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Production of Hydrogen for Fuel Cell: Microchannel Reactor Modelling for Combustion and Reform of Ethanol in Alternate Channels

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Microchannel reactors are miniaturized reaction systems containing reaction channels with characteristic dimensions in the range of 10-500 μm. One possible application of microchannel reactors is the production of hydrogen to generate electrical power for portable equipment. In this paper, we propose to study the use of a microchannel reactor to produce hydrogen from the reaction of steam reforming of ethanol. The proposed device has the following components: (1) channels for steam reforming of ethanol containing a catalytic bed of Ni/Al_2O_3, (2) channels for exothermic reaction to provide the heat required for the reforming reaction, (3) cell fuel (PEMFC) that consumes the hydrogen produced by the reaction of reform. This work involved the simultaneous modelling of components (1) and (2) with the aim of showing that the catalytic oxidation of part of the exhaust gases produced in the reform of ethanol provides the heat required for the reforming reaction of ethanol, and that in this case the temperature along the microchannel (both in the reforming channel as in the oxidation channel) is kept approximately constant. Two alternatives were studied: co-currents and crosscurrents flows in the channels of endothermic and exothermic reaction. By studying these two cases it was concluded that the configuration co-current presents best result, concluding that the system can be considered isothermal.

1. Introduction

In modern society, the rapid development of multifunctional portable devices such as laptops and cell phones leads to an inevitable consequence: these devices are increasingly within reach of a growing number of people. As a consequence, some important facts are happening worldwide: continuous increase of electricity consumption, the increasing need for providing efficient power sources for portable devices and the increase in environmental pollution due to the disposal of a large number of batteries.

There is, therefore a demand for long life and highly efficient batteries, which will result in less environmental impact. Microchannel reactors (Bineli et al., 2011, Kolb et al., 2011) can be used to obtain hydrogen to generate electrical power for portable devices.

The hydrogen produced by steam reforming of ethanol is used as feed to a fuel cell - PEMFC, through a highly efficient process which produces clean exhaust gases. Fuel cells are suitable for power supplies of portable equipment by its small size, light weight and provide energy for a range of 1 W to 100 W (Delsman, 2005). The reactions involved in the process of steam reforming of ethanol are endothermic, therefore it is necessary to supply heat to the system to maintain the temperature of reaction. This energy can be obtained from the oxidation of the products of the reaction of reforming of ethanol. In this paper, we propose the use of a microchannel reactor to produce hydrogen by the reaction of ethanol with water.

2. Proposed equipment - microreformer / microcombustor coupled

In this paper, we propose to explore the use of a microchannel reactor to produce hydrogen by the reaction of ethanol with water (Figure 1). The proposed device has the following components: (1) channels

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for steam reforming of ethanol containing a catalytic bed of Ni/Al_2O_3, (2) channels for exothermic reaction to provide the heat required for the reforming reaction.

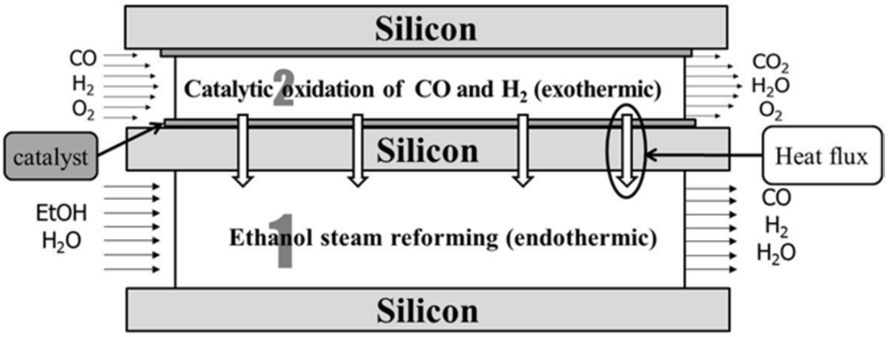

Figure 1: Proposed equipment.

Figure 2 shows schematically the equipment to be modelled. Operating conditions and assumptions of the model are: two-dimensional system; compressible fluid with ideal gas behaviour; channels with a length of 3.0 cm and a width of 1800 μm. The equipment to be studied can be divided into three subdomains (Ω_1, Ω_2, Ω_3), as shown in Figure 2, they are:

Subdomain Ω_1 (hot microchannel): In this channel the exothermic oxidation reactions of CO and H_2 occurs in the walls. The channel height is $h_1 = 50$ μm. The Knudsen number is the range of 10^{-1} and 10. In this range the flow can be considered as *slip flow* (Beskok and Karniadakis, 1999). Therefore, inside the microchannel the flow is free; operating in the slip flow regime and velocity profile is fully established. Beskok and Karniadakis (1999) proposed a model for laminar flow between parallel plates for slip flow regime showed below:

$$\frac{U(Y)}{\bar{U}} = \frac{-\left(\frac{Y}{H}\right)^2 + \left(\frac{Y}{H}\right) + \left(\frac{2-\sigma_v}{\sigma_v}\right)\frac{Kn}{1-bKn}}{\frac{1}{6} + \left(\frac{2-\sigma_v}{\sigma_v}\right)\frac{Kn}{1-bKn}} \tag{1}$$

With the parameters: σ_v = tangential momentum accommodation coefficient (TMAC), b = slip coefficient, U = fluid velocity in the X direction, H = distance between plates, Kn = Knudsen number, which is defined as $Kn = \lambda/h$, where λ is the mean free path of the molecules and h is a characteristic length scale.

The temperature at the entrance of the microchannel is $T_0 = 600$ K. The stream formed by the exhaust gases coming out from the microreformer are combined with the air flow with excess oxygen, comprising the feed of the microcombustor.

Subdomain Ω_2 (cold microchannel): In this subdomain, the endothermic reaction of steam reforming of ethanol occurs inside the channel. This channel is a packed bed, containing spherical particles (catalyst) of 50 μm in diameter and the channel height (h_2) is 500 μm. The temperature at the entrance of the microchannel is $T_0 = 600$ K, with feed molar ratio: $H_2O/EtOH = 4$. For this channel, we assume flat velocity profile. This assumption is reasonable because two conditions occur in this channel: (i) it is a packed bed; (ii) Flow with slip at the microchannel walls. With these two conditions combined, we can consider the flow of fluid as a plug flow.

Subdomain Ω_3 (wall separating the microchannels): silicon wall thickness with heath $h_3 = 50$ μm, impermeable to the chemical species.

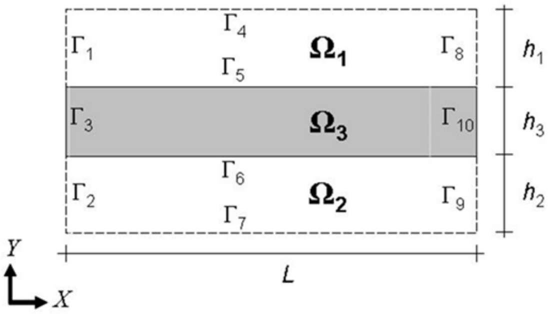

Figure 2: Schematic representation of ethanol microreformer with microcombustor coupled, identification and definition of the subdomains (Ω_1, Ω_2, Ω_3) and its boundaries (Γ_i).

In subdomain Ω_3 there is no mass transfer, whereas in the subdomains Ω_1 and Ω_2 the mass transfer can be represented mathematically by diffusion and convection equations (Bird *et al*., 2004). In these subdomains the boundary conditions are:

- Γ_1 and Γ_2: the concentrations of the reagents are known in the microchannel input;
- Γ_4 and Γ_5: in the channel walls, the reagents (CO and H_2) are consumed by the oxidation reaction;
- Γ_6 and Γ_7: the walls of the endothermic microchannel are impervious to chemical species;
- Γ_8 and Γ_9: in the microchannel outlet, the mass transport occurs only by convection

In subdomain Ω_3 the heat transfer is governed by conduction equation. In subdomains Ω_1 and Ω_2 the heat transfer phenomena is governed by conduction and convection equations (Bird *et al*., 2004). In Ω_1 there is no generation or consumption of thermal energy, since the reactions are occurring in the walls of the microchannel, whereas in Ω_2 occurs heat consumption due to the ethanol reforming reaction. The correspondent boundary conditions are:

- Γ_1 and Γ_2: the temperature of both input currents of the microchannels is 600 K;
- Γ_3, Γ_7 and Γ_{10}: there is no heat transfer through these boundaries;
- Γ_4 and Γ_5: in the channel walls, there are heat generation due to oxidation of the CO and H_2;
- Γ_6: it is considered the continuity of the temperature between subdomains Ω_2 and Ω_3;
- Γ_8 and Γ_9: in the microchannel outlet, the energy transport occurs only by convection

3. Results and discussion

The model solution was obtained by means of numerical finite element method using the software COMSOL *Multiphysics*^{TM}. Initially, the mass transfer equations were solved assuming isothermal process in both microchannels, because all of the heat generated by the oxidation reactions should be consumed by the reforming reaction so that no significant temperature variations occurs within the equipment. The result thus obtained was used as the initial estimate for the full model which is not isothermal.

The microchannel for the oxidation of CO and H_2 was modelled in three different configurations: In the first configuration it was considered a microchannel 50 µm height, with fluid fed cocurrent to the flow in the reform channel, at a speed of 0.2 m/s; in the second configuration, the feed was changed to counter-current flow and, finally, for the third configuration, the feed was countercurrent and with velocity of 0.05 m/s in a microchannel of height of 200 µm. The results are shown in Figures 3-a, Figure 3-b and Figure 3-c.

Analysing the figures, one can conclude that the configuration that has the smallest variation in the average cross temperature is the one in which the combustion fluid is fed in the co-current direction. This can be explained by the rate of chemical reactions: a higher rate of thermal energy generation occurs at the beginning of the exothermic reactions and higher rate of energy consumption also occurs at the beginning of endothermic reactions, therefore, the higher consumption and higher generation must occur in the same axial position. The temperature profile shown in Figure 4 shows that the higher variation occurs shortly after the entry of fluids in the microchannels and that this variation does not exceed 0.4% of the feed temperature.

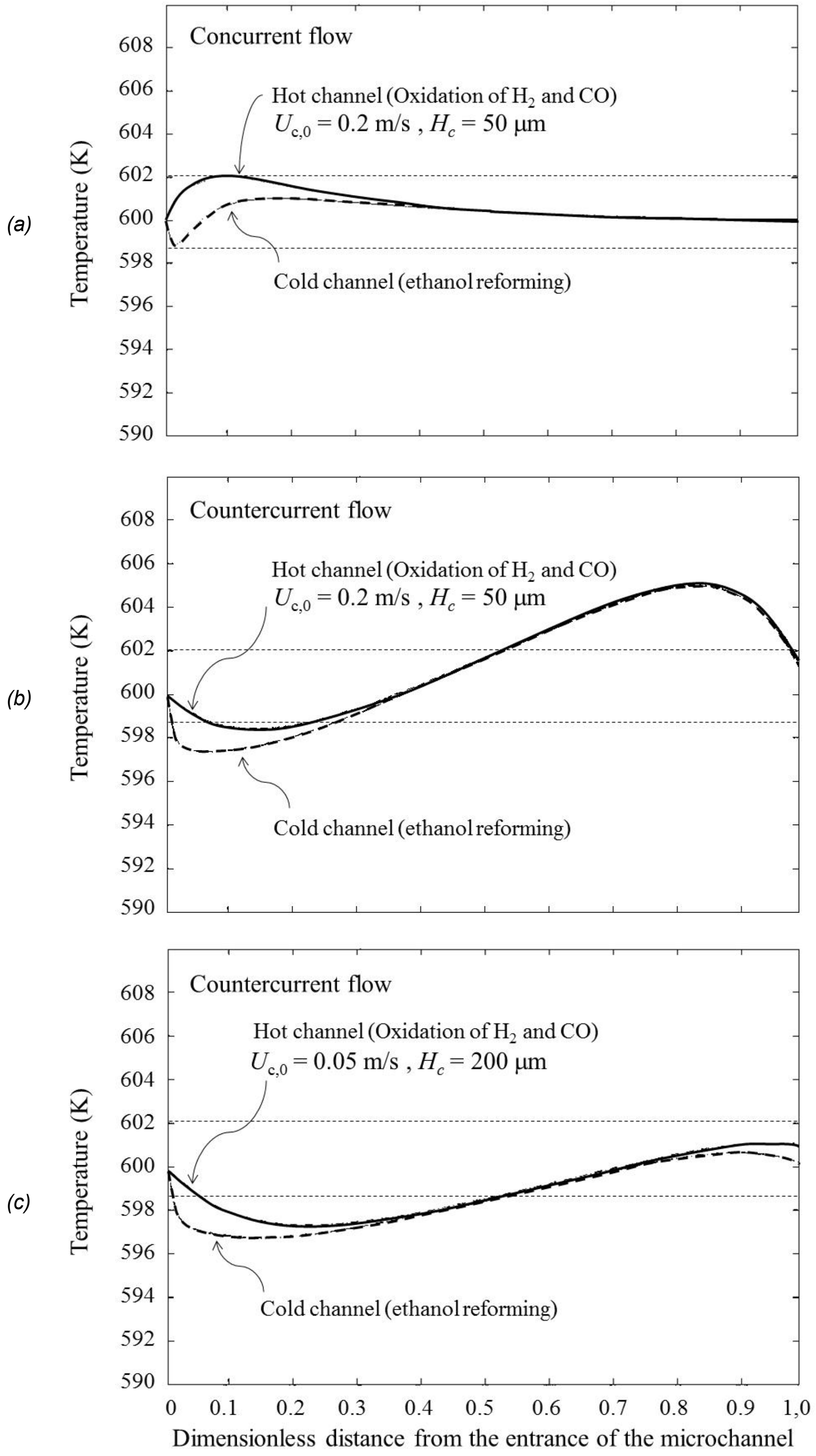

Figure 3: Local mean temperature distribution (*T*(x)) over a microchannel of reactor.

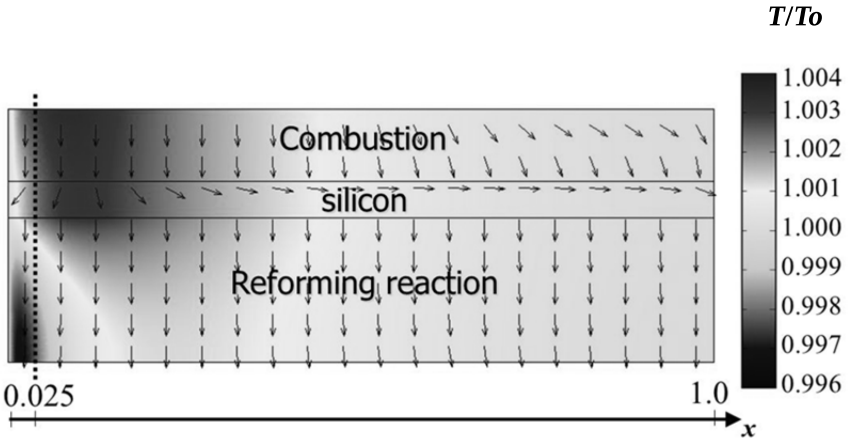

Figure 4: Distribution of temperature in the microreactor. $H_c = 50$ μm, $H_R = 400$ μm, $U_{c,o} = 0.2$ m/s, $U_{R,o} = 0.02$ m/s.

In Figure 5 it is possible to observe that the cross temperature profile at a distance of 625 μm from the entrance (2.5% of the length of the microchannel) is consistent with the expectations. It is possible to see that the temperature of the reformation channel is kept below the temperature of the combustion channel, also the shape of the temperature curve in the reformation channel is consistent with the direction of the heat flow and with the characteristic endothermic of the reaction. Due to the high thermal conductivity of silicon, the temperature of the wall that separates the microchannel remains practically constant in all axial positions. This result allows us to affirm that the use of microstructured equipment in which the resistance to heat transport is reduced, the isothermal process of steam reforming of ethanol is entirely feasible.
The microstructured reactors are mainly characterized by their high specific surface area compared to conventional chemical reactors.

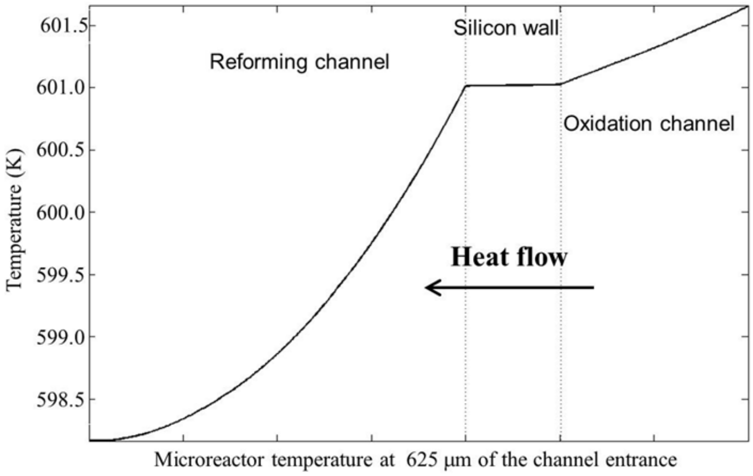

Figure 5: Distribution of temperature in the microreactor. Profile obtained at 625 μm from the entrance of the channel

4. Conclusions

The excellent rates of mass transfer and heat that exist in microchannel reactors enables exothermic and endothermic reactions occur in parallel in separate channels, simulating an autothermic process with little unwanted homogeneous reactions. This statement was confirmed by the model, in which the steam reforming of ethanol, highly endothermic, was processed in parallel with the exothermic oxidation reaction of hydrogen and carbon monoxide. The results obtained from this model provided important information, making it possible to formulate the subsequent models considering them as being isothermal. This result was made possible by the micrometric dimensions of the channels indicating that this parameter of the model strongly influences the temperature profile within the microreactor. Moreover, the oxidation of only 13.56% of the effluent from the cold channel generates the heat required by the steam reforming reaction of ethanol.

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