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ASTROPOWER LABORATORY REPORT SM-48457-Q2

SMALL FUEL CELL TO ELIMINATE PRESSURE CAUSED BY GASSING IN HIGH ENERGY DENSITY BATTERIES

PROGRESS REPORT FOR PERIOD
30 SEPTEMBER 1965 TO 30 DECEMBER 1965
UNDER CONTRACT NAS 5-9594

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SMALL FUEL CELL TO ELIMINATE PRESSURE CAUSED BY GASSING IN HIGH ENERGY DENSITY BATTERIES

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Contract NAS 5-9594

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SUMMARY AND CONCLUSIONS

The object of this program has been the evaluation of miniature fuel cells in preventing the build up of high pressures in sealed silver-zinc batteries. The miniature cells perform the function by electrochemically consuming the gases that are generated within these batteries during operation and stand. In the process the miniature cells also deliver electrical energy which may be used for an auditory warning or telemetering response indicating battery gassing.

Evaluation of the miniature fuel cell in this application has been continued during this quarter by conducting life tests in specially designed battery simulators. The tests are made to provide the necessary design data for specifying the appropriate size and configuration of miniature cells to handle the gassing rates encountered in any size silver-zinc battery under anticipated operating conditions.

Progress and significant results to date are summarized below.

1. Functionality of a miniature cell has been demonstrated for operating times of at least 3800 hours to date.

2. Performance of a miniature cell has been found to be independent of electrode cross-sectional area in the range of 0.25 to 5.0 cm^2.

3. Existing life tests in the "open flow" mode of operation were continued and additional tests in this mode were initiated on cells with modified configurations.

4. New life tests were initiated in the "dead end" mode of operation, wicking product water from the fuel cell.

5. Product water from a miniature fuel cell can be effectively wicked to the battery electrolyte to prevent electrode flooding. Maximum wicking rate or equivalent fuel cell current is between 15 and 50 ma/cm^2 for a cell with electrodes having a cross-sectional area of 1.0 cm^2. Corresponding hydrogen consumption rate is from 6.3 to 21.0 cc H_2/hour for a cell with 1.0 cm^2 cross-sectional area electrodes.

6. A miniature fuel cell with 1.0 cm^2 active electrode area can consume approximately 630 cc of H_2 at a rate of 4 cc/hour and

at least this amount of hydrogen at a rate of 0.4 cc/hour when operated in the "dead end" mode without wicks.

7. A program plan for the third quarter has been outlined and is presented.

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1.0 INTRODUCTION

The phenomena of gassing in silver-zinc cells is well know.[1, 2] Hydrogen, oxygen, or both, may be generated within these cells while in operation (especially during charge) and also while on stand.

Sealed silver-zinc cells are therefore subject to high internal pressures caused by this gassing. An uncontrolled pressure rise ultimately leads to rupture of the cell case and failure of the cell.

Several methods have been proposed for dealing with this problem in the past. These methods range from using catalysts, to promoting union of the gases (H_2 and O_2), absorbents for each of the gases such as pyrogallol for oxygen, to more recent methods based on stabistors, adhydrode and auxiliary electrodes. The adhydrode and auxiliary electrode techniques are based in part on the inherent electrochemical characteristics of high energy density batteries. In practice, these characteristics will vary under different modes of operation, including rate and degree of charge and discharge, and ambient temperature.

The objective of this program is to evaluate the use of miniature fuel cells in solving the problem of gassing and pressure rise in sealed silver-zinc cells. Operation of a miniature cell is not dependent upon electrochemical characteristics of the battery in which it is employed. Since its functions independently, it therefore provides independent reliable control for the battery.

Illustrations of miniature fuel cells and methods in which they may be incorporated in silver-zinc batteries are shown in Figures 1 through 4. Detailed descriptions are given elsewhere.[7, 8] Several arrangements are illustrated, including a jack type, screw type, and deep insert type. Sizes of these cells vary, depending upon the anticipated gassing rates within the battery. Nominal overall dimensions of those shown are 2 cm dia. x 2 cm long. They may be mounted in any convenient place near the top of the battery without interfering with its normal operation.

Work performed during the first quarter consisted of initiation of life tests in the open flow mode of operation and determining the effect of ambient

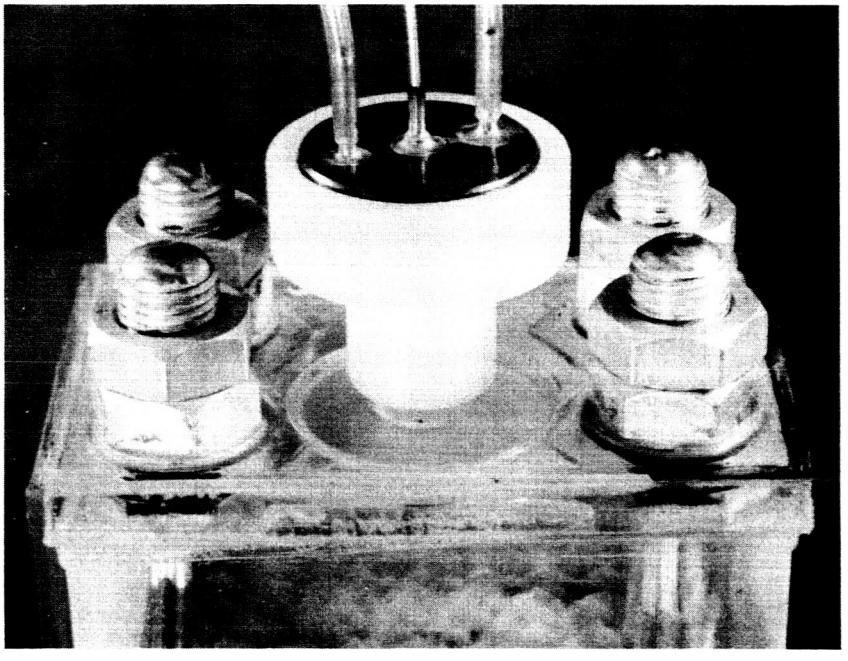

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Figure 1. Jack-Type (Press Fit) Connection of Miniature Fuel Cell to Actual Ag/Zn Battery

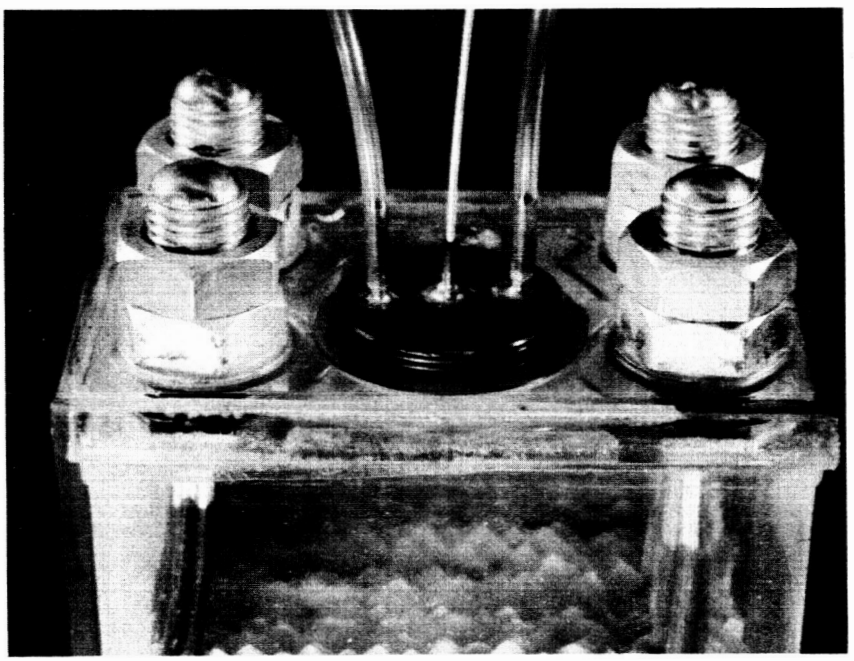

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Figure 2. Screw-Type Connection of Miniature Fuel Cell to Actual Ag/Zn Battery

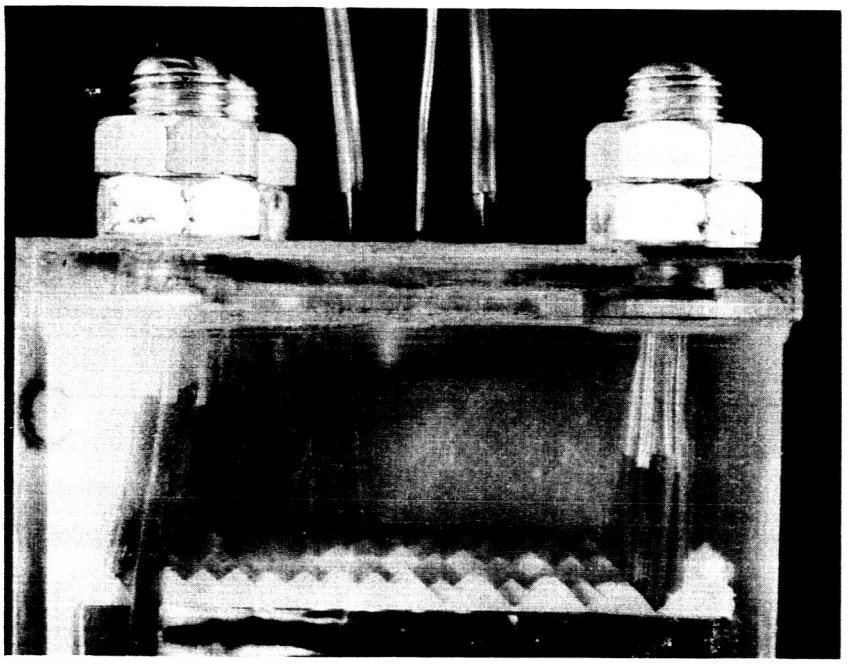

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Figure 3. Deep Insert Type of Miniature Fuel Cell Connection to Actual Ag/Zn Battery

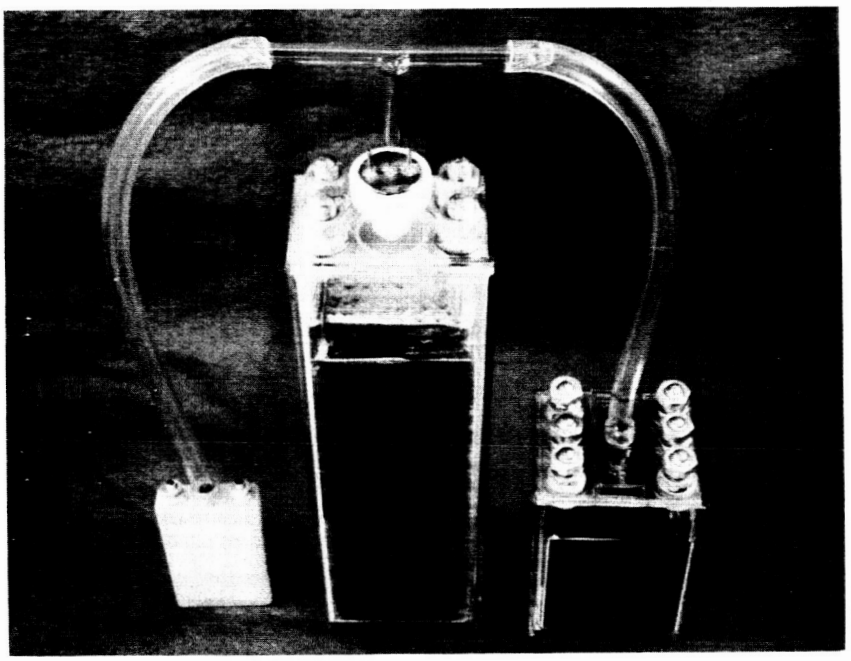

Figure 4. Application of Single Miniature Fuel Cell as Pressure Controller for Three Ag/Zn Batteries by Means of Manifolding Arrangement

temperature and gas composition on the performance of miniature fuel cells. Work performed during the past quarter consisted of continuing the above life tests and initiating several new ones in both the open flow and dead end mode of operation. Significant parameters examined here were electrode size and methods of removing product water from the fuel cell to prevent electrode flooding.

2.0 TECHNICAL DISCUSSION

2.1 Description of Miniature Fuel Cells

Three general methods of incorporating the miniature fuel cells in a silver-zinc battery were devised in a proprietary program[5] and are shown in Figures 1 through 4. The first of these is the jack type arrangement, as shown in Figure 1. In this case, the cell is designed for direct mounting on the battery vent tube. Battery gases must pass through the tube before entering the fuel cell. A screw type arrangement is shown in Figure 2. In this case, the outer frame of the fuel cell is threaded to be screwed into the top of the battery case. Path length through which the gases must travel to reach the fuel cell is somewhat shortened in this case. A deep insert type is shown in Figure 3. In this case, the fuel cell is mounted deep inside the battery case and exposed directly to the gases evolved from the electrodes.

A manifolding arrangement may also be employed as shown in Figure 4. In this case, gases from several batteries are transmitted through tubing to one miniature cell.

In all of the cases described above, the cathode side of the fuel cell is supplied with pure oxygen from an appropriately small oxygen cylinder or tank.

Key components of all miniature fuel cells used in this program have been the same for all tests. Potassium hydroxide solution has been employed as electrolyte. This solution is contained (immobilized) in an asbestos matrix. Platinum black has been employed as an electrocatalyst with both hydrogen and oxygen electrodes.

Two basic cell types have been employed. A description of each of these is given below.

2.1.1 Initial Cell Type

Components of the initial miniature fuel cell are shown in Figure 5.

Both hydrogen and oxygen electrodes are identical and consist of platinum black bonded under a pressure to a platinum plated silver

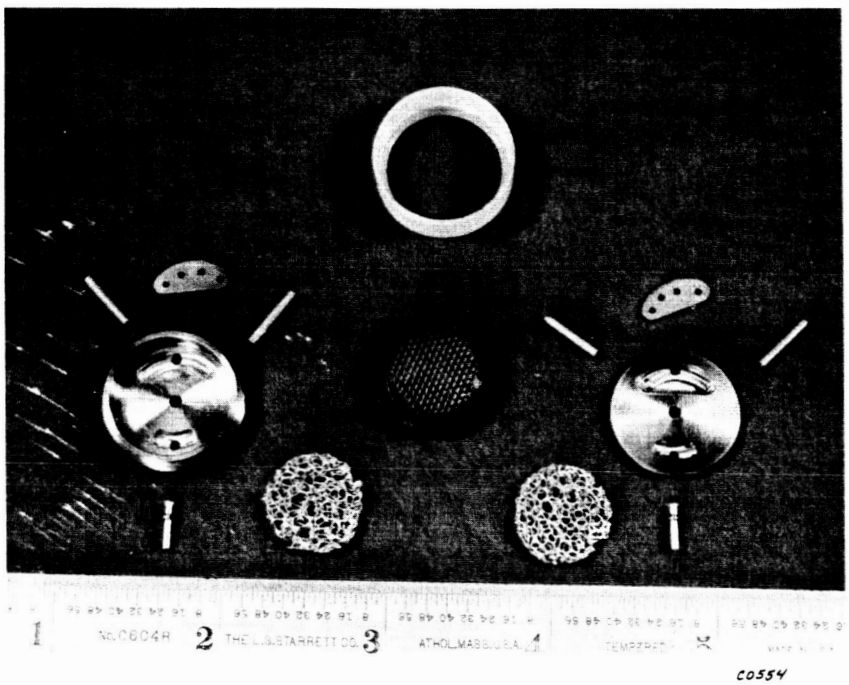

Figure 5. Initial Miniature Fuel Cell

screen. A few strands of each screen pass through the cell frame and serve as external electrical leads. The electrodes in turn are bonded under pressure to a disc of sheet asbestos to form a rigid electrode-electrolyte assembly. A polyurethane rim is mounted on the outer periphery of the membrane by means of the jig designed for this purpose. The purpose of this rim is to provide gas seals, both internally between compartments and externally to the atmosphere. The electrode-electrolyte assembly, along with the gas distribution plates, is mounted in the cell housing to form the complete miniature fuel cell as shown in Figure 4. The housing is made of nylon, and the distribution plates are made of stainless steel. One of the gas distribution plates contains inlet and outlet tubes for transmitting a gas (either hydrogen or oxygen) from an external supply tank. The other distribution plate has a machined center hole for transmitting gases from either a silver-zinc battery or one of the battery simulators to the electrode. The outside of this end plate is threaded for direct mounting onto either the battery or simulator.

Several sizes of miniature cells have been built with overall dimensions ranging from 5mm dia. x 5 mm long to 20 mm dia. x 10 mm long. Corresponding electrode diameters have ranged from 3mm to 15 mm.

2.1.2 Modified Cell Configurations

Electrodes for this cell (Figure 6) were obtained from the American Cyanamid Company. As in the cell described above, both hydrogen and oxygen electrodes are identical and contain platinum black as the electrocatalyst; however, they do differ in some details. In this case, the collector screen is made of nickel or tantalum and a Teflon waterproofing agent has been admixed with the platinum black. Potassium hydroxide impregnated asbestos is again employed as electrolyte.

Electrodes are spot welded to the gas distribution plates to provide good electrical contact. External electrical leads are then taken directly from external surfaces of the end plates. Both plates are very similar to those described above; one contains the inlet and outlet tubes for transmitting battery gases or simulator gases to the electrodes. The outside of the latter plate is again threaded for mounting in a battery or simulator. The construction material is stainless steel, as above.

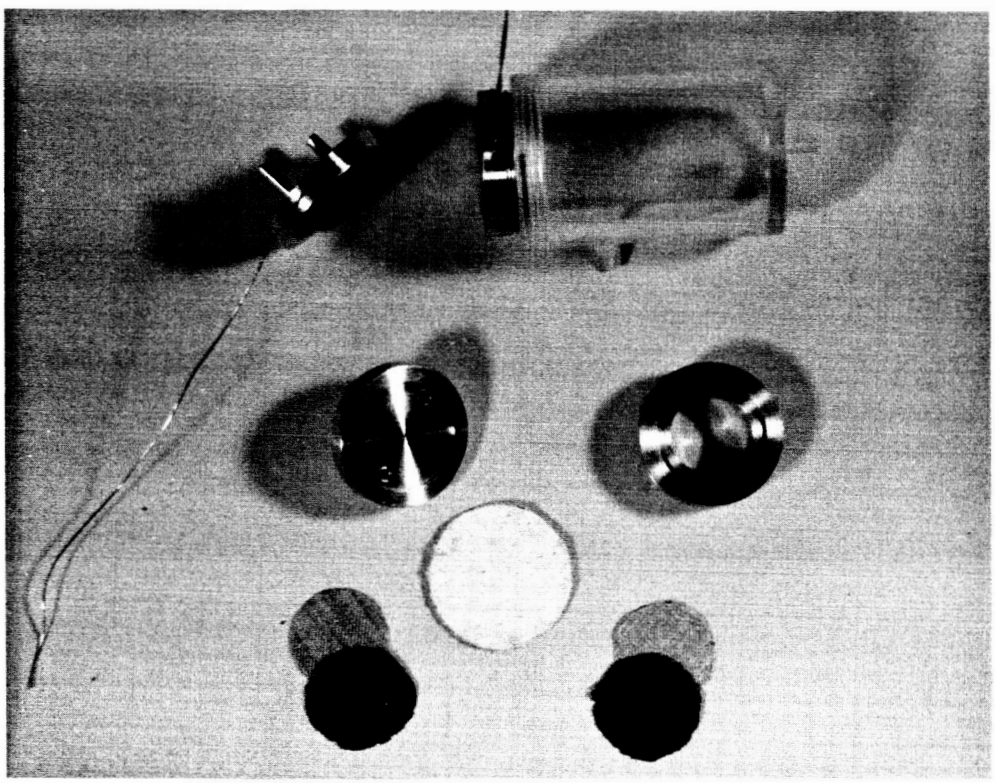

Figure 6. Modified Miniature Fuel Cell

Somewhat different types of gas seals are employed in this cell. Internal seal between compartments is provided by compression of the outer periphery of the asbestos layer between the surfaces of the gas distribution plates. External seal is provided by a bead of epoxy resin around the circumference where the cell halves meet.

Several sizes of these cells have also been built with the same range in overall dimensions as the initial cell (Section 2.1.1).

2.2 Life Tests

Existing life tests were continued in the open end mode of operation described in the first quarterly report.[4] Additional tests were initiated in this mode of operation on cells that contained both larger and smaller electrode diameters and on one that contained an ion exchange membrane in place of the previously used KOH-asbestos electrolyte system. Other tests were also initiated in the dead end mode of operation on cells employing wicks for transferring product water from the fuel cell to the battery electrolyte. The results of these tests are discussed below.

2.2.1 Open End Flow Without Wicks

The primary objective of these tests is to establish both electrochemical and mechanical functionality of miniature fuel cells for extended operating times to 5000 hours. The tests are carried out in previously described plastic battery simulators with slow flow of both hydrogen and oxygen to the cell (Figure 7). Excess gases that are not consumed in passing across the cell are vented to the atmosphere. The cells are run continuously across a fixed load resistance. Operating voltage as well as a complete polarization curve is determined on a daily basis.

Results of tests performed during this report period are given in Table I. Some but not all of these tests remain in progress. Test 1 described in the First Quarterly Report,[4] has continued to function satisfactorily throughout this report period for a total operating time of 3821 hours, to date. Maximum current output (or corresponding gas consumption capability) of this cell is quite limited in comparison with that of the other cells. The reason for this, as previously explained, is that the electrodes employed in

TABLE I

LIFE TESTS OF MINIATURE FUEL CELL IN OPEN FLOW MODE OF OPERATION

Test No.	Cell Type	Electrode Type	Electrode Area, (cm^2)	Load, Ohms	Time, Hrs.	Average Voltage, (Volts)	Average Current, (m.a.)
1	Initial	In-House	1.0	1000	3821	.70	.70
2	Modified	A.A.-1	1.0	1000	794	–	–
3	"	A.A.-1	1.0	1000	587	–	–
4	"	A.A.-1	1.0	1000	982	–	–
5	"	A.A.-1	1.0	1000	834	–	–
6	"	A.B.-6	1.0	1000	1869	.90	.90
9	"	A.B.-40	1.0	1000	1695	.95	.95
14	"	A.B.-40	1.0	100	1368	.85	.85
18	"	A.B.-6	1.0	100	1104	.80	.80
20	"	A.B.-40	6.25	4000	744	.95	.95
21	"	A.B.-40	5.0	200	744	.95	.95
24	Organic Membrane	A.B.-40	1.0	1000	456	–	–

(Continued)

TABLE I (Cont'd)

LIFE TESTS OF MINIATURE FUEL CELL
IN OPEN FLOW MODE OF OPERATION

Test No.	Notes
1	In progress, longest operating time to date, maximum current is limited.
2	Terminated, due to degradation of A. A. -1 electrodes, voltage dropped gradually from 0.9 to 0 volts.
3	Terminated, due to degradation of A. A. -1 electrodes, voltage dropped gradually from 0.9 to 0 volts.
4	Terminated, due to degradation of A. A. -1 electrodes, voltage dropped gradually from 0.9 to 0 volts.
5	Terminated, due to degradation of A. A. -1 electrodes, voltage dropped gradually from 0.9 to 0 volts.
6	In progress, longest time with A. B. -6 electrodes, stable to date.
9	In progress, longest time with A. B. -40 electrodes, stable to date.
14	In progress, lower voltage due to higher current and electrode flooding.
18	In progress, lower voltage due to higher current and electrode flooding.
20	In progress, stable to date, establishes functionality of smaller size.
21	In progress, stable to date, establishes functionality of larger size.
24	Terminated, due to deterioration of ion exchange membrane.

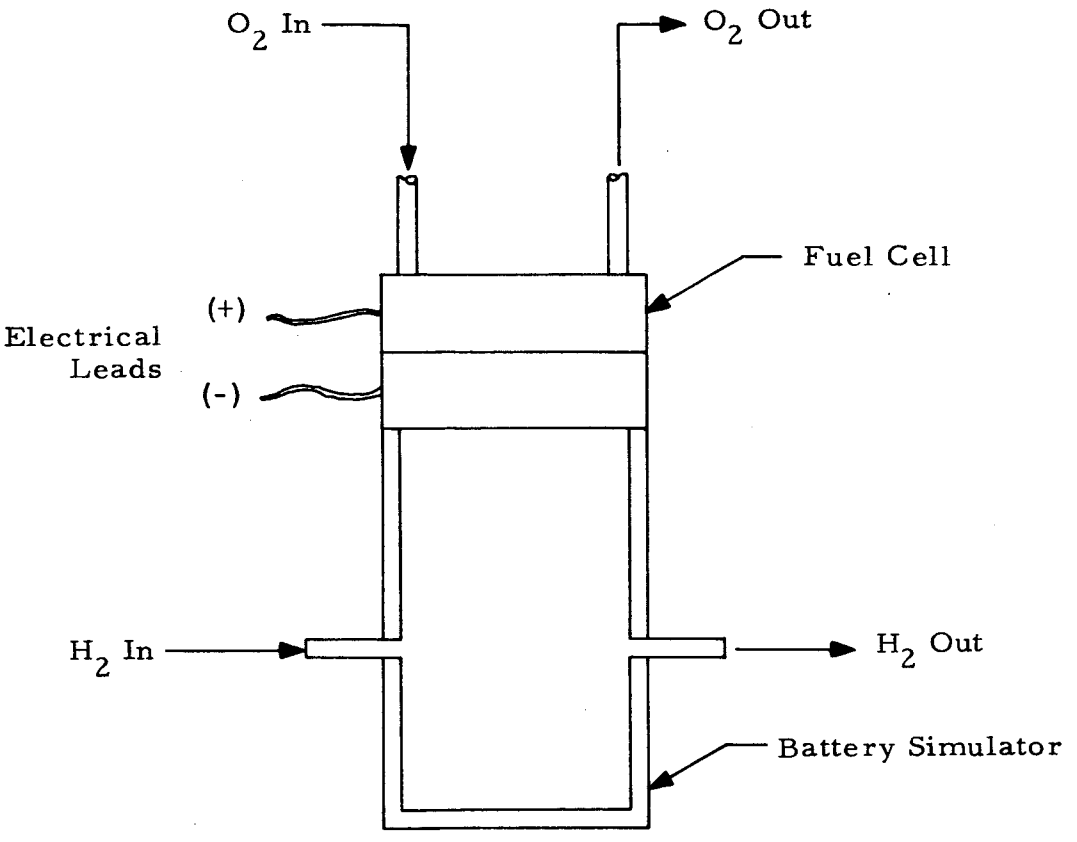

Figure 7. Schematic Diagram of Open Flow Tests

this cell are not as active as the American Cyanamid type employed in all other cells. Disregarding the limitation however, the cell has been found to function very well as evidenced by its stable voltage for an extended operating time.

Tests 2 through 5 inclusive, exhibited slow degradation in performance at various time intervals from 500 to 800 hours. The reason for this was attributed to chemical attack of the A. A-1 type electrodes. These contained a tantalum collector screen which is unstable in a strong base. Its use was discontinued thereafter in this application.

Tests 6 and 9 contain American Cyanamid electrodes Types AB-6 and AB-40, respectively, and are both run across a 1000 ohm load. Both have been found to exhibit stable performance throughout the test period with operating voltage of Test 9 being slightly higher (approximately 0. 1 volt).

Tests 14 and 18 also contain American Cyanamid electrodes Type AB-6 and AB-40 respectively and are both run across 100 ohm loads. Both have been found to exhibit stable but somewhat lower operating voltages than in Tests 6 and 9 above. The reason for this is attributed to a higher internal voltage drop (due to higher current) and also to possible electrode flooding (due to faster rate of water formation).

The significant parameter for Tests 21 and 22 is electrode diameter. Active electrode area in Test 21 is 0. 25 cm^2 or 1/4 of that employed in the above tests; active area in Test 22 is 5. 0 cm^2 or five times that employed in the above tests. External loads were selected to give equal current densities, as in the above tests, that is, 4000 ohms for Test 21 and 200 ohms for Test 22. Electrodes in both tests are American Cyanamid Type AB-40. Operating voltages of both cells have been found to be stable and nearly identical at the same current density. It can be concluded that performance characteristics of miniature cells are independent of electrode size within the range of 0. 25 cm^2 to 5. 0 cm^2.

An anion exchange membrane (Ionics Type III EZL) was employed as electrolyte in Test 24 in place of the KOH-asbestos electrolyte

used on all previous tests. This material is composed of copolymers of vinyl compounds containing quarternary ammonium groups and tertiary amine groups. Electrodes were the American Cyanamid type AB-40. Maximum current output was found to be rather limited, as in Test 1 above. The limitation in this case was attributed to the higher internal resistance of the membrane as compared with that of the KOH-asbestos system. Stable performance was obtained for over 400 hours when discharged across a 1000-ohm load. At this point performance dropped rapidly and the test was terminated. A possible explanation for the failure may be chemical oxidation of the membrane material. These results indicate that this particular membrane material is unsuitable for long term operation in a miniature fuel cell.

2.2.2 Dead End Flow Without Wicks

In Tests 7 and 8 the gases were supplied to the cell in the dead end mode (without venting excess gases) and without wicks, as in the following tests. Gases were stored in small containers and connected to each end of the cell with a short segment of tubing. For safety reasons, the size and pressure of the gas containers was kept to a minimum and consisted of 250 ml Ehrlenmeyer flasks at atmospheric pressure or below it. The flasks were refilled periodically to maintain the pressure level between atmospheric and approximately two inches of mercurcy vacuum. Electrodes were of the American Cyanamid Type AB-6 with an active area of 1.0 cm^2.

Stable performance has been observed on Test 7 for over 1500 hours when discharged across a 1000-ohm load with a corresponding current of approximately 1.0 ma. Hydrogen consumption rate in this case is approximately 0.4 c.c./hour. The test is being continued.

Test 8, which was discharged across a 100 ohm load with a corresponding current of approximately 10 ma, was terminated after 150 hours when performance began to decline rapidly. Hydrogen consumption rate in this case is approximately 4.0 cc/hour. The reason for the decline was attributed to electrode flooding as evidenced by an extremely wet condition upon disassembly.

TABLE II

LIFE TESTS OF MINIATURE FUEL CELLS IN DEAD END MODE OF OPERATION AND WITHOUT WICKS

Test Cell Number	Cell Type	Electrode Type	Electrode Area (cm^2)	Load (ohms)	Time (hrs)	Notes
7	Modified	A. B. -6	1. 0	1000	1500	In progress, voltage above 0. 8 v for majority of run
8	Modified	A. B. -6	1. 0	100	150	Terminated due to flooding, voltage above 0. 7 v for majority of run

These results signify that a miniature fuel cell without wicks and with 1.0 cm^2 area can consume approximately 630 cc of hydrogen at a rate of 0.4 cc/hour and at least this amount, if not more, at a rate of 0.4 cc/hour of hydrogen.

2.2.3 Dead End Flow With Wicks

The purpose of these tests is to evaluate the use of wicks in transferring product water from the fuel cell to the battery electrolyte. Such water transfer is a necessity for long term operation of the fuel cell where electrode flooding could limit performance.

The tests have been carried out in a simulated battery environment consisting simply of a small Ehrlenmeyer flask filled with hydogen containing a small amount of potassium hydroxide solution (see Figure 8). The fuel cell is mounted in a rubber stopper and fitted into the mouth of the flask. One end of the wick is in contact with the fuel cell electrolyte through a small hole cut in the middle of the hydrogen electrode and the other end is suspended in the simulated battery electrolyte, 35% KOH. Wick material is from the American Felt Company, Number 61NY185, and was cut in the form of a thin strip with dimension of approximately 1/16 inch x 1/8 inch x 6 inches. Electrodes are American Cyanamic Type AB-40 with active area of 1.0 cm^2.

Operation has been carried out in a similar manner to that described in Section 2.2.2 but with some modifications. In this case the flask is filled with a small positive pressure of hydrogen from an externally located cylinder. Pressure is then finely adjusted so that there was essentially no flow through the bottle as evidenced by bubbling from a vent line immersed in water. Oxygen pressure and flow were set in the same manner to achieve this dead end mode of operation. The cells have been run at constant current with daily adjustment of load on a decade resistance box.

Results to date indicate that these wicks function effectively in removing product water within given ranges of fuel cell current density. Operating voltage of the cell in Test 10 has remained constant within the narrow range of 0.89 to 0.91 volts at 5 ma (5 ma/cm^2) for over 1500 hours.

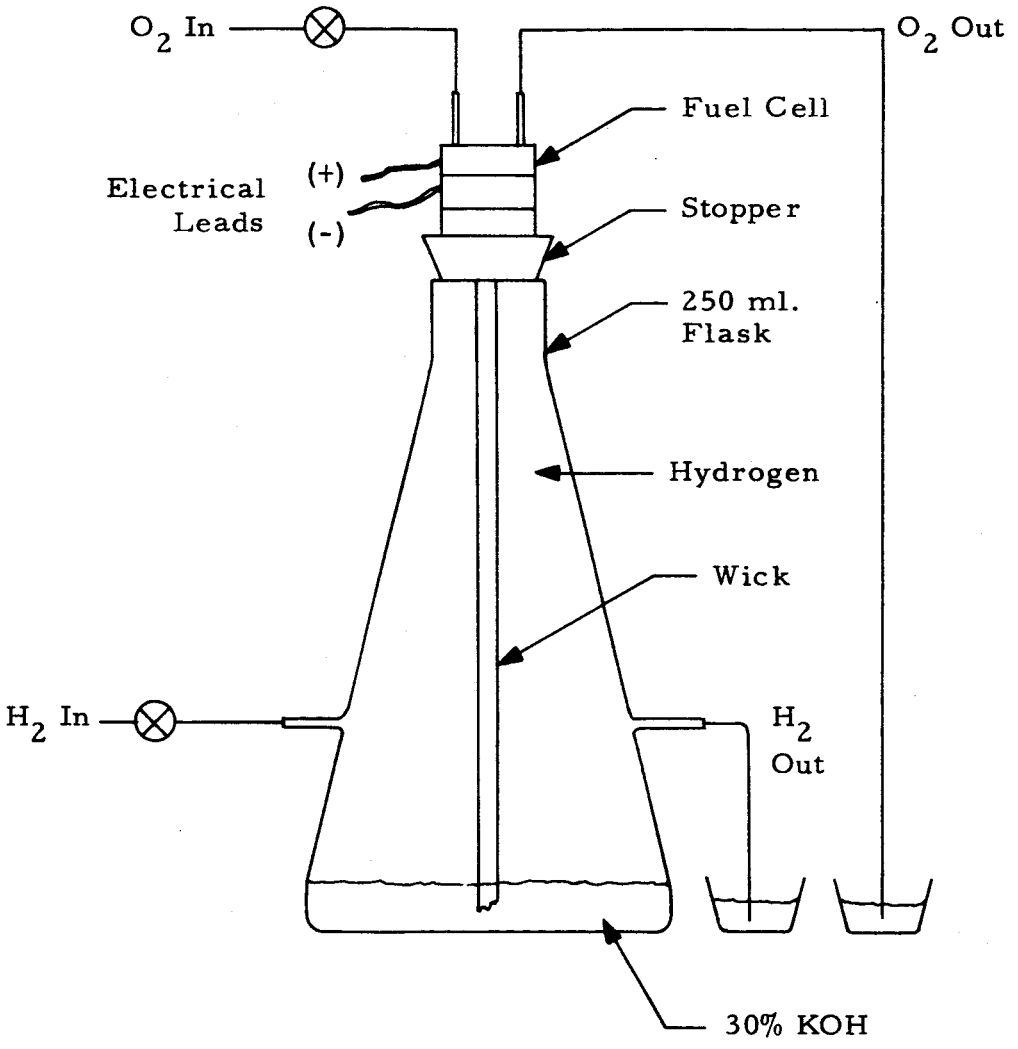

Figure 8. Schematic Diagram of Dead End Flow Tests with Wicks

TABLE III

LIFE TESTS OF MINIATURE FUEL CELLS IN DEAD END MODE OF OPERATION WITH WICKS

Test No.	Cell Type	Electrode Type	Electrode Area (cm^2)	Current Density (ma/cm^2)	Average Voltage	Time (hrs)	Notes
10	Modified	A. B. -40	1.0	5	.91	1540	In progress, stable to date
13	Modified	A. B. -40	1.0	50	–	72	Terminated, unstable voltage due to high current
17	Modified	A. B. -40	1.0	15	.80	1080	In progress, stable to date
22	Modified	A. B. -40	5.0	5	.93	672	In progress, stable to date
23	Modified	A. B. -40	0.25	5	.90	672	In progress, stable to date

At a much higher current of 50 ma (50 ma/cm^2) voltage declined gradually to less than 0.1 volt after 72 hours (see Test 13). In Test 17 current was set at 15 ma and voltage has remained relatively constant near 0.8 volts for over 1000 hours. Based on these results, it may be stated that the use of wicks permits stable operation without flooding at a current density of at least 15 ma/cm^2 but less than 50 ma/cm^2 for a cell with 1.0 cm^2 active electrode area. This corresponds to a hydrogen consumption rate of 6.3 to 21.0 c.c./hour. This would be sufficient to handle the gassing rates that would be encountered in a 10-15 AH silver-zinc battery under most operating conditions.

Additional tests with wicks and with both larger (5.0 cm^2 in Test 22) and smaller (0.25 cm^2 in Test 23) have been set up and are now in progress. Both have been run at a current density of 5 ma/cm^2 and each has exhibited stable voltage near 0.85 volts for over 600 hours. These results indicate that wicks may also be employed to give stable operation of miniature fuel cells with active electrode areas in the range of 0.25 to 5.0 cm^2 at currents of at least 5 ma/cm^2.

A final point regarding the dead end flow tests is its comparison with the open end flow tests in Section 2.2.1. Here it should be pointed out that current density at a given voltage is somewhat higher for the open end than for the dead end mode of operation. This may be explained on the basis of mass transfer limitations. In the dead end mode the gases are not transferred to their respective electrodes at a fast enough rate so that there is a voltage drop due to concentration polarization. No such limitation exists in the open end flow tests where an excess of gases are passed across the electrode surfaces.

2.2.4 Experimental Gas Consumption Rates

Until this report period, all estimates of gas consumption capability of the miniature fuel cell had been based only on cell current; it was assumed that coulomic efficiency was 100%. In order to establish this point, another life Test 16 was set up and run in a similar manner to that described in Test 7 (Section 2.2.2), with the addition of a mercury manometer connected to the flask. Pressure and current readings are recorded daily to check gas consumption. Typical results are given below.

Electrode Area	=	1.0 cm^2
Time of run	=	16.5 hours = 990 minutes
Initial current	=	4.9 ma
Final current	=	4.7 ma
Average current	=	4.8 ma
Current-time output	=	4.75 amp-min
Initial pressure	=	+0.2 cm Hg
Final pressure	=	-11.3 cm Hg (vacuum)
Change in pressure	=	11.5 cm Hg
Volume of flask	=	248 cm^3

On the basis of the pressure change the approximate amount of hydrogen consumed is 11.5 cm Hg x $\frac{248 \text{ cc } H_2}{76.0 \text{ cm Hg}}$ or 37.5 cc H_2.

On the basis of the current-time output, the amount of hydrogen consumed is 4.75 amp-min x $\frac{7.0 \text{ cc } H_2}{1.0 \text{ amp-min}}$ or 33.3 cc H_2.

These results signify that electrochemical reaction can account for only 89% of the total amount of hydrogen lost. The balance of the hydrogen may possibly have been lost by leakage or perhaps by chemical reaction with residual air (oxygen) which might have been present in the flask. This type of testing will be continued with improved gas seals and longer flushing periods to insure that initial content of the flask is pure hydrogen. At the present time the cell has been run in this mode for 25 days.

2.2.5 Operation with H_2-O_2 Mixtures

During the first report period a series of short term tests was carried out with mixtures of H_2 and O_2 in both the anode and cathode under open end flow conditions. This simulates certain operational modes of silver-zinc cells. Functionality of the miniature cells was demonstrated for operation under the above conditions on mixtures containing up to 50% oxygen in the anode and up to 10% hydrogen in the cathode.

Mixtures studies were continued during this quarter and apparatus and techniques for conducting long term tests in the dead end mode were set up. Actual tests have not as yet been initiated. The system and

mode of operation is similar to that used in other dead end tests (Section 2.2.2). In this case the flasks are filled each day with a mixture of H_2 and O_2 rather than pure H_2. Composition is checked after each filling by gas chromatography and necessary additions are made to bring the composition to the desired value. Intervals of time between fillings are adjusted to keep the concentration within a relatively narrow range. For example, a cell would be filled every day or two when it was run at 5 ma and consuming hydrogen in a 250 ml flask that contained an initial composition of 90.0% and 10.0% O_2. After one day the cell would have consumed approximately 7 cc of H_2 and the composition would have changed to 89.7% H_2 and 10.3% O_2 or a net change of only 0.3% H_2.

3.0 PROGRAM FOR THIRD QUARTER

1. All existing life tests will be continued throughout the next quarter or until failures occur.
2. Additional lifes tests will be initiated for operation on mixtures of hydrogen and oxygen in the fuel cell anode.
3. Tests will be initiated to measure pressure reduction capability of miniature fuel cells in representative commercial silver-zinc cells of 5 to 15 AH capacity.

4.0 PROJECT PERSONNEL

The following Astropower staff members are associated with this program at this time.

Dr. C. Berger, Principal Investigator

Dr. M. P. Strier

Mr. H. Frank

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