

## REGENERATIVE FUEL CELLS FOR SPACE APPLICATIONS

A. J. APPLEBY

*Center for Electrochemical Systems and Hydrogen Research, 310 ERC,  
Texas A&M University, College Station, TX 77843 (U.S.A.)*

### Summary

After several years of development of the regenerative fuel cell (RFC) as the electrochemical storage system to be carried by the future space station, the official stance has now been adopted that nickel-hydrogen batteries would be a better system choice. This paper compares RFCs with nickel-hydrogen and other battery systems for space platform applications.

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### Introduction

Originally the RFC was conceived as a suitable system for unmanned platforms in low-earth orbit (LEO) and in geosynchronous orbit (GEO), for the permanently manned LEO space station, and for orbital transfer vehicles between LEO and GEO. In addition, it might also be used for various military surveillance, command, and weapons applications connected with space platforms of various types. For these applications, primary power would be provided either by deployable photovoltaic (PV) arrays, solar thermal (dynamic solar) systems, or from a small nuclear reactor such as the proposed General Electric SP100 unit, which was originally to be rated at 100 kW, but which may grow (if developed) to 300 kW. At the present time, the potential for safe launching of nuclear units cannot be established, so that PV and solar thermal primary power sources must be looked upon as being the main contenders for these applications. The solar systems would require on-board storage for eclipse periods, and all would need storage for peak load.

The main technical objectives of the on-board storage power units would be light weight (and compactness) for the given mission, very long cycle life, and in-orbit maintainability. It was considered that initial electrochemical storage units would be in the 50 - 100 kW peak load class or less, and that modularity would be used to build up units of any desired future size.

### RFC concepts

LEO storage was examined from 1979 by NASA's Johnson and Lewis Research Centers. The RFC was at this time recognized as having the capa-

bility of meeting a 40 000 hour life requirement. This system, it was considered, could be ready for deployment by the end of 1986 [1]. It was to consist of fuel cells to supply peaking and eclipse power, with PV cells to supply power during sunlight periods, when electrolyzer cells would also be operated whose product, hydrogen and oxygen, would be stored for use in the fuel cell. The system was to consist of separate fuel cell and electrolyzer packages because these were already available, and this would also avoid the design of special electrodes that could operate in both charge and discharge modes. As we will see below, this is difficult for many systems. Finally, such a system using proven components avoids the problems of man-rating of new concepts and allows easy optimization of electrode areas and numbers of active cells in each mode.

Although the overall efficiency of the  $H_2/O_2$  fuel cell electrolyzer combination is only 50 - 60%, its waste heat can be thermally integrated at 70 - 95 °C to provide a useful source of spacecraft heating during both charge and discharge modes. On this basis, it is superior to competing battery systems operating close to 25 °C. Its disadvantage is that it would require a larger PV panel area than batteries, but the latter would probably require additional solar-thermal collector area for spacecraft life-support heating.

A further advantage of the RFC, compared with conventional batteries with a proven space ability in the late 1970s, was system lifetime for a given weight. As is well known, conventional secondary battery lifetime rapidly diminishes with increasing depth of discharge (DOD), since the active materials must undergo phase and/or crystalline alterations associated with volume changes and some degree of irreversibility. These lead to degradation in electrode structure and in active material utilization, often accompanied by destructive corrosion of the positive due to successive formation and breakdown of oxide films on each charge-discharge cycle.

Thus, in the 1970s, only one proven candidate secondary battery for space applications requiring many years of life was available — namely, nickel-cadmium. Most applications were for GEO communications satellites, for which there are two 45-day eclipse periods per year and one eclipse per eclipse day in a typically-inclined orbit. In a 24 h orbit the eclipse time might be about 1.2 h. Thus, 900 cycles would be required over a 10 year lifetime. These cycles would vary from medium discharge (60% DOD) to shallow discharge, with discharge at the  $C/2$  rate for 1.2 h. Accordingly, such cells are usually tested at 1.8 h charge,  $C/2$  (1.2 h) discharge at 60% DOD, to determine lifetime performance capability.

For typical aerospace Ni-Cd cells this exceeds 2000 cycles under these conditions. Hence, for GEO use, Ni-Cd cells were adequate. However, under aerospace conditions, when mounted in a battery, they are little better than 20 W h  $kg^{-1}$  at 100% DOD at the  $C/2$  rate.

For LEO applications, sunlight is typically 0.9 h in an equatorial orbit with eclipse for 0.6 h, with one cycle per orbit or 5840 cycles per year.

The calculated system weights for a 100 kW power storage plant with a 10 year orbit life have been quoted as: RFCs: 6000 kg; Ni-Cd batteries (50% DOD): 7000 kg; Ni-Cd batteries (25% DOD): 16 000 kg [2], where the orbit lifetime of Ni-Cd cannot be relied upon.

As a consequence of the above, a "proof of technology readiness" based on small General Electric SPE fuel cell-electrolyzer components [3] (developed since early 1984 by UTC's Hamilton Standard Division), and on UTC lightweight alkaline fuel cells (AFCs) combined with Life Systems, Inc. alkaline electrolyzers, was selected by NASA for demonstrations. Earlier programs supported by the U.S. Navy on submarine life-support systems using Nafion SPE membrane cells [3] demonstrated 35 000 h of life at 82 °C and indicated a potential electrolysis SPE membrane life in excess of 100 000 h. Similarly, the Life Systems alkaline electrolyzer also had a proven lifetime.

The SPE demonstrator consisted of an 8-cell stack of 1000 cm<sup>2</sup> fuel cells combined with a 22-cell stack 210 cm<sup>2</sup>-cell electrolyzer. The fuel cell operated at 112 A, 6.5 V, 71 °C. Cell voltages to sustain electrolyzer current densities of 200 and 800 mA cm<sup>-2</sup> have been reported as 1.59 and 1.78 V [4]. Hydrogen storage was at 880 kPa (8.8 atm) and oxygen at 780 kPa (7.8 atm). This unit was delivered to Johnson Space Center in February 1983, and it completed 3500 h before being terminated. No measurable degradation was observed. The alkaline system fuel cell consisted of the lightweight version [5, 6] of the space shuttle orbiter fuel cell [5] with later stack improvements. Stack lifetimes for the space shuttle system for over 10 000 h have been achieved [5]. Goals were determined to be 40 000 h life at 110 mA cm<sup>-2</sup> and 3000 h at 0.9 V, 1.1 A cm<sup>-2</sup> for pulsed power application [7].

The Life Systems electrolyzer technology used cells of about 90 cm<sup>2</sup> area, and has been tested over 13 000 h. The alkaline breadboard was delivered to Johnson Space Center in January 1984 and contained a 30-cell electrolysis unit rated at 1.5 kW. Voltages of 1.52 and 1.82 V were attained at 200 and 800 mA cm<sup>-2</sup>, respectively [4]. In April 1984, it was replaced by a 6 cell, 930 cm<sup>2</sup> area unit rated at 3 kW to provide a better match with the UTC fuel cell. The system was tested over many LEO cycles, the fuel cell showing less than 1 mV/1000 h voltage degradation.

In May 1986, the program changed, since NASA decided not to consider RFCs for the future space station, presumably as an economy measure. The projected power requirements of the manned space station started to change with its proposed size, and it was to have 25 kW of installed, deployable PV together with 50 kW of solar thermal system. The proposed polar orbital platform was to use nickel-hydrogen batteries as storage units for its PV array. It was felt that the same system could be used for both. Since that time, the space station PV requirement has risen, first to 37.5 kW of PV, and perhaps to 50 kW as specifications change.

## Comparison between Ni-H<sub>2</sub> and the RFC

The Ni-H<sub>2</sub> battery grew out of a proposed true RFC (*i.e.*, with one-piece fuel cell and electrolysis cell elements) concept for GEO use in the late 1960s [7]. This consisted of an AFC with high-loading noble metal electrodes in a lightweight pressure vessel internally divided in such a way that hydrogen and oxygen could be stored separately. The system was rapidly abandoned and replaced by various metal-gas battery concepts, such as zinc (or cadmium)-oxygen [8]. Metal-oxygen systems, particularly zinc-oxygen, are, in principle, attractive, since they have lightweight, high energy negatives (zinc has a *practical* energy density of 0.6 W h kg<sup>-1</sup> when combined with an oxygen electrode, whereas practical nickel positives combined with hydrogen negatives have 0.135 W h kg<sup>-1</sup> [9]). In addition, oxygen storage requires half the volume of hydrogen storage for the same number of A h, hence, a lighter container. As a result of the above, zinc negative-oxygen positive systems have a 50% higher energy density (on paper) than nickel positive-hydrogen negative systems. However, while their coulombic efficiency is good, they have poor voltage efficiency due to the irreversible characteristics of the oxygen electrode, even on Pt group metals or Au under pressure (overall efficiency 60%). Finally, the zinc electrode has a poor cycle life capability, and it can react explosively with compressed oxygen if the electrolyte dries out, for example, on overcharge at high rates. In consequence, the favored couple is Ni-H<sub>2</sub>, which operates at a coulombic efficiency of 95% and a voltage efficiency of about 85% (80% overall) under practical LEO conditions. Its average voltage at the C/2 discharge rate (based on 100% DOD) is 1.2 V to 60% DOD (cut-off at 1.0 V).

It is instructive to compare the specific energies of the Ni-H<sub>2</sub> battery and the true RFC on the single-cell level for a C/2 mission requirement (*i.e.*, all gas stored during charge to 100% degree-of-charge is consumed to a nominal baseline pressure representing container backfill).

The best breakdown to use for this purpose is that described for Ni-H<sub>2</sub> in ref. 9. While these cells, in the form of a 14-cell battery of nominal 35 A h capacity, were not necessarily as optimal as might be desired, they will serve, since the comparison with the H<sub>2</sub>-O<sub>2</sub> RFC is relative. The system used in an experiment in the Navigation Technology Satellite 2 (NTS-2) vehicle in 1976 was a development of that described in refs. 10 - 13. Each single unit consisted of 15 monopolar 38.5 A h (to 1.0 V at 23 °C) bicells each containing two Eagle-Picher electrochemically impregnated aerospace positives in contact (from the inside outwards) with reconstituted asbestos separators, Teflon-bonded platinum black hydrogen diffusion electrodes, and plastic gas diffusion screens to allow for gas passage to and from the hydrogen electrodes. The whole system was stacked on a center rod and held in a lightweight, Inconel 718 pressure vessel with appropriate mountings, busbars, and feedthroughs. Hydrogen pressure varied from 4.1 MPa (41 atm) fully charged to 700 kPa (7 atm) discharged to 1.0 V. The pressure vessel was somewhat over-designed, with a safety factor of 4. Each cell had a total

TABLE 1  
Cell weight breakdown

Component	Weight (g)	Percent. of total
Nickel electrodes	348.6	33.9
Hydrogen electrodes	72.0	7.0
Separators	35.1	3.4
Gas screens	8.0	0.8
Electrolyte	159.0	15.5
Electrode stack	622.7	60.6
Center rod and insulator	14.3	1.4
Busbars and tabs	41.4	4.0
Endplates	34.0	3.3
Nut and washers	4.6	0.4
Terminal conductors	79.0	7.7
Internal hardware	173.3	16.8
Pressure shell	179.0	17.4
Weld and support ring	35.0	3.4
Compression seals	18.4	1.8
Container	232.4	22.6
Cell total weight	1028.4	100.00

volume of 836 cm<sup>3</sup>, of which the internal components were 336 cm<sup>3</sup>. A weight breakdown is given in Table 1. It can be seen that energy density is 44.9 W h kg<sup>-1</sup>, and volume density is 0.055 W h cm<sup>-3</sup>.

It is easy to compare this with an RFC by replacing the nickel positives with oxygen electrodes, making the necessary volume adjustments to the stack and container, and making provision for electrolyte reservoir plates (ERPS) to store water on discharge (it should be noted that the overall process:  $\text{NiOOH} + 1/2 \text{H}_2 = \text{Ni(OH)}_2$  has the advantage of involving no change in electrolyte volume). On this basis, with a total lightweight ERP weight of 35 g, and with 72 g of electrolyte for a system consuming 12.6 g of water between full discharge and full charge, the stack weight will be 286 g and its volume 190 cm<sup>3</sup>. The container will require 1.5 times the free volume of that of the Ni-H<sub>2</sub> container to store the hydrogen and oxygen with divided storage (with technology to be decided, since the system described in ref. 7 developed cross-leaks). This gives a total system weight of 720.3 g for a volume of 940 cm<sup>3</sup> and the same capacity (in A h) as the Ni-H<sub>2</sub> system. Since the current density will be the same as that for the latter (with this design), *i.e.*, 22.5 mA cm<sup>-2</sup>, performance should be good with the oxygen and hydrogen electrodes now available, *i.e.*, 1.45 V average on charge, 1.0 V (average) on discharge, or 69% overall efficiency. Consequently, on discharge the system will provide 38.5 W h, giving 53.4 W h kg<sup>-1</sup>, or 0.041 W h cm<sup>-2</sup>, 19% higher and 25% lower, respectively, than those for

Ni-H<sub>2</sub>. In consequence, the attraction of the RFC appears marginal if it is designed for this application.

However, the above is not true if cycle life is considered. For GEO use, Ni-H<sub>2</sub> under normal 60% DOD cycling conditions with standard chemically impregnated electrodes was shown to be capable of more than 2000 cycles in 1976. The failure mode was expansion of the positive as the nickel sinter oxidized and broke up under the internal pressure conditions created. This could be alleviated if a low-loading (69%) of active material was used, when more than 4000 GEO cycles could be achieved [14]. Electrochemically impregnated electrodes [15] with standard loadings, which have a better distribution of active material in the sinter porosity than chemically impregnated electrodes, gave 4000+ cycles. Finally, 8000 - 12 000 cycles with low-loading, electrochemically-impregnated positives can now be routinely demonstrated [16]. If more than 50 000, 0.6 h cycles are required for the LEO application, then the Ni-H<sub>2</sub> system must be highly derated to ensure adequate long-term performance, *e.g.*, 50% DOD at  $C/1.2$  or even 40% DOD at  $C/1.5$ . In the former case, average discharge potential may be 1.10 V, in the latter 1.15 V. Overall real energy densities on the cell level will be 21.1 W h kg<sup>-1</sup> and 17.7 W h kg<sup>-1</sup>, respectively. The RFC can, however, be still discharged over many thousands of cycles at 100% DOD, thus delivering around 50 W h kg<sup>-1</sup> at the higher discharge rates.

The real capability of the fuel cell is, however, for fast discharge. The power ability of the "stack" within the model RFC considered here is 224 W kg<sup>-1</sup> at 1.67 C. This is quite a high figure, since it requires no systems components. For present combination fuel cell-electrolyzer RFCs, the figure will be generally lower. For example, an advanced alkaline fuel cell can deliver 250 W kg<sup>-1</sup> at 1.0 V, 300 mA cm<sup>-2</sup>. The complete system, with an advanced electrolyzer operating at 600 mA cm<sup>-2</sup> for 0.9 h charge, the total power system energy density will be 187 W kg<sup>-1</sup> or 112 W h kg<sup>-1</sup> for the 0.6 h mission requirement. With gases separately stored in Inconel 718 tanks with a safety factor of 4, propellant and tank weight is 5.73 kg(kW h)<sup>-1</sup> including a 17% gas reserve (*i.e.*, cycling between 4.1 MPa, 41 atm, and 700 kPa, 7 atm). Total system energy density will therefore be 68 W h kg<sup>-1</sup>. The system will be capable of pulsed power up to 380 W kg<sup>-1</sup> with only a 10% voltage loss, however, which is impossible for any type of Ni-H<sub>2</sub> battery delivering only 21 W h kg<sup>-1</sup>.

Since power system weights are reduced by the use of higher power density systems at equal or similar output voltage, the RFC system can only look better at higher rates. At the 0.6 h rate, the weight of the power system (in the above case 5.33 kg kW<sup>-1</sup> for nominal output) largely exceeds the weight of the storage system (5.73 kg(kW h)<sup>-1</sup>, representing 3.44 kg for the mission requirement). If long mission requirements (*e.g.*, 8 h) exist, however, the storage system weight greatly exceeds that of the power system, which results in a total of 6.40 kg(kW h)<sup>-1</sup>, or 156 W h kg<sup>-1</sup>. This then exceeds the energy available from any known secondary battery, including sodium-sulfur.

We should also note that at the 0.6 h rate, Ni-H<sub>2</sub> cells have a heat-rejection problem that also limits depth of discharge capability, and the system may require active cooling in advanced compact forms (e.g., bipolar systems), with increase in system weight and complexity. The fuel cell-electrolyzer combinations already contain all necessary subsystems.

### RFCs: the future

Already the RFC based on alkaline technology is capable of 1.0 V, 300 mA cm<sup>-2</sup> in a much lighter, this time bipolar, package than that of the Advanced Lightweight Fuel Cell of 1979. In a continuous mode, this system should now be capable of 2 kg kW<sup>-1</sup> at 1.0 V. A lightweight electrolyzer adding 1 kg kW<sup>-1</sup> of output is possible. Similarly, lightweight pressure vessel storage using composite materials and more reasonable safety factors (3 rather than 4) will further lower system weight. Hence, probable relative power system weights of 3 kg kW<sup>-1</sup> with 1.6 kg kW<sup>-1</sup> for a storage system for the 0.6 h mission are possible. This yields 130 W h kg<sup>-1</sup> for a system with 60% overall efficiency, far exceeding the capability of any H<sub>2</sub>/X system or of sodium-sulfur. In any case, the cycle life capability of the latter has yet to be proven to even 2000 cycles on a systematic basis, and the LEO mission requirement exceeds this by a factor of 30.

Another intriguing possibility is the use of a monolithic, solid oxide fuel cell (SOFC, [17]), either alone, or in combination with the lightweight alkaline fuel cell as the electrolyzer unit. As an electrolyzer it will add only 200 g kW<sup>-1</sup> to the fuel cell weight, and it will operate at 1.3 V and 500 mA cm<sup>-2</sup>. This approach has the advantage of decreasing system weight, so that in LEO mode 158 W h kg<sup>-1</sup> can be achieved, at the same time increasing system efficiency to 77%. Alternatively, if a lower round-trip efficiency can be tolerated with reduced weight, the monolithic SOFC can be used as a combined electrolyzer-fuel cell, with a stack weight of 200 g kW<sup>-1</sup> (output), and it would operate at 1.3 V (charge) and 0.8 V (discharge) at 500 mA cm<sup>-2</sup>, yielding (on paper) 330 W h kg<sup>-1</sup> in the LEO mode at 62% efficiency. However, systems (heat rejection) aspects of its use in the exothermic fuel cell mode are presently uncertain, and the technology may not be available before the year 2000.

A more conservative approach is the single-unit, low temperature system, either on the lines of the single cell with pressure vessel approach of ref. 7 (assuming that heat rejection on discharge is acceptable), or in a bipolar mode with a separate advanced storage system. The single cell can use either alkaline or SPE technology. The latter may again have the advantage of lowest weight, whereas the former should have higher efficiency (1.0 V fuel cell, 1.5 V electrolysis, yielding 67% overall). As an example, the SPE system has been recently shown to be capable of 150 g kW<sup>-1</sup> at 0.7 V, 1.75 A cm<sup>-2</sup> using proprietary lightweight stack components, with a low-resistivity 75 μm dry, 113 μm wet, Dow Chemical SPE membrane, using

data recently obtained by Ballard in Canada [18]. These results show 0.8 V at 500 mA cm<sup>-2</sup>, which as a baseline yields 427 g kW<sup>-1</sup> for the stack. Hence, total weight, including advanced storage, will be 2 kg kW<sup>-1</sup> for the mission requirement or 300 W h kg<sup>-1</sup>. The cathode catalyst and bipolar structure will be modified in a proprietary manner to permit electrolysis at 1.50 V, allowing round-trip efficiency of 53%. This can be increased at the expense of weight, if necessary.

The results above are summarized in Table 2, which compares the above RFC technologies, Ni-H<sub>2</sub>, and a hypothetical, long-life 100 W h kg<sup>-1</sup> Na-S battery for LEO application. The state-of-the-art weight of deployable PV is taken to be 55 W kg<sup>-1</sup> (25 W lb<sup>-1</sup>). All data are reduced to 1 kW output power levels. SOE refers to solid oxide electrolyser. The pacing element is clearly the mass of the PV array, although the weight penalty for the lower efficiency systems is quite small.

TABLE 2

Total LEO system reciprocal power density breakdown

Technology	kg kW <sup>-1</sup>	Efficiency	PV (kg kW <sup>-1</sup> )	Total (kg kW <sup>-1</sup> )
Ni/H <sub>2</sub>	30.0	78	15.5	45.5
RFC (baseline)	11.0	60	20.2	31.2
RFC (lightweight)	4.6	60	20.2	24.2
Adv. AFC-SOE	3.8	77	15.7	19.5
Regen. SOFC unit	2.0	62	19.5	21.5
Regen. SPE unit	2.0	53	22.6	24.6
Adv. Na-S	6.0	85	14.3	20.3

Finally, it would seem more profitable in future to manufacture H<sub>2</sub>/O<sub>2</sub> propellant in space for orbital transfer, since it is energetically more economical to transfer water from earth than to ship the same weight of cryogenic propellant. The fuel cell-electrolyzer combination therefore seems to be a more logical long-term solution to the problem of space energy storage than secondary batteries.

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