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Hydrodynamic Effect on the Iso-octane Steam-Reforming in a Monolithic Reactor Channel

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ABSTRACT

The production process of clean hydrogen by iso-octane steam reforming in a micro-reactor under atmospheric pressure, and the high temperature was investigated. The simulation is done using momentum conservation, mass conservation, and convection-diffusion equations, represented by the Navier-Stokes equations, the continuity equation, and the Stefan-Maxwell equation respectively. The resolution was performed using a differential equations discretization into their conservative by the finite element method with an unconditionally stable scheme. An analysis of the hydrodynamics of the flow effect shows that the hydrogen produced by iso-octane reforming is proportional to the mixed flow Reynolds number.

1. Introduction

Hydrogen combustion releases a large amount of energy (about three times more than that of gasoline at constant weight). This combustion generates only water, which makes it an important source of clean electricity for various applications, such as supplying power in remote areas not connected to power grids. Additionally, it can be used on sensitive sites as backup energy supply. Used as a fuel cell, it allows for electricity production directly on board a vehicle driven by an electric motor with the benefit of zero CO₂ emissions. The use of hydrogen is currently almost confined to an industrial application such as ammonia production, essential for producing fertilizers, and refining oil. It is also used in other

chemical products, plastics synthesis, as well as in some glass manufacturing processes.

Despite its abundance in the universe, we can't find hydrogen in its free state. Currently, this is achieved by two main methods: decomposition of water using an electric current of fossil, nuclear, or renewable origin, or by extraction from a hydrocarbon by partial oxidation, or a reduction of the water vapor. This method consists of transforming hydrocarbons into synthetic gas by reduction with steam in the presence of a catalyst. Also called steam reforming, takes place at high temperature and moderate pressure. The method is very effective and has been extensively investigated for several years. The ethane steam reforming at a temperature of 500 °C was studied by Jens

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(Rostrup-Nielsen 1973) to determine the few factors that could impact hydrocarbon steam forming. This experiment shows that the analysis procedure is influenced by the catalyst composition, unlike the ethane hydrogen and carbon monoxide methanation of ammonia that are unaffected by the support. Garcia and Laborde^[7] carried out a thermodynamic equilibrium analysis of ethanol vapor reforming for hydrogen production, in ranges of pressure (1 - 9 atm), temperature (400-800K), and a water-ethanol ratio (0:1 - 10:1). He observed that the best condition for this process is at $T > 650$ K and under atmospheric pressure. The same process was followed by Haryanto et al.^[3] who examined different types of catalysts and observed that CO/ZnO, ZnO, Rh/Al₂O₃, Rh/CeO₂ and Ni/La₂O₃-Al₂O₃ yield good results. Yu et al. (Yu, Aihara, and Antal 1993) studied the production of hydrogen by glucose gasification in supercritical water at $T = 600$ °C and a pressure $P = 34.5$ MPa, they found that the reactor wall strongly influences the yield of this reaction. Achanbach and Riensche^[1] conducted experiments to determine the kinetics of the process of reforming methane and steam in anode materials of a fuel cell, using a catalyst that consists of 80% ZrO₂ and 20% of Ni. This experiment was under a temperature which varies between 700 and 940 °C, a system pressure of order 1.1 to 2.8 bars, whereas that of CH₄ was from 0.11 to 0.33. Or he has proven that the partial pressure of H₂O does not influence the process of steam reforming. In order to study the kinetics of the thermal reforming reaction of liquid hydrocarbons and the performance of the used fuel processor. Methanol vapor catalytic reforming was studied as a means of generating hydrogen for a polymer electrolyte membrane fuel cell by Amphlet et al.^[4]. He developed a semi-empirical model of the kinetics of catalytic methanol steam reforming over a CuO/ZnO/Al₂O₃ catalyst. This model was capable to predict reformer performance against the various important parameters in the development of an integrated reforming polymer fuel cell system. They have found by this study that the improvement of catalyst activity and, in particular, of selectivity would be an important advantage for the optimization of the power/weight ratio of a fuel cell reformer plant intended to be used in automotive applications. A thermodynamic study of ethanol steam-reforming in a micro-reactor was carried out by Vasudeva et al. (Vasudeva et al. 1996). Adopting an approach that allows estimating the carbon formed with other gaseous species in the reforming process. Ethanol vapor, he found equilibrium hydrogen yields of up to 5.5 moles per mole of ethanol in the feed can be obtained compared to the stoichiometric value of 6.0. In order to maximize hydrogen production while discouraging reactions leading to undesirable products,

such as methane, acetaldehyde, diethyl ether or acetic acid, in competition with H₂ for hydrogen atoms. Mariño et al.^[12] carried out the procedure of steam reforming of bioethanol. The catalysts suitable for this reaction are Cu-Ni-K/γ-Al₂O₃ as they are capable of producing acceptable amounts of hydrogen operating at atmospheric pressure and at a temperature of 300 °C. It has proved that the addition of Ni to Cu/Ni/K/γ-Al₂O₃ catalysts promotes gassing of ethanol, increases the gas yield and reduces the production of acetaldehyde and acetic acid. As the presence of Ni slightly increases the production of hydrogen by a slight increase in ethanol conversion since it promotes the segregation of Cu₂⁺ ions on the catalytic surface. Garcia et al.^[8] treated the aqueous fraction of bio-oil, resulting from the rapid pyrolysis, catalytically reformed with steam at 825 and 875 °C, a high space velocity (up to 126 000 h⁻¹) and a residence time short (26 ms), in a fixed bed microreactor interfaced with a molecular weight spectrometer. In this study, several nickel-based research and commercial catalysts were tested; these catalysts were prepared by impregnating a support α-Al₂O₃ with nickel and additives. Two strategies have been applied to improve catalyst performance. The first approach was to improve the vapor adsorption to facilitate partial oxidation, i.e., the gasification of coke precursors. The second attempted to slow surface reactions leading to the formation of coke precursors due to cracking, deoxygenation and dehydration of the adsorbed intermediates. To improve steam adsorption, while cobalt and chromium additives were applied to reduce coke-forming reactions, Garcia used magnesium and lanthanum. Cobalt-activated nickel and chromium-activated nickel on MgO-La₂O₃-α-Al₂O₃ catalysts gave the best results in laboratory tests; it also observed a gradual deactivation of the catalyst under the reaction conditions. Resulting in decreased hydrogen and carbon dioxide yields and an increase in carbon monoxide. The loss of activity has also resulted in the formation of larger quantities of methane, benzene, and other aromatic compounds. He has proved by this study that commercial catalysts developed for steam reforming of natural gas and crude oil fractions have been more efficient for the production of hydrogen from bio-oil than most catalysts. research, mainly because of the greater water-gas transfer activity. Dimitris et al.^[11] investigated the catalytic performance of supported noble metal catalysts for steam reforming (SR) of ethanol in the temperature range of 600 to 850 °C with respect to the nature of the active metal phase (Rh, Ru, Pt, Pd), the nature of the support (Al₂O₃, MgO, TiO₂) and the metal filler (0-5% by weight). He found that for weakly charged catalysts, Rh is significantly more active and selective with respect to hy-

drogen formation than Ru, Pt, and Pd, which exhibit similar behavior. The catalytic performances of Rh and in particular Ru are considerably improved with the increase of the metal charge, which leads to higher ethanol conversions and hydrogen selectivities at given reaction temperatures. The catalytic activity and selectivity of the highly charged Ru catalysts are comparable to those of Rh and, therefore, ruthenium has been further investigated as a less expensive alternative. Dimitris has also proved that under certain reaction conditions, the 5% Ru/Al₂O₃ catalyst is capable of completely converting ethanol with selectivities. more than 95% hydrogen, the only by-product being methane. Pacheco et al. [14] developed a mathematical model of aspen plus process simulator, this model has been validated by an isooctane steam reforming experiment using a platinum-cerium oxide catalyst. Jeong et al. [9], studied the production of hydrogen by methanol steam reforming on Cu/Zn catalysts (Cu/ZnO, Cu/ZnO/Al₂O₃, Cu/ZnO/ZrO₂/Al₂O₃). The catalysts based on Cu/Zn are derived from precursors of the hydrotalcite type prepared by a co-precipitation process. These catalysts are characterized by measurements of N₂O chemisorption, XRD and BET surface. ZrO₂ added to the Cu/Zn catalyst improves the copper dispersion on the catalyst surface. It has proven that among the catalysts tested, Cu/ZnO/ZrO₂/Al₂O₃ has the highest methanol conversion and the lowest CO concentration in the exit gas. Adhikari et al. [2] treated Steam reforming of glycerol for the hydrogen production. The hydrogen yield of this procedure depends on several process variables, such as system pressure, temperature, and the reactants ratio. To understand the effects of these variables, Adhikari performed a thermodynamic equilibrium analysis for the glycerol Steam reforming process in the following variation ranges: pressure 1-5 atm, temperature 600-1000 K and feed ratio water-glycerol from 1:1 to 9:1. The equilibrium concentrations of different compounds were calculated by the method of direct minimization of Gibbs free energy. He found that the best conditions for the production of hydrogen are at a temperature > 900 K, at atmospheric pressure and at a water/glycerol molar ratio of 9: 1. Under these conditions, the production of methane is minimized and the Carbon formation is thermodynamically inhibited. Men et al. [13] studied the reaction between ethanol and water in the temperature range of 400 to 600 °C at atmospheric pressure and using a catalyst in a micro channel reactor. Men have found that the nature of the metal, the metal charge, and the type of media influence the catalytic activity and selectivity of the catalysts. Compared to the Co and Ni catalysts, he found that the Rh-based catalysts had the highest catalytic activity, whereas the Rh-Ni bimetallic catalysts showed a sig-

nificant improvement in terms of conversion. Ethanol and hydrogen selectivity The CeO₂-activated bimetallic Rh-Ni catalyst was stable for at least 100 hours with no detectable degradation in performance. Rabe et al. [15] analyzed the kinetics of catalytic gasoline reforming deactivation processes, the results of this modeling indicated that this reaction is strongly affected by the presence of sulfur in the alimention Du et al. [5] handled steam reforming of methanol with micro-channels. Studying the choice of catalyst, the operating parameters, and the micro-channel configuration, he found that the corresponding active Cu surface is proportional to its concentration, which improves the activity of the catalyst. The reaction temperature, the feed rate, the temperature of the mixture and the molar ratio of H₂O to CH₃OH on the conversion rate of methanol also has a significant impact. It has also been found that the increase in the length of the micro-channels can significantly improve the conversion rate of methanol. Kolb [10] provided an overview of research activities in the field of energy-using microreactors. He noted that major research efforts in this research area are currently focused on treating fuel as a source of hydrogen, primarily for distributed use through fuel cells. The development of catalysts, the reforming and removal of carbon monoxide by water-gas displacement, the design of reactors, preferential oxidation, selective methanation and membrane separation are therefore under analysis. Simsek et al. (Simsek et al. 2013) studied the steam oxidative reforming of methane to synthesis gas over an alumina supported bimetallic Pt Rh catalyst using two configurations of the coated and packed microchannel reactors. In the first geometry, thin layers of catalysts are applied to the opposite walls of a single microchannel, whereas the second geometry is described by particulate catalysts encapsulated in an empty microchannel and of the same dimensions as the first one. The two configurations are compared on the basis of methane conversion and selectivity to carbon monoxide measured at different parameters, namely reaction temperature, steam/carbon and oxygen/carbon molar ratios in feed and time of contact. They found that the coated catalyst gave CO selectivities significantly higher than the packed counterpart over the entire range of parameters. The production of clean hydrogen fuel via reformation of hydrogen sulfide in the presence of methane has been studied by El-Melih et al. [6] et al. From a small-scale reactor, the experimental results are presented on the quantities of hydrogen produced from a mixture of methane diluted in nitrogen and hydrogen sulphide at temperatures of 1273-1673 K The results provide essential information on this method of treatment and its ability to clean hydrogen production. The experimental setup was supported by

numerical simulations in the same range of experimental conditions, helped to determine the main reaction process. Experimental results and simulation results showed a potential hydrogen recovery of more than 95% from the mixture of hydrogen sulphide and methane resulting from the destruction of hydrogen sulphide.

This work aims to study hydrogen production by iso-octane reforming with water steam via an endothermic reaction in a micro-reactor using a Ni/Al₂O₃ catalyst.

2. Method and Governing Equations

In a micro-reactor of diameter $d = 0.001$ m and length $L = 0.02$ m. The iso-octane steam reforming reaction is carried out. This requires an energy $\Delta H = 1275$ kJ/mol. Figure 1 shows the design of the studied micro-reactor.

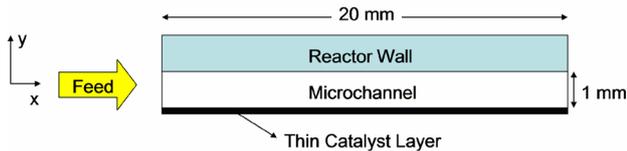
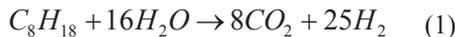


Figure 1. representative design of micro-reactor

According to the following reaction:



Considering a laminar flow, homogeneous, irrotational, and weakly compressible, the numerical study is based on a model provides the evolution of quantity of movement and conservation of mass, represented by the Navier-Stokes equations (2), and by the equation of continuity (3). On the other hand, it shows the evolution of the chemical species involved in the simulated reaction described by the diffusion-convection equations of Stefan-Maxwell (4)

$$\frac{\partial(\rho v)}{\partial t} + \nabla(\rho v \cdot v) = \nabla P + \rho g \quad (2)$$

$$\frac{\partial \rho}{\partial t} + (\rho \vec{u}) = 0 \quad (3)$$

$$\sum x_i x_j \left(\frac{J_j}{C_j} - \frac{J_i}{C_i} \right) = \frac{\nabla \mu_i}{RT} = \nabla x_i \quad (4)$$

For the turbulent flow, we used the RANS approach, based on two models; the k - ϵ model, and the k - ω model. Each model is represented by tow equations: the kinetic energy equation, and the dissipation rate equation.

The k - ϵ turbulence model:

$$\rho \frac{\partial \log k}{\partial t} + \rho U \cdot \nabla k = \nabla \cdot \left[\left(\eta + \frac{\eta_T}{\sigma_k} \right) \nabla \log k \right] + \left(\eta + \frac{\eta_T}{\sigma_k} \right) (\nabla \log k)^2 + \eta_T e^{-\log k} P(U) - \frac{2}{3} \rho \nabla \cdot U - \rho C_{\epsilon 2} e^{(\log \epsilon - \log k)} \quad (5)$$

$$\rho \frac{\partial \log \epsilon}{\partial t} + \rho U \cdot \nabla \log \epsilon = \nabla \cdot \left[\left(\eta + \frac{\eta_T}{\sigma_\epsilon} \right) \nabla \log \epsilon \right] + \left(\eta + \frac{\eta_T}{\sigma_\epsilon} \right) (\nabla \log \epsilon)^2 + C_{\epsilon 1} \left[\eta_T e^{-\log k} P(U) - \frac{2}{3} \rho \nabla \cdot U \right] - \rho C_{\epsilon 2} e^{(\log \epsilon - \log k)} \quad (6)$$

While

$$P(U) = \frac{1}{2} [\nabla u + (\nabla u)^T]^2 - \frac{2}{3} (\nabla \cdot U)^2 \quad (7)$$

Table 1. The turbulence model constants.

C_μ	Turbulent modeling constant (=0.09)
C_{d1}	Turbulent modeling constant (=1.14)
C_{d2}	Turbulent modeling constant (=1.92)
σ_k	Turbulent modeling constant (=1.00)
σ_ω	Turbulent modeling constant (=1.30)
k	Von Karman constant (=0.42)
C^+	Logarithmic wall fraction constant (=5.5)
k	Turbulent kinetic energy
ω	Turbulent dissipation rate

The k - ω turbulence model:

$$\rho \frac{\partial \log k}{\partial t} + \rho U \cdot \nabla k = \nabla \cdot \left[\left(\eta + \sigma_k \eta_T \right) \nabla \log k \right] + \left(\eta + \sigma_k \eta_T \right) (\nabla \log k)^2 + \eta_T e^{-\log k} P(U) - \frac{2}{3} \rho \nabla \cdot U - \beta_k \rho e^{\log \omega} \quad (8)$$

$$\rho \frac{\partial \log \omega}{\partial t} + \rho U \cdot \nabla \log \omega = \nabla \cdot \left[\left(\eta + \sigma_\omega \eta_T \right) \nabla \log \omega \right] + \left(\eta + \sigma_\omega \eta_T \right) (\nabla \log \omega)^2 + \alpha \left[\eta_T e^{-\log k} P(U) - \frac{2}{3} \rho \nabla \cdot U \right] - \rho \beta e^{\log \omega} \quad (9)$$

Table 2. The turbulence model constants

C_μ	Turbulent modeling constant (=0.09)
α	Turbulent modeling constant (=13/25)
β_0	Turbulent modeling constant (=9/125)
β_{0k}	Turbulent modeling constant (=0.09)
σ_ω	Turbulent modeling constant (=0.5)
k	Von Karman constant (=0.42)
C^+	Logarithmic wall fraction constant (=5.5)
k	Turbulent kinetic energy
ω	Specific Turbulent dissipation rate

The simulation was carried out with solving the differential equations in the variation form by the finite element method using an unconditionally stable (numerically) schema.

3. Results and Discussion

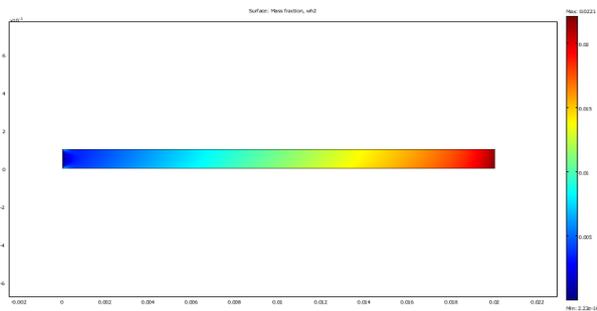


Figure 2. micro- channel simulation results

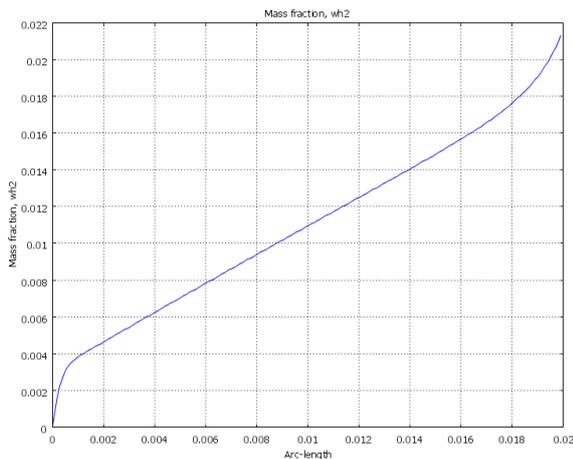


Figure 3. Mass fraction of hydrogen produce

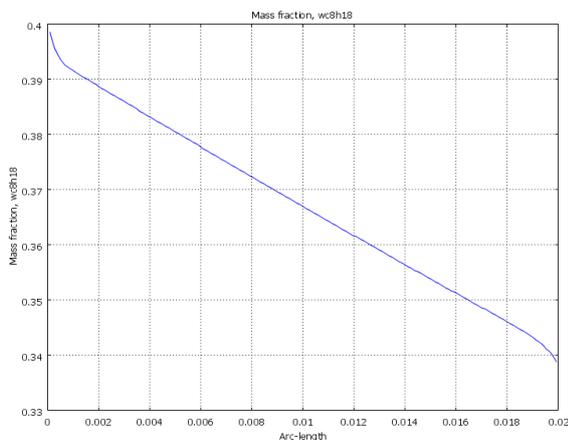


Figure 4. Mass fraction of iso-octane consumed

The mass fraction of hydrogen produced, from iso-octane steam reforming reaction in a conventional micro-reactor, increases with the length of the micro-channel where this reaction takes place, and it reaches a max: $wh_{2max} = 0.021$. This increase is related to a decrease in

iso-octane mass $w_{c8h18max} = 0.339$.

3.1 Mesh Test

The mesh test was realized using a triangular form, or we tested several sizes which correspond to the different degrees of freedom and a time resolution. The following table represents the details of the test:

Table 3. the mesh test characteristics

Test Number	Number of element (-)	Number of Freedom (-)	Time resolution (s)
Test N° 01	548	7780	1.391
Test N° 02	2192	29253	2.109
Test N° 03	8768	113299	5.757
Test N° 04	35072	445791	301.091

The results obtained are represented by figure 5 and figure 6

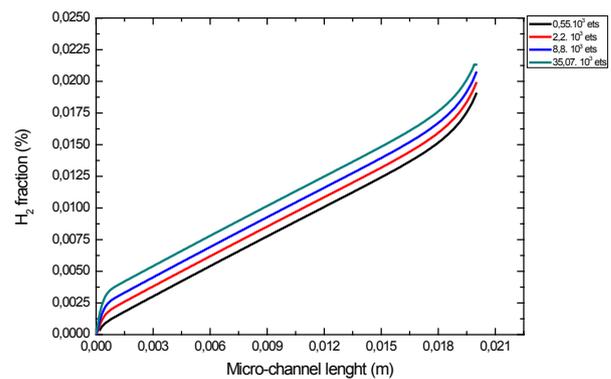


Figure 5. Mass fraction of hydrogen produce for the different mesh used

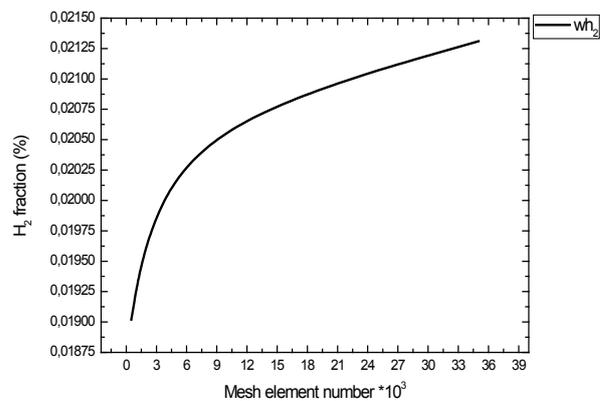


Figure 6. Mass of H₂ max produced as a function of the number of mesh element

For these different mesh, the hydrogen fraction re-

covered at the micro-channel output increases from $Wh_2 = 0.019$ using a mesh of 548 element number and 7780 freedom number, to $Wh_2 = 0.021$ for a mesh of 35072 number of elements, and a freedom number of 445791. Marking a difference of $\Delta Wh_2 = 0.002$ for a difference of 34524 element.

The following simulations, we used a triangular mesh, of 8768 number of elements.

3.2. Laminar Flow

An inertial study was conducted in order to highlight the influence of hydrodynamics on the isoctane steam reforming process. For the laminar regime, the mass of the catalyst is varied from $m = 0.01$ kg to $m = 0.9$ kg, and for each mass, we tested Somme Reynolds number, to follow the critical "Re" that characterizes it. The mass fraction of hydrogen increases with the mass of the catalyst, and it represents a saturation at $wh_2 = 1.8$. Same to the critical Reynolds number; it increases with the mass of the catalyst, to have saturation at $Re = 1900$ (Figure 8).

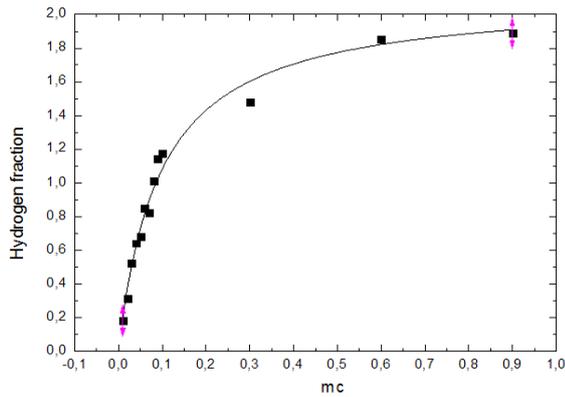


Figure 7. Mass of H_2 max produced as a function of the mass of the catalyst

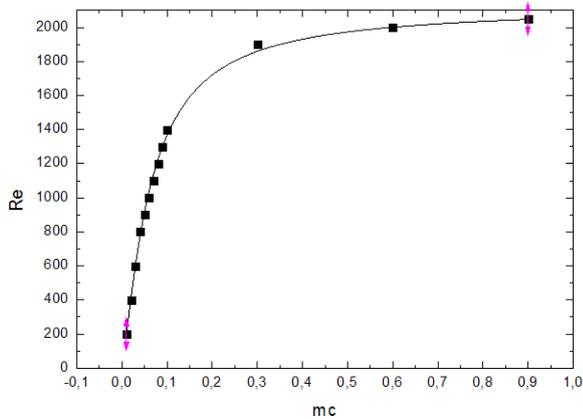


Figure 8. Critical Reynolds number for each mass of the catalyst

3.3. Turbulent Flow

Variants Reynolds number from $Re = 2000$ to $Re = 10000$ for both $k-\epsilon$ (Figure 9) and $k-w$ (Figure 10) turbulence models. For this regime, the fraction of hydrogen increases proportionally to the Reynolds number (Figure 11), with the maxima of 1.2 for a maximum Reynolds number of 10000, that is (63,15) times higher than the reference case (laminar). Similar to the "k- ϵ " turbulence model, the hydrogen mass fraction produced increases to 1.4. Comparing the two models; we can see a difference of 20% (Figure 12)

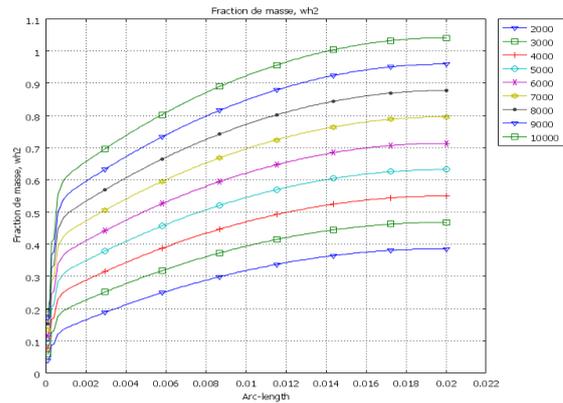


Figure 9. hydrogen mass for the different Reynolds numbers: model $k-\epsilon$

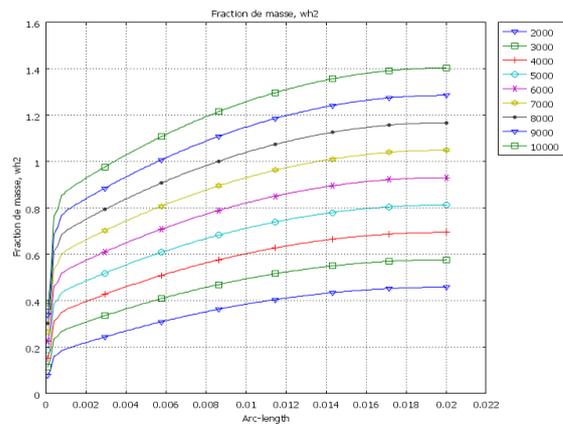


Figure 10. hydrogen mass for the different Reynolds numbers: model $k-w$

Table 4. Correlations

Model	equation
Turbulence $k-w$ $Wh_2=f(Re)$	$Wh_2=0.20733+1.2.10^{-4}*Re$
Turbulence $k-\epsilon$ $Wh_2=f(Re)$	$Wh_2=0.21665+8.25.10^{-5}*Re$
Laminaire : $Rec=f(mc)$	$Rec=(2118.40758*mc^{1.25})/(0.031*mc^{1.25})$
Laminaire : $wh_2=f(mc)$	$Wh_2=(2.11025*mc)/(0.095+mc)$

4. Conclusion

Isooctane steam reforming in a micro-reactor is one of the promising processes for producing clean hydrogen. The turbulence of the flow in the micro-channel further enhances this technique. Two turbulence models have been used to simulate the iso-octane steam reforming on a monolithic reactor with a Ni/Al₂O₃ catalyst. Depending on the turbulent model the C₈H₁₈ conversion and the H₂ production vary greatly. The hydrogen fraction at the output of the micro-channel is linearly proportional to the Reynolds number; while the turbulent KW model is found to be more compatible with this technique than the K-eps.

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