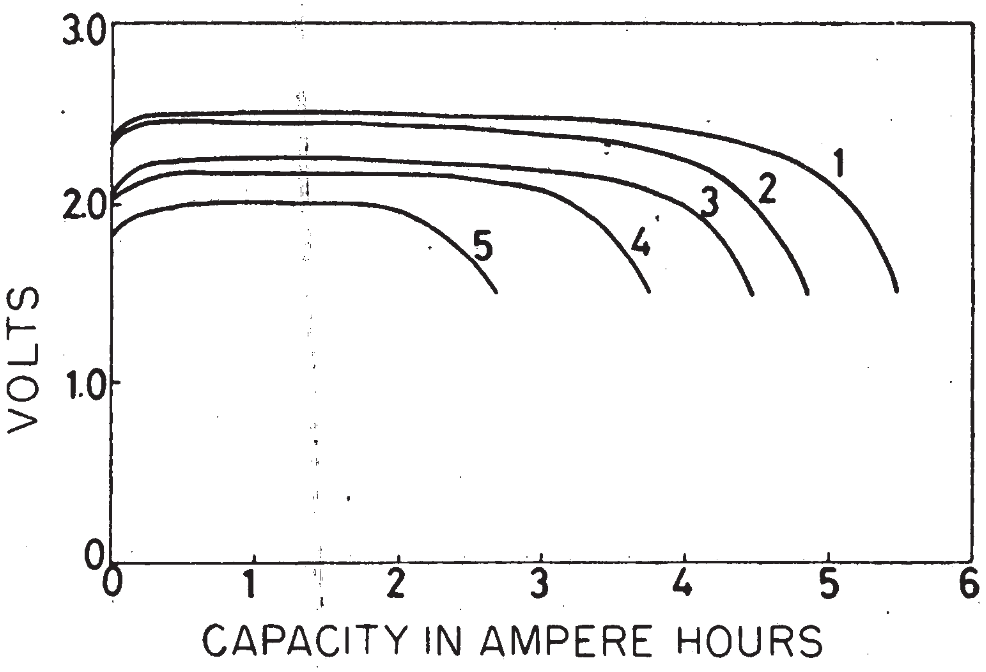

Figure 1: Discharge of Li/$(CF)_n$ cells with an 8 ohm load at the following temperatures; 1: 50°C, 2: 20°C, 3: 0°C, 4: -10°C, 5: -20°C (ref 74).

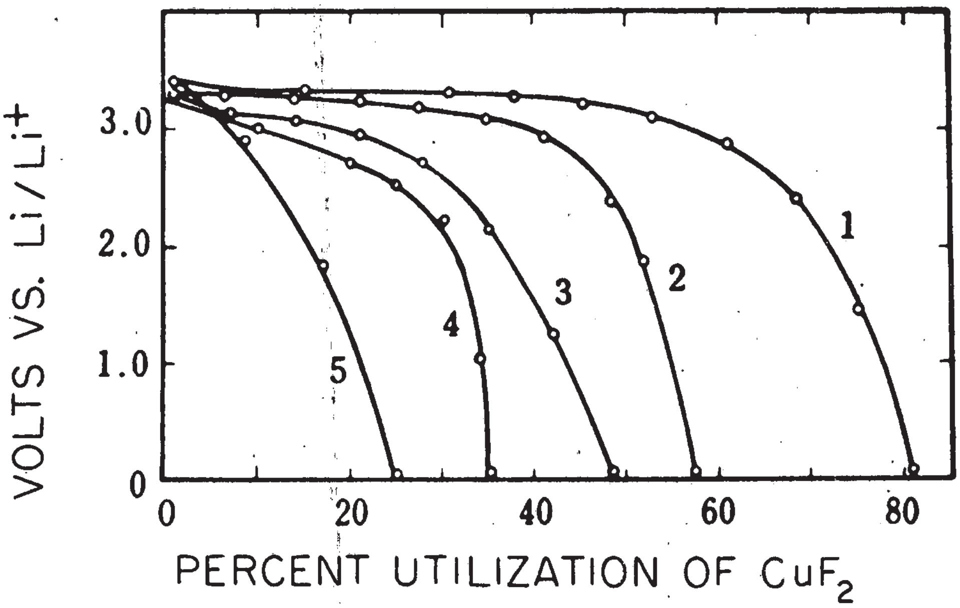

Figure 2: Discharge of CuF_2 electrodes at current densities of 1 to 5 mA/cm^2 as indicated on curves (ref 62).

250 Wh/lb (14-16). Fluorinated carbons which have a formula C_xF where x can be between one and 7.5 have been reported to yield virtually 100% utilization (59, 62).

Figure 2 shows plots of voltage versus percent utilization for copper fluoride electrodes for different current densities (62). Because of its low conductivity, the utilization falls rapidly with current density (63). However, the high voltage has resulted in reports of energy densities as high as 225 Wh/lb exclusive of the case material. (12, 54, 64-66). Electrode structures that would maximize the contact of copper fluoride particles with electronically conductive surfaces have been tried to improve performance at high currents (62, 65, 66). Graphite and silver flakes give some improvement.

Attempts to make secondary batteries have not been too successful. Copper fluoride is not easily formed anodically and energy densities have been under 20 Wh/lb (67).

Figure 3 shows the performance of lithium/cupric chloride cells discharged at the ten hour rate for different temperatures; good utilization is obtained down to -55°C (68). The two plateaus for curves 1 and 2 are for cupric chloride reduction to cuprous chloride or copper, followed by the simultaneous reduction of the two halides to copper. Sixty to one hundred Wh/lb have been obtained (68, 69). One disadvantage is the wet-stand capability. Losses of up to 20% over one month have been reported. The couple must be used as a reserve battery if long term storage is desired.

Secondary batteries in which the active material is cuprous chloride have yielded energy densities of the order of 60 Wh/lb and can also operate between -50 and +55°C (70, 71).

OXIDE COUPLES

Table 5 shows some lithium/oxide couples. While most have been built as experimental cells, the most highly developed is the lithium/sulfur dioxide system (19,72-74). More than one manufacturer is producing these and they come in a variety of sizes. I will say more about these later.

Silver oxide gives high utilization but suffers from a high solubility in propylene carbonate electrolyte (75-77). Manganese dioxode can be discharged to about 90% above 2.0V (75, 78), but the voltage tends to decay rapidly on discharge (79). As a secondary electrode, recharge capabilities are poor (77). The tungsten oxide electrode like the carbon monofluoride and carbon cells give more than the theoretical utilization (approximately 20%) in phosphorous oxychloride (15). Mercuric oxide cells give flat voltage curves at about 2.0V and utilization is of the order of 60% (80).

I have listed the silver chromate couple here, but others such as copper, mercury, lead and cobalt chromates can be used (81, 82). Operating voltages are between 2.0 and 2.8 volts, with silver chromate giving the

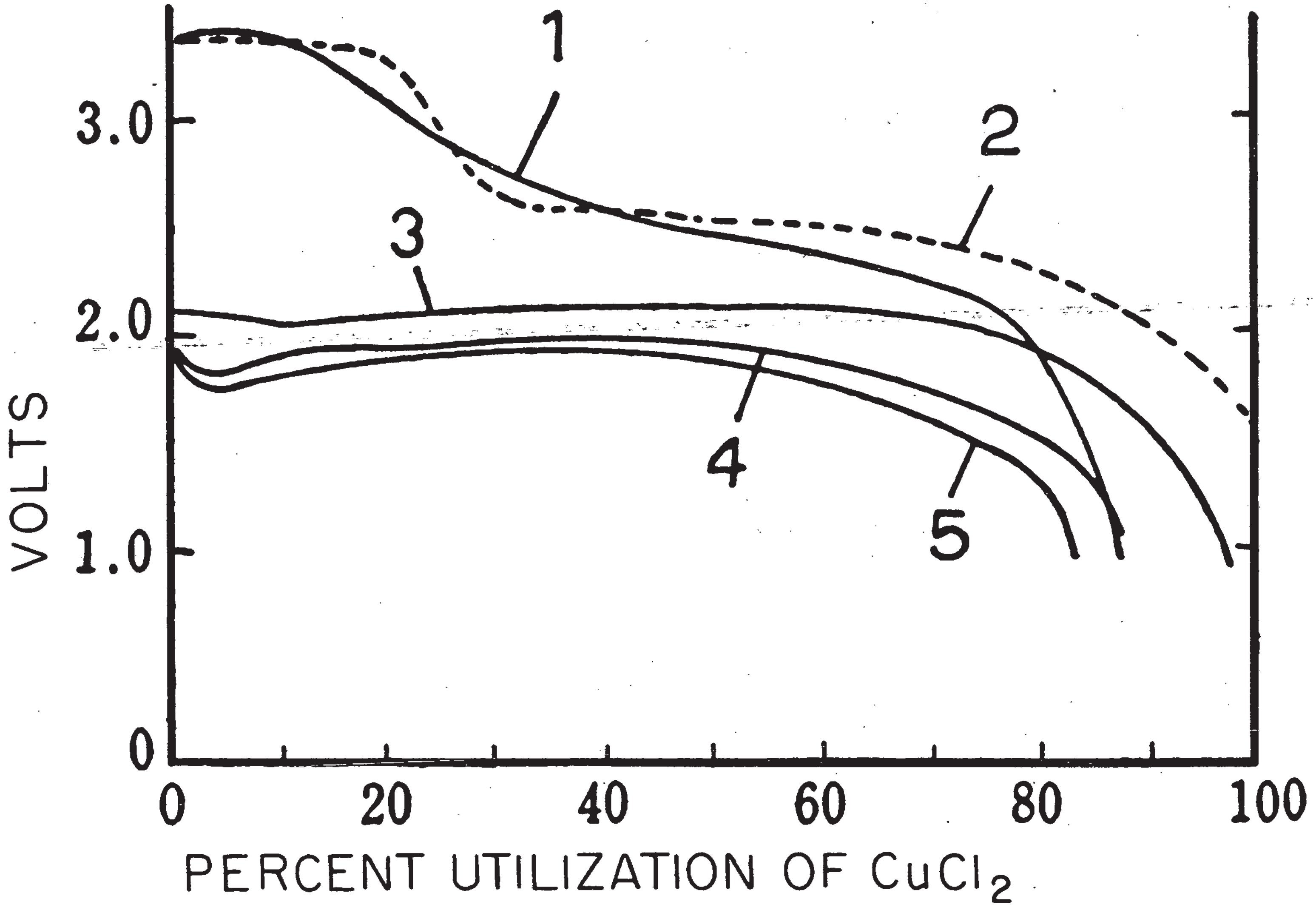

Figure 3: *Discharge of 2Ah Li/$CuCl_2$ cells, 200 mA, at the following temperatures; 1: 75°C, 2: 25°C, 3: -40°C, 4: -50°C, 5: -55°C (ref 68).*

best utilization and highest voltage. Lithium/mercuric chromate cells come a close second. The cells with chromates of silver and mercury give energy densities in the order of 100 Wh/lb (82).

TABLE 5

Lithium/Oxide Couples

Reaction	Theoretical Cell Voltage	Theoretical Energy Density Wh/lb
$2Li + CuO \rightarrow Li_2O + Cu$	2.24	583
$2Li + AgO \rightarrow Li_2O + Ag$	2.96	523
$2Li + 2SO_2 \rightarrow Li_2S_2O_4$	2.95	501
$2Li + MoO_3 \rightarrow Li_2O + MoO_2$	2.90	447
$2Li + 2MnO_2 \rightarrow Li_2O + Mn_2O_3$	2.69	349
$2Li + HgO \rightarrow Li_2O + Hg$	3.13	348
$2Li + WO_3 \rightarrow Li_2O + WO_2$	2.95	292
$2Li + PbO_2 \rightarrow Li_2O + PbO$	2.74	263
$2Li + Ag_2CrO_4 \rightarrow Li_2CrO_4 + 2Ag$	3.35	235

Figure 4 shows the performance of "D" size sulfur dioxide cells discharged at 0.25A for various temperatures. At -40°C considerable utilization is obtained. The cathodes consist of a high surface area carbon electrode and the depolarizer is sulfur dioxide which is injected under pressure, roughly equal to its liquifying pressure at room temperature. Experimental cells using nitrogen dioxide instead of sulfur dioxide have been built. These gave good discharge capabilities at about 3.5 volts (43).

Figure 5 shows a comparison of room temperature discharges for sulfur dioxide, molybdenum trioxide and copper sulfide "D" size cells. As can be seen, the molybdenum trioxide cells behave very much like the sulfur dioxide cells. They give utilizations of the order of 70 to 80% and energy densities

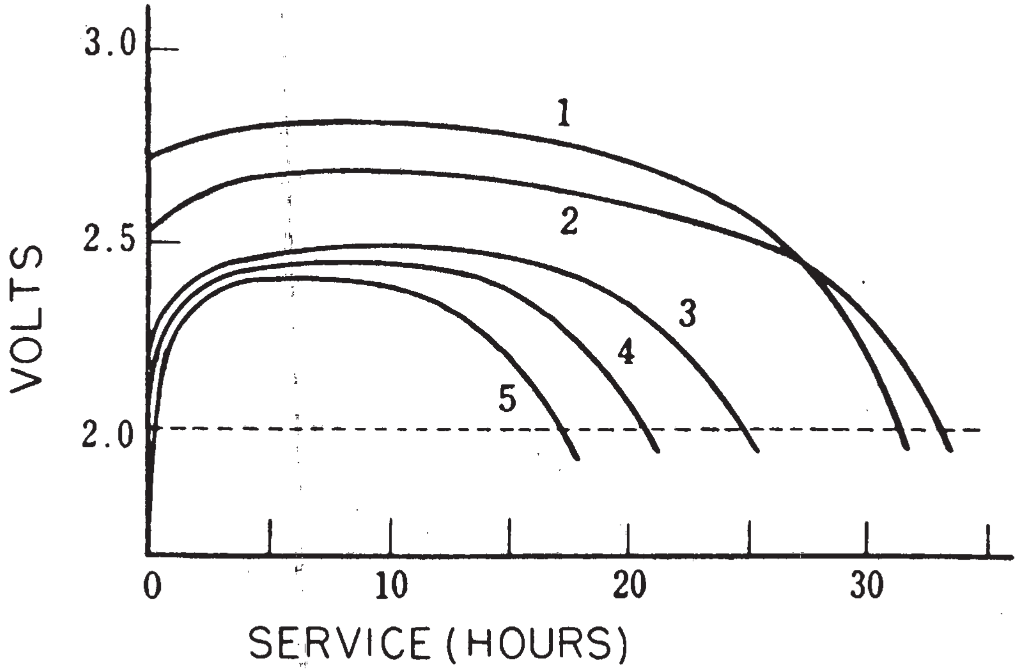

Figure 4: Discharge of "D" size Li/SO_2 cells, 250mA, at the following temperatures; 1: 51.7°C, 2: 23.9°C, 3: -17.8°C, 4: -28.8°C 5: -40°C (ref 19).

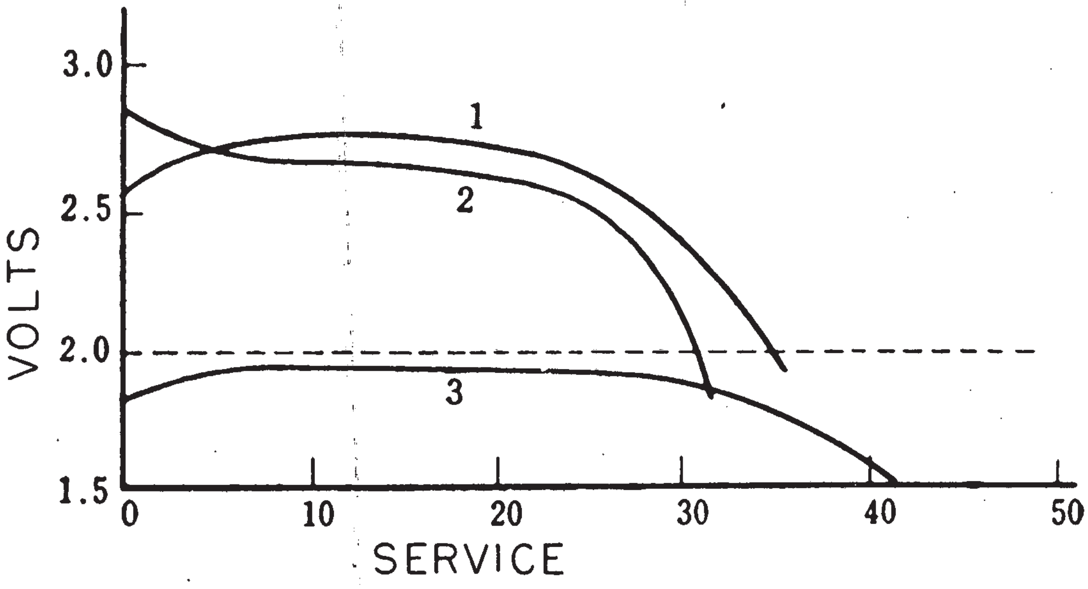

Figure 5: Discharge of "D" size lithium cells at 23.9°C and 250mA. 1: Li/SO_2, 2: Li/MoO_3, 3: Li/CuS (ref 74).

of 85 to 90 Wh/lb. A secondary version of this system has been demonstrated to be feasible (77, 84, 85). Experimental primary lithium/vanadium pentoxide cells have been built and behave much like the molybdenum trioxide cells (78).

SULFIDE COUPLES

We see on Figure 5, the discharge curve for the lithium/copper sulfide couple. The run-out is below 2.0 volts and below that for the other two systems (26, 70, 86). In general, the sulfide couples discharge at a low voltage, below 2.0 volts as can be seen by the theoretical cell voltages in TABLE 6. In spite of the low voltage, high utilization of the copper sulfide makes possible energy densities close to that of the lithium/sulfur dioxide system. However, the utilization does depend greatly on the structure of the electrode and the electrolyte (87).

TABLE 6

Lithium/Sulfide Couples

Reaction	Theoretical Cell Voltage	Theoretical Energy Density Wh/lb
$2Li + CuS \rightarrow Li_2S + Cu$	2.27	504
$2Li + CuBS \rightarrow Li_2S + Cu + B$	1.99	403
$4Li + Ni_3S_2 \rightarrow 2Li_2S + 3Ni$	1.80	327
$12Li + Ni_7S_6 \rightarrow 6Li_2S + 7Ni$	1.80	405
$4Li + NiS_2 \rightarrow 2Li_2S + Ni$	1.80	571
$8Li + Ni_3S_4 \rightarrow 4Li_2S + 3Ni$	1.80	521
$2Li + NiS \rightarrow Li_2S + Ni$	1.80	421

The copper borosulfide cell is a relatively new experimental system (88, 89). Utilization at room temperature is of the order of 50% and it tends to drop off more rapidly than the sulfur dioxide cells at low temperature.

Of the five nickel sulfides listed, NiS and NiS_2 are insulators while the rest are conductors (90, 91). The lithium/Ni_3S_2 couple appears to be the most promising. Cells have been built that give approximately 100 Wh/lb. Low temperature performance down to -40°C has been studied and reasonable behaviour is obtained in butyrolactone. About -20°C is the limit for propylene carbonate. The output voltage tends to drop rapidly with temperature for all the sulfides.

HAZARDS

One of the aspects of lithium batteries that we should not forget is the hazards. All of these batteries have high energy densities, that is a lot of energy in a small package. In some fields energy densities of this size would be classified as explosives.

Lithium, although not as explosive as sodium or potassium when in contact with water is none the less very reactive, especially if hot or in the presence of oxidants, and should not be regarded lightly. Perhaps the ultimate in safety with regard to lithium electrodes may be ones which contain coarse lithium powder mixed with a low thermal conductivity additive and which can decompose and blanket the cell if it becomes exposed to water or oxygen at high temperatures. Energy density need not suffer significantly because of the small amount of lithium present.

You all know that care must be taken with the halogens, but possibly little is appreciated about the toxicity of other components. Sulfur dioxide is dangerous to life at 400 to 500 ppm. Fortunately one ppm can be detected by taste and 20 ppm is irritating to the eyes, nose and throat. Thionyl chloride is similar to sulfur dioxide in odor and detectibility, but more toxic. As low as 20 ppm is fatal to some animals (92).

It is therefore necessary to take precautions when using these cells. I am not trying to overplay the hazards, but we must not overlook them. I am confident that with the right approach these cells can be made as safe as dry cells.

CONCLUSIONS

I have tried to give a short outline of lithium batteries using various electrolytes and cathodes. As you can see they have the potential of offering high energy densities. Excellent performance at low temperatures will undoubtedly expand their applications.

The field of lithium batteries in a little over a decade has come from a laboratory curiosity to commercially available products. We are still on the threshold and I think we can optimistically look to the future for continued rapid growth. The more highly developed systems today are at the top of the energy density spectrum for batteries, yet if one recalls some of the slides shown, these batteries are really at the low end of the energy density spectrum for lithium batteries.

I would be remiss if I did not offer my congratulations to those who are working in this field and an exhortation to continue to strive for improved and safe lithium batteries.

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