

LITHIUM/FeS BATTERY DEVELOPMENT PROGRAM

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The objective of this project is to develop lithium-alloy/iron sulfide batteries for electric vehicles and other applications. The major objective for the electric-vehicle battery is to provide long-range, rapid acceleration, safety, and moderate cost for the long-term change to electric vehicles. The effort in 1982 was concerned specifically with the development of Li-Al/FeS cells for use in electric passenger automobile batteries. The battery performance goals for this application are a specific energy of 100 W h/kg at the 3-h rate, a peak specific power of 150 W/kg at 50 percent state of charge, and a lifetime of 500 deep-discharge cycles. Two subcontractors, Eagle-Picher Industries Inc., and Gould Inc., are developing and fabricating full-sized cells for testing and evaluation. Their technical approaches to cell design differ significantly in that the Eagle-Picher cells have boron nitride felt separators and are flooded with electrolyte, while the Gould cells have magnesium oxide powder separators and are operated in an electrolyte-starved condition. The effort at ANL includes cycle-life and performance testing and post-test examinations of the contractors' cells, as well as cell design studies aimed toward increased specific power. The cell design studies include the development of mathematical correlations that can be used to estimate the performance of new cell designs.

The primary effort up to June 1982 was directed to lifetime testing and characterization of Gould and Eagle-Picher status cells, determining the specific power and specific energy of development cells, resistance mapping studies to determine the power limiting components of state-of-the-art cell designs, electrode polarization measurements to determine the limiting electrodes and electrode bed conductivities, cell modeling studies to determine new designs with improved specific power and specific energy to meet the cell performance goals, freeze-thaw testing to determine the effect of temperature changes (from room temperature to 465 °C) on cell performance, failure analysis of the cells, electrode morphology studies, and in-cell corrosion studies. The accomplishments during this period were as follows:

- The power of state-of-the-art Gould and Eagle-Picher cells was improved by using lower resistance electrode connector and cell terminal designs. Resistance mapping studies showed that nearly half the cell resistance was in these components. One-quarter- to 0.5-in-diameter copper cores were used for the positive and negative terminals and 2.25-in-wide nickel electrode connectors replaced approximately 0.4-in-wide low carbon steel connectors used in earlier cells.

- Tests of the latest status cells demonstrated a mean time to failure of 330 cycles and a specific energy of 80 W h/kg at the 4-h discharge rate for the Gould MgO powder separator cells and 345 cycles at 90 W h/kg for the Eagle-Picher BN felt separator cells. A statistical comparison with previous status cell groups showed that progress had been made in improving the specific energy while sustaining cycle life above 300 cycles.
- Testing of a 10-cell module fabricated by Eagle-Picher demonstrated 150 cycles. A good correlation was found between the predicted number of cell failures in the module at a given number of cycles based on a statistical analysis of the status cell cycle life and the measured number of cell failures.
- In the ANL cell development program, a Li-Al/FeS cell that achieved a specific energy of 103 W h/kg at the 3-h discharge rate and a specific power of 136 W/kg at 50 percent state of charge was designed. This cell achieved the highest combination of specific energy and specific power of any Li-Al/FeS cell tested to date. Major differences in this cell and previous cells were the successful development of electrodes capable of operation with a negative-to-positive capacity ratio of 0.8 compared to 1.3 for Eagle-Picher cells and 1.6 for Gould cells and the achievement of good performance with 0.7-cm-thick positive electrodes compared to the 0.2-cm and 0.3-cm thickness for previous cells.
- Modeling programs were developed to assist in identifying cell designs with the potential of meeting the cell performance goals. Preliminary results for 0.3-cm-thick positive electrodes and electrode sizes of 12.7 cm × 12.7 cm, 12.7 cm × 17.8 cm, and 17.8 cm × 17.8 cm show an increase in the specific energy and specific power with increased usable capacity (i.e., increased number of plates) and the optimum specific energy and specific power in the direction of smaller electrode size and larger number of plates for a given usable capacity. These studies have been continued to determine the effect of increased positive electrode thickness on performance.
- Li-Al/FeS bicells with BN felt separators have been thermally cycled from room temperature to 465 °C for up to 68 times over 440 deep discharges with no effect on performance. Post-test examinations of these cells showed that the BN felt separator was not affected by the thermal cycles. Of four cells tested, no cell failures could be attributed to the thermal cycling. The capacity loss rate for these cells was typically 0.02 percent per cycle.
- Ni/Ni₃S₂ reference electrodes were developed for polarization studies in the Li-Al/FeS cells. These studies were conducted to determine voltage losses and voltage-loss processes within the separator and electrode beds. For cells with a negative-to-positive capacity ratio of 1.3, these

studies showed that the positive electrode polarization was several times greater than that for the negative electrode near the end of discharge.

- Post-test analyses of Gould and Eagle-Picher status cells determined the causes of short circuit failures for cells with both BN-felt and MgO-powder separators.
- The electrode morphology studies showed that the Li-Al electrode typically forms a stable, reproducible microstructure for greater than 1000 cycles.
- Iron powder additions to FeS electrodes were shown to reduce the corrosion of iron current collectors.

The plans for 1983 include the following:

- Testing of Eagle-Picher and Gould Li-Al/FeS development cells for performance and cycle life will be completed.
- Post-test examinations of the cells to determine failure mechanisms, microstructural properties of the electrodes, reaction distribution, and in-cell corrosion rates will be completed.
- Post-test examinations will be conducted on sodium/sulfur (beta) cells to identify reactions of the seal materials during cell operation, determine the corrosion products and their behavior, and investigate the characteristics of the phases in the sulfur electrode.

Recent publications

- 1 D. L. Barney and A. A. Chilenskas, The impact of lithium on the cost and availability of lithium-aluminum/iron sulfide batteries, in A. Kozawa *et al.*, (eds.), *Progress in Batteries and Solar Cells 4*, JEC Press Inc., Cleveland, OH, 1982, pp. 271 - 274.
- 2 D. L. Barney, R. K. Steunenberg and A. A. Chilenskas, Lithium/iron sulfide batteries for advanced vehicle propulsion, *Proc. Electric and Hybrid Vehicle Advanced Technology Seminar, Pasadena, CA, December 8 - 9, 1980*, pp. 189 - 216.
- 3 A. A. Chilenskas, D. L. Barney, R. K. Steunenberg and E. C. Gay, Engineering characteristics of secondary lithium/molten salt batteries, *Proc. 30th Power Sources Symposium, Atlantic City, NJ, June 7 - 10, 1982*.
- 4 A. A. Chilenskas, E. C. Gay, K. Kawahara *et al.*, Design considerations for a Li-Al/FeS E.V. battery, *Proc. 17th Intersoc. Energy Conversion Eng. Conf., Los Angeles, CA, August 8 - 13, 1982*.
- 5 M. M. Farahat, A. A. Chilenskas and D. L. Barney, Heat generation in lithium/iron sulfide cells, 158th Electrochem. Soc. Mtg., Hollywood, FL, October 5 - 10, 1980, *Proc. Lithium Batteries Symp.*, 81-4 (1981) 412 - 420.
- 6 M. M. Farahat, A. A. Chilenskas and D. L. Barney, Quick-charge feasibility of Li-Al/FeS batteries, *Proc. 16th Intersoc. Energy Conversion Eng. Conf., August 9 - 14, 1981*, pp. 744 - 746.
- 7 E. C. Gay, W. E. Miller and T. D. Kaun, Electric vehicles — a strategy for reduced utilization of critical fuels in transportation, *Proc. 8th Annual NOBCCHE Mtg., Chicago, IL, April 22 - 25, 1981* (in press).

- 8 E. C. Gay, W. E. Miller, V. M. Kolba and H. Shimotake, Lithium/iron sulfide cell development for electric-vehicle propulsion, *Proc. High Temp. Secondary Batteries Session of the 29th Power Sources Symp.*, Electrochem. Soc., 1981, pp. 173 - 176.
- 9 E. C. Gay, W. E. Miller and F. J. Martino, Use of multiple regression analysis to develop equations for predicting Li-Al/iron sulphide cell performance, *J. Applied Electrochem.*, 11 (1981) 423 - 431.
- 10 F. J. Martino, W. E. Moore and E. C. Gay, Effect of thermal cycling on the performance and lifetime of LiAl/FeS cells, *Electrochem. Soc. Mtg., Montreal, Canada, May 9 - 14, 1982, Extended Abstracts, 82-1* (1982) 559 - 560.
- 11 W. E. Miller, E. C. Gay and D. J. Kilsdonk, Performance of a LiAl/FeS battery and cells, *Proc. 30th Power Sources Symposium, Atlantic City, NJ, June 7 - 10, 1982*.
- 12 L. Redey and D. R. Vissers, Construction of reference electrodes for long term testing of compact Li-Al/FeS cells, *J. Electrochem. Soc.*, accepted for publication, 1982.

ANL Reports

- 1 D. L. Barney, R. K. Steunenberg, A. A. Chilenskas *et al.*, Lithium/iron sulfide batteries for electric-vehicle propulsion and other applications, Progress Report for October 1979 - September 1980, *ANL-80-128*, February 1981.
- 2 D. L. Barney, R. K. Steunenberg, A. A. Chilenskas *et al.*, Lithium/iron sulfide batteries for electric-vehicle propulsion and other applications, Progress Report for October 1980 - September 1981, *ANL-81-65*, February 1982.
- 3 J. E. Battles, F. C. Mrazek and N. C. Otto, Post-test examinations of Li-Al/FeS_x secondary cells, *ANL-80-130*, December 1980.
- 4 V. M. Kolba, J. E. Battles, J. D. Geller and K. Gentry, Failure analysis of Mark IA lithium/iron sulfide battery, *ANL-80-44*, October 1980.

DESIGN, DEVELOPMENT, AND FABRICATION OF THE MARK II LITHIUM/IRON SULFIDE ELECTRIC AUTOMOBILE BATTERY

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The objectives of the project are to develop a cell having the following performance and lifetime characteristics: specific energy, 95 W h/kg; peak specific power, 120 W/kg at 50 percent state of charge; and cycle life, 400 cycles mean time to failure. The cell performance goals are expected to be achieved with a stabilized capacity of 335 A h at the C/3 rate.

Development work was continued on batteries and on multiplate cells with BN felt separators. Two groups of status cells were produced at Eagle-Picher and tested at ANL. The second group of cells raised the state-of-the-art specific energy and power for production-type cells to 90 W h/kg and 80 W/kg, respectively, and demonstrated a mean time to failure of 350 cycles