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Finite time analysis of an endoreversible fuel cell

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Abstract

The aim of this paper consists in a detailed thermodynamical description of a fuel cell, using finite time thermodynamics (FTT). Starting from the comparison beetween a reversible fuel cell and a Carnot heat engine driven by a perfect chemical reaction, we remind that – contrary to a common opinion – both systems have the same thermodynamical performances. Thereby, we evolve the comparison beetween these two systems to the area of finite time thermodynamics. The main results is the definition of an endoreversible fuel cell characterized by a maximum-power efficiency.

Keywords : Fuel cell ; Heat engine ; Efficiency ; Finite time thermodynamics ; Entropy ; Optimization.

1 Introduction

The fuel cell is an electrochemical device that generates electrical power by a direct conversion of chemical energy [1]. It is one of the most promising low pollution power source and provides an alternative to common systems based on fossil fuels. This system is also well known to be very efficient from a thermodynamical point of view : assuming that its performances are not limited by Carnot's efficiency. It is usual to compare two perfect systems in order to demonstrate it : the reversible fuel cell (RFC) and the Carnot Heat Engine (CHE) driven by a reversible combustion system.

In 1959, Liebhafsky [2] argued to the superiority of RFC on CHE performances, considering that the former directly convert into work the consumed chemical energy, contrary to heat engine that previously convert it into heat. Cowden *et al.* [3] used the same type of arguments to explain the absence of Carnot's thermodynamical efficiency limitation for fuel cells, *i. e.* the fundamental difference beetween an electrochemical dissociation and a common combustion process. These works concluded to the non-subservience of the RFC to the Carnot limitation and therefore to its superiority on the CHE. We will demonstrate that this conclusion is a misinterpretation of fundamental principles enacted by Carnot.

Actually, Carnot explained in 1824 [4] that the efficiency of a thermal engine operating beetween two heat reservoirs at different temperatures is always less than the one of a perfect engine (the CHE) running in the same conditions. It leads to the famous Carnot's efficiency limitiation, that must be applied only to thermal engines, *i. e.* to systems operating beetween two different temperature heat sources. Fuel cell could be considered as an isothermal system and operating in a completely different manner than a thermal engine. Hence, Carnot's efficiency do not have to be used to quantify performances of a fuel cell, reversible or not [5, 6]. This alone conclusion can not allow us to conclude on a superiority of one of our two systems on the other.

However, the non-validity of the Carnot efficiency to analyse fuel cell performances do not prevent us to compare it with heat engine. Appleby and Foulkes [7] and later Lutz *et al.* [8] proposed the first thermodynamical comparison beetween RFC and CHE. They conclude on the equivalence of both systems in considering a combustion reaction as the source of the high temperature reservoir. Hence, they showed that a RFC have the same thermodynamical performances as a CHE operating beetween two temperatures scales. The higher one is calculated from the equivalent combustion process (and named "combustion temperature", noted T^*) and the

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lower one is the operating temperature of the fuel cell, noted T. Then, thermodynamical efficiency η of a RFC could be written in the same form as one of a CHE :

$$\eta = 1 - T/T^* \tag{1}$$

with T the temperature of the fuel cell and T^* the combustion temperature, both will be detailed and redefined later in this work. Wright [5] used an exergy analysis to obtain same conclusions and formulated the efficiency of a fuel cell with an exergy flow. At last, Ro and Sohn [9] obtained similar results and proposed an hybridization beetween RFC and CHE.

Here, we propose to evolve comparison beetween CHE and RFC in describing both with the finite time thermodynamics (FTT) [10]. Consequently, we propose to consider an endoreversible fuel cell (ERFC), *i. e.* operating reversibly in exchanging heat flux irreversibly with its surrounding [11]. The entropy production due to heat flux exchanges with ambiance is represented by a finite thermal conductance. Then, according to the Carnot's principle, electrical power produced depends on heat flux rejected to surroundings and consequently to operating temperature of the fuel cell. Finally, performances of the ERFC are expressed and found to be stronlgy influenced by difference beetween operating temperature and the ambiance one.

2 Energetic and entropic balance

Let us consider an open and steady state thermodynamical system operating at constant temperature T and constant pressure p. It exchanges molar flow rates $\dot{N}_i$, work transfer rate $\dot{W}$ and heat flux $\dot{Q}$ with surrounding. This system, represented by the diagram of Fig. 1., could be any of our two power conversion devices (RFC or CHE driven by a chemical reaction). Indeed, both of them could run in consuming fuel and producing usefull power and heat flux. Likewise, in our system occurs an exothermic chemical reaction described by the followed relation :

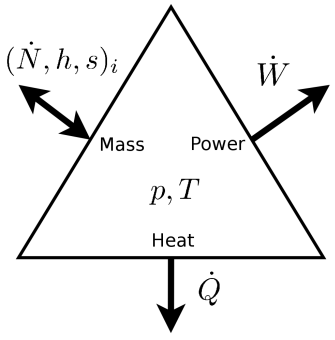

Fig. 1. Schematic diagram of the considered open system.

$$\sum_{j\in\mathcal{R}} \nu_j \cdot A_j \rightarrow \sum_{k\in\mathcal{P}} \nu_k \cdot A_k \tag{2}$$

with A_i the chemical species and ν_i their related stoichiometric coefficients, for $\mathcal{R}$ the group of reactants and $\mathcal{P}$ the group of products of reaction (2). As the whole system, previous reaction occurs coherently at temperature T and pressure p. Moreover, we suppose that both reactants and products are perfect gases. Accounting for the first law of thermodynamics leads us to the energy balance of the system :

$$\sum_{j\in\mathcal{R}} \dot{N}_j \cdot h_j(T) = \dot{W} + \dot{Q} + \sum_{k\in\mathcal{P}} \dot{N}_k \cdot h_k(T) \tag{3}$$

with h_i molar enthalpies of reactants and products, both considered at the system temperature T. In combining relations (2) and (3), the provided power could be written :

$$\dot{W} = -\Delta_{\mathrm{r}}\dot{H}(T) - \dot{Q} \tag{4}$$

with $\Delta_{\mathrm{r}}\dot{H} < 0$ the change in enthalpy flux across the chemical reaction (2). Considering the reaction progress coordinate ξ defined by [14] :

$$\mathrm{d}\xi = \left(-\frac{\mathrm{d}N_j}{\nu_j}\right)_{j\in\mathcal{R}} = \left(\frac{\mathrm{d}N_k}{\nu_k}\right)_{k\in\mathcal{P}} \tag{5}$$

previous chemical power could be expressed as :

$$\Delta_{\mathrm{r}}\dot{H}(T) = \sum_{k\in\mathcal{P}} \dot{N}_k \cdot h_k(T) - \sum_{j\in\mathcal{R}} \dot{N}_j \cdot h_j(T) = \left(\sum_{k\in\mathcal{P}} \nu_k \cdot h_k(T) - \sum_{j\in\mathcal{R}} \nu_j \cdot h_j(T)\right) \cdot \frac{\mathrm{d}\xi}{\mathrm{d}t} = \Delta_{\mathrm{r}}h(T) \cdot \dot{\xi} \tag{6}$$

with $\Delta_{\mathrm{r}}h$ the change in molar enthalpy through the chemical reaction and $\dot{\xi} = \mathrm{d}\xi/\mathrm{d}t$ the reaction rate representing the derivative of reaction progress regarding to time t. Entropy balance of the system is obtained by a similar usage of the second principle of thermodynamics :

$$\sum_{j\in\mathcal{R}} \dot{N}_j \cdot s_j(p_j, T) + \dot{\Theta} = \frac{\dot{Q}}{T} + \sum_{k\in\mathcal{P}} \dot{N}_k \cdot s_k(p_k, T) \tag{7}$$

s_i are molar entropies of reactants and products, depending on partial pressure p_i and temperature T. The rate of entropy production inside the system, noted $\dot{\Theta} = \mathrm{d}_{\mathrm{i}}S/\mathrm{d}t$ [15] represents its own internal irreversibilities. Considering both conversion devices as reversibles ($\dot{\Theta} = 0$), previous relation allow us to formulate a new expression of the exchanged heat flux :

$$\dot{Q} = -T \cdot \Delta_{\mathrm{r}}\dot{S}(\mathbf{p}, T) = -T \cdot \Delta_{\mathrm{r}}s(\mathbf{p}, T) \cdot \dot{\xi} \tag{8}$$

with $\Delta_{\mathrm{r}}s$ the change in molar entropy across reaction (2) and $\mathbf{p} = \{p_i\}$ the vector of partial pressures of both reactants and products. Net rate of work supplied by the system could be expressed in a new form by combination of relations (4) and (8) :

$$\dot{W} = -\Delta_{\mathrm{r}}\dot{G}(\mathbf{p}, T) = -\Delta_{\mathrm{r}}g(\mathbf{p}, T) \cdot \dot{\xi} \tag{9}$$

with $\Delta_{\mathrm{r}}g$ the change in molar Gibbs energy due to chemical reaction. Thus, we obtain the well known expressions of molar work supplied by a RFC [1, 7] :

$$\mathrm{w} = -\Delta_{\mathrm{r}}g(\mathbf{p}, T) \tag{10}$$

Considering the change in enthalpy flux $\Delta_{\mathrm{r}}\dot{H}$ as chemical power provided to the system, we can write the thermodynamical efficiency as [1, 7] :

$$\eta_{\mathrm{RFC}} = \frac{\dot{W}}{\dot{Q}} = \frac{-\Delta_{\mathrm{r}}g(\mathbf{p}, T)}{-\Delta_{\mathrm{r}}h(T)} \tag{11}$$

that is the famous form of a perfect fuel cell efficiency.
Previous expression is in fact related to any steady state open system containing exothermic, isothermal and isobaric chemical reactions like (2), and producing heat power and usefull rate of work. As we have seen, the link beetween efficiency (11) and RFC is obvious but need to be explained a little bit more in the case of a thermal engine. For this, let us consider our previous system presented on Fig. 1. but now divided into two subsystems named respectively (I) and (II), as presented on Fig. 2. Like the first one, the whole system (I ∪ II) is supposed to be reversible, *i. e.* (I) and (II) are reversibles and no entropy is produced by the heat power $\dot{Q}_{\mathrm{h}}$ transfered from (I) to (II).

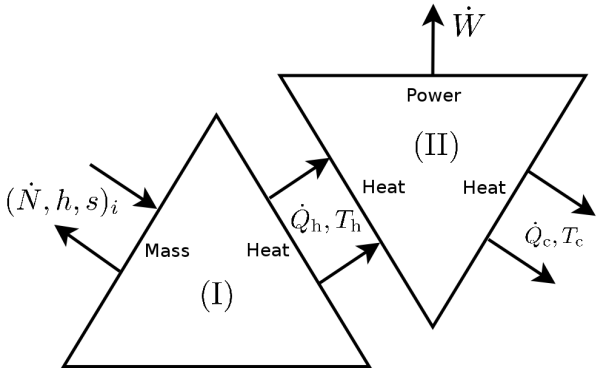

Fig. 2. A schematic diagram of a thermal engine.

Subsystem (I) is similar as first one, but produce only heat power $\dot{Q}_{\mathrm{h}}$ and no rate of work $\dot{W}$. In fact, it could represent a combustion system based on the chemical reaction (2). Applying an energy balance as (3) to (I), we obtain :

$$\dot{Q}_{\mathrm{h}} = -\Delta_{\mathrm{r}}\dot{H}(T) \tag{12}$$

subsystem (I) operating reversibly at temperature T_{h}, its balance of entropy gives [5] :

$$\dot{Q}_{\mathrm{h}}/T_{\mathrm{h}} = -\Delta_{\mathrm{r}}\dot{S}(\mathbf{p}, T) \tag{13}$$

with the entropic temperature [16] obtained from combination of two previous relations :

$$T_{\rm h} = T^*(T) = \frac{\dot{Q}_{\rm h}}{-\Delta_{\rm r}\dot{S}(\mathbf{p},T)} = \frac{\Delta_{\rm r}h(T)}{\Delta_{\rm r}s(\mathbf{p},T)} \tag{14}$$

that and is explicitely function of the operating temperature T. We can note that some authors [5, 7–9] have defined combustion temperature differently, e. g. Lutz *et al.* [8] with the followed definition :

$$\Delta_{\rm r}g(\mathbf{p},T^\diamond) = 0 \Rightarrow T^\diamond = \frac{\Delta_{\rm r}h(T^\diamond)}{\Delta_{\rm r}s(\mathbf{p},T^\diamond)} \tag{15}$$

that corresponds to the exact definition of a combustion temperature but suppose tacitly that reactants and products are both draged and rejected at temperature $T^\diamond$. Contrary to this hypothesis, considering that chemical reactants/products are draged/rejected at the temperature T of the system leads to the use of first definition (14). Later, we will prefer to called T^* entropic temperature, to make much of the difference with combustion temperature $T^\diamond$.

The second subsystem (II) of Fig. 2 consumes the heat flux $\dot{Q}_{\rm h}$, convert it partially into power $\dot{W}$ and reject the remained heat flux $\dot{Q}_{\rm c}$ to a cold reservoir at the temperature $T_{\rm c}$: it is heat engine. Running reversibly, it is in fact a Carnot heat engine (CHE) operating beetween constant temperatures $T_{\rm h} = T^*$ and $T_{\rm c} = T$. Therefore, its provided rate of work could be written as :

$$\dot{W} = \dot{Q}_{\rm h} - \dot{Q}_{\rm c} = \dot{Q}_{\rm h} \cdot \eta_{\rm CHE} \tag{16}$$

with the Carnot efficiency :

$$\eta_{\rm CHE} = 1 - \frac{T_{\rm c}}{T_{\rm h}} \tag{17}$$

combining with expression (14) of entropic temperature, we obtain efficiency of the whole system :

$$\eta_{\rm I\cup II} = 1 - \frac{T}{T^*} = 1 - \frac{T\cdot\Delta_{\rm r}s(\mathbf{p},T)}{\Delta_{\rm r}h(T)} = \frac{\Delta_{\rm r}g(\mathbf{p},T)}{\Delta_{\rm r}h(T)} \tag{18}$$

that is identical to the efficiency (11) of the first reversible system presented on Fig. 1. and consequently of the RFC [5] :

$$\eta_{\rm RFC}(T) = \eta_{\rm CHE}(T,T^*) = 1 - \frac{T}{T^*} \tag{19}$$

Considering the provided rate of work, we obtain :

$$\dot{W} = \dot{Q}_{\rm h}\cdot\eta_{\rm CHE} = -\Delta_{\rm r}\dot{H}(T)\cdot\left(1 - \frac{T\cdot\Delta_{\rm r}s(\mathbf{p},T)}{\Delta_{\rm r}h(T)}\right) = -\Delta_{\rm r}\dot{G}(\mathbf{p},T) \tag{20}$$

that is identical to relation (9). In conclusion, an RFC running at the temperature T and a CHE operating (with the help of a reversible combustion device) beetween temperatures T and T^* (defined by relation (14)) have identical thermodynamical performances. Therefore, we can remark that efficiencies of both CHE and RFC are maximum when cold temperature is equal to the surrounding one. We will see later that this situation is physically incongruous if our conversion device have to exchange heat fluxes by finite areas.

3 Endoreversible FC and FTT

Previously, different power conversion devices (presented in Fig. 1 and Fig. 2.) exchanged heat fluxes reversibly with surrounding. It means that heat exchanges occurs through infinite size areas or during infinite periods [10, 17]. To realize it, we can consider the followed Fourier's law of thermal conduction :

$$\mathrm{d}Q = -\lambda\cdot\nabla T\cdot\mathrm{d}\mathcal{S}\cdot\mathrm{d}t \tag{21}$$

with Q the thermal energy and λ an equivalent thermal conductivity. In previous relation, we can easily conclude that a non-zero heat quantity ($\mathrm{d}Q \neq 0$) can be transfered in an isothermal phenomena ($\nabla T \rightarrow 0$) only accross an infinite area ($\mathrm{d}\mathcal{S} \rightarrow \infty$) and/or during an infinite period ($\mathrm{d}t \rightarrow \infty$).

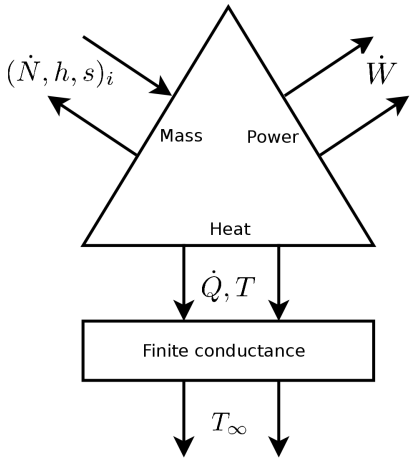

Fig. 3. Schematic diagram of an endoreversible fuel cell.

The aim of finite time thermodynamics (FTT) is to analyse perfect (or real) systems exchanging heat flux irreversibly, *i. e.* across finite size devices or during finite time lengths. Here, we consider once again the first open and reversible system (Fig. 1.), but hence exchanging irreversibly heat flux $\dot{Q}$ with surrounding. This type of system (presented on Fig. 3.) is called endoreversible, *i. e.* reversible but exchanging heat irreversibly [11]. Entropy production due to heat transfer is represented by the finite conductance $K\,[\mathrm{W}\cdot\mathrm{K}^{-1}]$:

$$\dot{Q} = K \cdot (T - T_\infty) \tag{22}$$

Practically, K could be considered as effect of the system's external surface or of its cooling device (heat exchangers, fans, etc.). These conductance modify expression (4) of the provided rate of work :

$$\dot{W} = -\Delta_\mathrm{r}\dot{H}(T) - K \cdot (T - T_\infty) \tag{23}$$

and of the entropy flux created by chemical reaction :

$$\Delta_\mathrm{r}\dot{S}(\mathbf{p}, T) = -K \cdot \left(1 - \frac{T_\infty}{T}\right) \tag{24}$$

introducing entropic temperature T^* in combination of two previous relations, we obtain :

$$\dot{W} = -T^* \cdot \Delta_\mathrm{r}\dot{S}(\mathbf{p}, T) - K \cdot (T - T_\infty) = K \cdot \left(1 - \frac{T_\infty}{T}\right) \cdot (T^* - T) \tag{25}$$

that is a function of running temperature T. The first remark we can do is that the rate of $\dot{W}$ is equal to zero for two different values of temperature T :

1. If $T = T_\infty$, the system operates as the same temperature as surrounding and no heat flux can be exchanged irreversibly with ambiance ($\dot{Q} = 0$). In accordance with the second principle of thermodynamics, no rate of work can be produced. Thermodynamical efficiency η is equal to the Carnot's one (1).

   Here is an important consequence of the finite time point of view : a reversible system operating in exchanging heat flux trough finite size devices can not produce any usefull power if its efficiency is equal to the Carnot's one. This conclusion is also available in the case of an ERFC.

2. If $T = T^*$, rate of work and thermodynamical efficiency are both equal to zero : no difference in Gibbs energy can be produced by chemical process and all the consumed chemical energy is converted into heat.

Beetween theses two extremes cases, provided rate of work $\dot{W}$ is positive and function of operating temperature T. We can look for the value of T that corresponds to a maximum (optimum) value of produced power, hence :

$$T_\mathrm{opt} = \arg\max_T \left\{\dot{W}(T)\right\} \tag{26}$$

that leads us to the followed nonlinear equation :

$$\frac{\partial \dot{W}}{\partial T} = 0 \Rightarrow T_\mathrm{opt} = \sqrt{T_\infty \cdot T^*(T_\mathrm{opt})} \tag{27}$$

Previous result was obtained in first on thermal engines by Chambadal (and independently by Novikov) in 1957 [12, 13], and later by Curzon and Ahlborn [18]. These optimal value of running temperature leads us to the correspondant efficiency :

$$\eta_{\mathrm{ERFC}} = 1 - \frac{T_{\mathrm{opt}}}{T^*(T_{\mathrm{opt}})} = 1 - \sqrt{\frac{T_\infty}{T^*(T_{\mathrm{opt}})}} \tag{28}$$

With $T^*(T_{\mathrm{opt}})$ the optimal entropic temperature, *i. e.* entropic temperature corresponding to the optimal operating one. The most oustanding result of previous calculation is that the maximum-power efficiency η_{CN} is independent from value of thermal conductance K. It is a fundamental result of FTT analysis when applied to power conversion devices [10]. Maximum power provided by our system gets :

$$\dot{W}_{\max} = \dot{W}(T_{\mathrm{opt}}) = K \cdot \left(\sqrt{T^*(T_{\mathrm{opt}})} - \sqrt{T_\infty} \right)^2 \tag{29}$$

that depends explicitely on thermal conductance K. Consequently, $\dot{W}$ could be practically increased with conductance K. Now, we can apply previous relations to the case of a reversible hydrogen fuel cell.

4 Application hydrogen RFC

As a practical example of previous results, we can consider the case of an RFC operating in consuming hydrogen as fuel. Chemical reaction (2) gets :

$$\mathsf{H}_2 + 1/2 \cdot \mathsf{O}_2 \rightarrow \mathsf{H}_2\mathsf{O} \tag{30}$$

Considering this reaction in standard conditions and producing water vapor only, evolutions of entropic temperature T^* (definition (14)) and thermodymical efficiency η (relation (1)) regarding to operating temperature T are presented on Fig. 1. In accordance with classical form of the RFC thermodynamical efficiency, both are decreasing with temperature T. The reduced provided rate of work $\dot{W}(T)/\dot{W}_{\max}$ is drawn on Fig. 5, regarding to temperature T. As explained previously, this power is null for $T = T_\infty$ and $T = T^*_{\max}$ and maximum for $T_{\mathrm{opt}} \simeq 1\,012\,\mathrm{K}$. Here, $T^*_{\max} \simeq 4\,178\,\mathrm{K}$ is the maximum value of entropic temperature. The usefull curve $\dot{W}/\dot{W}_{\max} = f(\eta)$ is drawn on Fig. 6. Once again, we can see that power $\dot{W}$ is actually null for maximum efficiency ($T = T_\infty$) and have a maximum value for $T = T_{\mathrm{opt}}$ and $\eta_{\mathrm{ERFC}} \simeq 77,4\%$. This curve is also usefull to make the difference beetween low and high-temperature fuel cells. A low-temperature fuel cell is characterized by an high value of its thermodynamical efficiency. However, its weak temperature difference with surrounding prevent to reject important heat flux $\dot{Q}$, and according to Carnot principle, to produce important rate of work $\dot{W}$. On the contrary, high-temperature fuel cells can easily evacuate generated thermal power, because of high temperature differences with ambiance, and are able to produce high values of electrical power.

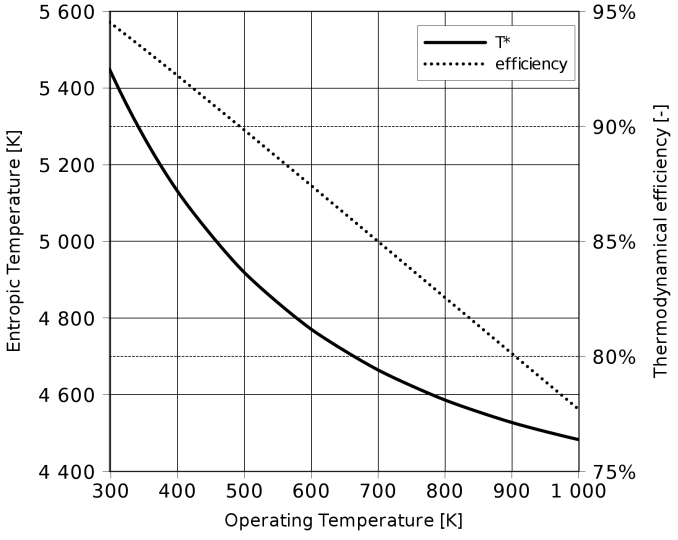

Fig. 4. Entropic temperature T^* and efficiency η.

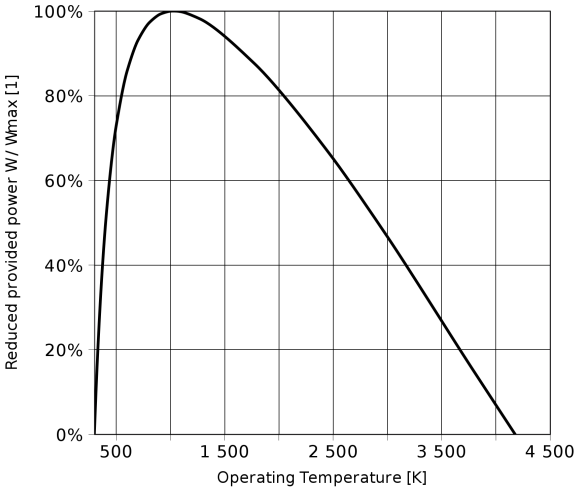

Fig. 5. Reduced provided rate of work $\dot{W}(T)$.

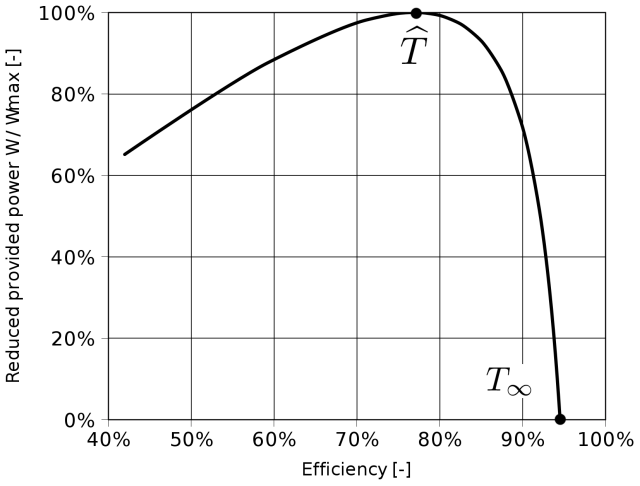

Fig. 6. Reduced provided power regarding to efficiency.

5 Conclusions

Considering comparison and equivalence beetween a reversible fuel cell and a Carnot engine (driven a reversible combustion process) allowed us to descibe the former with the finite time thermodynamics (FTT) approach. The main results was the definition of an endoreversible fuel cell, operating reversibly but exchanging heat irreversibly with its surrounding, through finite thermal conductances. The introduction of a finite conductance beetween fuel cell and its ambiance have showed that the theorical well known maximum efficiency situation – corresponding to a thermal equilibrium beetween both – is physically incongruous. Even if the famous efficiency limitation (1) is impractical to qualify performances of an isothermal system as the fuel cell, the latter is submit to the second principle of thermodynamics and can not produce any electrical power without rejecting heat flux to the surrounding. Then, a temperature difference beetween the system and its ambiance is essential to produce any rate of electrical work. The optimization of the power output regarding to the fuel cell temperature have allowed us to highlight the existence of an optimal one, practically calculated for a hydrogen-oxygen reaction in standard conditions. For the moment, water produced is supposed to be only in a vapor form, that is a weak hypothesis at low operating temperatures.

Our present endoreversible fuel cell is based on an unique finite conductance, thermal one and due to the heat flux exchange with surrounding. It would be significant to also consider a non reversible chemical reaction, using results of chemical thermodynamics in finite time [17]. Finally, different types of internal entropy production could be progressively took into account.

As a matter of fact, design and optimization processes of fuel cell systems have to take into account also fundamental Carnot principles. Heat flux rejected by the system to the surronding is an essential point and condition the imaginable produced electrical power.

Nomenclature

Acronyms

CHE	Carnot heat engine
ERFC	EndoReversible Fuel Cell.
RFC	Reversible Fuel Cell

Notations

G	Gibbs energy $[\mathrm{J}]$.
g	Molar Gibbs energy $[\mathrm{J} \cdot \mathrm{mol}^{-1}]$.
H	Enthalpy $[\mathrm{J}]$.
h	Molar enthalpy $[\mathrm{J} \cdot \mathrm{mol}^{-1}]$.
p	Pressure $[\mathrm{bar}]$.
Q	Heat energy $[\mathrm{J}]$.
S	Entropy $[\mathrm{J} \cdot \mathrm{K}^{-1}]$.
$\dot{S}$	Entropy flow $[\mathrm{W} \cdot \mathrm{K}^{-1}]$.
T	Temperature $[\mathrm{K}]$.

Greek symbols

Δ_r Difference due to chemical reaction. η Thermodynamical efficiency.

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