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Alkaline fuel cells running at elevated temperature for regenerative fuel cell system applications in spacecrafts

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ABSTRACT

The energy supply is one of the most important subsystems of spacecrafts. Its layout depends on factors like size or mission profile and contributes strongly to the spacecraft mass. Nowadays, secondary batteries are the common energy storage for space missions. However for certain applications regenerative fuel cell systems (RFCS) offer mass savings due to their higher energy density. A RFCS is composed of an electrolyser and a fuel cell. In a first step water is electrolysed by electrical energy. Hydrogen and oxygen are stored in tanks. Later they are supplied to the fuel cell. There they recombine to water providing electrical energy.

The total efficiency of the process and the mass of the complete RFCS are strongly affected by the fuel cell efficiency. It can be increased by elevating the fuel cell operation temperature. This leads to a higher energy density and enables mass reductions of the radiator which is necessary in space to get rid of the waste heat. In this article the effect of the temperature on the radiator area is calculated. The impact on efficiency and fuel cell mass is outlined. Tests results of the operation of an alkaline fuel cell between $60 \,^\circ$ C and $140 \,^\circ$ C are presented.

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1. Introduction

Fuel cells are energy converters that transform chemical into electrical energy. Advantageously, the energy conversion is not subject to the Carnot factor. Therefore, theoretically high efficiencies can be reached.

1.1. Alkaline fuel cell

Different types of fuel cells exist, grouped by parameters like operation temperature or the electrolyte used. In alkaline fuel cells (AFC) the electrolyte is a liquid potassium hydroxide solution (KOH). Alkaline fuel cells are operated at low temperatures, usually below 100 °C. They are characterised by fast reaction kinetics and low overvoltages. By this they enable a high efficiency and the use of cheap non-precious metal catalysts. AFC have a long heritage in space applications, e.g. they have been chosen as energy supply for the Apollo Program and the Space Shuttle Orbiter [1]. The electrolyte can either be mobile and circulated in a separate electrolyte cycle or immobile and bound to a central matrix. The immobile or fixed solution offers some advantages, especially for spacecrafts. The direction of gravity has no influence and no KOH will be set free in case of the impact of a micrometeoroid. There are less moving parts and therefore fewer oscillations which would otherwise influence the microgravity environment. The basic assembly and the working principle of a fixed AFC are given in Fig. 1 [2]. Each cell comprises a diaphragm, enveloped by the electrodes (EDE, electrode-diaphragm-electrode assembly). A homogenous gas supply is guaranteed by flow fields. Hydrogen and oxygen are supplied to the reaction zones and diffuse into the electrode pores. The molecules are bound to the catalyst surface and split. The released electrons flow to the opposing electrode supplying an external load. The ions are conducted through the electrolyte closing the circuit. The produced water is released from the cell via an excess gas flow. Endplates compress the cell(s) to minimise contact resistances and achieve leak tightness. Multiple cells can be combined to a stack by placing them on top of each other and connecting them via bipolar plates in between (bipolar design, cathode of one cell facing anode of the next one).

1.2. Alkaline electrolyser

Reversing the working principle of a fixed AFC gives a fixed alkaline electrolyser (FAE). Electrical energy and water are supplied to the cell and the water is split into its elements by electrolysis.

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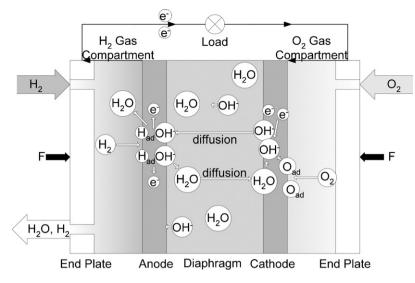


Fig. 1. AFC assembly.

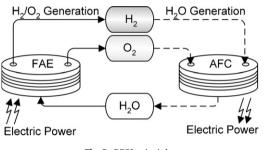


Fig. 2. RFCS principle.

Since a few years, a FAE has been investigated at Astrium, Friedrichshafen. The alkaline fuel cell used for the tests described hereafter was developed based on the design of the fixed alkaline electrolyser.

1.3. Regenerative fuel cell system

The combination of both aggregates leads to a regenerative fuel cell system (RFCS). If a surplus of energy is available, the electrolyser is used to produce H_2 and O_2 from stored water (charge mode, Fig. 2, continuous lines). The gases are stored in tanks. With decreasing power supply the RFCS is switched into discharge mode (Fig. 2, dashed lines). The gases are supplied to the fuel cell and recombine under the formation of water which is stored again. In a RFCS no mass flows over the system boundaries occur. Only energy (electrical energy, heat) enters or leaves the system.

RFCS have several advantages compared to batteries. Especially for applications with high power demand and long discharge cycles they offer a smaller system mass. The provided power is decoupled from the amount of stored energy. If more energy has to be stored for a certain task, the mass of the actual energy converter remains constant and only the tank mass increases. The dimensioning and optimisation of single elements of RFCS-based energy systems depending on the mission is currently investigated by Astrium and the University of Technology Dresden [3]. Some of the investigated applications are given in Fig. 3. Below the continuous line in Fig. 3 applications are shown where batteries are beneficial, above, such missions with RFCS benefits. The most promising applications are geostationary satellites, exploration missions or HALE (high altitude long endurance) aircrafts. For example, future communication satellites in geostationary orbits (GEO) will require electrical power in the range of larger 20 kW [4]. Due to the estimated higher energy density of the RFCS compared to Li-ion batteries, mass savings can be achieved. One further advantage of the RFCS lies in its possible synergies with other spacecraft subsystems (Fig. 4) [2]. The RFCS is a part of the electrical power system (EPS). The electrolysed gases can also be used as fuel for the propulsion system (PS) or attitude and orbit control system (AOCS). Product water of the fuel cell can be routed to the environment control and life support system (ECLSS). Treated ECLSS waste water can be used as source for the electrolyset for the crew. The waste heat can be used for the thermal control system (TCS).

The RFCS technology has also some drawbacks like the increased complexity and a reduced technology readiness level compared to nowadays batteries. Much ancillary equipment is needed.

Mission profiles with very short discharge durations and/or low power requirements like low earth orbit (LEO) satellites or small rovers may further favour batteries (no mass reductions possible due to disproportional high amount of ancillary equipment). The round-trip-efficiency (charge + discharge efficiency) of a RFCS is – and in near future will remain – smaller compared to a battery system. At equal electrical power more waste heat is produced. This has to be considered during the dimensioning of a spacecraft to achieve a minimum overall system mass.

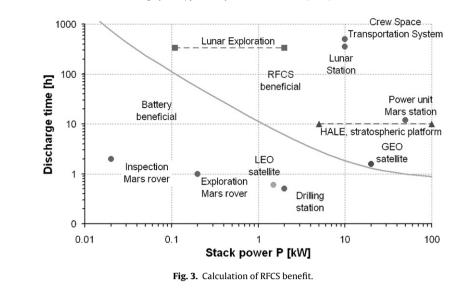
A terrestrial spin-off of the RFCS technology could be the energy storage for the smoothing of the power output of regenerative power sources like wind parks or solar arrays which usually provide a strongly oscillating load.

2. Calculations

The following calculations focus on the influence of the operation temperature of the alkaline fuel cell on the RFCS mass. The electrolyser is omitted as so far it has not been investigated in detail under the aspect of system mass reductions.

2.1. AFC temperature vs. radiator size

Fig. 5 shows the calculated influence of the electrical power of a geostationary satellite on the specific energy. The specific energy of batteries remains nearly constant as their mass increases proportionally with power. For RFCS it improves with increasing power demand (power and energy decoupled). The upper boundary of the respective areas for batteries and RFCS characterises the possible



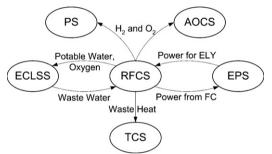


Fig. 4. RFCS synergies to other spacecraft subsystems.

value without thermal hardware. The area itself gives resulting specific energies if the mass of the thermal equipment is considered.

To achieve the maximum specific energy it is necessary to use as little thermal hardware as possible. Therefore an integrated mechanical/thermal architecture (including radiators) is needed to accommodate RFCS stacks, tanks and auxiliaries [4].

The missing atmosphere in a satellites orbit or on the lunar surface limits the heat rejection to radiation cooling. The theoretical size of the radiators can be calculated by Stefan–Boltzman's law:

 $P = \varepsilon \cdot \sigma \cdot A \cdot T^4$

with

- *P*. the waste heat to be radiated.
- ε, the emissivity.
- σ , the Stefan–Boltzman constant (5.67051 × 10⁻⁸ W m⁻² K⁻⁴),
- *A*, the radiator size and
- *T*, the surface temperature of the radiator.

For the detailed layout of the complete system different additional parameters like heat intake by solar flux on the orbit plane or infrared and albedo radiation from earth (or moon for lunar applications) have to be taken into account. Further factors are the heat conduction and distribution within the radiator and its structures or the view factor (shadowing by other surfaces).

It is obvious, that the temperature has a large impact on the radiation capability. By increasing the working temperature of the AFC, the necessary radiator area can be drastically decreased. One example for a geostationary communication satellite with a RFCS as energy storage is given in the following.

During eclipse an electrical power of 20 kW should be provided by the alkaline fuel cell. At 80 °C a discharge efficiency of 60.9% and at 130 °C of 74.6% is assumed. These values have already been reached during tests (see chapter 3). From these values the waste heat can be calculated to 12.8 kW (80 °C) and 6.8 kW (130 °C).

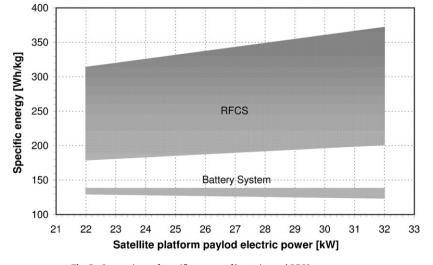


Fig. 5. Comparison of specific energy of batteries and RFCS over power.

Table 1
Comparison of the necessary radiator area for battery and AFC.

	Battery 20°C	AFC		
		80°C	130°C	
Electrical power		20		kW
Discharge efficiency	90.0	60.9	74.6	%
Thermal power	2.22	12.84	6.81	kW
Radiator area	6.63	18.20	5.68	m ²
Radiator surface weight	3.3	3.5	4.0	kg m ⁻²
Radiator mass	21.9	63.7	22.7	kg

Further an emissivity of 0.8 (8 mil quartz mirror [5]) is assumed. Using Stefan-Boltzman's law the radiator area at 80 °C can be calculated to 18.2 m². If the temperature is increased to 130 °C, the area decreases to 5.7 m², which equals a reduction by 68%. A comparison of the radiator area for a battery system and an AFC within a RFCS at different temperatures is given in Table 1. The radiator mass at low temperature is larger than for a battery system. However, by increasing the temperature, the mass decreases, also if a higher radiator surface weight is used for the calculation due to the more stringent properties of the high-temperature-radiator. At 130 °C the theoretical radiator mass is only slightly above the one for the battery system. The electrical power system contributes strongly to the overall spacecraft mass (typically 25-30% of the dry mass [5]). Therefore, for a geostationary satellite with a typical mass of more than 1000 kg the mass reduction as a consequence of the higher temperature is significant and desirable.

2.2. AFC efficiency vs. system mass

An increased AFC temperature affects the mass of an RFCS-based electrical power system in a second way. With increasing temperature reaction kinetics accelerate. As a consequence the operational voltage increases enabling a higher efficiency. The thermoneutral cell voltage E_{tn} of an AFC can be calculated from:

$$E_{tn} = \frac{\Delta H_R}{z \cdot F}$$

with

- ΔH_R , the reaction enthalpy,
- *z*, the number of exchanged electrons per reaction (2) and

• *F*, the Faraday constant (96,485 $C \mod^{-1}$).

It gives back the maximum achievable voltage without consideration of any losses that is an efficiency of 100%. ΔH_R depends on the temperature and is determined from the enthalpies of formation of the involved species (H₂, O₂, H₂O). A calculation of the enthalpies of formation can be found at [6].

The operational voltage of a fuel cell E_{op} deviates from the thermoneutral voltage. It is influenced by parameters like design, electrodes, catalysts, applied current density, etc. It is decreased by entropic losses, the limited speed of diffusion processes of the involved species, ohmic losses within the cell and wiring and other factors. The efficiency of a fuel cell is defined as the ratio of the operational voltage E_{op} reached (with all losses) to E_{tn} . If the fuel cell is part of a RFCS it equals the discharge efficiency of the RFCS.

$$\eta = \frac{E_{op}}{E_{tn}}$$

A higher operational voltage leads to further mass reductions as shown in the following example. Based on:

 $P = n \cdot E_{op} \cdot I$

with

Table 2

Calculation of AFC cell number for different temperatures.

	80 ° C	130°C	
Electrical power	2	20	kW
Current	30		А
Thermoneutral voltage	1.24	1.23	V
Operational voltage	0.755	0.918	V
Discharge efficiency	60.9	74.6	%
Number of cells	883	727	-

• *P*, the electrical power,

• *n*, the number of cells in a fuel cell stack and

• *I*, the cell current.

the required AFC cell number for the electrical power system of a satellite can be calculated. Table 2 presents the operational conditions and the number of necessary cells for a GEO satellite with 20 kW electrical power. The influence of the temperature on the thermoneutral voltage via the enthalpy of formation is taken into account [6]. An increase in operational voltage by 165 mV was assumed for the calculation. This has already been achieved during tests (see chapter 3). The number of cells and therefore the stack mass decreases by 17% which leads to a corresponding increase of the energy density.

3. Experimental setup

A test bench has been assembled at Astrium, Friedrichshafen (see Fig. 6) to provide information on the influence of the temperature of alkaline fuel cells for RFCS applications in spacecrafts.

The AFC test rack comprises sensors and actuators for the gas and water management, including heaters, pumps and pipework. Parameters like temperature, pressure, flow rates and gas contamination are monitored at different points of the rack and analysed. The data is sent to a programmable logic controller for the controlling of the operation parameters. The evaluation of the AFC behaviour is done via a data acquisition and control unit. To achieve temperatures of up to 150°C an additional thermostat with oil as heat transfer fluid is used. The pressure within the gas cycles is maintained at 1-3 bar. Hydrogen and oxygen are provided from external bottles. The produced water is transported from the cell via an excess gas flow. It is condensed and collected. The excess oxygen is vented to the ambient environment. Hydrogen can be vented or also recycled. This recycling option represents an important part of a closed-loop-RFCS. For test purposes and in case of technical failures the system can be purged with nitrogen. Safety features like an automatic emergency shutdown (ESD) for dedicated failure cases are included. Most important is the monitoring of the gas concentrations within the adjacent gas compartments. Due to diffusion processes H₂ can diffuse into the O₂ compartment or vice versa. At 2% cross contamination an automatic ESD and system purge are initiated. A summary of the operation parameters of the AFC within the test rack is given in Table 3.

Table 3

Operation parameters of the AFC within the test rack.

	Nominal	Range	
Cell number	4	1-4	-
Temperature	80-130	20-150	°C
Pressure	1.2-2.5	1-3	bar
Electrolyte molarity	6	6-14	mol l ⁻¹
Voltage (4 cells)	~3.2	0-4.8	V
Current	30	0-60	А
Current density	$\sim \! 170$	0-350	mA cm ⁻²



Fig. 6. AFC test rack.

4. Results and discussion

The tested fuel cell (Fig. 7) had a circular design with an effective diameter of 150 mm. The anode was composed of a PTFE-carbonpowder-mixture on a nickel mesh with a palladium catalyst. The cathode was made of a PTFE-silver-mixture on a nickel mesh. A porous Zirfon[®] separator (ZrO₂ in a polysulphone matrix) [7] filled with liquid potassium hydroxide was used as diaphragm. The cell frames were manufactured from polyether ether ketone (PEEK). Proprietary metallic flow fields were used. All materials were chosen from the electrochemical series to tolerate the challenging conditions at up to 140 °C and 14 mol1⁻¹ KOH. During the tests most cell materials showed no difference in degradation between $60 \,^{\circ}$ C and $140 \,^{\circ}$ C, except for the Zirfon diaphragm. It tolerated the high temperature only during the short term tests but dissolved after 500 h in a separate oven test at $140 \,^{\circ}$ C with increasing KOH molarity (=higher corrosiveness). For the future a new diaphragm on the basis of ceramic NiO will be investigated to achieve higher operation durations.

The bipolar assembly was tested in single cell configuration or as a stack of up to four cells. The identical design was used throughout the whole temperature range to provide comparability of the tests and to show, that the system has the capability to operate at all temperature levels (especially interesting during transient phases like run up and shut down or non-operation).

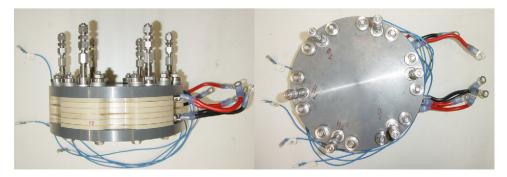


Fig. 7. Side and top view of AFC 4 cell stack.

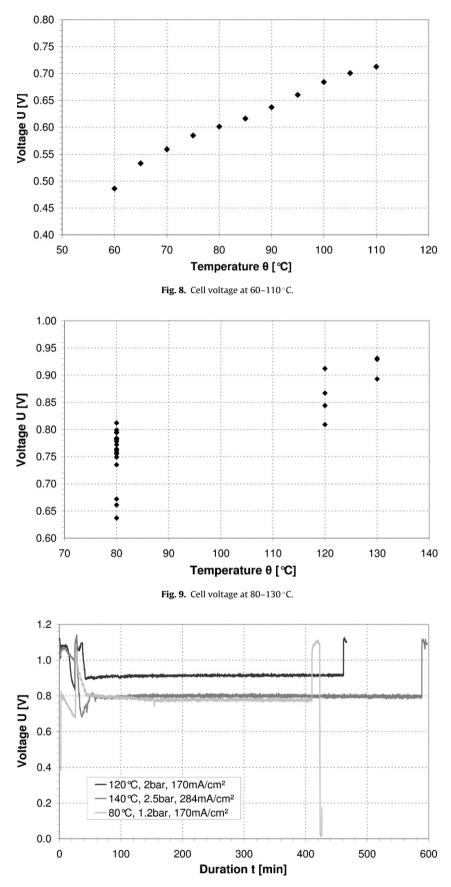


Fig. 10. Stability at 80 °C, 120 °C and 140 °C.

The operation conditions during the tests were chosen dependent on the respective temperature. The performance of an AFC is strongly influenced by the humidification of the cell. With increasing temperature the ability of the gas flow to transport water from the cell rises. To avoid a dry-out of the diaphragm (which would lead to gas breakthroughs) the operation parameters were calculated before each test to guarantee an equal humidification. The changed parameters included:

- molarity: an increased molarity of the electrolyte leads to a lower H₂O vapour pressure and less water loss,
- gas pressure: an increased pressure decreases the gas volume and therefore the H₂O transport from the cell,
- λ_{H_2} and λ_{O_2} : the smaller the amount of excess gas (λ = ratio of supplied gas to stoichiometric consumption) the smaller is the vapour transport.

The main objective of the described tests was to check the positive influence of the temperature rise on the efficiency and to prove the feasibility of the operation at temperatures of 60-140 °C.

Fig. 8 depicts the result of a test at a constant current density of 170 mA cm^{-2} over a temperature range from $60 \circ \text{C}$ to $110 \circ \text{C}$. Differing from the adjustments stated above, molarity ($6 \text{ mol} \text{ I}^{-1}$) and pressure (ambient) were kept constant during this test. The quite low efficiency was a result of these missing adjustments. Nevertheless, a significant rise of the cell voltage with operation temperature was identified. The efficiency related to a thermoneutral voltage of 1.23 V increased from 39% to 57%.

Fig. 9 comprises several measurements under adjusted operation parameters. Each measurement stands for a single test over several hours (1-9 h) at a constant current density of 170 mA cm^{-2} . The voltage was measured and a mean value formed. The operation parameters were adjusted and continuously controlled during the whole test to guarantee an equal humidification of the cells. The average voltage measured at $80 \,^{\circ}$ C was 0.755 V. At $120 \,^{\circ}$ C it was 0.858 V and at $130 \,^{\circ}$ C it was 0.918 V. This equals a rise of efficiency from $80 \,^{\circ}$ C to $130 \,^{\circ}$ C from 60.9% (compared to $1.24 \,^{\circ}$ V) to 74.6% (compared to $1.23 \,^{\circ}$ V). From the different tests it is obvious, that the efficiency could be significantly increased by increasing the temperature (see also Fig. 10).

At higher temperature levels the AFC showed an equal or even slightly better stability than at 80 °C (Fig. 10). At 80 °C high current densities often led to instable behaviour. As shown in Fig. 10 the measured voltage at 140 °C and a significantly increased current density of 284 mA cm^{-2} is equal to the one at 80 °C and 170 mA cm^{-2} and remains constant over several hours. Therefore,

the operation at higher temperatures seems feasible and even recommendable.

5. Conclusion

The article describes the influence of the operation temperature on the performance of an alkaline fuel cell for RFCS applications in spacecrafts. The basic working principles of an AFC and a RFCS are outlined in brief. The benefits of a RFCS over batteries for certain space missions are summarised. The consequences of an increased temperature of the fuel cell on the system mass of an RFCS-based EPS are calculated. Two effects are identified. The higher temperature leads to a better radiation characteristic resulting in a lower radiator mass. Secondly the better efficiency at higher temperatures enables mass savings due to a decreased number of necessary cells. At Astrium, Friedrichshafen a test facility has been assembled to verify the previous calculations and assumptions by tests. An alkaline fuel cell was tested at various temperature levels. It was shown, that by increasing the temperature an increase in cell voltage could be achieved. For the future even further improvements seem possible if the temperature is increased even more.

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