

COMPARISON OF MAN-PORTABLE POWER GENERATION ALTERNATIVES BASED ON FUEL-CELL SYSTEMS

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ABSTRACT

Micro-fabricated fuel cell systems are a potential consumer product with the promise to outperform batteries for man-portable power generation in terms of the achievable energy density. There is a plethora of potential civil and military applications and a great variety of potential devices are being considered in various research institutions. We have developed a systematic methodology for the comparison of alternatives and examination of the influence of technological parameters and have implemented this in a web-based tool. We consider a variety of fuel/chemical choices, including hydrocarbons, methanol, ammonia and hydrides; oxygen options considered are atmospheric air, compressed air, compressed oxygen, and oxygen generators; fuel cells types considered are Solid Oxide Fuel Cells, hydrogen operated Polymer Electrolyte Membrane Fuel Cells, Direct Methanol Fuel Cells, Proton Ceramic Fuel Cells and Single Chamber Fuel Cells. In this paper we present and motivate the resulting process alternatives and demonstrate the use of the methodology, in case studies, showing the scaling of the process performance and the influence of water management.

1 INTRODUCTION

The ever-increasing use of portable electric and electronic devices increases the need for efficient autonomous man-portable power supplies [Jacobs et al.(1996), Dyer(2002)]. Currently, batteries are the predominant technology in most applications. However, batteries have a large environmental impact, high cost and relatively low gravimetric (Wh/kg) and volumetric (Wh/l) energy densities. State-of-the-art primary batteries reach up to 1300Wh/l and 700Wh/kg and rechargeable up to 400Wh/l and 300Wh/kg [Linden(2001), Brodd(1999)]. The upper limit on battery performance is now being reached as most of the materials that are practical for use as ac-

tive materials in batteries have already been investigated and the list of unexplored materials is being depleted [Dyer(2002), Linden(2001)].

There are two main approaches for fuel cell systems, namely direct fuel cells running on stored hydrogen, methanol, formic acid, or medium sized hydrocarbons, as well as fuel processing for hydrogen or syngas generation and subsequent oxidation of these intermediates in a fuel cell. Micro power generation devices based on either approach are products that comprise a more or less complex chemical process. There is a plethora of possible processes and process combinations, as well as a wide variety of applications and consumers, ranging from cellular phones and laptops for home use to the power needs of the dismounted soldier, thus it is plausible that the optimal device configuration will depend on the product specifications characterizing particular applications. This necessitates a flexible methodology for the comparison of different technology alternatives that can facilitate product engineering of these devices.

In macro-scale processes design is performed in stages, e.g., [Biegler et al.(1997), Douglas(1988)], and plant layout is typically addressed independently of and subsequently to the design of the flowsheet. At the micro-scale the process components are highly integrated and heat losses significantly influence the process performance [Mitsos et al.(2004a)] and possibly the optimal design, so that the problems of flowsheet design, physical layout and integration of heat sinks and heat sources need to be addressed simultaneously. A very promising approach is to couple two or more unit operations thermally in a near-isothermal stack [Arana et al.(2003)].

2 BRIEF DISCUSSION OF METHODOLOGY

We propose a methodology that allows the comparison of different flowsheet alternatives as well

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as the influence of operating conditions, such as temperature, and unit efficiencies, such as conversion. The alternative processes considered, were chosen with the constraint that the realization of the processes is either currently under investigation or foreseeable in the short term future (coming years). While kinetic data exist for special catalysts, e.g., [Deschmukh et al.(2004), Idem and Bakhshi(1996)], that allow for detailed modeling, our intention is to have a general methodology that can cover various geometries and reactor types (PFR, CSTR, packed bed, etc.) and be independent of the specific catalysts used. Our models are therefore based on user specified efficiency parameters in the various units like conversion, electrochemical efficiency, separation efficiency, etc. Once these parameters, as well as the operating conditions have been specified, the performance of the system is calculated. A variety of metrics can be used for the performance evaluation in our methodology; depending on the potential application different objectives are more important; typically gravimetric and volumetric energy density are most important. The scope of this paper does not allow for a detailed description of the modeling equations.

Most power consuming devices are not operated constantly and have rapidly changing power demands, and therefore the dynamics and operation of power generation devices are very important. Similar to the electric vehicle application [Weber and Ivers-Tiffée(2004)], a fast start-up procedure, at most on the order of minutes, is required. In this paper we consider the steady-state performance under the assumption that the devices will be able to respond to power demands rapidly and therefore the average performance will most likely be dominated by the steady-state behavior of the devices. For the calculation of energy density, we include the mass/volume of the device, an auxiliary battery as well as the fuel cartridge for a given mission duration (time between refueling).

The methodology presented here is complemented with more detailed modeling and the study of transient performance and optimal operation, currently under investigation [Chachuat et al.(2004)]. We have implemented the methodology as a tool available in the form of a web-interface [Mitsos et al.(2004b)], which allows for facile use by remote users, who are unfamiliar with the modeling language and the details of the models. Upon request and subject to approval this web-interface can be made available for academic purposes.

3 CONSUMER SPECIFICATIONS

There are a large number of devices, with different characteristics, that currently require man-portable power production and have both military and civilian application. Electronic devices, such as cellular phones, digital camcorders and laptop computers have a small power requirement during stand-by and a high power requirement during actual operation; state-of-the-art batteries are used and although the battery takes up a significant portion of the device volume and mass, only a few hours of actual operation are possible; these devices have strict requirements on the voltage of the electrical power supplied. Simpler power consuming devices, such as flashlights are more flexible regarding the power supply; cheaper and less efficient batteries are typically used for such applications. On the limit of portability are electrical vehicles for the elderly and handicapped, which typically use lead-acid rechargeable batteries with a mass of several kg and a mission duration of many hours. It is to be expected that in the near future new power consuming devices, with possibly drastically different specifications on the power demand, will come to the market. An example is so called exoskeletons (also dubbed power pants, power elbows, etc.): robotic suits with the promise of multiplying the force of soldiers or rescue workers or even allowing motion to disabled people [Huang(2004a), Huang(2004b)]. These devices will probably be characterized by a very low power demand during stand-by operation, for monitoring purposes, and a spike in the power demand, reaching tens or hundreds of Watts during the actual operation. Other power consuming devices that are likely to become interesting applications for man-portable power generation include portable medical devices and robots or even small unmanned air vehicles.

Not only the power consuming device, but also the customer, influence the specifications on the power generation device, and since potential customers range from children to a dismounted soldier there is a great variety of needs. In typical civilian applications safety and non-toxicity are a necessary condition; an important aspect is that power generation devices can be carried and operated on airplanes, and refueling in different countries must be possible; also there is a strong trade-off between price and performance. For military applications safety is less important, since soldiers can be trained for safety and are already exposed to dangerous materials; here performance and reliability are the main

criteria and cost considerations are almost negligible; there are very important logistic considerations for refueling. Design constraints for military applications may include operation without noise generation or a thermal signature; operation under extreme conditions is possible, e.g., very low or very high temperatures, under water operation.

4 ALTERNATIVES CONSIDERED

As possible fuel/chemicals we consider hydrocarbons, methanol, ammonia and hydrides; for the oxygen supply we consider atmospheric air, compressed air, compressed oxygen and oxygen generators; fuel cells types considered are Solid Oxide Fuel Cells, hydrogen operated Polymer Electrolyte Membrane Fuel Cells, Direct Methanol Fuel Cells, Proton Ceramic Fuel Cells and Single Chamber Fuel Cells. We graphically represent the alternatives considered by a flow-sheet superstructure (Figure 1). We want to emphasize that the superstructure is only conceptual, and several of the "units" can actually be physically combined and our models account for thermal integration of the processes.

The first design choice is to choose the fuel that will be used for power production and whether to perform fuel processing in a reactor, or to directly feed the fuel to a fuel cell. Based on the process design heuristic for simplicity [Saha and Rinard(2000)] the postulated superstructure contains only one reactor. The next design choice is whether this fuel or a secondary fuel will be fed into a burner for heat generation. The heat produced from burners serves to compensate for stream preheating, heat losses, endothermic reactions or even heating of the system at startup.

Depending on the fuel processing reaction a secondary feed of water or oxygen to the reactor is necessary. If desired, we can split part of the reactor products and burn it to supply heat, in which case a stream split is necessary. As described in [Mitsos et al.(2004a)], simple recycle of the reactor products does not seem a promising option and no recycling after separation is considered here.

Certain components, such as carbon monoxide have deleterious effects on some fuel cells, e.g., PEM, and it may therefore be necessary to perform a gas purification. We assume that the purification will lead to two streams, one of essentially pure hydrogen

along with a waste stream. We consider a partial loss of the hydrogen in the waste stream, but we neglect any energetic penalty for the purification and the effect of a sweep stream, which may be necessary for the operation [Franz et al.(1999)]. The purification could either be sequential to the reactor, or the reactor and the membrane could be combined into one unit, allowing for higher selectivity of the reactions towards hydrogen [Shu et al.(1991), Franz et al.(1999)]. The separation waste can be either discarded or burned. If desired, the purification product (H_2) can be split, and a part can be fed into a burner.

We consider a variety of fuel cells, namely either a hydrogen operated Polymer Electrolyte Membrane Fuel Cell (PEM), a Solid Oxide Fuel Cell (SOFC) with the option of internal reforming, a Proton Ceramic Fuel Cell (PCFC), a Single Chamber fuel cell operating with hydrogen and carbon monoxide, or finally a Direct Methanol Fuel Cell (DMFC). PEMs are run at low temperatures but cannot tolerate impurities, and water management is an issue. A SOFC has the benefit of fuel flexibility, but it is operated at high temperature which leads to large heat losses and problematic start-up. Single Chamber fuel cells are potentially easier to fabricate [Dyer(1990)], but have the drawback that they are operated with premixed gases which potentially can lead to explosions and require catalysts with high selectivity. A PCFC is a relatively new concept [Coors(2003)], which has the potential of fuel flexibility while operating at slightly lower temperatures than SOFCs. A DMFC is a PEM based fuel cell in which a dilute methanol solution in water is reformed at a relatively low temperature, around 350K; major technical challenges include methanol crossover and water management. The reader is referred to the literature for extensive discussions about the technology differences in the fuel cells, e.g., [Song(2001), Haile(2003)].

The conversion in the fuel cells (often also denoted "fuel utilization") is not complete, and the unreacted part of the fuel can either be burned or recycled; this basic recycling option was analyzed in [Mitsos et al.(2004a)]. A more promising recycling option would be recycling after separation, e.g., separate the hydrogen of the fuel cell effluent and recycle it to the fuel cell, or separate the steam/water and use it for reforming reactions and to prevent coking. These options are very appealing from the point of view of minimizing the mass, but separation might be very difficult to implement in the general case. We allow for the option of separating the liquid and gaseous components of the anode and cathode effluents in a flash at a given temperature, most likely

near-ambient, and recycling a fraction of the liquids (mainly water and methanol) to the reactor or the fuel cell anode. Depending on the implementation of the recycle stream, a pressure increase mechanism may be necessary, e.g., a micro-fabricated pump, and we consider an energetic penalty in terms of a compression power. The remaining liquid components constitute a purge stream. The gaseous components can be recycled to the reactor, membrane, or fuel cell as in [Mitsos et al.(2004a)].

The cathode effluent stream of the fuel cell can be reused to provide oxygen for a burner because it is plausible that the fuel cells will be operated at a relatively large oxygen excess. Reusing excess oxygen is most advantageous in volume-critical applications where the oxygen cartridge may occupy a large fraction of the total system volume. In addition, the temperature of the cathode effluent stream is higher than the ambient, so this reduces the energetic requirement of preheating the oxygen feed to the burner. However, in circumstances where the fuel cell discard temperature is substantially lower than the operating temperature of the burner (i.e., for a PEM or DMFC), preheating is still necessary. The cathode effluent also contains nitrogen, and in some cases, e.g., a PEM, also steam, and heating of these components to the burner operating temperature may outweigh the advantage of using preheated oxygen.

5 CASE STUDIES

5.1 Scaling of Processes

The scalability of micro power generation devices is particularly interesting since there are two major scales. One is the nominal power output, which is mainly associated with the device size, and the other is the time between refueling (mission duration), which is associated with the fuel cartridge size. In this case study we present the influence of these two scales on achievable system performance; We use the parameters in Table 5.1 and as metrics for the scalability we use the volumetric and gravimetric system energy densities, where the system includes the power generation device and the stored fuel. Figure 2 shows the achievable energy density in Wh/(l system) and Wh/(kg system).

For low power outputs the heat losses dominate over the exothermicity of the fuel processing and

burning of the fuel cell effluents as well as part of the fuel is needed (Design I). Since the heat generation scales linearly with power output while heat losses scale sublinearly (with a power of 2/3) the achievable energy density increases significantly with the power output. At a power output of about 0.6W a kink is observed, because for higher power output the heat generation from burning the fuel cell effluents is sufficient (Design II). Above approximately 1.6W the process is exothermic enough, so that the fuel cell effluents need not be oxidized (Design III). The system energy density increases with mission duration and approaches the energy density with respect to the fuel volume/mass because the device size becomes negligible. This case study demonstrates that the influence of scale on process performance is significant; since different processes scale in general differently, the optimal design is also likely to be influenced by the scale.

Table 1: Parameters for the comparison in Figure 2.

Ambient temperature	$T_{amb} = 298K$
Power output	$PW = 1W$
Reactor temperature	$T_{op} = 1000K$
Reactor outlet temperature	$T_{out} = 500K$
Conversion in reactor	$\zeta = 0.9$
SOFC temperature	$T_{op} = 1000K$
Residence time in reactor	$\tau = 1ms$
Discard temperature from SOFC	$T_{out} = 500K$
Conversion in burners	$\zeta = 0.95$
PEM temperature	$T_{op} = 350K$
Residence time in burners	$\tau = 1ms$
Discard temperature from PEM	$T_{out} = 350K$
Air excess in burners	$\Phi = 1.2$
Conversion in fuel cell	$\zeta = 0.8$
Residence time in fuel cell	$\tau = 20ms$
Overall heat loss coefficient	$U = 3W/m^2/K$
Emissivity (incl. view factor)	$\epsilon = 0.2$
Efficiency of fuel cell	$\eta_{FC} = 0.7$
Air excess in fuel cell	$\Phi = 1.2$
Air feed penalty	$K_C = 10J/mol/K$
Burner temperature	$T_{op} = 1000K$
Burner discard temperatures	$T_{out} = 500K$
Water factor in fuel cell	$\Psi = 1$
Propane molfraction in feed	0.5
No air excess in reactor	$\Phi = 1.0$
Device density	$\rho = 1kg/l$
Fuel cartridge thickness	$d = 1mm$
Cartridge density	$\rho = 1.5kg/l$
Startup time	$\tau_{startup} = 60s$
Energy density for battery	200Wh/kg, 200Wh/l.
Device volume / reaction volume	10

Water Management in Water Reforming Reactions

Steam reforming is widely used in stationary applications, e.g., [Farooque and Maru(2001)]; this is an interesting alternative because water/steam are relatively inexpensive, and in the case of fuel cells operating at high temperature the heat excess from the fuel cell can be used for the fuel processing reaction. In portable applications, on the other hand, carrying the water strongly decreases the energy density, and therefore steam reforming reactions are not necessarily the optimal fuel processing path. Water separation and recycling is a task that may be impossible to implement at the micro-scale, but it has the promise of significant improvements in performance because it can minimize the size of the water cartridge. On the other hand, reforming reactions are operated at relatively high temperatures, and therefore recycling of water is associated with a large energetic penalty for vaporization and heating; also recycling an excess of water dilutes the fuel and increases the required device volume, resulting in increased heat losses. The tradeoff between these considerations leads to an optimum recycling ratio for a given requirement for water in the reactor feed.

Here we consider a process based on the combination of a hydrocarbon reforming reaction with a SOFC. The water requirement in the reactor feed is specified according to an excess factor Ψ , relative to a complete reforming reaction:

$$N_{\text{H}_2\text{O},in} \geq \Psi (3 N_{\text{C}_3\text{H}_8,in} + 4 N_{\text{C}_4\text{H}_{10},in}). \quad (1)$$

The gaseous components after the flash separation are combusted, and the energy balance is closed by burning hydrocarbons. The reactor, fuel cell and burner are assumed to be in thermal contact, and carbon monoxide and hydrocarbons are assumed to be consumed in reforming reactions in the SOFC. Table 2 summarizes the parameters used. Figure 3 shows the effect of recycling for different stoichiometric compositions of the reactor inlet as a function of the recycling ratio. Depending on value of the water factor Ψ , the optimum is observed at a recycling ratio of about 0.5 – 0.8. It should also be noted that while external recycling, if at all possible, can improve the process performance, there is a tradeoff of increase in the device size and complexity. Whether recycling should be pursued depends on the product specifications and objectives, e.g., the power output and mission duration and the requirements on process performance, as well as on advances in the technology, e.g., the catalysts used.

Table 2: Parameters for water reforming study in Figure 3.

Ambient temperature	$T_{amb} = 298\text{K}$
Power output	$PW = 1\text{W}$
Reactor temperature	$T_{op} = 1000\text{K}$
Reactor outlet temperature	$T_{out} = 1000\text{K}$
Conversion in reactor	$\zeta = 0.9$
SOFC temperature	$T_{op} = 1000\text{K}$
Residence time in reactor	$\tau = 10\text{ms}$
Discard temperature from SOFC	$T_{out} = 500\text{K}$
Conversion in burners	$\zeta = 0.95$
Residence time in burners	$\tau = 1\text{ms}$
Air excess in burners	$\Phi = 1.2$
Conversion in fuel cell	$\zeta = 0.8$
Overall heat loss coefficient	$U = 3\text{W}/\text{m}^2/\text{K}$
Residence time in fuel cell	$\tau = 20\text{ms}$
Emissivity (incl. view factor)	$\epsilon = 0.2$
Efficiency of fuel cell	$\eta_{FC} = 0.7$
Air excess in fuel cell	$\Phi = 1.2$
Compression parameter for air feed	$K_C = 10\text{J}/\text{mol}/\text{K}$
Burner temperature	$T_{op} = 1000\text{K}$
Discard temperature from burner	$T_{op} = 500\text{K}$
Water factor in fuel cell	$\Psi = 1$
Pump parameter	$K_P = 100\text{J}/\text{l}$
Propane molfraction in feed	0.5

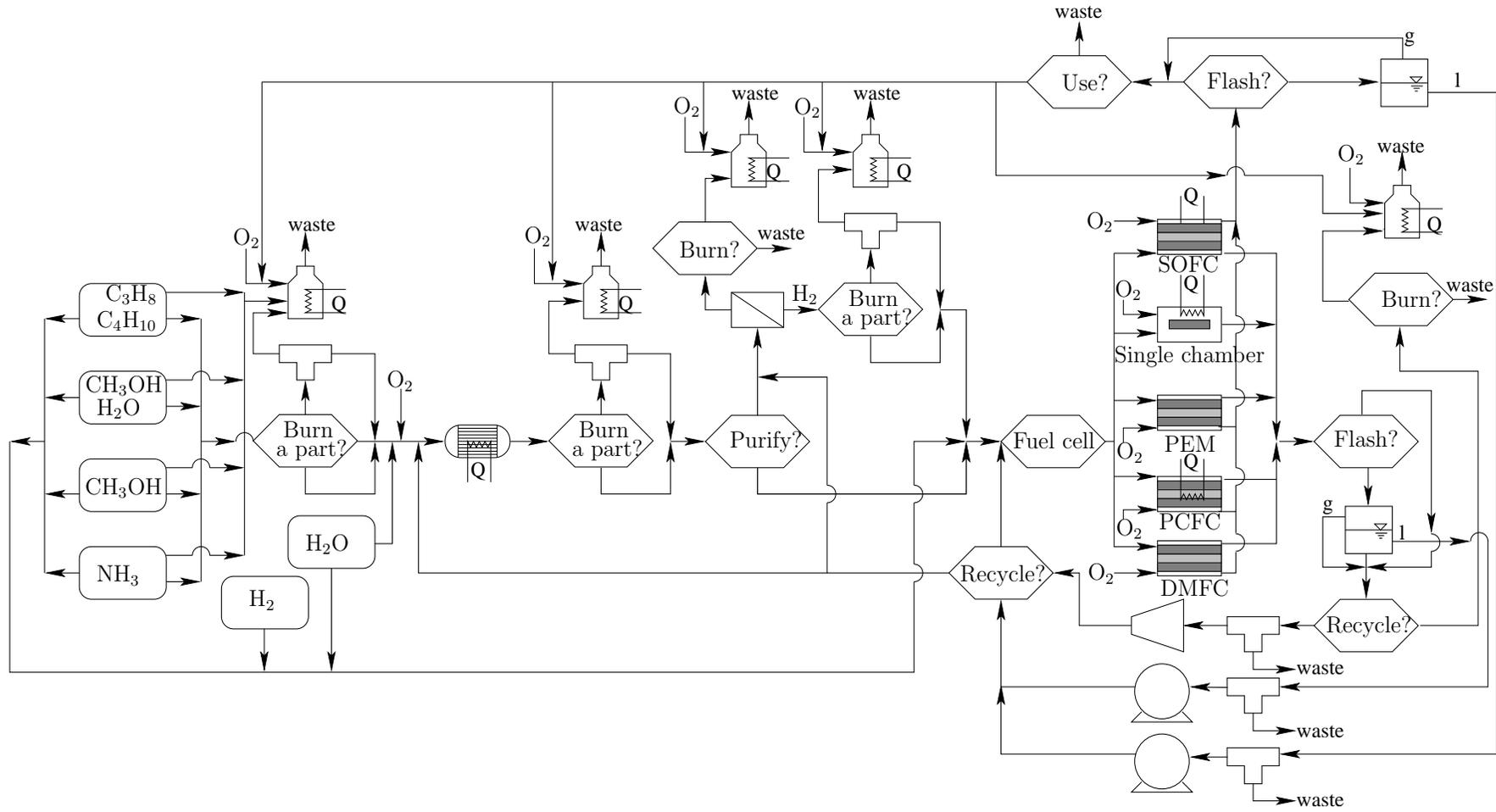
6 CONCLUSIONS AND FUTURE WORK

Portable power generation devices based on fuel cells have the potential of outperforming batteries for man-portable power generation by an order of magnitude in terms of energy density. There is a plethora of conceivable applications and processes and this results in the need for product engineering. We have presented a methodology for the comparison of the alternatives and investigation of the influence of technological parameters. We have implemented the methodology as a tool available in the form of a web-interface [Mitsos et al.(2004b)], which allows for facile use by remote users, who are unfamiliar with the modeling language. Upon request and subject to approval this web-interface can be made available for academic purposes.

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Figure 1: Conceptual process superstructure.



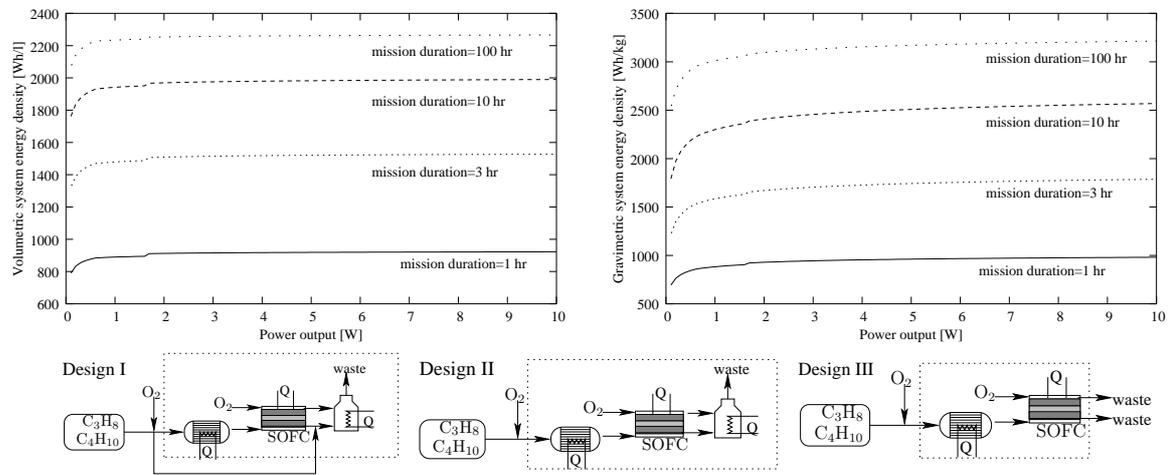


Figure 2: Volumetric and gravimetric system energy density of hydrocarbon partial oxidation in combination with a SOFC as a function of mission duration and power output.

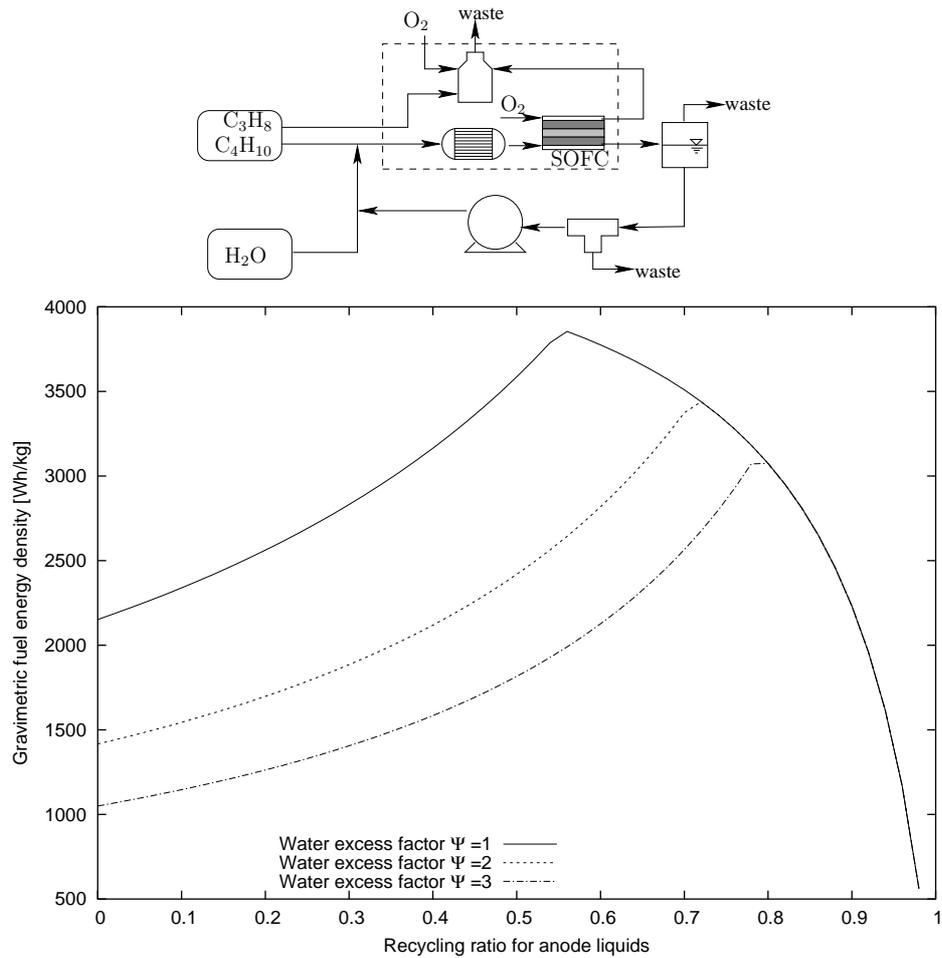


Figure 3: Effect of water recycling in the reforming reaction of hydrocarbons.

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