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OPTIMAL DESIGN AND OPERATION OF MICRO POWER GENERATION PROCESSES

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Abstract — This paper presents the optimal design and operation of a micro power generation process that consists of a reactor, a solid-oxide fuel cell (SOFC) and two burners in a stack fed with ammonia and butane fuels. Hydrogen is produced from ammonia decomposition, while butane is catalytically oxidized to produce heat and maintain the stack at a sufficiently high temperature. In the first part, a novel model is developed for predicting the steady-state performance of the process. Subsequently, this model is used as a basis to study and determine the optimal design and operation of the system. The optimization problem is formulated so that the consumption rate of utilities (ammonia and butane) is minimized, while meeting a specified power demand and maintaining the stack at its thermal equilibrium. The dependence of the optimal design and operating strategy on the operating temperature and the heat losses of the system is emphasized.

Key words: Man-portable power; micro power generation; optimal operation and design; dynamic optimization

1 Introduction

The widespread use of portable electric and electronic devices increases the need for efficient autonomous man-portable power supplies (up to 50 W). 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 1300 Wh/l and 700 Wh/kg and rechargeable up to 400 Wh/l and 300 Wh/kg and the upper limit on performance is now being reached. Out of the alternatives that are possible, we are focusing on power generation devices based on the electrochemical conversion of common fuels and chemicals in fuel cells. These micro processes have the potential to yield much higher energy densities than state-of-the-art batteries, because on one hand the above mentioned fuels have very high energy contents (up to 7000 Wh/l and 13000 Wh/kg), and on the other hand fuel cells can in principle achieve very high efficiencies.

In a previous work, Mitsos *et al.* [1] have proposed a methodology for the comparison of different alternatives for micro power generation processes based on a process superstructure, including hundreds of different designs, and identified the conditions under which the technologies considered are a promising alternative to batteries. Among those promising alternatives, we have selected a micro power generation process that consists of a reactor, a solid-oxide fuel cell (SOFC) and two burners in a stack fed with ammonia and butane fuels. Hydrogen is produced from ammonia decomposition, while butane is catalytically oxidized to produce heat and maintain the stack at a sufficiently high temperature. Note that although hydrocarbons such as propane or butane have higher theoretical energy densities than ammonia [1], propane/butane partial oxidation for hydrogen production has never been demonstrated thus far in microreactors, whereas ammonia decomposition has been successfully performed in microfabricated reactors with conversions exceeding 90% [2, 3]. These considerations therefore justify the choice of ammonia fuel in our initial study. Hydrogen generation from the partial oxidation of hydrocarbon fuels will be the topic of future work.

In this contribution, we report on the development of a model for predicting the steady-state performance of the micro power generation system. Subsequently, we use this model for determining the design and operating variables that optimize the process in terms of fuel consumption, while satisfying a given power demand. This work illustrates how operational considerations influence the optimal design of micro power generation processes, following the paradigm of interaction of design and operation.

2 Steady State Modeling and Simulation

The conceptual flowsheet of the process is represented Figure 1. During steady-state operation, the process operates as follows:

1. the catalytic decomposition of ammonia (NH_3) is first performed in the reactor to produce hydrogen (H_2);

2. the reactor effluent is then fed into the anode of the SOFC, whereas a first air stream is fed to the cathode, and electrical power is produced from the electrochemical reaction;

3. both the anode and cathode effluents are finally mixed and fed into the hydrogen burner,

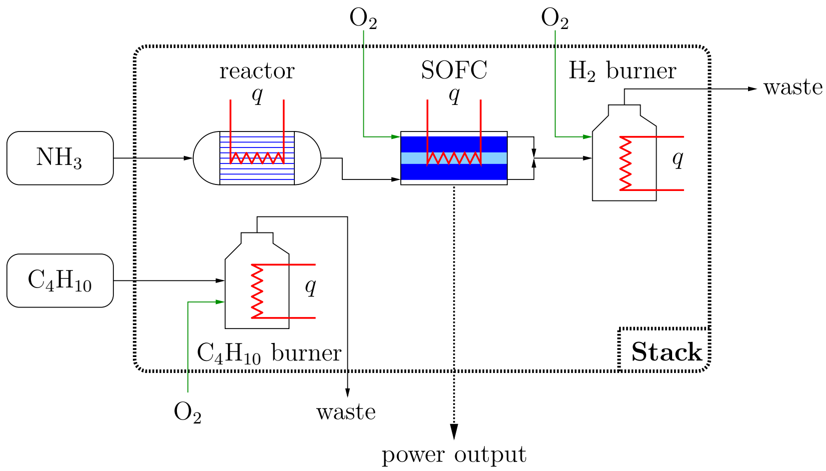

Figure 1: Conceptual process flowsheet.

along with a second air stream, for catalytic oxidation;

4. in parallel, a mixture of butane (C_4H_{10}) and air is fed into a second burner for catalytic oxidation to produce heat, thus maintaining the stack at a desired, sufficiently high temperature, despite the heat losses and the endothermicity of the ammonia decomposition reaction.

All four units are fabricated in a single silicon stack. It should be noted that silicon is an extremely good thermal conductor, so the temperature throughout the stack can be anticipated to be near uniform.

A one-dimensional model has been derived, which describes the steady-state behavior of the micro power generation device. It is based on the following major assumptions:

- **the gases behave ideally**;
- **the pressure P inside the stack is uniform** (atmospheric pressure): this assumption requires that the pressure drop along the gas channel remains relatively small;
- **the four units operate at a common temperature T**: this assumption requires that heat transfer is fast enough within the stack, as it is typically observed at the micro-scale for silicon based reactors and was also confirmed by finite element simulations; it allows one to not postulate a particular geometry for the unit operations and their arrangement in the stack;
- **the outlet temperature of the waste streams T^{out} is allowed to be different from the operating temperature T**: this assumption reflects the possibility for heat recovery between the inlet and outlet gas streams [4];
- **the cell voltage U is uniform throughout the electrodes**: this last assumption is generally satisfied in practice as the electrodes are normally good electrical conductors.

The steady-state model consists of three different sets of Differential Algebraic Equations (DAEs) corresponding to the mass-balances along the axial length of the reactor, the fuel cell, and the hydrogen burner (hybrid discrete/continuous system with fixed mode sequence and explicit transition conditions [5]). The mass-balances equations assume convective flow in the gas channels, neglecting axial diffusion. The chemical species considered are NH_3, H_2, N_2, H_2O and O_2. The kinetic rate for the ammonia decomposition reaction is given by the reduced kinetic expression derived in [6]. The electrochemical model accounts for ohmic losses across the solid-oxide electrolyte, as well as losses due to the anode and cathode activation overpotentials. The kinetic rates at the electrodes are taken from [7]. It is assumed that only H_2 is electrochemically oxidized in the SOFC and, subsequently, oxidized in the burner, while reaction of NH_3 is neglected in these units due to its generally low residual concentration. Also note that, unlike the reactor/fuel cell/hydrogen burner unit models, the butane burner is described by simple (mass and energy balance) algebraic relations based on fixed conversion and air excess number; this is motivated by the fact that butane catalytic oxidation is generally fast.

The resulting steady state simulation model has been implemented using the software package `DAEPACK` [8]. For the complete solution of the model, in addition to all the chemical/thermodynamical properties, geometry data and inlet compositions, one still needs to specify five degrees of freedom. This is done here by specifying the cell voltage U, as well as the ammonia inlet flow rate in the reactor $F^{\mathrm{r}}_{\mathrm{NH_3}}$, the air inlet flow rates in the fuel cell cathode $F^{\mathrm{ca}}_{\mathrm{air}}$ and in the hydrogen burner $F^{\mathrm{b_I}}_{\mathrm{air}}$, and the butane inlet flow rate in the second burner $F^{\mathrm{b_{II}}}_{\mathrm{C_4H_{10}}}$.

Figure 2 presents the characteristics curves for cell voltage and power versus intensity in the micro SOFC. They were obtained at three different operating temperatures $T = 1100\,\mathrm{K}$, $T = 1200\,\mathrm{K}$ and $T = 1300\,\mathrm{K}$, and are based on the following specifications for the micro power generation device[1]:

reactor:	$F^{\mathrm{r}}_{\mathrm{NH_3}} = 15\,\mathrm{sccm}^a$	length: $L^{\mathrm{r}} = 10\,\mathrm{mm}$	height: $H^{\mathrm{r}} = 0.48\,\mathrm{mm}$	width: $W^{\mathrm{r}} = 0.2\,\mathrm{mm}$
fuel cell:	$F^{\mathrm{ca}}_{\mathrm{air}} = 50\,\mathrm{sccm}$	length: $L^{\mathrm{fc}} = 10\,\mathrm{mm}$	height: $H^{\mathrm{fc}} = 0.48\,\mathrm{mm}$	width: $W^{\mathrm{fc}} = 10\,\mathrm{mm}$
H_2 burner:	$F^{\mathrm{b_I}}_{\mathrm{air}} = 0\,\mathrm{sccm}$	length: $L^{\mathrm{b_I}} = 10\,\mathrm{mm}$	height: $H^{\mathrm{b_I}} = 0.48\,\mathrm{mm}$	width: $W^{\mathrm{b_I}} = 0.2\,\mathrm{mm}$

[a]sccm: Standard Cubic Centimeters per Minute

One sees that the power output reaches a peak value of about 0.45 W, 0.85 W and 1.25 W for an operating temperature of 1100 K, 1200 K and 1300 K, respectively. The corresponding cell voltages are around 0.32 V, 0.37 V and 0.45 V, respectively. In all three situations, the activation losses are the most important factor governing the performance of the cell; the ohmic losses are also important at low temperature. Since the calculations have been performed based on the exchange current density expressions given in [7], the activation losses predicted by the model might however be overestimated; indeed, recent improvements in connection to the electrodes microstructure have resulted in reduced polarization losses, allowing better electrochemical performances [9].

Figure 3 presents the component mole fractions along the gas channels in the reactor, anode, cathode and hydrogen burner. The values correspond to an operating temperature of 1300 K and

[1]Note that there is no need to specify the butane flow rate $F^{\mathrm{b_{II}}}_{\mathrm{C_4H_{10}}}$ for the simulations since the operating temperature is fixed, *i.e.*, T is not calculated from energy balance considerations.

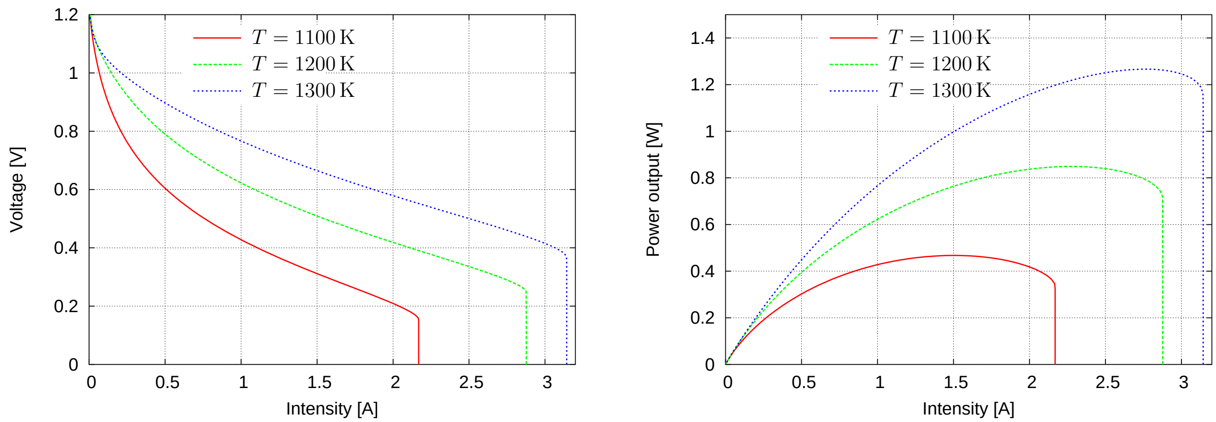

Figure 2: Cell voltage (left plot) and Power (right plot) vs. Intensity at different operating temperatures.

a cell voltage of 0.65 V. Based on the fuel cell characteristics presented above, one sees that these conditions correspond to an electrical power production close to 1 W. Based on the gas composition and velocity (not shown on Figure 3), performance factors can be easily calculated for each unit of the device. Here, it is found here that (i) NH_3 conversion in the reactor is greater than 98%, (ii) H_2 conversion in the SOFC is around 50%, with an electrochemical efficiency close to 69%, and (iii) H_2 combustion in the burner is around 90%. It immediately appears that the conversion of hydrogen is very low for the current process configuration, hence indicating that the process performance could be greatly improved under a more suitable choice of either design and operation parameters. These aspects are developed in the next section.

3 Optimal Operation and Design

Mathematical models are essential tools in the design of fuel cell systems since they provide a picture of the gas composition and velocities, potential and current density in the system for various process configurations and operating conditions. They can be used to examine the effects of change on one or more variables and the relative system sensitivity to relevant design parameters. In addition, they allow one to use this information to improve the system performance through the application of systematic optimization methods, as will be illustrated in the present section.

Optimization considerations in stationary, macro scale power generation processes generally consist of improving the system efficiency in terms of its power production, without paying much attention to the size of the system itself. In contrast, since micro power generation processes are intended to be used in man-portable applications, the focus is more on the dimensions/weight of the devices, while the power output of the system is dictated by the type of application, generally in the range 0.1 W – 10 W. Qualitatively, the optimization problem for optimal operation and design of micro power generation processes can therefore be stated as:

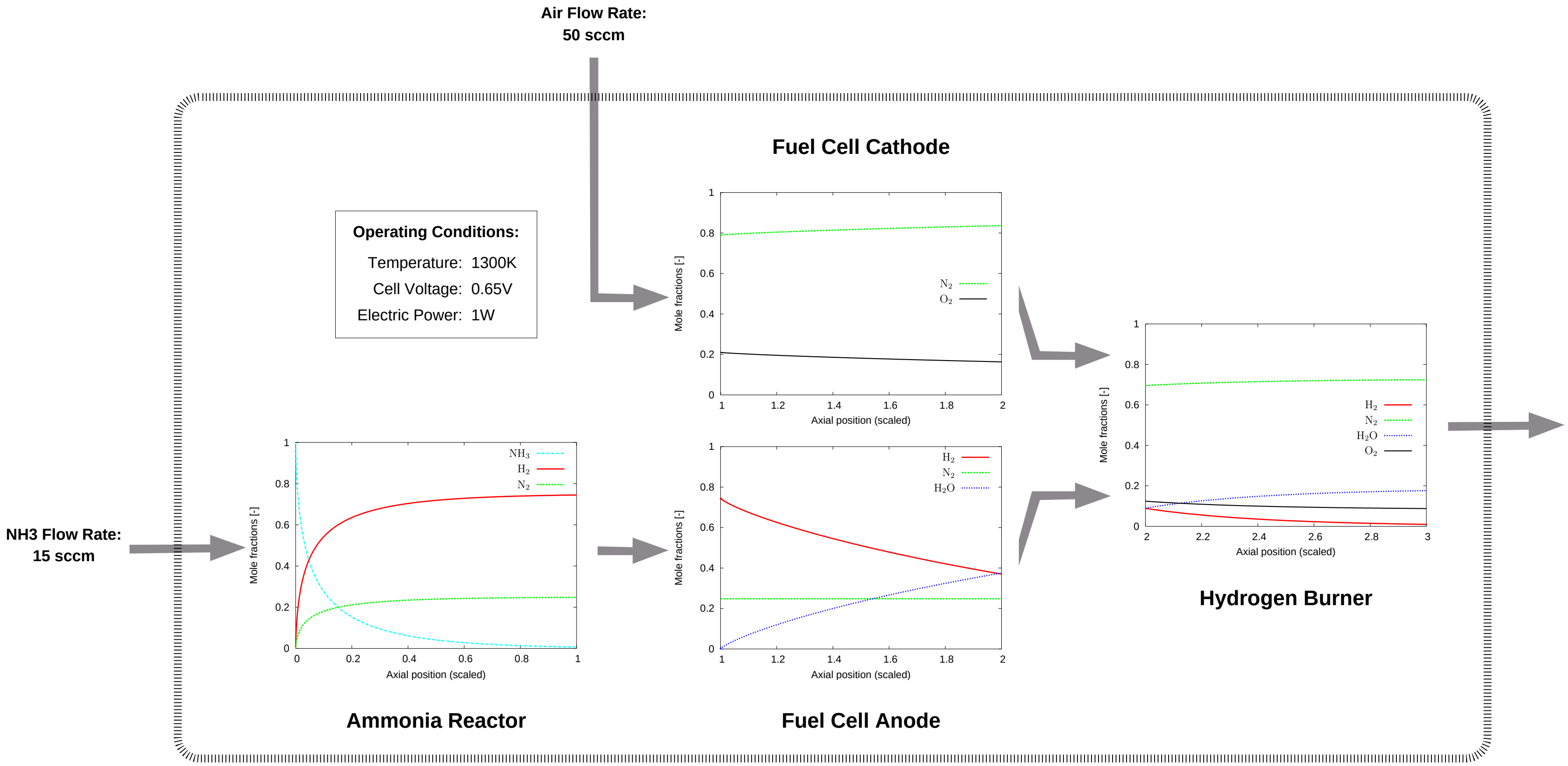

Figure 3: Gas composition along the stack at $T = 1300\,\mathrm{K}$ and $U = 0.65\,\mathrm{V}$.

> *"Find the optimal operational and design decision variables that minimize the quantity of fuels (ammonia and butane) needed to satisfy the nominal power demand and maintain the device at its thermal equilibrium."*

Mathematically, this problem gives rise to a challenging optimization problem with hybrid discrete/continuous DAEs embedded. The hybrid DAE model has been described previously in Section 2; the objective and constraint functions are detailed subsequently.

Objective Function – the function to be minimized corresponds to the instantaneous mass consumption of ammonia and butane fuels by the system; note in particular that the mass of the device itself is not considered here, which is a valid assumption for long mission durations.

Nominal Power Generation Constraint – the amount of power $\mathscr{P}^{\mathrm{fc}}$ generated by the micro SOFC must meet the specified nominal power demand $\mathscr{P}^{\mathrm{nom}}$, *e.g.*, 1 W.

Thermal Equilibrium Constraint – at steady state, the total heat load Q^{tot} in the device must be equal to zero for the system to be in thermal equilibrium (closed energy balance). For the micro power generation process shown in Figure 1, Q^{tot} can be calculated as the sum of the following contributions:

$$Q^{\mathrm{tot}} = \left(\dot{H}^{\mathrm{r}}_{\mathrm{in}} + \dot{H}^{\mathrm{ca}}_{\mathrm{in}} + \dot{H}^{\mathrm{b_I}}_{\mathrm{in}} - \dot{H}^{\mathrm{b_I}}_{\mathrm{out}}\right) - \mathscr{P}^{\mathrm{fc}} - Q^{\mathrm{loss}} - Q^{\mathrm{b_{II}}}$$

where $\dot{H}^{\mathrm{r}}_{\mathrm{in}}$, $\dot{H}^{\mathrm{ca}}_{\mathrm{in}}$, $\dot{H}^{\mathrm{b_I}}_{\mathrm{in}}$ and $\dot{H}^{\mathrm{b_I}}_{\mathrm{out}}$ are the enthalpy streams in the reactor inlet, in the fuel cell cathode inlet, in the hydrogen burner inlet, and in the hydrogen burner outlet streams, respectively; Q^{loss} denotes the overall heat losses to the environment, which accounts for conductive as well as radiative losses; and $Q^{\mathrm{b_{II}}}$ is the heat production term from butane combustion, calculated by assuming a fixed conversion and air excess number for the burner.

The optimization parameters for the system are a mixed set of (*i*) **design decision variables**: lengths of the reactor, fuel cell and hydrogen burner gas channels L^{r}, L^{fc} and $L^{\mathrm{b_I}}$, respectively; and (*ii*) **operational decision variables**: temperature T, cell voltage U, and reactor, cathode, hydrogen burner and butane burner feed flow rates $F^{\mathrm{r}}_{\mathrm{NH_3}}$, $F^{\mathrm{ca}}_{\mathrm{air}}$, $F^{\mathrm{b_I}}_{\mathrm{air}}$ and $F^{\mathrm{b_{II}}}_{\mathrm{C_4H_{10}}}$, respectively.

The described hybrid optimization problem has been solved using the SQP solver `NPSOL` [10]. We have used the software `DAEPACK` for consistent initialization and integration of the DAE model, as well as for the calculation of the first-order state sensitivities (integrator `DSL48S` and nonlinear solver `BLOCKSOLVE`). Furthermore, all the necessary differentiations (DAE model, objective and constraint functions) have been generated by using the automatic differentiation capabilities of `DAEPACK`.

The optimal operation and design results obtained for the micro power generation device shown in Figure 1 are given in Table 1. The nominal power demand was set to $\mathscr{P}^{\mathrm{nom}} = 1\,\mathrm{W}$ and the geometry parameters (other than the channel lengths) were given the same values as in the simulations described in Section 2.

One sees that the optimal temperature minimizing the consumption of ammonia and butane fuel is high, around 1500 K; operating the system at such a high temperature is however unrealistic

Table 1: Optimal operation and design results.

Design decision variables		Operation decision variables	
$L^{\mathrm{r}} =$	$4.06\,\mathrm{mm}$	$T =$	$1499\,\mathrm{K}$
$L^{\mathrm{fc}} =$	$3.11\,\mathrm{mm}$	$U =$	$0.503\,\mathrm{V}$
$L^{\mathrm{b_I}} =$	$3.26\,\mathrm{mm}$	$F^{\mathrm{r}}_{\mathrm{NH_3}} =$	$10.91\,\mathrm{sccm}$
		$F^{\mathrm{ca}}_{\mathrm{air}} =$	$48.83\,\mathrm{sccm}$
		$F^{\mathrm{b_I}}_{\mathrm{air}} =$	$0\,\mathrm{sccm}$
		$F^{\mathrm{b_{II}}}_{\mathrm{C_4H_{10}}} =$	$1.78\,\mathrm{sccm}$
Objective function: $0.212\,\mathrm{mg{\cdot}s^{-1}}$			

from a practical point of view, mainly because of material constraints. Rather, we remove the temperature from the list of decision variables, and conduct a parametric study by varying the operating temperature in the range [1000 K, 1300 K]. The results are presented in Figure 4. The optimal utilities consumption rate is displayed *vs.* temperature on the left plot, and the optimal design parameters (gas channel lengths) are displayed *vs.* temperature on the right plot. As temperature increases, so do the reaction rates in the reactor, in the fuel cell and in the burner; in other words, increasing the operating temperature allows one to obtain the same conversions while significantly reducing the size of the units. On the other hand however, the heat losses per unit area are substantially increased at high temperature[2]. Accordingly, operating the system at a higher temperature is only beneficial if the additional heat losses can be compensated by the reduction of the device size.

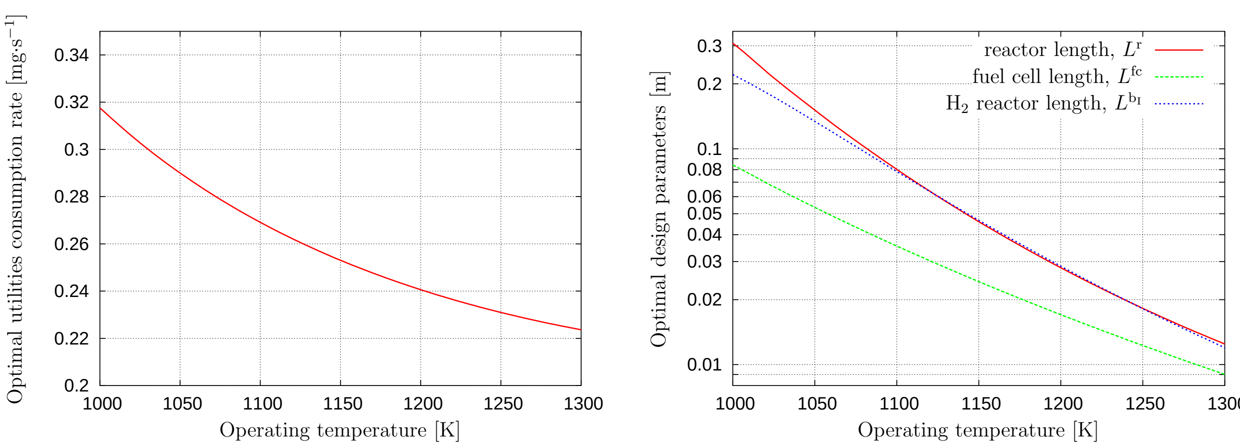

Figure 4: Optimal utilities consumption rate (left plot) and optimal design parameters (right plot) *vs.* operating temperature.

The previous considerations also illustrate that heat losses are one of the key issues in the development of micro power generation processes [1]. A simplified estimation of heat losses has been

[2]This is true in particular for the radiative heat losses which are proportional to T^4.

implemented thus far in the model, which accounts for both conductive/convective and radiative heat losses by defining an overall heat transfer coefficient U^{loss} and an overall emissivity coefficient ε (including the view factor) as:

$$Q^{\text{loss}} \quad = \quad A\left[U^{\text{loss}}\left(T - T_{\text{amb}}\right) + \varepsilon\sigma_{SB}\left(T^4 - T_{\text{amb}}^4\right)\right] \tag{1}$$

where A denotes the equivalent surface (A is calculated by assuming a fixed aspect ratio of the device as $A = 6V^{\frac{2}{3}}$, with V corresponding to the necessary volume for the gas in the stack). However, several micro devices and components of the proposed process are not yet fully developed, and the aforementioned overall heat transfer and emissivity coefficients are not accurately known. A parametric study has therefore been conducted in Figure 5, which shows the effects of changing those parameters on the optimal design and operation of the system. Because of the high operating temperatures, the influence of radiative heat losses (right plot) is more important than conductive/convective heat losses (left plot). Also note that the effect of radiative heat losses is dramatic as the utilities consumption rate is increased by up to 300% when varying the emissivity coefficient ε from 0.1 to 0.9.

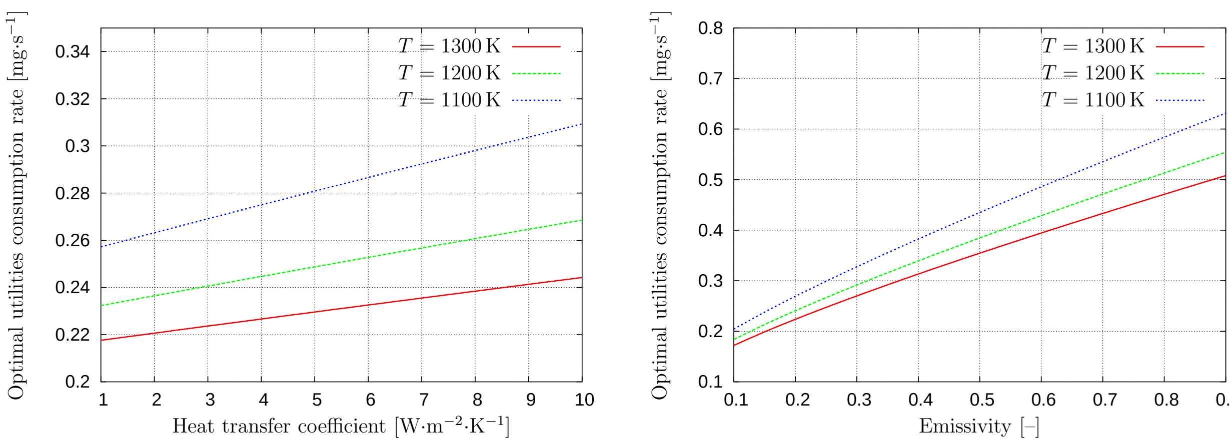

Figure 5: Optimal utilities consumption rate *vs.* overall heat transfer coefficient (left plot) and overall emissivity coefficient (right plot).

4 Conclusion

In this paper, a novel model has been presented for predicting the steady-state performances of a micro power generation process that consists of a reactor, a SOFC and two burners in a stack. Hydrogen is produced from ammonia decomposition, while butane is catalytically oxidized in a separate burner to produce heat and maintain the stack at a sufficiently high temperature.

In the second part of the paper, the developed steady state model has been used as a basis to study and determine the optimal design and operation of the system. The optimization problem is formulated so that the consumption rate of utilities (ammonia and butane) is minimized, while meeting a specified power demand and maintaining the stack in thermic equilibrium. This optimization problem with hybrid discrete/continuous DAEs embedded has been solved using `NPSOL`, in conjunction with the software `DAEPACK`. The dependence of the optimal design and operation on the operating temperature of the system has been studied, as well as the dependence on the heat losses. In particular, these results illustrate that radiative heat losses play a central role in the operation and design of micro power generation processes.

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