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**DEVELOPMENT OF LITHIUM/METAL SULFIDE BATTERIES
AT ARGONNE NATIONAL LABORATORY:
SUMMARY REPORT FOR 1976**

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MASTER

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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PREFACE

This report is a summary of the accomplishments during 1976 in Argonne National Laboratory's program on high-temperature secondary batteries. The work is carried out principally in Argonne's Chemical Engineering Division, and involves the efforts of many scientists and technicians. Some of the efforts of these individuals are reflected in the list of publications at the end of the report, and in other publications planned for the near future. Although acknowledgment of the contributions of individuals is beyond the scope of this brief report, their efforts are essential to the success of the program.

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DEVELOPMENT OF LITHIUM/METAL SULFIDE BATTERIES
AT ARGONNE NATIONAL LABORATORY:
SUMMARY REPORT FOR 1976

ABSTRACT

This report consists of highlights for 1976 of Argonne National Laboratory's program on the development of lithium/metal sulfide batteries. The intended applications for these high-performance batteries are stationary energy storage for electric utility systems and electric vehicle propulsion. The battery cells consist of a lithium-aluminum alloy negative electrode, an FeS or FeS₂ positive electrode, and a molten LiCl-KCl electrolyte, which requires an operating temperature of 400-450°C. Most of the cells tested during the year were of a prismatic design, with capacities in the range of 100-150 A-hr.

Contracts were continued with three industrial firms on the development of cell fabrication techniques and the fabrication of cells for testing at Argonne. Industrial contracts were also continued on the development and fabrication of electrical feedthroughs and electrode separators. In addition, the Atomics International Division of Rockwell International is conducting a more general research and development effort on lithium/metal sulfide batteries under a contract with Argonne. These contracts have proved to be an effective means of transferring this battery technology to industry.

A design study was initiated on a 6 MW-hr battery module for stationary energy storage. This module is expected to be ready for testing in the Battery Energy Storage Test (BEST) Facility in 1981 or 1982. A conceptual design for a 30-kW-hr electric-vehicle battery, which has been completed, will serve as the basic design for the Mark I battery that is to be tested in a van.

In the base technology development effort at Argonne, the program has been separated more clearly between the stationary energy storage and electric vehicle applications because of the different performance and lifetime requirements. The engineering work has been concerned mainly with the effects of modifications in cell designs and materials on performance and lifetime. The assembly of cells in the uncharged state was demonstrated successfully and found to offer some advantages. A large proportion of the Argonne effort was directed to the testing of industrially fabricated cells of various designs.

Materials development studies have continued on electrical feedthroughs and new types of separators, including papers, felts and powders. Chemistry studies have involved alternative electrolyte compositions, liquid lithium electrodes, reaction mechanisms and preparative methods for electrode materials.

I. OVERVIEW OF BATTERY PROGRAM

The lithium/metal sulfide battery program at ANL has the dual objective of developing batteries for use as power sources for electric vehicles, and large-scale, stationary energy-storage devices for applications such as load-leveling on electric-utility networks. During the past year, the research and development efforts on batteries for the two applications have been combined into a single program activity. At the same time, the performance and cost goals and the technical approaches for the electric-vehicle and stationary energy-storage batteries have become much more clearly separated. These goals--for both near-term and more advanced batteries--are summarized in Table 1.

Table 1. Performance and Cost Goals for Lithium-Aluminum/Metal Sulfide Batteries

Battery Goals	Electric Vehicle Propulsion		Stationary Energy Storage	
	Mark I 1978	Prototype 1985	BEST ^a 1981	Prototype 1985
Power				
Peak	30 kW	60 kW	1.5 MW	25 MW
Sustained Discharge	12 kW	25 kW	1 MW	10 MW
Energy Output	30 kW-hr	45 kW-hr	5 MW-hr	100 MW-hr
Discharge Time, hr	4	4	5	5-10
Charge Time, hr	8	5-8	8	10
Cycle Life	300	1000	1000	3000
Cost of Cells, \$/kW-hr	-	35	25-30 ^b	20

^a Battery Energy Storage Test Facility (discussed below in the text).

^b Projected cost for a production rate of 2000 MW-hr/yr.

The divergence between the development work on the electric-vehicle battery and that on the stationary energy-storage battery results from the different requirements of the two applications. The electric-vehicle battery must have a high specific energy (about 150 W-hr/kg) to provide an adequate driving range and sufficiently high peak and sustained specific power to maintain normal highway speeds and to allow passing and hill-climbing. For the stationary energy-storage battery, the specific energy and specific power requirements are less stringent, but the batteries must have a lower cost and longer lifetime.

These different requirements are reflected in the cell designs for the two applications. The electric-vehicle battery cells are smaller (13 by 18 cm), and the active material in the positive electrode is FeS₂, which gives higher specific power and specific energy than FeS, but requires molybdenum

current collectors. The cells for stationary energy storage are larger (25 by 25 cm) and the material in the positive electrodes is FeS, which is compatible with iron or steel current collectors. Because of the relative ease of joining iron or steel current leads, it may be possible to use multiple-electrode cells for stationary storage instead of the one-positive, two-negative electrode arrangement used in the electric-vehicle cells. Other design differences, such as the electrode thicknesses, are also anticipated for the two applications.

The passage by Congress of the Electric and Hybrid Vehicle Research, Development and Demonstration Act of 1976 has heightened national interest in the electric-vehicle battery. Our plans call for testing an electric-vehicle battery (Mark I)--first in the laboratory and then in a van--during 1978. The performance goals for the Mark I battery (Table 1) do not include a cost goal. However, the goals for more advanced electric-vehicle batteries include realistic cost goals, as well as more stringent performance goals. A conceptual design for a 30-kW-hr electric-vehicle battery, completed during the past year, will serve as the basic design for the Mark I battery.

A design study has also been started on a 6 MW-hr battery module for stationary energy storage; the design provides a module size that can be transported by truck. This module is expected to be ready for testing in the Battery Energy Storage Test (BEST) Facility in 1981 or 1982. The BEST Facility is a joint undertaking by ERDA and the Electric Power Research Institute and will be used to test advanced energy-storage batteries on a utility network.

The present effort in the program is directed toward the development of prismatic cells with vertically oriented electrodes. Most of the cells have two negative electrodes of a solid lithium-aluminum alloy (~50 at. % Li), a central positive electrode of either FeS_2 - CoS_2 or FeS - Cu_2S , a separator for electrical isolation of the electrodes, and an electrolyte of molten $LiCl-KCl$ eutectic (mp, 352°C) that fills the pores in the electrodes and the separator. The use of this electrolyte requires operation of the cell at about 400 to 450°C. Boron nitride fabric separators, which are used in most of the cells, provide satisfactory electrical isolation of the electrodes. However, the boron nitride cannot be woven finely enough to completely prevent the escape of the particulate active materials from the electrodes. For this reason, finely woven zirconium oxide fabric is usually placed over the faces of the positive electrode and fine steel or stainless steel screen over the faces of the negative electrodes. Development work is in progress on lower-cost, thinner separators such as papers, which will serve the dual function of isolating the electrodes electrically and retaining the particulate material in the electrodes. Work is continuing on the development of cells with initially uncharged electrodes,* but the development of cells with initially charged electrodes is also being pursued.

The participation of industrial firms in the development and fabrication of cells and components, begun in 1975, is continuing. Under contracts with ANL, three firms--Gould Inc., Eagle-Picher Industries, Inc., and Catalyst

*These cells are fabricated with negative electrodes of porous aluminum and positive electrodes of Li_2S and iron; upon first charge, $Li-Al$ alloy is formed in the negative electrode and FeS_2 (or FeS) is formed in the positive electrode.

Research Corporation--are developing manufacturing procedures and fabricating test cells. By the end of December 1976, about 44 cells of various designs had been produced by these three firms. In addition, the Atomics International Division of Rockwell International is conducting a more general research and development program, mainly on Li-Si/FeS cells and components, under a contract with ANL. In the area of materials and components for Li-Al/FeS_x cells, the Carborundum Corporation, the University of Florida,* Fiber Materials, Inc., and Zircar Products, Inc. are fabricating paper and felt electrode separators. Ceramaseal, Inc. and Coors Porcelain are developing electrical feedthroughs and insulators.

The cell and battery engineering effort at ANL is concerned with the design and testing of cells fabricated by the industrial contractors. The strategy being applied is for each industrial contractor to develop a baseline cell design, using both the base technology developed at ANL and their own experience. When the baseline design has been tested and proved acceptable, various modifications are evaluated in an effort to improve the specific energy, specific power, and reliability of the cells. The industrially produced cells are also being tested in series and parallel arrangements to determine their behavior in battery configurations.

The cell and battery engineering work also includes further development of base technology. Modifications in the design and materials used in electrodes, separators, and other cell components are being evaluated to determine their effects on performance and lifetime, with the advances made being incorporated as quickly as possible into cells being fabricated by contractors.

In advanced engineering studies, more innovative approaches to cell designs and materials are explored. Some of these include the design of multiple-electrode cells, the use of additives to improve the performance of lithium-aluminum electrodes, and the design of new types of negative-electrode structures.

A continuing emphasis is placed on the development and evaluation of materials used in the cells and batteries. Development work at ANL on electrical feedthroughs and electrode separators complements the work being done under contracts. In the separator studies, improvements in the properties of felt and paper separators are being sought and new types of separators, e.g., powders and rigid structures, are being developed. The materials work also includes corrosion testing of candidate materials of construction and postoperative examination of cells tested in the engineering programs.

Cell chemistry studies during the past year included assessment of alternative electrolytes, design of liquid-lithium electrodes, identification of the mechanism of discharge in FeS electrodes by metallography, and improvements in preparative methods for electrode materials.

Under a separate budget activity, but related to the lithium/metal sulfide battery program, are continuing investigations of alternative high-temperature cell systems. The objective of this work is to develop a battery that would use low-cost, abundant materials, but still approach or equal the performance of other high-temperature batteries. In these investigations, cells having calcium-alloy rather than lithium-alloy electrodes are showing promise and are currently being scaled up for engineering development.

*Funded directly by ERDA.

II. COMMERCIAL DEVELOPMENT

The commercial development effort is directed to the establishment of a self-sustaining industry capable of producing lithium/iron sulfide batteries in sufficient numbers to meet national needs. This objective is to be accomplished through the use of normal market forces, with a minimum of federal support. One phase of the commercialization effort comprises evaluations of economic factors, such as potential markets and manufacturing costs. An important part of the commercialization effort is the rapid transfer of battery technology to industry. This is being achieved by having representatives of industrial firms participate in the battery program at ANL and by contracting industrial firms to undertake the development and fabrication of cells, the design and development of components, and the design of batteries.

A. Commercialization Studies

A commercial development plan, presently being formulated, consists of a market study, an analysis of battery cost as a function of production rate, a manufacturing plan, a financial plan, and an analysis of competing technologies. The first phase of the market study* is being directed to the identification of special markets, *e.g.*, military, aerospace, and certain industrial users, for the period from about 1981 to 1985. The study is based on the assumption that the batteries would be produced in pilot-scale facilities at a cost of about \$100-300/kW-hr. Preliminary results indicate that these special markets might support several pilot plants with capacities of 100-200 MW-hr/yr.

B. Fabrication of Cells by Industrial Firms

Three industrial firms, under contracts with ANL, are developing manufacturing procedures and fabricating electrodes and cells. These firms are Gould Inc., Eagle-Picher Industries, Inc., and Catalyst Research Corporation. Performance testing of these cells is described in Section IV.

For each new batch of cells, the industrial contractor is first given design guidelines by ANL. Working within these guidelines, the contractor produces a detailed design, which is evaluated by ANL. After design details are mutually agreed upon, a few cells are built by the contractor and tested at ANL. When the design has proved acceptable, a larger batch of cells of this baseline design is fabricated and the cells are tested at ANL. These test results serve as the basis for evaluating the reproducibility in performance of cells of a given design and for modifying cell designs to provide better cell performance.

1. Gould Inc.

The first batch of Gould cells were 13 by 13 cm cells with electrodes made by vibratory loading of active powders into porous current-collector structures. As a result of problems encountered in loading the active powders into the porous structures and in handling the BN separators during cell

*Assistance with this study is being given by personnel from ANL's Division of Energy and Environmental Systems and from three battery developers, namely, Gould Inc., Eagle-Picher Industries, Inc., and Atomics International.

assembly, the cells did not achieve the expected performance. A decision was made to discontinue work on cells with powder-loaded electrodes, and a second contract was made with Gould to produce cells having uncharged electrodes formed by hot-pressing. The cells have molded BN separators (described below) and are larger (13 by 18 cm) than the first cells.

By the end of 1976, Gould had fabricated six uncharged FeS cells of the new design. Four of these are being tested at ANL and the other two will be tested at Gould. Efforts at Gould are now directed toward the design and fabrication of FeS₂ cells. These cells are designed for operation on the upper voltage plateau.*

2. Eagle-Picher Industries, Inc.

Under the initial contract, Eagle-Picher fabricated 24 cells with electrodes made by cold-pressing active powders and electrolyte into metal "honeycomb" current collectors. Tests of the first few FeS₂ cells revealed a weak connection between the molybdenum current collector and terminal lead of the positive electrode; this connection was redesigned before the remainder of the FeS₂ cells were fabricated. A molded BN separator was also incorporated into the cell design. To make this separator, the BN fabric is wetted with an alcohol-electrolyte mixture, formed in a die, and dried. The molded separator is smooth and easy to handle, and can be readily inspected for flaws. (As stated above, this type of separator is also being used for the second batch of Gould cells.)

Work at Eagle-Picher is presently directed toward the fabrication of 58 additional cells, under an addendum to their contract. The 13 by 13 cm cells have cold-pressed electrodes and are assembled in both the charged and uncharged states. The cells are divided into nine groups, each group having a different modification of the baseline design. The modifications include a variation in the ratio of the theoretical capacities of the positive and negative electrodes, an increase in the diameter of the positive lead, and the use of yttria felt instead of boron nitride fabric as the separator material. Provisions are also being made at Eagle-Picher to produce larger (13 by 18 cm) cells.

3. Catalyst Research Corp.

Work at Catalyst Research is directed to the development of procedures for manufacturing cells in a dry-room atmosphere (<1% relative humidity) and for casting lithium-aluminum electrodes in porous metal substrates. Cell testing is also being carried out at Catalyst Research. The cells tested thus far have exhibited excessive degassing, particularly on start-up; the possibility that degassing is related to the fabrication of electrodes in air is being investigated. Progress has been made recently in the development of the cast Li-Al electrode, and Catalyst Research is fabricating cells with cast negative electrodes and FeS₂ electrodes supplied by ANL.

* Two voltage plateaus (FeS₂ \rightarrow FeS and FeS \rightarrow Fe) are obtained in the complete discharge of a Li-Al/FeS₂ cell; these occur at about 1.6 and 1.3 V, respectively. In an upper-plateau FeS₂ cell, either charged or uncharged, the proportions of active materials in the electrodes are chosen so that the cell operates on the 1.6-V plateau (FeS₂ \rightarrow FeS).

C. Development of Cell Components by Industrial Firms

The participation of industrial firms in the development and fabrication of cell components and selected materials is continuing. The main emphasis in this area is on (1) the development of separator materials and the fabrication of these materials into practical separators, (2) the development of insulated feedthroughs, and (3) the production of materials of special importance to cell fabrication, *e.g.*, Li-Al alloys, LiCl-KCl, and Li₂S. Work at ANL on separators and feedthroughs is discussed in Section VI.

Carborundum Corporation is producing boron nitride fiber for near-term use and is also conducting a design and cost analysis for a plant capable of large-scale production of BN fiber. As part of a separate contract, Carborundum produced paper separators of BN fibers bonded with BN; these were subjected to in-cell testing at ANL and appear promising. At the University of Florida, a paper-making machine is being set up for a pilot-plant run to produce BN paper bonded with 10 wt % asbestos. This material is being produced for engineering-scale cell tests. In other work, Fiber Materials, Inc. has developed a process for the manufacture of BN felt, and Zircar Products, Inc. has developed a Y₂O₃ felt separator.

Ceramaseal, Inc. has developed a ram-type feedthrough that has exhibited excellent leak-tightness; attempts are now under way to reduce the weight and size of the feedthrough. The reduction in size is necessary to make it compatible with the thin cells planned for the Mark I electric-vehicle battery. Coors Porcelain has developed a method of fabricating cold-pressed yttria insulators and is working on a nonmetallic braze for the yttria parts.

III. SYSTEMS DESIGN

A. Conceptual Design of a 30-kW-hr Electric-Vehicle Battery

A conceptual design has been completed for the 30 kW-hr electric-vehicle battery (Mark I) that is scheduled for testing in a van in 1978. A sectional view of this battery is shown in Fig. 1. The battery housing provides space for about 200 prismatic (13 by 18 cm) cells. These cells are connected in a parallel/series arrangement to give 100 cell couples connected in series (each cell couple consists of two cells in parallel). This arrangement gives an average battery voltage of about 145 V, with an energy output of about 30 kW-hr at the 5-hr discharge rate. A thermal-insulation system consisting of a vacuum annulus filled with 40-80 reflective surfaces (multifoil) is employed. The total heat loss of the 30-kW-hr battery at an operating temperature of 435°C is projected to be 100-200 W, including the losses through penetrations in the insulation at the end of the battery. Cooling with ambient air controls the battery temperature during vehicle operation; internal resistance heaters are provided to heat the battery from ambient to operating temperature, and to maintain the battery temperature during brief periods of nonuse.

A joint effort was recently undertaken by ANL and Eagle-Picher Industries, Inc. to prepare a more detailed design of the battery housing and to fabricate a prototype housing for testing at ANL. The results of thermal-efficiency tests and heating and cooling tests under simulated conditions of battery operation will provide input to the design of the Mark I battery housing.

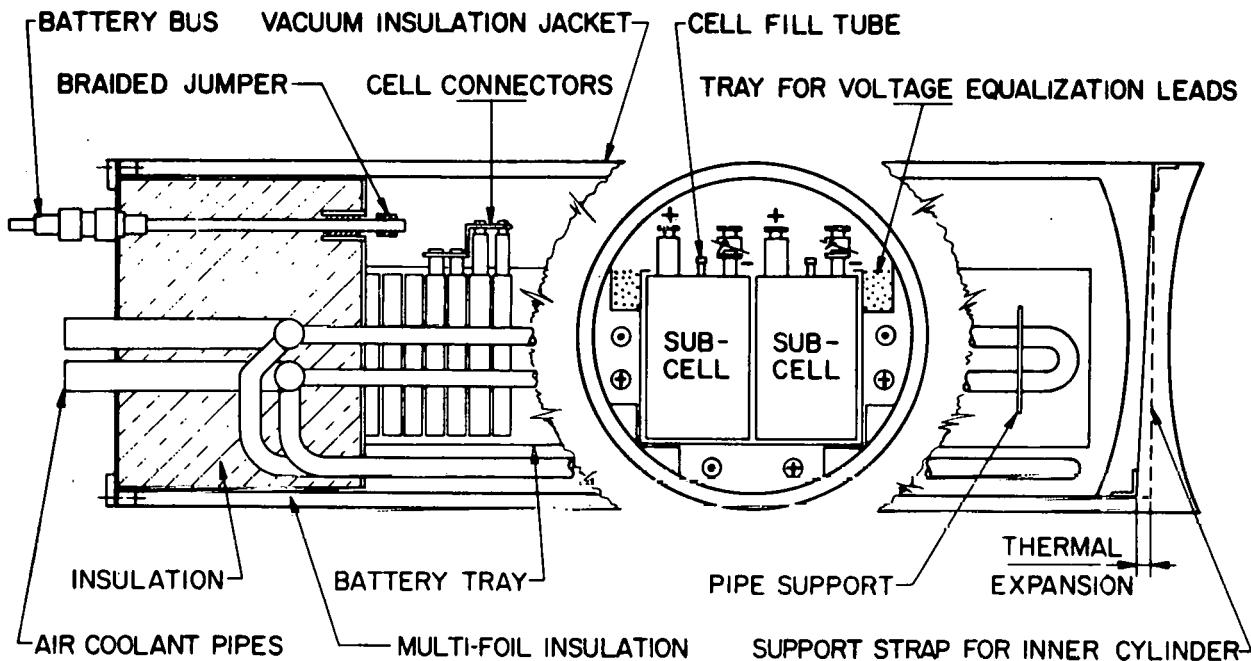


Fig. 1. Conceptual Design of 30-kW-hr Electric-Vehicle Battery (sectional view)

B. Conceptual Design of a Battery Module for Stationary Energy Storage

A conceptual design and cost study for a battery module for stationary energy storage was started in December 1976 as a joint effort between the Chemical Engineering and Engineering Divisions of ANL. This design will serve as a reference for the detailed design and construction of a 6 MW-hr module to be tested in the BEST Facility in 1981 or 1982.

Effort is presently being directed toward the design and cost analysis for the module housing and auxiliaries. The bases for the cost analysis include (1) the use of inexpensive materials,* (2) construction of module housings in a semiautomated plant, and (3) a module size that permits delivery of the housing to the utility site by truck.

Weather-tight construction is specified for the housing to eliminate the need for additional outer protection at the site. About 15 cm of inexpensive, commercially available insulation (e.g., fiberglass or foamed silica brick) is adequate for temperature maintenance. Cooling to remove heat generated by battery inefficiency is provided by the use of once-through ambient air. Work is under way to devise a method for removing faulty cells without cooling the module to ambient temperature.

C. Development of a Charger for an Electric-Vehicle Battery

The feasibility of a battery charger employing charge equalization of the cells was demonstrated in the laboratory early in 1976. A contract was

* The preliminary target goal for cost of the housing is \$5-7/kW-hr of stored energy.

then made with Gulton Industries, Inc. to design and construct an economical and lightweight cell-equalization system suitable for use on mass-produced electric vehicles. The main charger is controlled by the battery terminal voltage. At a specified voltage level, the main charger is deactivated, and the equalizer completes the charging process. The equalizing charger provides an independent constant-voltage source to each cell so that each cell may come to a full charge independently.

Gulton Industries produced two prototype six-cell equalizer systems, one for FeS cells and the other for FeS₂ cells. Both units provide effective cell equalization during a weekend battery charge. In mass-produced quantities, the cost of a charge equalization system is estimated to be about one dollar per cell. This type of charging system also shows promise for use with other battery systems under development for electric-vehicle propulsion.

IV. CELL PERFORMANCE AND LIFETIME TESTING

A. Industrial Cells

The cells produced under contracts with industrial firms are tested at ANL, using a set of standard procedures that permit a comparison of the performance characteristics of cells of different designs. The test procedures include measurements of the capacity and specific energy of the cells as a function of discharge rate and the specific power as a function of discharge current and state of discharge. In the lifetime tests, charge/discharge cycling is continued until the performance (usually capacity or coulombic efficiency) declines to an unacceptable level.

The first cells of the second batch being produced by Gould (see Section II.B) were delivered in December 1976. These cells, which are 13 by 18 cm FeS cells having hot-pressed electrodes fabricated in the uncharged state, are now being tested.

The cells produced by Catalyst Research Corp. were fabricated in a dry-air atmosphere to evaluate the possibility of avoiding the expense and complexity of inert-atmosphere fabrication facilities. The cells showed considerable degassing, especially during start-up; the possibility that the degassing is related to fabrication of the electrodes in air is being investigated. In tests of these cells at Catalyst Research, the most recent result was obtained for a 13 by 18 cm cell with negative electrodes of Li-Al cast in a porous-metal current collector and an FeS₂ electrode supplied by ANL. This cell achieved a peak capacity of about 70% of the theoretical value (144 A-hr); after about 1000 hr of operation, the capacity had declined by ~13%, but the coulombic efficiency remained high (99%).

Eagle-Picher produced both FeS and FeS₂ cells that are 13 cm square and of two thicknesses. The Type A (thin) cells are ~2 cm thick and have theoretical capacities of 70 A-hr; the Type B (thick) cells are ~3.5 cm thick and have capacities of 150 A-hr. The capacities of the positive and negative electrodes are matched in both types of cells.

A comparison of the utilization of active material (percent of theoretical capacity) in Type A and Type B FeS₂ cells is shown in Fig. 2. The results

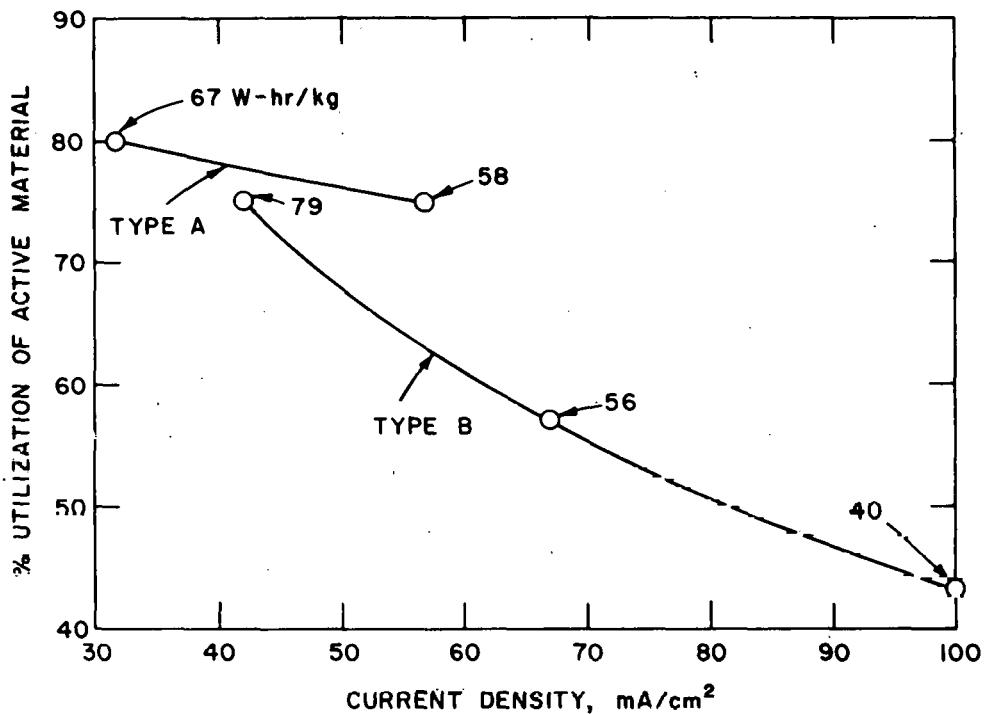


Fig. 2. Utilization of Active Materials in Thick (Type B) and Thin (Type A) FeS_2 Cells as a Function of Current Density

show that thin electrodes effect a higher utilization of the active materials, particularly at higher current densities. In the thicker cells, not all of the active material is utilized, and the added weight of the unused material decreases the specific energy and specific power.

Three FeS_2 cells (2B4, 2B5, and 2B7; Type B) were subjected to deep cycling* at constant current of 13 A (current density, 42 mA/cm^2) to test the effect of charge cutoff voltage on the utilization of active materials. The results, shown in Fig. 3, demonstrate that increasing the charge cutoff voltage from 2.0 to 2.2 V produced an increase in the utilization of active materials from 80 to 114 A-hr (or from 53 to 76% of the theoretical capacity). The corresponding increase in specific energy was from 60 to 79 W-hr/kg. In contrast to these results, the thinner Type A FeS_2 cells have shown no effect of charge cutoff voltage on the utilization of active materials.

The shapes of the curves in Fig. 3 are typical for the Type B FeS_2 cells; the capacity increases markedly during the first few break-in cycles, reaches a maximum value within about the first 30 cycles, and then decreases to a stable value. After extended cycling, the capacity begins to decline and the test is terminated. (Cell 2B7, for which the curve in Fig. 3 is incomplete, is discussed below.)

*Discharge cutoff voltage, 1.0 V. A deep cycle is generally considered to be one in which more than 50% of the theoretical capacity is discharged.

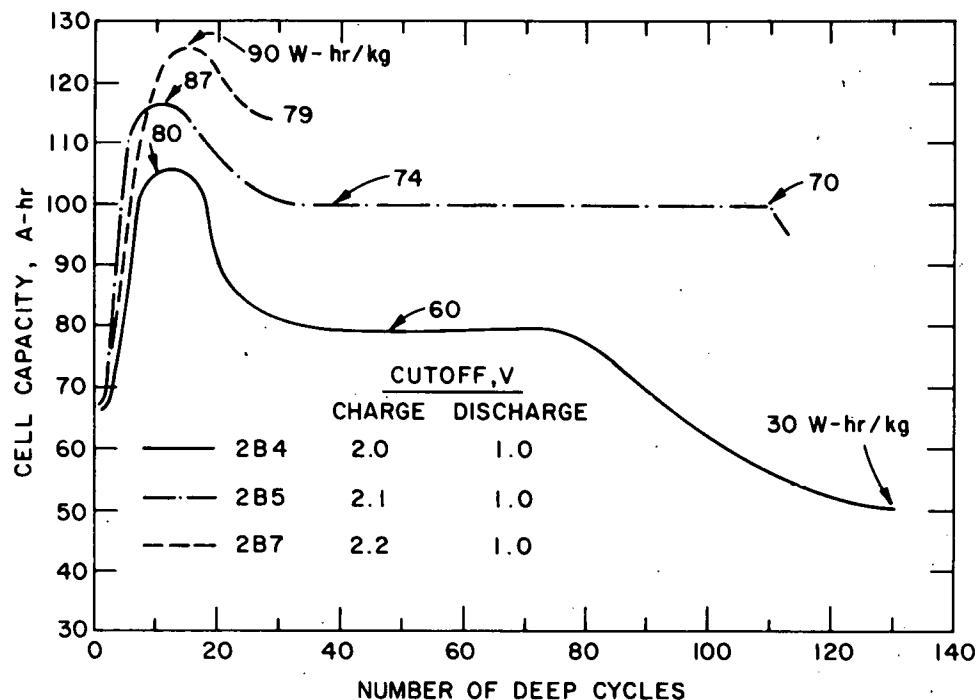


Fig. 3. Cycling Tests of Eagle-Picher Thick (Type B) FeS₂ Cells at 42 mA/cm²

The testing of Cell 2B4 was concluded after 1700 hr and 130 cycles because its capacity declined steadily after the 80th cycle. However, the coulombic efficiency of this cell remained high, even at the end of its life. The operation of Cell 2B5 was terminated after 127 cycles and 2175 hr when the coulombic efficiency decreased markedly (an indication of a partial short circuit). Cell 2B7, after 30 deep cycles, was placed on a modified cycling schedule that was designed to resemble the duty cycle of an electric-vehicle battery and consisted of a combination of shallow and deep cycles at the 2- and 4-hr rates. Its specific energy was 40 W-hr/kg at the 2-hr rate. Under the new cycling schedule, Cell 2B7 also exhibited the characteristic period of stable performance discussed above. When the capacity began to decline, operation of this cell was terminated; at this point, it had completed 2550 hr of operation and 525 (shallow plus deep) cycles. The major conclusions from this series of cell tests are that increasing the charge cutoff voltage of FeS₂ cells from 2.0 to 2.2 V increases their capacity and has no adverse effects on their lifetime.

Preliminary tests of Eagle-Picher FeS cells indicated that they would have considerably longer lifetime than the FeS₂ cells. Because of the limited availability of cell cycling equipment, the decision was made to conduct lifetime tests of these cells as batteries consisting of two or three cells connected in series or parallel, rather than as single cells.

The evaluations of the Eagle-Picher cells have indicated that (1) cells of a given design showed reproducible performance in terms of cycle life, specific energy, and specific power, (2) the performance of the cells was similar to that obtained with cells developed at ANL, and (3) improvements

in cell design and performance are needed to meet the program goals for the electric-vehicle and stationary energy-storage applications.

Late in 1976, the first cells of the second batch being produced by Eagle-Picher were delivered to ANL. By the end of the year, testing of cells having Y_2O_3 felt separators had been started.

In addition to the single-cell tests described above, the performance of cells in series and parallel arrangements is being evaluated, start-up and conditioning methods are being investigated, and various charging procedures and operational schemes are being developed. These tests are being performed using the first batch of Eagle-Picher cells; the FeS cell tests are oriented toward stationary energy-storage applications and the FeS_2 cell tests toward the electric-vehicle battery. Thus far, the cell voltages have been monitored individually, and charge or discharge has been terminated when any one cell in the battery configuration reached its voltage limit. Some of the cells comprising these batteries had been cycled previously in single-cell tests. For instance, the cells in Battery B7-S (discussed below) were operated for about 850 hr and 75 cycles before they were connected in series as a two-cell battery.

The performance of the FeS batteries has been very stable. In Battery B7-S, the capacity of the Type B FeS cells has declined by only about 10% after more than 5000 hr and 375 cycles of operation in series. The cell equalization procedure, which is used in charging the cells, has proven effective in bringing the cells to the same state of charge. The performance of FeS_2 batteries has been much less stable than that of the FeS batteries. The capacity of Battery B8-S, which consisted of two Type A FeS_2 cells in series, declined gradually throughout its lifetime of 2250 hr and 380 cycles. When the test was concluded, the capacity had declined by about 25%.

The results of the battery tests that have been performed to date may be summarized as follows. (1) Both FeS and FeS_2 cells operate satisfactorily when connected either in parallel or in series. (2) The charge equalization procedure is effective in bringing slightly mismatched cells to the same state of charge. (3) The FeS batteries exhibit more stable performance and longer lifetimes than the FeS_2 batteries. (4) Further battery development work is required to meet the previously stated program goals.

B. ANL Cells

The development work presently being carried out at ANL on engineering-scale prismatic cells has the objective of identifying and testing cell and electrode designs that provide improved cell performance. Advances in technology that have been demonstrated in ANL cells are incorporated as quickly as possible into the designs of cells being produced by industrial firms.

1. Li-Al/ FeS_2 Cells

A new design for uncharged cells, shown in Fig. 4, is being tested. The main design features are as follows: (1) the central (positive-electrode) terminal rod is connected to the central current collector by means of a "door-hinge" structure,¹ (2) a thin mat of BN fibers is used as the separator

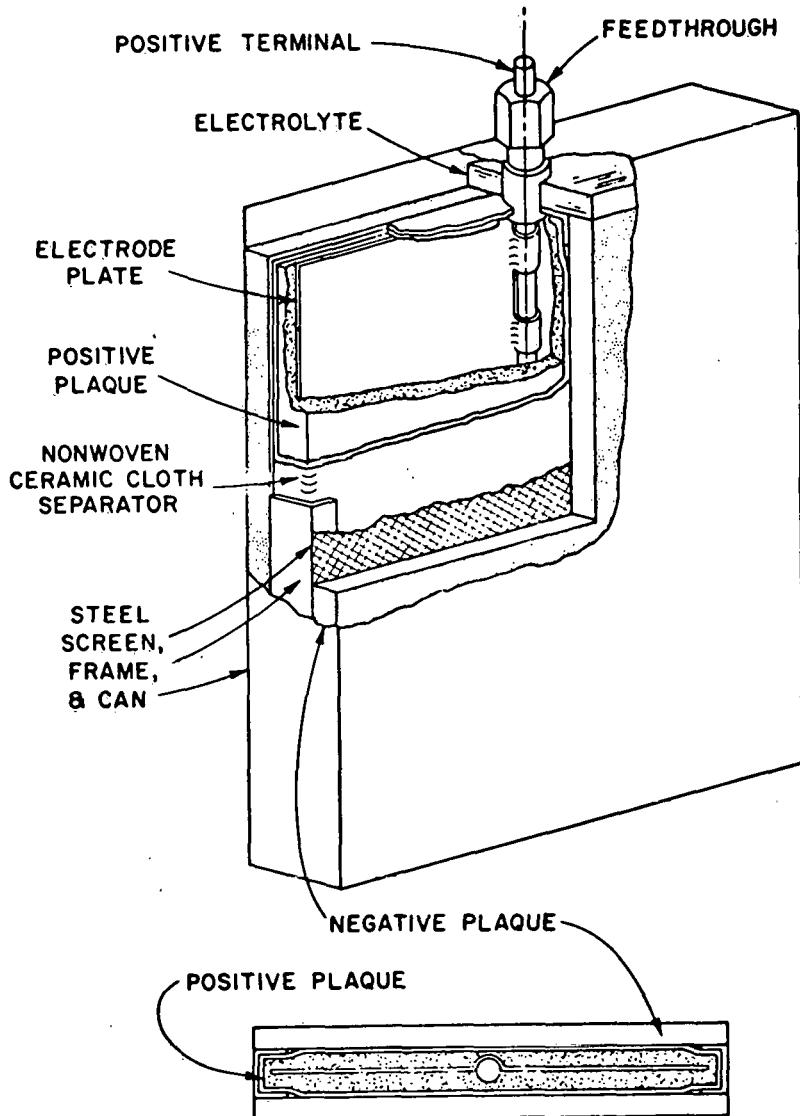


Fig. 4. Design of Engineering-Scale, Uncharged Cell

and particle retainer, and (3) the negative electrodes are cast plates of 25 at. % Li-Al.*

Cell EC-1, an uncharged, two-plateau FeS_2 cell with a hot-pressed positive electrode was operated to test this design. Performance curves for Cell EC-1 are given in Fig. 5. The cell achieved specific energies of 70 and 100 W-hr/kg at the 5- and 10-hr rates, respectively. These high specific energies were attributed to the compact design of the cell and its consequent low weight. However, a short circuit developed early in cell life as a result of the extrusion of active material from the electrodes. Accordingly, improvements in design are being sought which would provide better retention of the active materials in the electrodes and thereby prolong cell life.

* There is some evidence to indicate that excess lithium capacity improves cell performance.

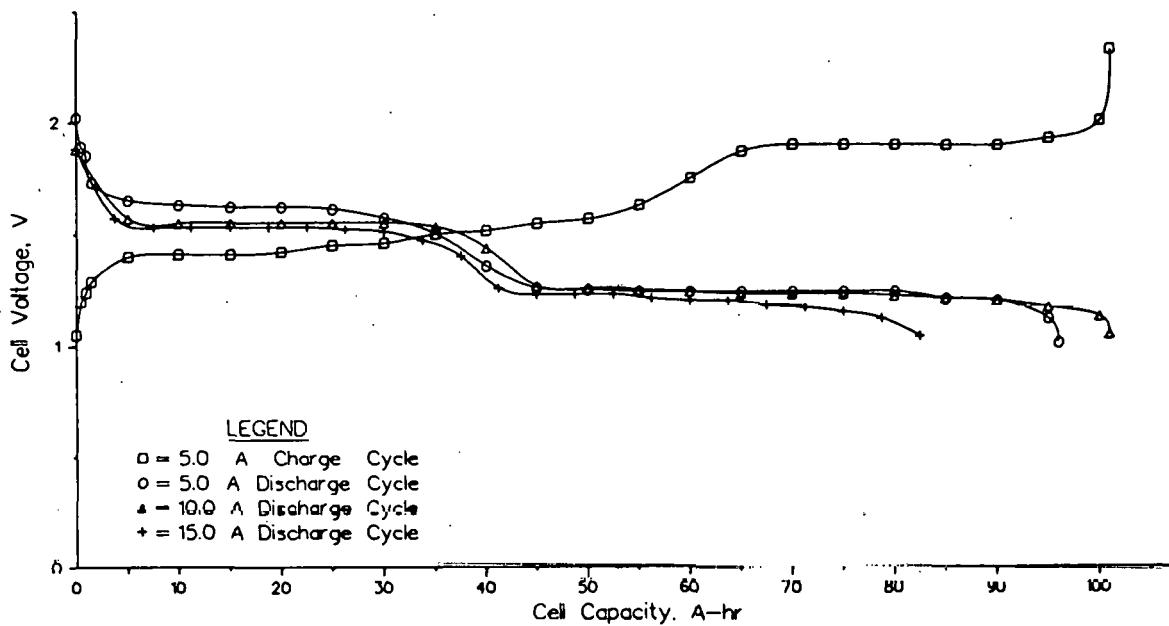


Fig. 5. Performance of an Uncharged, Two-Plateau Li-Al/FeS₂-CoS₂ Cell (Cell EC-1; theoretical capacity, 120 A-hr)

Uncharged FeS₂ cells operated on the upper voltage plateau only are being considered for use in electric-vehicle batteries. The advantages of upper-plateau cells and a demonstration of the feasibility of fabricating and operating an uncharged cell of this type are described later in Section V. Experiments were also undertaken to study the design and performance of these cells more fully. Performance curves for one of these cells (R-24) are presented in Fig. 6. The power output of the cell was stable and utilizations were high (e.g., 83%* at a high discharge current of 40 A, which is equivalent to a 2-hr discharge rate). Because of problems with short-circuiting, the lifetime of Cell R-24 was short. However, the high utilizations achieved in early cycling are encouraging, and efforts are being focused on improving the cell design.

Long cycle life with good retention of capacity has been demonstrated by Cell YF-1. This cell is a charged, intermediate-size cell (about 8 by 13 cm, ~75 A-hr theoretical capacity) having negative electrodes of Li-Al powder in a porous iron structure and an FeS₂ positive electrode in a carbon-bonded structure.† At year's end, this cell had been in operation for more than 4400 hr and 410 cycles at the 5-hr rate, which no decline in capacity from the original value (45 A-hr, 60% of theoretical). A significant feature of cell operation was the use of current-limited, constant-voltage charging (2.1 ± 0.1 V, 10-A maximum current) rather than the constant-current charging presently used for most cells. Constant-voltage charging appears to be a better charging technique for attaining stable cell capacity over large

* Based on FeS₂ \rightarrow FeS (112 A-hr).

† Carbon-bonded positive electrodes, which are suitable for use with both FeS₂ and FeS, are described in the next subsection.

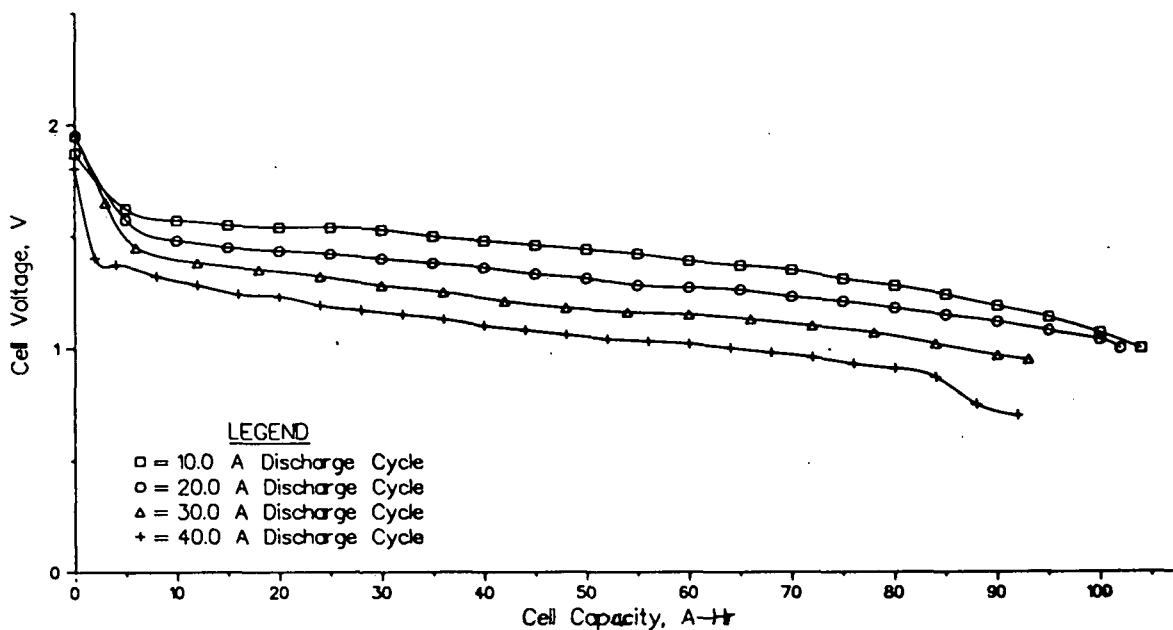


Fig. 6. Performance of an Uncharged, Upper-Plateau Li-Al/FeS₂-CoS₂ Cell (Cell R-24; design capacity, 112 A-hr)

numbers of cycles. Investigations of the effect of various charging techniques on cell performance are being continued.

2. Li-Al/FeS Cells

The effort in this part of the program is directed toward development of cells for stationary energy-storage batteries. Cell SS-1 was the first cell of this type to be scaled up to a larger size (from 13 by 13 cm to 24 by 35 cm). This cell, which was assembled in the charged state, had a carbon-bonded FeS-Cu₂S positive electrode and two negative electrodes of Li-Al in a porous iron structure; its theoretical capacity was 650 A-hr. At the 10-hr rate, the cell achieved a utilization of about 70%, with an energy output of 577 W-hr. The utilization and coulombic efficiency of this cell were as high as those achieved by previous smaller cells, thereby indicating that the scale-up effort was successful.

Development of carbon-bonded electrodes, begun in late 1975, is continuing. This type of electrode is formed from a paste-like mixture of active material, a volatile constituent (ammonium carbonate) to produce porosity in the final structure, and a binder (graphite cement). When this mixture is cured, the resulting electrode structure is a porous current collector in intimate electrical contact with the active material. A charged Li-Al/CuFeS₂ cell, Cell CB-1, with a carbon-bonded positive electrode and a hot-pressed Li-Al electrode has been in operation for more than 550 cycles and 7600 hr. The coulombic efficiency has remained at about 98%, and the capacity is very stable with cycling. In the continuing operation of Cell KK-5, which has a carbon-bonded, uncharged FeS electrode, the coulombic efficiency remains at 98%, and the capacity has declined only 3% after 305 cycles and 5300 hr. The carbon-bonding technique is a promising method for fabricating positive electrodes, and effort will be continued to improve these electrodes.

V. ADVANCED ENGINEERING

During the past year, advanced engineering studies have been focused mainly on (1) the development of the uncharged, upper-plateau FeS_2 cell for the electric-vehicle battery, (2) the development of a multiple-electrode cell for the stationary energy-storage battery, and (3) studies aimed at improving the cycle life of the Li-Al negative electrode. In addition, an advanced cell design has been developed which may permit the use of a variety of separator materials, including papers, felts, fabrics, etc., in compact, high-performance cells.

Upper-plateau FeS_2 cells are of interest for electric-vehicle batteries because of the following potential advantages: (1) high power capability throughout discharge, (2) high average voltage, (3) simplified control of battery voltage owing to the use of a single voltage plateau, and (4) better protection from overcharge.

In earlier work, described in ANL-76-45 (Rev.), upper plateau FeS_2 cells fabricated in the charged state had exhibited high power capabilities. In more recent work, Cell A-2 was built and operated to demonstrate the feasibility of initially uncharged FeS_2 cells of the upper-plateau type. In the fabrication of this cell, a new technique was used for introducing "excess lithium capacity" into uncharged FeS_2 cells by adjusting the $\text{Li}_2\text{S}:\text{FeS}_2$ ratio. This technique may avoid the need for introducing lithium into the negative electrode during fabrication. The specific energy achieved by Cell A-2 was high, namely, 100 W-hr/kg at the 5-hr rate.

A multiplate cell containing three negative and two positive electrodes was also developed and tested. This cell (A-1) was an uncharged two-plateau FeS_2 cell weighing about 3 kg; it was designed to have an energy output of about 400 W-hr, which is believed to be about 40% of full scale for the stationary energy-storage application. This cell achieved a specific energy of about 110 W-hr/kg at the 8-hr rate.

All of the compact engineering cells tested have experienced a common problem, namely, the tendency of the electrodes to expand into the gas/electrolyte chamber at the top of the cell. This expansion often results in short-circuiting. Cells A-1 and A-2 both had operating lifetimes of less than 1400 hr because of this type of short-circuiting. To avoid this problem, a new design was developed in which the upper chamber was eliminated and the cell housing served as a support for the electrodes. This design, shown in Fig. 7, employs a single-piece molybdenum current collector and a single-sheet separator/particle retainer.

Morphological studies of Li-Al negative electrodes have suggested that their cycle life may be limited by the formation of very finely divided Li-Al particles, with a resulting decrease in electronic conduction. Therefore, we are exploring the possibility of controlling the morphological changes by the addition of a third metal to form a ternary alloy. Additions of lead or tin did not improve the performance or cycle life of the electrode. Copper appeared to extend the cycle life, but with some decrease in performance. In contrast, indium had a strongly beneficial effect. For example, a Li-Al electrode containing 3.9 wt % indium achieved high lithium utilization and

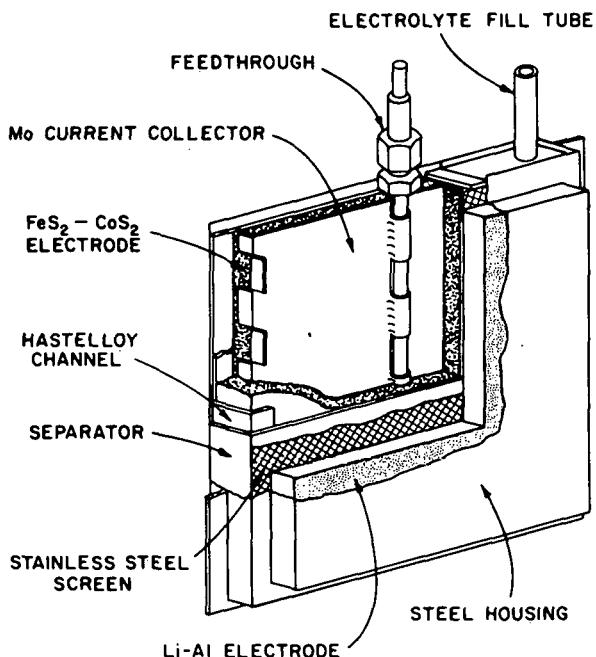
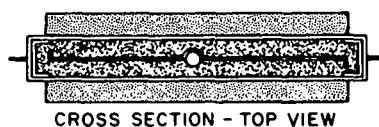


Fig. 7.
Design for Advanced FeS_2 Cell



retained capacity throughout its operation for 136 cycles; concentrations as low as 1 wt % were also effective. Metallographic examination of the Li-Al-In electrodes indicated that the particle size distribution was unusually uniform. The effectiveness of indium as an additive to the Li-Al electrode was demonstrated subsequently in an engineering-scale cell (Li-Al-6 wt % In/FeS-Cu₂S, Cell FM-1), which operated for 225 cycles and 2500 hr with less than 5% decline in capacity.

Two new types of negative electrodes were evaluated as part of the effort to improve current collection. The first type was an uncharged (lithium-free) electrode fabricated from a woven fabric of aluminum ribbon, copper-coated iron wire, and copper ribbon; the electrical performance of this electrode has been excellent. The second type was an uncharged electrode formed by carbon-bonding aluminum powder. This electrode yielded electrical performance equivalent to that of more conventional Li-Al electrodes, with no evidence of lithium carbide formation. Additional, long-term tests are planned for both new types of electrodes to determine their cycle life capabilities.

VI. MATERIALS STUDIES

A. Electrical Feedthroughs

An electrical feedthrough is required to isolate the positive electrode terminal from the cell housing, which is at negative potential. The feedthrough must be leak-tight, light in weight, and inexpensive. The only commercially available feedthroughs that have adequate corrosion resistance are mechanical seals, which are not sufficiently leak-tight, and are unacceptably bulky. A brazed-type feedthrough will probably prove to be the most satisfactory for long-term use, and new and innovative brazed seals are being sought. However, thus far, none has been found that is completely satisfactory. Accordingly, work is in progress to improve the mechanical seals.

The feedthroughs used in most of our test cells have been modified versions of the Conax thermocouple seal. With the addition of BN powder as a sealant and the use of Y_2O_3 as a lower insulator, leak rates as low as about $10^{-6} \text{ Pa}\cdot\text{m}^3/\text{s}$ have been obtained. More recently, by the use of a crimping procedure, the massive retainer nut at the top of the housing was eliminated, thereby reducing the mass of the housing by about 50%. In addition, a secondary solder-glass seal has been added; feedthroughs of this type have demonstrated leak rates as low as $10^{-10} \text{ Pa}\cdot\text{m}^3/\text{s}$, i.e., the feedthroughs are essentially leak-tight. Presently, the feedthrough is being scaled down to an outside diameter of 1.9 cm for compatibility with the designs of the Mark I vehicle cells.

B. Electrode Separator Development

A compact, lightweight cell requires an electrode separator that is thin, inexpensive, and resistant to corrosion in the cell environment. The principal function of the separator is to provide electrical isolation of the electrodes from each other, while permitting the electrolyte to serve as a medium for the transport of lithium ions between the electrodes. It would be desirable to develop a separator material that would also serve as a retainer to confine the particulate active materials in the electrodes--a function now performed by cloths or screens placed over the electrode faces.

Paper, felt, and powder separators are being developed and several of these candidate separators have been successfully tested in special Li-Al/LiCl-KCl/FeS test cells. The following paper and felt separators performed satisfactorily in 1000-hr cell tests: (1) a composite paper of Y_2O_3 fiber/asbestos fiber developed at the University of Florida, (2) a BN felt bonded with BN developed by the Carborundum Corporation, and (3) a Y_2O_3 felt developed by Zircar Products, Inc. An all-powder Y_2O_3 separator was also tested for 2000 hr. These materials all show promise for use as separators; however, tests in engineering cells are needed to verify their utility.

Development work is continuing on both fibrous and powder separators. Efforts on the fibrous separators are devoted to the development and testing of papers and felts having improved properties. Work on powder separators includes assessment of the cell design requirements for powder separators, testing of different types of powders such as MgO, CaO, AlN, BN, and Si_3N_4 , and identification of the optimum particle size.

Porous, rigid separators are also being developed.* The relatively simple fabrication procedures required for these separators may be economically attractive. To date, porous Y_2O_3 structures have been prepared by four different methods. These structures are being characterized by measurements of flexural strength, pore-size distribution, and resistance to gas flow. In-cell testing will be conducted in laboratory cells of the type currently used to evaluate flexible separators.

C. Static Compatibility Tests

Because corrosive conditions differ in the Li-Al, FeS_2 , and FeS electrode environments, static corrosion tests are used extensively to screen candidate materials of construction for compatibility with the appropriate environment. These tests also aid in identifying the mechanisms of various electrochemical reactions.

Static corrosion tests of metals and alloys in equal-volume mixtures of FeS or FeS_2 and LiCl-KCl continued; the tests were conducted for 1000 hr at temperatures of 400, 450, and 500°C. Some of the materials had been tested previously, *e.g.*, molybdenum, nickel, several steels, and Hastelloy B and C; some were tested for the first time, *e.g.*, Inconels and Incoloy. The tests verified the expected correlations between increased temperature and accelerated corrosion rate. An important result of these tests was that the sulfide scales formed on some alloys (Hastelloy, Inconel, stainless steel) in the FeS_2 environment were similar in morphology and growth characteristics to those reported for alloys exposed to H_2S and sulfur-vapor environments. As a result, similar mechanisms for sulfidation of the alloys in the FeS_2 -salt system and in the gaseous environments are postulated.

The scope of the corrosion program was also broadened to study the effects of cell cycling on the corrosion rate of metals and alloys.† The corrosion tests were conducted under conditions that simulated an FeS_2 electrode at various states of charge, namely, 25, 50, 75, and 100%. The results showed that 80% or more of the total corrosion of an alloy (*e.g.*, the Hastelloys and Inconels) occurred between 75 and 100% charge. Estimates of the extent of corrosion of Hastelloy B in an operating cell, based on the test results, were in general agreement with results obtained in postoperative cell examinations.

Compatibility tests were also conducted on metallized Y_2O_3 components being considered for use in brazed-type feedthroughs. Five different metallizing techniques were used to metallize the surfaces of yttria samples. The samples were then exposed to LiCl-KCl saturated with Li-Al, after which they were subjected to fracture tests to evaluate bond strength. The optimum metallizing process was selected on the basis of corrosion resistance, bond integrity, and metallizing composition. Brazed feedthroughs employing this concept will be evaluated further in half-cell tests.

* This work is being performed by the Materials Science Division of ANL.

† The most corrosive environment to which metals and alloys are exposed is a fully charged FeS_2 electrode; in an operating cell, the FeS_2 electrode is at full charge only a small fraction of the time.

D. Postoperative Examinations

Postoperative examinations are conducted on cells to determine materials behavior and to identify causes of cell failure. These examinations employ microscopic, X-ray diffraction, chemical, and electron- and ion-microprobe analyses. The results are evaluated and recommendations are made to engineering personnel on means for avoiding or alleviating the specific problems that lead to poor performance or failure of the cells. During the past year, postoperative examinations were conducted on about 20 engineering-scale (13 by 13 or 13 by 18 cm) prismatic cells. These cells had been subjected to cycling tests lasting from several hundred to several thousand hours.

Examinations of Li-Al/FeS₂ cells yielded the following information. Chemical analyses confirmed earlier findings that the positive electrodes were reasonably homogeneous in sulfide content from top to bottom with no settling of the active materials. Analyses of the positive and negative electrodes showed no indication of cross-migration of the active materials between the electrodes. Metallographic and microprobe analyses of FeS₂ cells showed the presence of crystalline Li₂S and iron in the separator. This phenomenon has been observed only in FeS₂ cells, and additional studies will be required to determine its effect on cell performance, the mechanism by which it occurs, and possible preventive measures if those become necessary.

The most common cause of cell failure was short circuiting resulting from (1) extrusion of active material from one electrode to the extent that it contacted the counter electrode or (2) the presence of metallic or iron sulfide particle stringers in the separator. The extrusion of active material results from swelling of the electrodes, which can be controlled by appropriate design changes. The presence of particles in the separators may be avoided by improving the particle-retention characteristics of the separator and retainers.

The postoperative examinations also indicated that design modifications were needed to avoid brittle fracture of the connection between the molybdenum lead and the current collector in FeS₂ electrodes and to prevent ZrO₂ retainer cloths from being cut by honeycomb structures in positive electrodes. Examination of a cell that had been operated in an air atmosphere showed no significant differences from those operated in helium. The importance of maintaining proper operating conditions was shown by the observations that aluminum had reacted with the current collector of the negative electrode in cells operated above 450°C and that overcharge of a cell causes excessive corrosion of the positive current collector.

VII. CELL CHEMISTRY

The cell chemistry studies have been concerned mainly with (1) various problems arising in the cell development work, (2) improvements in the active materials and electrolyte used in the Li-Al/metal sulfide cells, and (3) a better understanding of fundamental cell processes.

Because of the increased interest in the fabrication of cells in the uncharged state, the availability of high-quality Li₂S for use in the positive electrode has assumed greater importance. Eagle-Picher Industries, Inc. has

developed a process for preparing Li_2S by the reaction of H_2S with Li_2CO_3 at 700-800°C; this method of preparation was suggested by ANL on the basis of literature data. The Li_2S product is whiter and more dense than that produced by the more conventional reaction of n-butyllithium with H_2S in an organic solvent. Analyses of four lots of Li_2S produced by the new process indicated a purity of 99%.

A variety of studies have been conducted on alternative electrolyte compositions. The addition of 10 at. % potassium ions to the LiF-LiCl-LiBr electrolyte system had the desired effect of lowering the melting point well below its original value of 445°C, but the phase relationships of the modified electrolyte were not favorable for its use in cells. Additions of up to about 20 mol % NaCl to the LiCl-KCl eutectic electrolyte did not prevent the formation of "J" phase ($\text{LiK}_6\text{Fe}_{24}\text{S}_{26}\text{Cl}$) during the discharge of FeS electrodes, but they did appear to promote conversion of the J phase to Li_2FeS_2 .* However, an addition of LiCl to LiCl-KCl eutectic, which increased the LiCl concentration from 58 to 70 mol %, did eliminate the J phase. The presence of 10 mol % NaCl in the LiCl-KCl eutectic appeared to have no effect on the phases formed during the discharge of FeS_2 electrodes.

The discharge mechanism of the FeS electrode in LiCl-KCl electrolyte was investigated by metallographic techniques. Different discharge paths were observed at the surface and in the interior of the FeS particles. The surface was first discharged from FeS to a mixture of J phase and iron. The interior of the particle was then discharged from FeS to Li_2FeS_2 , and finally to a mixture of Li_2S and iron. After the interior was fully discharged, the reaction resumed at the surface and the J phase was converted to Li_2S and iron.

The ready wettability of separators and particle retainers by the molten-salt electrolyte is a desirable feature in providing good ionic conduction between the electrodes. Accordingly, wettability tests have been continued. Advanced and receding contact angles of drops of molten LiCl-KCl on Y_2O_3 , Type 304 stainless steel, ZrO_2 , graphite, Al_2O_3 , and BN surfaces correlated well with the ease of penetration of fabrics or screens made from these materials. Graphite, Al_2O_3 , and BN fabrics were penetrated with difficulty. The other materials were easily penetrated.

One of the problems encountered with liquid lithium electrodes is the dewetting of the negative electrode substrate after repeated cycling. A possible solution is the use of a low-surface-area wick that transports a liquid lithium alloy from an isolated pool into the electrolyte. In a survey study, various combinations of liquid lithium alloys, molten-salt compositions, and substrate materials were investigated to determine whether preferential wetting of the substrate by the liquid metal could be achieved. The most promising combination found was a lithium alloy containing 1.2 at. % (10 wt %) Cu, LiCl-KCl eutectic containing 4.2 mol % LiF , and substrates of corrugated stainless steel sheets or screens. In a test of these materials, a Li-Cu/FeS cell having a central cylindrical FeS electrode surrounded by an annular Li-Cu wick electrode was constructed and operated through 50 cycles without any evidence of dewetting or short-circuiting.

* There are some indications that the kinetics of the discharge reaction in FeS electrodes are improved by eliminating the J phase.

VIII. ALTERNATIVE CELL SYSTEMS

Work has continued on investigations of alternative high-temperature cell systems. The objective of this effort is to develop a second-generation battery that is fabricated from low-cost, abundant materials and has performance characteristics that approach or equal those of other high-temperature batteries.

Lithium-silicon electrodes were studied in Li/LiCl-KCl/Si and Li/LiF-LiCl-LiBr/Si cells. Measurements of the emfs and capacities of the four Li-Si voltage plateaus showed that the capacities corresponding to the plateaus declined with repeated cell cycling. This decline resulted from a reaction of silicon with the steel electrode housing and porous nickel current collector. The decline in capacity was about 20% after six cycles at 425°C. After the plateau-capacity data were corrected for the loss of silicon, the compositions of the compounds corresponding to the four plateaus were determined; these values were in good agreement with the results of an earlier study at Atomics International.²

A brief study was conducted on a TiS₂ electrode, which discharges by an intercalation mechanism to LiTiS₂. In Li/LiCl-KCl/TiS₂ cell tests, full utilization of the TiS₂ electrode was obtained at high current density (100 mA/cm²) and low volume fraction of electrolyte (0.15) in an electrode 0.4 cm thick. However, the high equivalent weight and cost of TiS₂ are expected to limit its use to special applications.

A variety of calcium-alloy/LiCl-KCl-CaCl₂/FeS cells were fabricated and tested in an effort to find a calcium alloy having good polarization characteristics. The negative electrodes that were tested were the binary intermetallics CaAl₂, Ca₂Si, and CaMg₂, and the ternary compounds of calcium with Mg₂Si. The calcium magnesium silicide electrode had the best polarization characteristics and a satisfactory capacity (1 A-hr of Ca per gram of Mg₂Si). The Ca-(Mg₂Si)/LiCl-KCl-CaCl₂/FeS cell has performance capabilities similar to those of the Li-Al/FeS cell, and, with further development, may be suitable for stationary energy-storage systems. Although most of the cell studies were directed toward development of the calcium-alloy/FeS system, promising results were obtained in preliminary tests of two other systems employing alkaline-earth alloys. Magnesium dendrite formation caused short-circuiting of a Mg/NaCl-KCl-MgCl₂/FeS₂ cell (cell voltage, 1.53 V) after four cycles, but in other respects the cell performance was acceptable. Studies of methods for avoiding the formation of dendrites have been initiated. A test of a Ca-Al/LiCl-KCl-CaCl₂/calcium ferrate cell, which was terminated voluntarily after 60 cycles of operation, demonstrated that the calcium ferrate positive electrode is reversible and has an acceptable capacity (greater than 0.5 A-hr per gram of CaO). Laboratory studies of this new positive electrode are continuing.

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