



Methane steam reforming in a microchannel reactor for GTL intensification: A computational fluid dynamics simulation study

G. Arzamendi^a, P.M. Diéguez^a, M. Montes^b, J.A. Odriozola^c, E. Falabella Sousa-Aguiar^d, L.M. Gandía^{a,*}

^a Departamento de Química Aplicada, Edificio de los Acebos, Universidad Pública de Navarra, Campus de Arrosadía s/n, E-31006 Pamplona, Spain

^b Departamento de Química Aplicada, Facultad de Ciencias Químicas de San Sebastián, Universidad del País Vasco, Pº Manuel de Lardizábal 3, E-20018 San Sebastián, Spain

^c Instituto de Ciencia de Materiales de Sevilla, Centro Mixto CSIC-Universidad de Sevilla, Avda. Américo Vespucio 49, 41092 Sevilla, Spain

^d CENPES/PDEDS/Petrobras, Av. Horácio Macedo, 950 - Ilha do Fundão CEP 21941-915 Rio de Janeiro, Brazil

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ABSTRACT

The integration of the steam reforming and combustion of methane in a catalytic microchannel reactor has been simulated by computational fluid dynamics (CFD). Two models including 4 or 20 square microchannels of 20 mm of length and 0.7 mm of side have been developed. It has been assumed that a thin and homogeneous layer of an appropriate catalyst has been uniformly deposited onto the channels walls. The kinetics of the steam reforming of methane (SRM), water-gas shift (WGS) and methane combustion in air have been incorporated into the models. This has allowed simulating the effect of the gas streams space velocities, catalyst load, steam-to-carbon (S/C) ratio and flow arrangement on the microreformer performance. The results obtained illustrate the potential of microreactors for process intensification: complete combustion of methane is achieved at gas hourly space velocities (GHSV) as high as 130,000 h⁻¹. As concerns the SRM, methane conversions above 97% can be obtained at high GHSV of 30,000 h⁻¹ and temperatures of 900–950 °C. Under these conditions selectivity for syngas is controlled by the WGS equilibrium.

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1. Introduction

Gas-to-liquid (GTL) process is the conversion of natural gas (NG) to high quality liquid syngas in several stages: syngas obtainment, production of long-chain hydrocarbons by the Fischer–Tropsch synthesis (FTS), and hydrocracking of the heaviest fractions for naptha, diesel and lubricants production [1]. Renewed interest in GTL processes is mainly driven by factors as [2]: (i) the steady increase in global energy demand; (ii) the existence of large volumes of stranded natural gas in remote areas (GTL is the main alternative to liquefied NG for monetizing these reserves); (iii) GTL syngas are almost free of sulfur and aromatic hydrocarbons; (iv) the need of reducing associated gas flaring for economic and environmental reasons (GTL is more interesting than associated gas re-injection or piping); (v) the high price of crude oil, that has improved the economics of the GTL process which has the drawback of very high investment costs; (vi) concerns on energetic security. A global production of GTL syngas of the order of 250,000 barrels per day (b/d) can be estimated for 2020. This is a low amount compared with the expected middle distillates consumption of about 40 million b/d assuming an annual consumption growth of 1.9–3%

[2]. It should be noted that conventional GTL has some limitations: it is only viable for onshore applications and the investment costs are very high making it profitable only when exploiting very large gas fields. Microreactors-based GTL plants can help to overcome some of these limitations. They are suitable for gas associated to relatively small oil fields situated in remote or deepwater areas and can be adapted to onshore, offshore (platforms) and mobile/onboard (tankers) applications. Microreactors have high surface-to-volume ratio, they are intrinsically safe, give enhanced heat and mass transfer rates, produce extremely low pressure drop and allow easy thermal integration of the processes involved [3–5]. Microreaction technology for GTL is at the precommercial stage and the developments in this area are being led by companies such as Velocys® Inc. (Plain City, OH, USA) [6] and CompactGTL plc (Abingdon, Oxfordshire, UK) [7]. A number of issues, mainly economic, feedstock availability and H₂/CO ratio, have to be taken into account when considering the production of syngas for GTL applications [1,8]. Currently, the preferred technology in conventional GTL is the autothermal reforming (ATR) of methane, in which suitably pre-treated natural gas is mixed with steam and pure oxygen, fed first to a combustor and then flown through a bed of a Ni-based catalyst where the steam- and CO₂-reforming reactions take place. This is an energetically efficient process that allows obtaining under proper operation syngas with H₂/CO ratio close to two which is the optimum composition for the low-temperature FTS [8]. However, the

* Corresponding author. Tel.: +34 948 169605; fax: +34 948 169606.
E-mail address: lgandia@unavarra.es (L.M. Gandía).

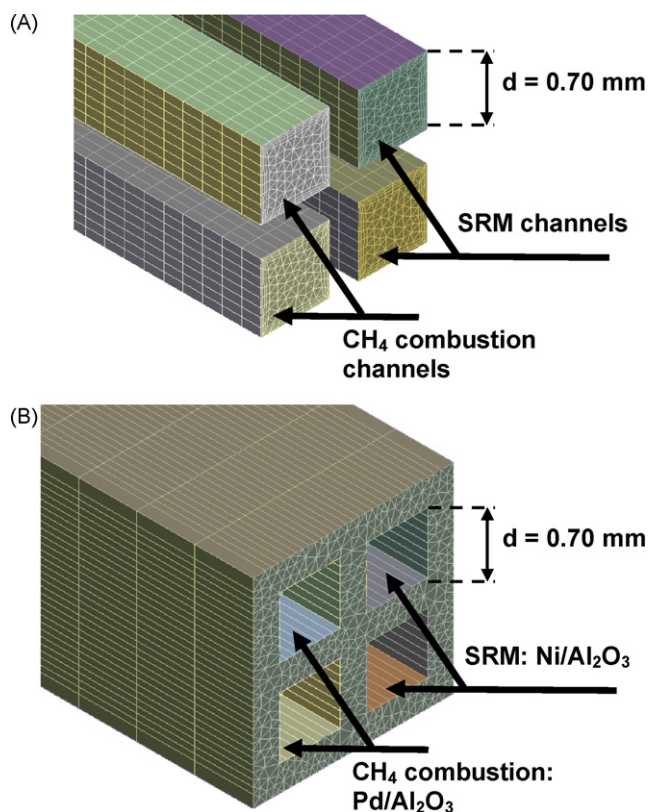


Fig. 1. Fluidic (A) and solid (B) domains of the CFD model consisting of four square parallel channels of 20 mm of length and 0.7 mm of side.

high capital costs and the need of an oxygen plant make the ATR as well as the catalytic or non-catalytic partial oxidation technologies non-competitive with respect to the steam reforming for the monetization of stranded natural gas in remote areas. As a matter of fact, the above-mentioned companies leading the application of microreactor technology to GTL have adopted methane steam reforming as the method for syngas production [6,7]. Of course, the H_2/CO ratio is well above 2 but, in the integrated process, light hydrocarbons produced by the FTS are combusted to preheat the reformer feed or to generate steam.

The aim of this computational fluid dynamics (CFD) simulation study is to guide the design of a microreactor for the steam reforming of methane (SRM) reaction through the study of the effect of relevant operating variables: space velocities, catalyst load, steam-to-carbon (S/C) ratio and flow arrangement. As natural gas will be available at the point of application, it is proposed to couple the endothermic SRM reaction ($\Delta H^\circ_{298K} = 205.6 \text{ kJ/mol}$) with the exothermic combustion of methane ($\Delta H^\circ_{298K} = -802.6 \text{ kJ/mol}$) to reach a suitable reaction temperature and supply the required heat of reaction.

2. Microreformer model

Three-dimensional (3D) simulations involving fluids flow, heat transfer and catalytic chemical reactions demand long computation times. Therefore, two physical models have been considered in this study: a first simple model to study of the influence of the operating variables, and a second more complex model closer to the final design. The first model consisted of four square parallel microchannels of 20 mm of length and 0.70 mm of side (d). The SRM reaction takes place in two of the channels and the combustion of methane in the other two, as illustrated in Fig. 1A and B, which show the fluidic and solid (steel) domains, respectively. A preliminary study

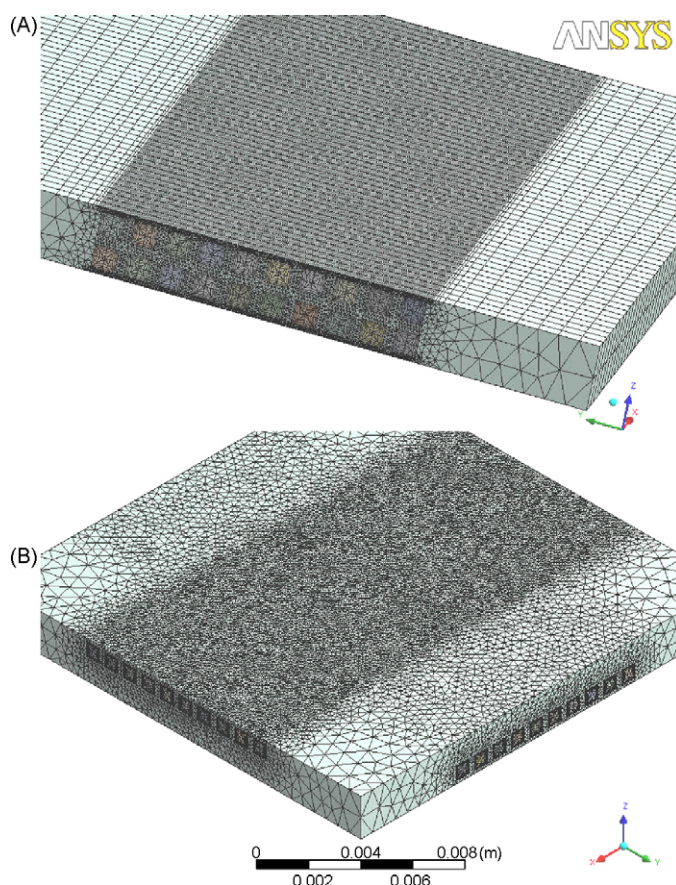


Fig. 2. Geometry corresponding to the parallel cocurrent (A) and cross flow (B) arrangements of the CFD model consisting of two superposed plates.

with channel sides in the 0.35–2.8 mm range showed that 0.7 mm is a sufficiently low dimension for SRM intensification, so the channel size was set at 0.7 mm in this work. This simple model was scaled up in order to obtain a more realistic description of the microreactor. The new geometry consisted of two superposed steel plates resulting in a block which is 1/10 of the microreactor that is now under construction by stacking and welding of micromachined steel plates. Each sheet has 10 square parallel microchannels of 0.70 mm of side and 20 mm of length which are separated by 0.30 mm fins. This model was used to investigate the effect of the flow arrangement: parallel cocurrent or cross flow, as illustrated in Fig. 2A and B, respectively. In this case, one sheet is for the reforming process and the other for the combustion reaction. After the generation of the geometries, their physical space was divided into an unstructured mesh with a number of control volumes where the governing equations are solved iteratively until the established criteria of convergence are fulfilled.

Catalytic reactions were modelled considering the microchannels walls as sources of products and sinks of reactants. It has been assumed that a thin and homogeneous layer of an appropriate catalyst has been uniformly deposited onto the channels walls. In the case of the SRM channels, a typical Ni catalyst and the rate expressions (Eq. (1), P in MPa) of the Temkin's group [9] were adopted.

$$r_{\text{SRM}} \left[\frac{\text{mol}_{\text{CH}_4}}{\text{s} \cdot \text{m}_{\text{Ni}}^2} \right] = \frac{3.06 \times 10^5 \exp(-15.6 \times 10^3/T)}{1 + 0.5(P_{\text{H}_2\text{O}}/P_{\text{H}_2}) + 20P_{\text{CO}}} \times P_{\text{CH}_4} \quad (1)$$

The water-gas shift (WGS) reaction was also considered and the CO_2 concentrations at the microchannels outlet were very close to the equilibrium values, which is a typical behaviour of SRM [10]. For the combustion of methane in air, a Pd/Al₂O₃ catalyst and

the kinetic expression reported by Kolaczowski and Serbetcioglu [11] were selected. As concerns the homogeneous combustion of methane, simulations carried out with the rate expression reported by Gosiewski et al. [12] for the combustion of lean methane-air mixtures in monolithic inserts revealed that this process is not significant under the conditions prevailing in this work. This can be explained by the very large surface-to-volume ratio of the microchannels (5714 m^{-1}) since whereas the homogeneous reaction takes place in the void space of the channel the heterogeneous reaction develops on the inner catalytic walls. Kinetic equations were implemented in the CFD codes in a catalyst weight basis; then, a given loading of catalyst is assumed to be deposited onto the walls. The loading was set at 1 mg/cm^2 for the Pd/Al₂O₃ catalyst, whereas it was changed between 0.5 and 8 mg/cm^2 for the SRM catalyst.

The validity of the continuum model was checked through the estimation of the Knudsen number. Values were between 6×10^{-5} and 5×10^{-3} which are sufficiently low as to assure the validity of the Navier–Stokes equations for the systems considered in this work [13]. As concerns the Reynolds numbers, maximum values were about 100 and 800 for the SRM and the methane combustion channels, respectively, so the flow regime is laminar. All the simulations were carried out considering steady state, adiabatic external walls and constant total pressure of 1 atm at the channels outlet. Simulations were performed with ANSYS CFX software, which is based on the finite volume method for spatial discretization of the governing Navier–Stokes equations [14]. Criteria of convergence were based on the residuals and the imbalance level of the conservation equations. Simulations were performed on a Dell Precision PWS690 workstation running MS Windows XP® × 64 with an available RAM of 16.0 GB. The duration of the simulations varied between 24 and 72 h depending on the complexity of the case.

3. Results and discussion

3.1. Four channels model

This model has been used to investigate the effects of the gas streams flow rate, SRM catalyst loading and steam-to-carbon (S/C) ratio. The inlet temperature of the gas streams has been set at $600\text{ }^\circ\text{C}$, a typical SRM inlet temperature [15], and the flow was arranged in parallel cocurrent. In the case of the fuel, the inlet composition has been established at 2% (wt.) CH₄ in air because, for safety reasons, it is considered more suitable for practical applications.

The effect of the gas streams flow rates has been examined for S/C molar ratio of two and SRM catalyst loading of 4 mg/cm^2 . Values of the reforming gas hourly space velocity (GHSV)_{SRM} in the $10,000\text{--}30,000\text{ h}^{-1}$ (STP) range were considered. This is equivalent to contact times of 120–360 ms or feeding each microchannel with $1.6\text{--}4.8\text{ cm}^3/\text{min}$ (STP) of the methane-steam mixture. This is also equivalent to space-times in the range 1.5–4.5 ($g_{\text{catalyst}}\text{ h})/\text{mol}_{\text{methane}}$ or weight hourly space velocities (WHSV) based on the total mass of SRM catalysts and feed mass flow rate of $34\text{--}102\text{ h}^{-1}$. As the methane concentration in the combustion gas stream is low, the flow rate of the methane-air mixture had to be significantly higher than that of the reforming gas. The results of these simulations are shown in Fig. 3A and B where the methane conversion and mean temperature at the SRM channels outlet as well as the selectivity for syngas are plotted against the ratio between the combustion and SRM streams mass flow rates for three distinct values of the (GHSV)_{SRM}. As the fuel flow rate increases, both the SRM conversion and mean outlet temperature increase. The conversion of the CH₄ combustion reaction was complete in all cases. In the range of (GHSV)_{SRM} considered, it was necessary to reach $920\text{--}960\text{ }^\circ\text{C}$ to obtain methane conversions above 96%. To this end,

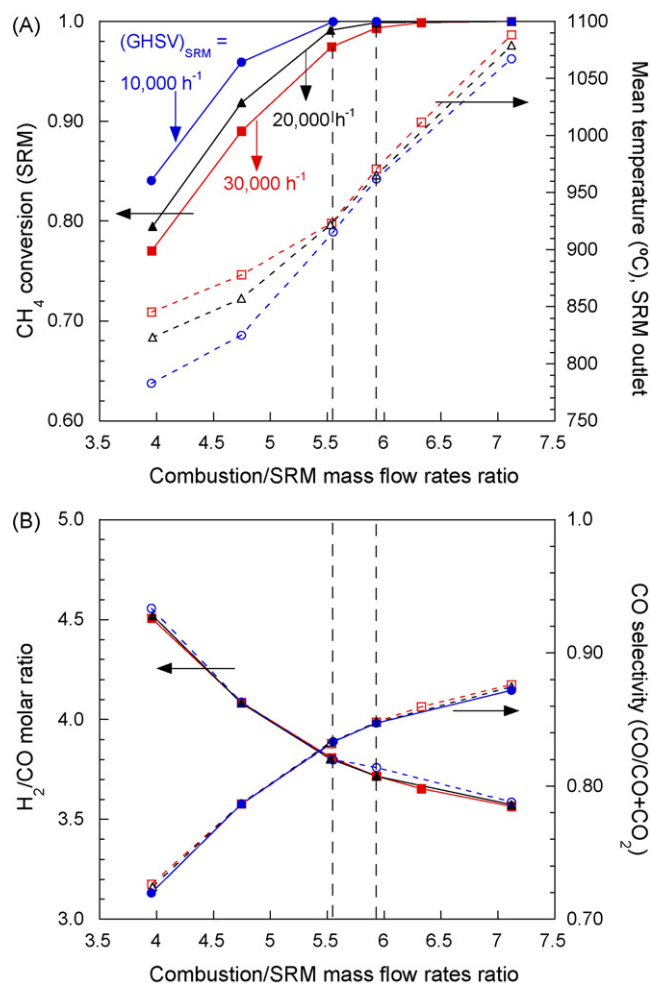


Fig. 3. SRM conversion (filled symbols), mean reforming outlet temperature (open symbols) (A) and syngas selectivity (B) as a function of the ratio between the combustion and SRM streams mass flow rates. Simulations were carried out with the four channels model at S/C=2, 4 mg/cm^2 of SRM catalyst and (GHSV)_{SRM} of 10,000 (circles), 20,000 (triangles) and $30,000\text{ h}^{-1}$ (squares).

the mass flow rate of the combustion stream should be 5.5–6 times higher than that of the reforming gas. Under these operating conditions, the syngas selectivity defined as the CO produced divided by the sum of the produced CO and CO₂ is about 84% on a molar basis (see Fig. 3B). On the other hand, the H₂/CO molar ratio is 3.75 which can be compared with the limiting value of three corresponding to 100% selectivity for syngas that can be obtained through methane steam reforming. CO₂ is formed through the WGS reaction, which is a reversible exothermic process favoured by relatively low operating temperatures. As a matter of fact, it can be appreciated in Fig. 3 that the selectivity for syngas rapidly decreases at temperatures below $900\text{ }^\circ\text{C}$ due to the combined effect of the temperature and methane conversion decrease on the WGS reaction. Indeed, as the methane conversion decreases the water concentration increases which also favours the shift reaction.

The SRM reaction is conducted by the Velocys® group at very low contact times, between $900\text{ }\mu\text{s}$ to 2–25 ms, and temperatures in the $800\text{--}900\text{ }^\circ\text{C}$ range, obtaining methane conversions close to equilibrium. In this case, a very active Rh/MgO–Al₂O₃ reforming catalyst on FeCrAlY felt substrate is used [16,17].

3.1.1. SRM catalyst loading and steam-to-carbon ratio

The influence of the SRM catalyst loading deposited onto the reforming channels walls has been investigated in a series of sim-

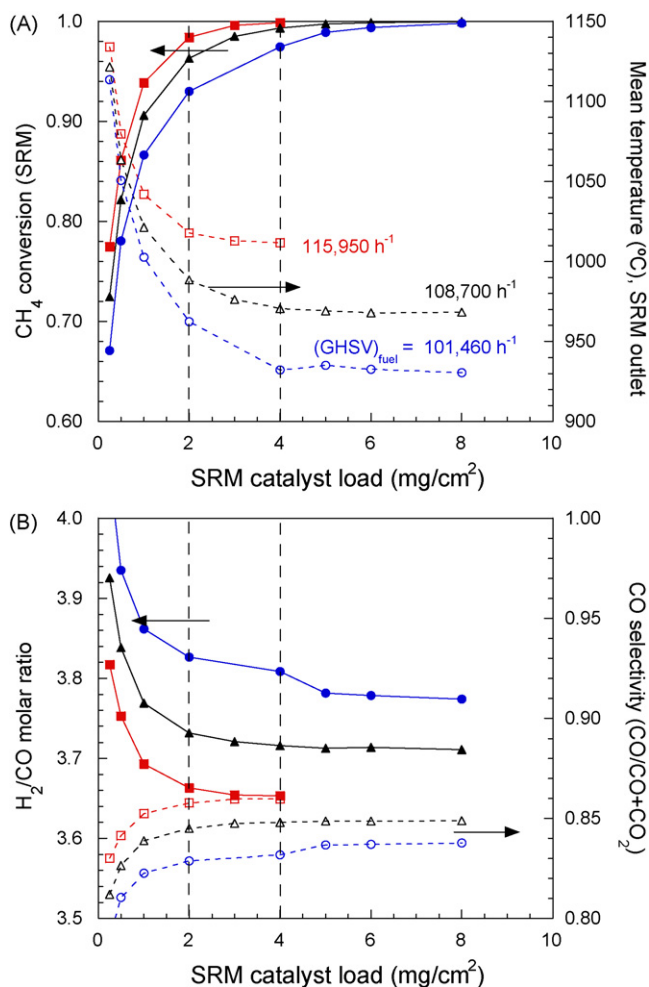


Fig. 4. SRM conversion (filled symbols), mean reforming outlet temperature (open symbols) (A), H₂/CO ratio (filled symbols) and selectivity for CO (open symbols) (B) as a function of the SRM catalyst loading. Simulations were carried out with the 4 channels model at S/C = 2, (GHSV)_{SRM} of 30,000 h⁻¹ and (GHSV)_{fuel} of 101,460 (circles), 108,700 (triangles) and 115,950 h⁻¹ (squares).

ulations run with S/C molar ratio of two and high (GHSV)_{SRM} of 30,000 h⁻¹ (STP). The results are shown in Fig. 4A and B where the methane conversion and mean temperature at the SRM channels outlet as well as the selectivity for syngas are given for three values of the methane-air stream gas hourly space velocity, (GHSV)_{fuel}, in the 101,460–115,950 h⁻¹ (STP) range. This is equivalent to space-times in the range 0.6–0.7 (g_{catalyst} h)/mol_{methane} and ratios between the combustion and SRM streams mass flow rates between 5.5 and 6.25. As can be seen in Fig. 4A, depending on the temperature, it is sufficient with 2–4 mg/cm² of reforming catalyst to obtain methane conversions above 97%. Obviously, the amount of catalyst required decreases as the temperature increases. Whereas at 930 °C a CH₄ conversion of 97% is reached with a catalyst load of 4 mg/cm², the temperature has to be increased up to about 1000 °C to get this conversion level if the load is reduced to 2 mg/cm². On the other hand, at sufficiently high SRM catalyst load, the selectivity for syngas is controlled by the temperature, in accordance with the fact that the WGS reaction is equilibrated. Thus, the H₂/CO ratio decreases from 3.82 at 930 °C to 3.65 at 1020 °C. Nevertheless, at low values of the catalyst load, the selectivity for syngas is particularly poor in spite of the high temperatures prevailing in these conditions. This is due to the low methane conversions achieved. As little heat is consumed by the SRM, the heat released by the complete combustion of methane in the neighbouring channels rises

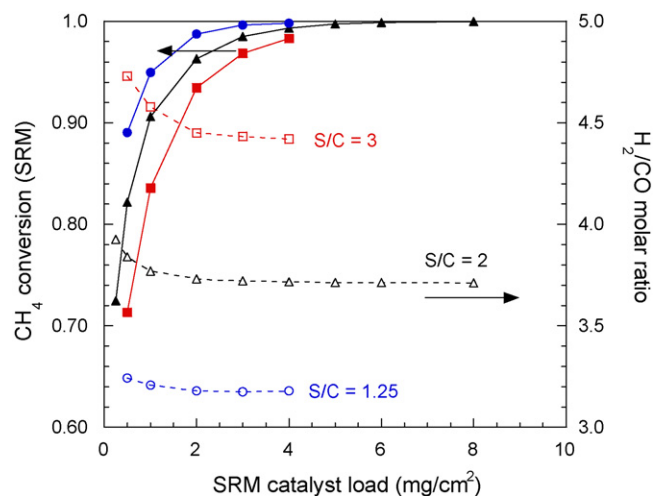


Fig. 5. SRM conversion (filled symbols) and H₂/CO ratio (open symbols) as a function of the SRM catalyst loading. Simulations carried out with the four channels model at S/C of 1.25 (circles), two (triangles) and three (squares).

the reforming gas temperature; however, water is in considerable excess with respect to CO, which favours the shift reaction to CO₂ and H₂.

As concerns the steam-to-carbon (S/C) ratio, its value may vary in SRM between about 1.5 and 4.5 depending on the desired final product (e.g. H₂, methanol, hydrocarbons, etc). There is a trend to decrease the S/C ratio [9], although it should be noted that the potential for carbon formation increases at lower S/C. In this work, three values of the S/C ratio, 1.25, 2 and 3 have been considered. Due to the different concentration, the volumetric feed flow rate was adjusted for each simulation in order to maintain constant the space-time based on the total mass of SRM catalysts and methane fed to the microchannels. As shown in Fig. 5, the steam-to-carbon ratio has a strong influence on the syngas selectivity which is greatly reduced as water in excess increases; thus the H₂/CO ratio decreases from 4.4 for S/C = 3 to only 3.2 at S/C = 1.25. This is of course due to the fact that water promotes the WGS reaction. Regarding the conversion of SRM, higher values were obtained at decreasing S/C ratios in spite of the facts that the space-time was constant and that temperature differences were not significant since the fuel mass flow rate was the same in all the simulations of this series. On the other hand, at the high SRM temperatures involved, the equilibrium conversion is similar in all cases and close to one, so the influence of the S/C ratio on the SRM conversion lies on the dependence of kinetics on the concentrations. In this regard, according to the adopted kinetic expression for the SRM on Ni [9] (see Eq. (1)), the reaction rate is first order with respect to CH₄ and decreases with the concentration of water. Therefore, the SRM rate increases as the S/C ratio decreases which explains the trend of the conversion results depicted in Fig. 5.

3.1.2. Case analysis

This section has been included with the aim of illustrating some of the capabilities of CFD as a simulation tool through the analysis of the most relevant results of a typical case. Simulation conditions were as follows: (GHSV)_{SRM} = 30,000 h⁻¹ (STP), S/C molar ratio = 2, SRM catalyst loading = 2 mg/cm² and ratio between the combustion and SRM mass flow rates of 6. Fig. 6A and B show the solid steel block and gas streams temperatures, respectively. Temperatures can be read in the coloured scale included in each figure. As can be seen, the maximum difference of temperature in the solid block is limited to less than 25 °C. The highest temperatures in the solid (close to 1010 °C) are reached near the fuel channels entry, which in this case are the two on the top (Fig. 6A). Fig. 6B shows the

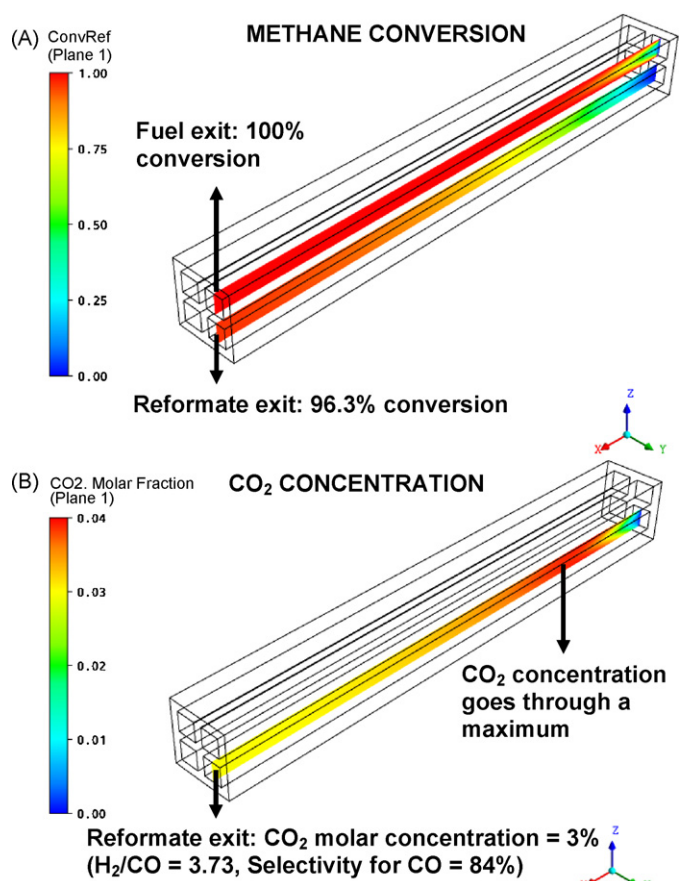
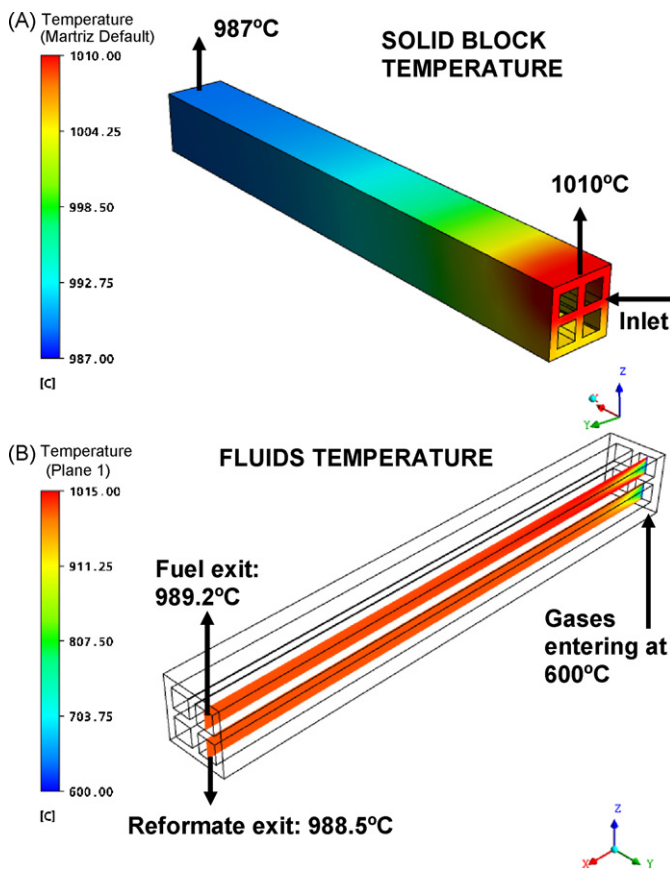


Fig. 6. Solid block (A) and fluids temperature (B) from a simulation carried out with the four channels model. $(GHSV)_{SRM} = 30,000 \text{ h}^{-1}$, S/C molar ratio = 2, SRM catalyst loading = 2 mg/cm^2 and ratio between the combustion and SRM mass flow rates of six.

Fig. 7. Methane conversion (A) and CO₂ molar fraction (B) for a simulation carried out with the four channels model. Simulation conditions as for Fig. 6.

temperature of the gaseous streams in a plane parallel to the fluids flow in the centre of the microchannels. It can be appreciated that the fuel passes in a very short reactor distance from the inlet temperature of $600 \text{ }^\circ\text{C}$ to a maximum temperature of about $1015 \text{ }^\circ\text{C}$, which means that CH₄ combustion on Pd is a very fast reaction. Then, the fuel slightly cools down to the mean outlet temperature of $989.2 \text{ }^\circ\text{C}$. Regarding the reformat, it is rapidly heated to about $988 \text{ }^\circ\text{C}$ resulting afterwards in an almost isothermal flow along the microchannels. The exit temperatures of both, the fuel ($989.2 \text{ }^\circ\text{C}$) and SRM ($988.5 \text{ }^\circ\text{C}$) are very close.

Fig. 7A shows the evolution of the methane conversion for the combustion and SRM reactions. As above-mentioned, the combustion is very fast and complete conversion is reached in a very short reactor distance coinciding with the maximum fuel stream temperature. The increase of the SRM conversion is much more gradual. Thus, CH₄ conversion is about 75% at half channel length and the exit conversion is 96.3% in this case. Fig. 7B shows the evolution of the molar fraction of CO₂ in the reformat. As can be seen, the concentration of this product goes through a maximum (4% mol/mol) near the entry, where the CH₄ conversion is relatively low and the excess of water favours the CO shift reaction. Then, as the methane and water conversion increase, the WGS equilibrium is displaced resulting in lower CO₂ concentrations (3%) at the reforming gas outlet. This exit value gives rise to a H₂/CO ratio of 3.73 and selectivity for syngas of 84%. It should be noted that as the reformat flow is almost isothermal in great part of the channel length, the shift equilibrium is controlled by the availability of water, which depends on the methane conversion.

3.2. Two plates model

In this model, a total of 20 microchannels are considered, resulting in a high number of volume elements which greatly increases the computation time exigency. In order to check the reliability of our approach, a first series of simulations was performed with the two plates model in parallel cocurrent flow arrangement to compare the results with that of the four channels model. Simulation conditions were as for the preceding section although several values of the SRM catalyst loading were considered. The results show a reasonably good accordance between the models, although the two plates system gives SRM mean outlet temperatures slightly higher ($10\text{--}30 \text{ }^\circ\text{C}$). This results in higher SRM conversion and syngas selectivity when using the two plates model; differences, however, were not significant. For example, at sufficiently high catalyst load, selectivity for CO was 85% with the four channels model and 86% with the two plates. A similar trend was found when using this approach to model the integration of the steam reforming and combustion of methanol in a microreactor [18].

The two plates system has been used to study the effects of the flow arrangement by comparing the parallel cocurrent and cross flow dispositions. Simulation conditions were $(GHSV)_{SRM} = 30,000 \text{ h}^{-1}$ (STP), S/C molar ratio = 2, SRM catalyst loading = 2 mg/cm^2 and ratio between the combustion and SRM mass flow rates of six. The results are shown in Fig. 8A and B, where coloured scales for both, solid block temperature and CH₄ conversion at the SRM channels outlet, are included. As expected from the different flow arrangements, the two dispositions differ in the distribution of temperatures. In the case of the parallel flow, the temperature in the block is very homogeneous, with a mean value

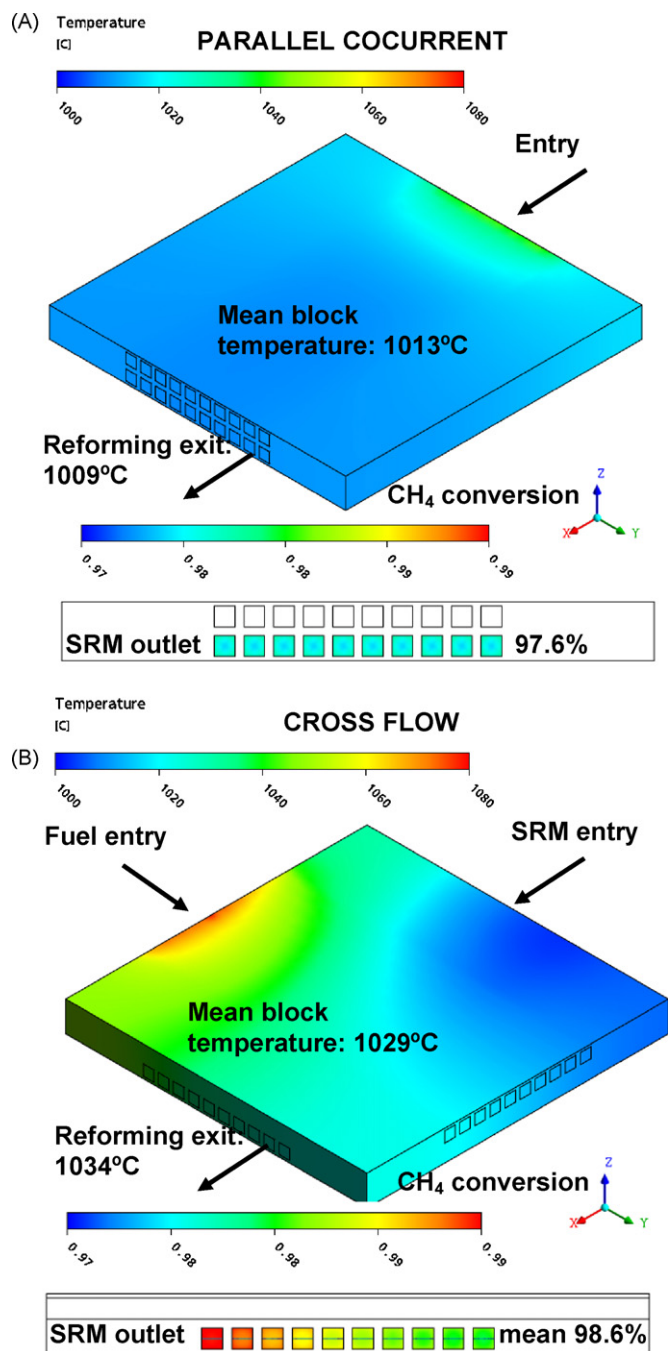


Fig. 8. Comparison between the performance of the parallel cocurrent (A) and cross flow (arrangements). Simulations carried out with the two plates model.

of 1013 °C and a maximum temperature difference of 40 °C. The reformat exits the channels at 1009 °C and the CH₄ conversion is the same (97.6%) at the outlet of the 10 reforming microchannels. In contrast, the cross flow arrangement gives higher mean block temperature (1029 °C) and maximum temperature difference (80 °C). The mean exit temperature of the reformat is also higher (1034 °C) which illustrates the better heat transfer characteristics of this arrangement compared with the parallel cocurrent flow. Nevertheless, there is a smooth distribution of temperatures, so the channels that are closer to the fuel entry are hotter and, as shown in Fig. 8B, the SRM conversion at the outlet is higher as well. Differences are very small ranging between 98% and 99%, leading to a mean methane conversion of 98.6%, which is only 1% higher than the one achieved with the parallel cocurrent flow arrangement.

Additional simulations have shown that the thermal behaviour and SRM performance of the cross flow arrangement is intermediate to that of the parallel cocurrent and countercurrent dispositions.

4. Conclusions

Computational fluid dynamics (CFD) is an useful tool for modeling and simulation purposes. In this work, two CFD models with different degree of complexity have been developed and used to simulate the integration of the steam reforming of methane (SRM), including the water-gas shift (WGS) reaction, and the combustion of methane in a catalytic microchannel reactor. The study has shown the great potential of microreactors for process intensification. It is possible to achieve SRM conversions above 96% at 930–1,000 °C and 2–4 mg/cm² of catalyst at high gas hourly space velocities (GHSV) in the 10,000–30,000 h⁻¹ range. Under these conditions (S/C=2) the resulting H₂/CO ratio is 3.65–3.84 and the selectivity for CO 83–86%. Selectivity is controlled by the WGS equilibrium. In this regard, selectivity for syngas increases as the temperature and methane conversion increase and the steam-to-carbon ratio in the feed decreases. The results have shown that complete combustion of methane takes place in a very short reactor distance at the channel entry. In this zone the reforming gas is rapidly heated and then, its flow is almost isothermal. Temperature in the solid block is significantly homogeneous, especially for the parallel cocurrent flow arrangement. In the case of the cross flow disposition, maximum temperature differences were generally below 80 °C. The flow arrangement did not affect significantly the reforming performance of the microreactors, so there is no definitive reason based on performance criteria for selecting one of them as the most suitable. Nevertheless, the cross flow arrangement allows an easier feed streams distribution and assembly of the connectors and tube fittings. This is the arrangement selected for the SRM microreformer that is now under construction.

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