

Methane Steam Reforming Reaction Behaviour in a Packed Bed Membrane Reactor

Lemnouer Chibane and Brahim Djellouli

Abstract—In this work, the performance of the reaction of steam reforming of methane carried out in a pd-membrane reactor was theoretically studied and analyzed. In order to determine the optimal conditions leading to the improvement of the hydrogen production from this reaction, an isothermal steady-state model was developed to simulate the operating parameters. The main results show that the model can predict the performance of the membrane reactor. It was found that the conversion of methane was significantly enhanced by the removal of hydrogen from the reaction side under the following conditions: temperature ranging from 580-600°C, pressure=300-600kPa, steam to methane ratio=3 and sweeping ratio=3. Under those conditions, the H₂/CO ratio obtained is satisfactory, a nearly complete conversion of methane and highly hydrogen recovery were achieved.

Index Terms—Hydrogen production, Methane steam reforming, Palladium membrane reactor.

I. INTRODUCTION

In our current life two energy vectors are usually used: electricity and natural gas (as a progressive replacement of petroleum). The latter would be gradually replaced by another one having at least the same qualities: This new vector was identified and accepted, it will be hydrogen. Actually, hydrogen is the most important compound of chemical and petrochemical industries. This is used in various applications, such as the production of certain chemical products, especially methanol, oxo synthesis or in the Fischer-Tropsch process [1, 2] and also for ammoniac synthesis. So, hydrogen is an energy vector which can be stored, and does not generate any pollutants such as carbon dioxide. For all these reasons, vast hopes are placed on this vector. Recently, several works were oriented towards the use of hydrogen as a source of energy for prototypes of vehicles with hydrogen and stationary generators of electricity [3]. So, the hydrogen and its corollary fuel cell have been promoted for some years to the rank of alternative energy. The fuel cell constitutes an extremely important axis of research [4]. Currently, this energy vector is produced by water electrolysis [5], by natural gas reforming [6-8] or by steam reforming of liquid hydrocarbons [9-11]. Moreover, hydrogen can be produced by other means, such as from the biomass [12] or by the solar thermal dissociation of water at high temperature [13]. The reaction of methane steam

reforming appears as an alternative process due to its low raw material cost and the favourable balance energy of the considered reaction especially in membrane reactors. This reaction has been investigated by several studies in conventional reactors [14] or in the new generation of membrane reactors [6, 15-21], and also in fluidized bed membrane reactors [8].

The aim of this contribution is to analyze the steam methane reforming reaction behaviours in a packed bed membrane reactor at moderate temperatures and pressures. The aim is to investigate the adequate operating parameters which allow the shifting of equilibrium in the hydrogen production direction by the immediate evacuation of hydrogen across the membrane at moderate temperatures and other investigated parameters.

II. BRIEF REVIEW

Steam reforming is a highly endothermic process which is carried out by passing a mixture of steam and natural gas over a catalyst [16]. In order to achieve near equilibrium conversion, steam reforming in conventional technology is conducted on a supported nickel catalyst in a multitubular reactor operated at a temperature of 850°C, a pressure ranging from 1.6 to 4.1MPa, and a steam to methane ratios between 2 and 4 [16, 22]. Methane conversion is usually around 78% for it is limited by the thermodynamic equilibrium [16].

The MSR was studied in packed bed reactor [23], conducted under the following conditions: supported nickel catalyst (Ni/Al₂O₃), the catalyst mass=0.012-0.020g, pressure=1atm, temperature=500°C (moderate pressure and temperature) and with steam to methane ratio in the range of 2-6. Under these conditions, the conversion of methane always increases with steam to methane ratio. For the lowest catalytic mass the negative order of water is always observed, even for steam/methane molar ratio as low as 2. This negative effect of the water is due to the relative small number of active sites present in the lower catalyst load used. It was showed that hydrogen and carbon dioxide production increases with S/C ratios while CO production decreases. The explication that was made for the effect of water is that the reaction rate corresponding to the reaction of reforming (CH₄+H₂O=CO+3H₂) has non-monotonic dependence on water partial pressure while the dependence of the global reaction rate (CH₄+2H₂O=CO₂+4H₂) on water partial pressure is a monotonic function. This means that there would be an optimum inlet water concentration that gives a maximum performance in terms of hydrogen and CO₂ selectivity and simultaneously a minimum CO selectivity.

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Supported nickel catalysts are used in industry despite the fact that these catalysts require a high steam to methane ratio (3.0-3.5) in the feed in order to favour the gasification of carbon species and to avoid the carbon formation reaction ($C+H_2O=CO+H_2$) which would lead to the catalyst deactivation [24]. Other metals such as Rh, Ru, Pd, Pt and Co are also active for the methane steam reforming [25].

The methane steam reforming was studied by [26] in a fixed bed tubular quartz micro reactor with Cobalt catalyst. The catalyst showed stability and activity at a feed molar ratio $S/C=2/1$, $W/F=0.35\text{g.h.mol}^{-1}$ and $T=750^\circ\text{C}$. Under these conditions, the catalyst presents the average H_2/CO molar composition and the amount of carbon formed on catalysts after 6 hours of reaction with $S/C=2$ and produced high H_2/CO molar ratios. To enhance the reforming conversion, the nickel catalysts can be used [6]. The mean reaction (MSR) was studied using a nickel catalyst (12wt% Ni/ Al_2O_3) [27]. It was showed that at $T=750^\circ\text{C}$ and a steam to methane ratio equal to 3, a nearly complete conversion was obtained, while only 30% conversion was achieved when the reaction temperature was decreased to 550°C . Since methane is the major compound in natural gas, it is likely that complete conversion of natural gas also requires a reaction temperature of about 750°C . At 750°C , hydrogen concentration reaction equilibrium at about 77%, at the same temperature, the equilibrium composition for CO and CO_2 is 14% and 8.5%, respectively. However, the natural gas steam reforming at $T=760^\circ\text{C}$ with S/C ratio of 3 and $P=1\text{atm}$, showed that the product gas concentration was close to the equilibrium composition. No carbon formation was observed on the used catalysts. It was showed that in general it is not possible to obtain satisfactory conversions of methane at moderate temperatures, because the reforming reaction is endothermic and thermodynamically favoured by a high temperature and a low pressure [19]. Therefore, the development of membrane based separation process could make it possible to increase the conversion in the reforming process [28].

One possible way to liberate this reaction from the equilibrium limitation and thus attain a high methane conversion at lower temperatures is by the use of membrane reactors [16]. Membrane reactors could be used in the process to overcome the equilibrium limitation by selectively removing hydrogen from the reaction zone [29]. The membrane continuously removes the hydrogen produced in the catalytic reaction zone thus pushing the chemical equilibrium and allowing higher methane conversion at a lower temperature [18]. The use of membrane reactors appears to be a possible way to improve hydrogen yield at lower temperatures because the removal of hydrogen from the reaction environment prevents the equilibrium to be achieved [30]. The membranes used in catalytic reactors are generally characterized with a high permeability, a good selectivity of separation and are stable to the temperature of the reaction especially in the presence of gas. Among hydrogen selective membranes, Pd membranes remain the most promising. Pd-based membrane reactors were considered in most of the studies, because they would produce pure hydrogen thus simplifying the conventional operation which includes extensive hydrogen purification

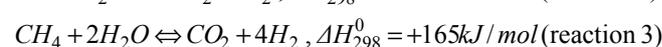
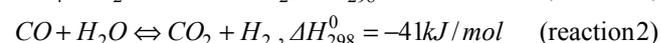
steps [22].

A numerical study [28] of the methane steam reforming in a Pd-based membrane reactor, showed that the membrane thickness plays a major role both in methane conversion and H_2/CO . A membrane thickness, less than 1 micrometer increases conversion even at moderate temperatures, so the reactor could be operated at milder temperatures with high conversion rates at $P=29\text{atm}$. While allowing an enhancement in carbon monoxide production and a reduction of the H_2/CO ratio. A calculation for a modified methane steam reforming process with integrated hydrogen separation was performed by [31]. It was obtained that almost 90% of methane conversion could be achieved below 850°C by using a palladium membrane with a thickness of 50 micrometer.

However, the concept of membrane reactor is complex and depends on a number of factors including reaction pressure, sweeping gas flow rate, permeation zone pressure, feed properties, and steam to carbon ratio.

III. MATHEMATICAL MODEL

Methane steam reforming involves two reversible reactions: the reforming reaction 1 and the Water-Gas-Shift reaction 2. The third one 3 is the global reaction. Reactions I and III are endothermic, but the water gas shift reaction is exothermic. The reaction scheme studied is as follows [15], [19], [21], [32-35]:



The mathematical model is based on the Langmuir-Hinshelwood mechanism, where the reaction rates are given by [7]:

$$r_1 = \frac{k_1}{P_{H_2}^{2.5}} \left(P_{CH_4} P_{H_2O} - \frac{P_{H_2}^3 P_{CO}}{K_1} \right) \text{ (1)}$$

$$r_2 = \frac{k_2}{P_{H_2}} \left(P_{CH_4} P_{H_2} - \frac{P_{H_2} P_{CO}}{K_2} \right) \text{ (2)}$$

$$r_3 = \frac{k_3}{P_{H_2}^{3.5}} \left(P_{CH_4} P_{H_2O}^2 - \frac{P_{H_2}^4 P_{CO_2}}{K_3} \right) \text{ (3)}$$

$$DEN = 1 + K_{CO} P_{CO} + K_{H_2} P_{H_2} + K_{CH_4} P_{CH_4} + K_{H_2O} \frac{P_{H_2O}}{P_{H_2}} \text{ (4)}$$

where r_j ($j=1-3$) are the reaction rates of reactions (1, 2, 3) respectively. k_j is the reaction rate constant defined by Arrhenius $k_j = k_{0j} \exp(-E_j/RT)$, K_j ($j=1-3$) are the constant equilibrium, K_i ($i=CH_4, H_2O, H_2, CO$ and CO_2) are the species adsorption constants of any species i defined

as $K_i = K_{0i} \exp(-\Delta H_i / RT)$.

It is clear that only the first and second reactions are linearly independent. Thus, we can describe this problem by two measures of conversion:

$$X_{CH_4} = \frac{F_{CH_4}^0 - F_{CH_4}}{F_{CH_4}^0} \quad (5)$$

and

$$X_{CO_2} = \frac{F_{CO_2}^0 - F_{CO_2}}{F_{CH_4}^0} \quad (6)$$

The various expressions of the partial pressures for each species are expressed as a function of conversions:

$$P_{CH_4} = \frac{(1 - X_{CH_4})}{\sigma} \quad (7)$$

$$P_{CO} = \frac{(X_{CH_4} - X_{CO_2})}{\sigma} \quad (8)$$

$$P_{CO_2} = \frac{X_{CO_2}}{\sigma} \quad (9)$$

$$P_{H_2O} = \frac{(m - X_{CH_4} - X_{CO_2})}{\sigma} \quad (10)$$

$$P_{H_2} = \frac{(P_{H_2,0} + 3X_{CH_4} + X_{CO_2} - Y_{H_2})}{\sigma} \quad (11)$$

$$\sigma = \frac{(1 + m + P_{H_2,0} + 2X_{CH_4} - Y_{H_2})}{P_r} \quad (12)$$

The membrane considered [6] is a Palladium dense membrane which has a thickness of 20 micrometer and is only permeable to hydrogen. The studied reactor (Fig. 1) has an internal diameter $ID = 0.851$ cm, and a length $l = 5.1$ cm. This reactor consists of two concentric tubes. The external one is a stainless steel tube and the internal tube is an inert membrane. The weight of the Nickel catalyst (12%)/ γ Al_2O_3 used is $W = 11$ g. It is regularly arranged inside the internal tube. The reactants are axially injected inside the inner tube through the catalyst. After the catalytic reaction, the hydrogen formed diffuses through the membrane into the outside tube, where it is evacuated by a sweep gas (or with an inert gas). The simulated operating conditions are presented in Table I.

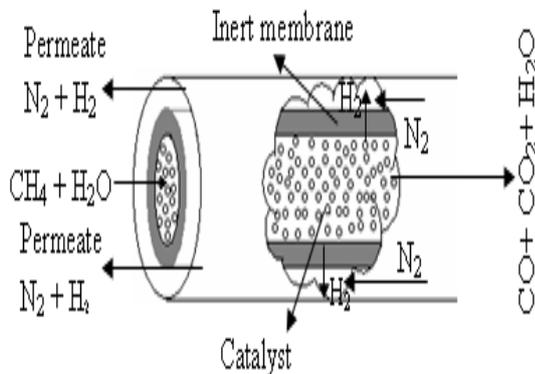


Fig. 1: Schematic representation of membrane reactor

The model which we established is based on the following assumptions: a steady-state condition and the reaction takes place under isothermal and isobaric conditions. The catalyst deactivation by coke formation is negligible. For the membrane, we assume that it is only permeable to hydrogen and there is no boundary layer on membrane surfaces.

TABLE I: KINETIC AND THERMODYNAMIC CONSTANTS OF THE REACTION

Constants	Pre-factors	E or H (kJ/mol)
k_1 (kmol.kPa ^{0.5} /kg.h)	4.225x10 ¹⁶	240.1
k_2 (kmol/kPa.kg.h)	1.955x10 ⁴	67.13
k_3 (kmol.kPa ^{0.5} /kg.h)	1.02x10 ¹⁶	243.9
K_1 (kPa ²)	4.70x10 ¹⁶	224.0
K_2 (-)	1.14x10 ⁻²	-37.3
K_3 (kPa ²)	5.37x10 ¹⁴	186.7
K_{CO} (kPa ⁻¹)	8.23x10 ⁻⁷	-70.65
K_{H_2} (kPa ⁻¹)	6.12x10 ⁻¹¹	-82.90
K_{CH_4} (kPa ⁻¹)	6.64x10 ⁻⁶	-38.28
K_{H_2O} (-)	1.77x10 ⁵	88.68

The hydrogen permeation through the palladium membrane with a thickness δ follows a particular mechanism of molecular hydrogen dissociation and atomic diffusion form. The hydrogen permeated depends not only on the membrane properties but also on a linear function of the driving force [36].

The limiting step is the atomic hydrogen diffusion in the dense membrane and the flux follows the Sieverts' law. The rate of hydrogen permeation can therefore be expressed as a function of the difference in the square root of hydrogen partial pressures on both sides of the membrane.

A permeation law resulting from a mechanism of solution-diffusion, allows us to express the term of permeation by the following equation [28]:

$$F_{CH_4}^0 dY_{H_2} = \frac{2\pi r_m dl}{\delta} Q (P_{H_2,r}^{0.5} - P_{H_2,p}^{0.5}) \quad (13)$$

The permeation coefficient of hydrogen depends greatly on temperature and can be described by an Arrhenius type of equation, as below:

$$Q = Q_0 \exp(-E_p / RT) \quad (14)$$

By introducing the dimensionless form, we get:

$$\frac{dY_{H_2}}{dz} = \frac{2\pi r_m l}{\delta F_{CH_4}^0} Q_0 \exp(-E_p / RT) (P_{H_2,r}^{0.5} - P_{H_2,p}^{0.5}) \quad (15)$$

The pressure of hydrogen in the permeation zone ($P_{H_2,p}$) and inert ratio (I) are defined respectively as:

$$P_{H_2,p} = \frac{Y_{H_2} P_p}{Y_{H_2} + I} \quad \text{with } I = \frac{F_1^0}{F_{CH_4}^0} \quad (16)$$

It is obvious that the hydrogen partial pressure in the permeation zone depends on the sweep ratio. Any increase in the sweep ratio leads to a decrease of the hydrogen partial pressure in the permeation side.

The mass balance in the gaseous phase was given for each component by the expression:

$$\frac{dF_i}{dz} = \rho \Omega \sum_{j=1}^{N_R=3} \eta_j \nu_{ij} r_j \quad (17)$$

For the methane and the carbon dioxide, we can write:

$$\frac{dF_{CH_4}}{dz} = \rho \Omega (-\eta_1 r_1 - \eta_3 r_3) \quad (18)$$

$$\frac{dF_{CO_2}}{dz} = \rho \Omega (+\eta_2 r_2 + \eta_3 r_3) \quad (19)$$

where Ω is the reactor section (m^2), ρ is the catalyst density (kg/m^3) and η_1, η_2, η_3 are the effectiveness factors defined as the ratio of the observed reaction rate and to the reaction rate calculated at external catalytic surface conditions (or at bulk fluid conditions in the absence of external mass transport resistance). Analytical expressions of η are very useful to evaluate the effective reaction rate to be used in mass balances.

$$\eta_i = \frac{\int_0^V r_i(P_s) \rho \frac{dV}{V}}{r_i(P_s^s) \rho} \quad (20)$$

The variations of the conversion of methane and the carbon dioxide along the reactor length are:

$$\frac{dX_{CH_4}}{dz} = \frac{\rho \Omega (\eta_1 r_1 + \eta_3 r_3)}{F_{CH_4}^0} \quad (21)$$

$$\frac{dX_{CO_2}}{dz} = \frac{\rho \cdot \Omega (\eta_2 \cdot r_2 + \eta_3 \cdot r_3)}{F_{CH_4}^0} \quad (22)$$

With, $X_{CH_4} = X_{CO_2}$ at $z = 0$

The mass balance in the catalytic solid is given by :

$$D_{e,CH_4} \frac{d^2 P_{s,CH_4}}{d\xi^2} + \rho R T (-r_1 - r_3) = 0 \quad (23)$$

$$D_{e,CO_2} \frac{d^2 P_{s,CO_2}}{d\xi^2} + \rho R T (+r_2 + r_3) = 0 \quad (24)$$

ξ : The dimensionless radial position in the solid particle.

With the boundary conditions:

$$\frac{d^2 P_{s,CH_4}}{d\xi^2} = \frac{d^2 P_{s,CO_2}}{d\xi^2} = 0 \quad \text{at } \xi = 0 \quad (25)$$

$$P_{s,CH_4} = P_{CH_4} \quad P_{s,CO_2} = P_{CO_2} \quad \text{at } \xi = 1 \quad (26)$$

$D_{e,i}$: is the effective diffusion coefficient, which is estimated for each reaction species as follows [37]:

$$D_{e,i} = \frac{2}{3} r \sqrt{\frac{8RT}{\pi M_i}} \quad (27)$$

r is the radius of pores, M_i is the molar mass for each species i .

The partial pressures of species H_2 , CO and H_2O are related to the pressure of CH_4 and of CO_2 by the following relations:

$$D_{e,H_2} \frac{d^2 P_{s,H_2}}{d\xi^2} = 3D_{e,CH_4} \frac{d^2 P_{s,CH_4}}{d\xi^2} - D_{e,CO} \frac{d^2 P_{s,CO}}{d\xi^2} \quad (28)$$

$$D_{e,CO} \frac{d^2 P_{s,CO}}{d\xi^2} = -D_{e,CH_4} \frac{d^2 P_{s,CH_4}}{d\xi^2} - D_{e,CO_2} \frac{d^2 P_{s,CO_2}}{d\xi^2} \quad (29)$$

$$D_{e,H_2O} \frac{d^2 P_{s,H_2O}}{d\xi^2} = -D_{e,CH_4} \frac{d^2 P_{s,CH_4}}{d\xi^2} - D_{e,CO_2} \frac{d^2 P_{s,CO_2}}{d\xi^2} \quad (30)$$

Integrating (27), (28) and (29) yields:

$$D_{e,H_2} (P_{s,H_2} - P_{H_2}) = 3D_{e,CH_4} (P_{CH_4} - P_{s,CH_4}) - D_{e,CO_2} (P_{CO_2} - P_{s,CO_2}) \quad (31)$$

$$D_{e,CO} (P_{s,H_2} - P_{H_2}) = D_{e,CH_4} (P_{CH_4} - P_{s,CH_4}) - D_{e,CO_2} (P_{CO_2} - P_{s,CO_2}) \quad (32)$$

$$D_{e,H_2O} (P_{s,H_2} - P_{H_2}) = D_{e,CH_4} (P_{CH_4} - P_{s,CH_4}) + D_{e,CO_2} (P_{CO_2} - P_{s,CO_2}) \quad (33)$$

The kinetic and thermodynamic parameters of the reaction are given by [7], as shown in Table II.

TABLE II: OPERATING CONDITIONS AND REACTOR PARAMETERS

Parameters	Values
Temperature ($^{\circ}C$)	500-600
Reaction pressure zone (kPa)	140-600
Permeation pressure zone (kPa)	101
Reactor length (m)	0.036
Tube internal radius (m)	0.475×10^{-2}
Tube external radius (m)	0.85×10^{-2}
Membrane radius (m)	0.475×10^{-2}
Membrane thickness (m)	20×10^{-6}
Methane flow rate (kmol/h)	1.07×10^{-4}
Steam to methane ratio	1-4
CO_2 /methane ratio	0.004
H_2 /methane ratio	0.001
Nitrogen/methane ratio (sweep ratio)	1-4
Weight catalyst (g)	11
Catalyst density (g/m^3)	2355
Pores diameter (m)	30×10^{-10}
Particle diameter (m)	7.5×10^{-3}

IV. RESULTS AND DISCUSSION

The aim of this work is to study the behavior of the reaction of methane steam reforming in membrane reactor (Pd/SS) at moderate temperatures and pressures. The reactor performance was evaluated through the following quantities: conversion of methane, hydrogen recovery Y_{H_2} and H_2/CO ratio. The procedure followed to study that performance begins by the computation of the hydrogen recovery through the palladium membrane (15), and the calculation of the methane and carbon dioxide conversions using the equations obtained from the mass balances in the fluid phase (21) and (22). These equations were solved numerically by the Runge-Kutta method of order 4 [38], with the initial conditions:

$$\text{at } z = 0 : X_{CH_4} = X_{CO_2} = 0$$

The numerical solution requires the knowledge of the effectiveness factors, which are computed from the mass balance equations in the solid catalytic phase (23 and 24). The symmetric orthogonal collocation method is recommended [38]. This method reduces the set of differential equations (23) and (24) to nonlinear algebraic equations. The routine DNEQBF (IMSL package) uses a secant algorithm to solve a system of nonlinear equations and the finite-difference method is used to estimate the Jacobian.

The kinetics of steam reforming reaction present a critical

parameter, which is the hydrogen partial pressure. This parameter is adjustable and we can compute it by trying values from zero to some values that are limited by industrial constraints. Because of kinetic limitations we cannot use zero as a value for hydrogen partial pressure because this will give us infinite kinetics. To overcome this problem, we have to use a small quantity of hydrogen by considering arbitrary low values of the ratio: $F_{H_2}^0 / F_{CH_4}^0$. The adjustable $F_{CO_2}^0 / F_{CH_4}^0$ ratio is determined by varying it from 0.004 to 0.05. We have found that low values of this ratio give high conversions. In the following studies, the optimal parameters are used: $F_{H_2}^0 / F_{CH_4}^0 = 0.001$ and $F_{CO_2}^0 / F_{CH_4}^0 = 0.004$.

A. Effect of Steam to Methane Ratio

An operating parameter has a large effect on methane steam reforming which is the steam to methane ratio (S/C). It is the key parameter in the methane steam reforming reaction. The effect of molar steam to methane ratio on methane conversion was examined by varying the S/C ratio from 1 to 4 under various conditions. At a fixed value of temperature, pressure and sweep ratio, it is obvious that the methane conversion keeps increasing with the steam to methane molar ratio and the H_2/CO ratio increases when the steam to methane ratio increases, but the hydrogen recovery does not change monotonically with increasing the steam to methane ratio. The effect of steam to methane ratio is very remarkable on hydrogen recovery at any temperature, pressure and sweeping gas.

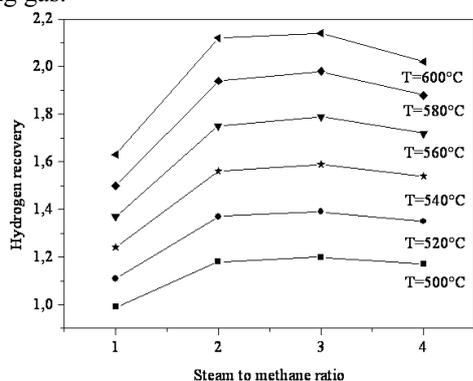


Fig. 2: Effect of steam to methane ratio on hydrogen recovery ($I=3$, $P=140\text{kPa}$, $P_p=101\text{kPa}$)

In general, the hydrogen recovery is a function of the molar steam to methane ratio. Fig. 2 shows that an optimum value of steam to methane ratio must be employed. The maximum hydrogen recovery was found at steam to methane ratio equal to 3 in the investigated conditions. On the other hand we have found that the hydrogen recovery increases with increasing temperature. This is well illustrated in Fig. 2 with the following conditions: sweeping gas=3, pressure=140kPa). From these results, we can conclude that the optimum value of the steam to methane ratio is 3. The choice of this parameter (S/C) is independent of temperature. Some other results obtained with several conditions showed that the hydrogen recovery is dependent on steam to methane ratio. In the same case, it is shown (Fig. 3) that at a fixed temperature, sweep ratio and pressure, the conversion increases with increasing the S/C ratio and H_2/CO ratio increases when the S/C ratio increases. Whereas the H_2/CO

ratio decreases at a fixed temperature, fixed sweep gas and pressure ranging from 140-600kPa.

The results obtained under those conditions ($I=3$, $P=140\text{kPa}$), showed that limited performances were obtained at a low temperature (500°C): the maximum conversion of methane achieved for S/C equal to 3 is 50%, and a high H_2/CO ratio (17.01) was obtained. It should be noted that the conversion predicted under those conditions is in agreement with the conversion of methane obtained by [6]. At the same conditions ($I=3$, $P=140\text{kPa}$, $S/C=3$), an important conversion of methane was achieved (84%) when the temperature was increased to $T=600^\circ\text{C}$. In this case, the H_2/CO ratio was reduced to 5.84 (Fig. 4). The H_2/CO ratio increases when the S/C ratio increases, but decreases when increasing the temperature.

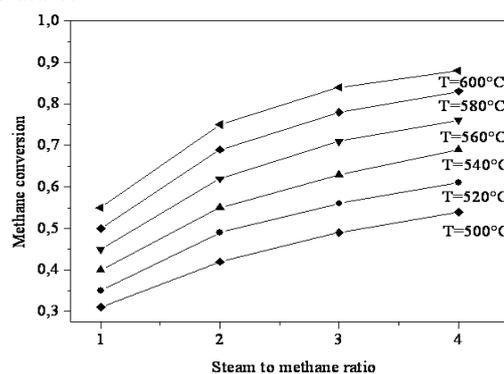


Fig.3: Effect of steam to methane ratio on methane conversion ($I=3$, $P=140\text{kPa}$, $P_p=101\text{kPa}$)

In order to avoid carbon formation, higher steam to methane ratio is recommended [39]. The steam to methane ratio has a positive effect on reforming and water gas shift reactions: the effect is more pronounced if the space velocity is fixed because of the unavoidable increase of the overall inlet flow rate and the consequent reduction of residence time with steam to methane ratio at a fixed methane inlet feed [20], [30]. At the same time, increasing steam to methane ratio reduces the hydrogen partial pressure in the reaction zone and then the pure hydrogen flow rate recovered. We can see that, at a fixed methane inlet ($1.07 \times 10^{-4} \text{ kmol.h}^{-1}$) and a steam to methane ratio varying from 1 to 4 under the following conditions ($T=500^\circ\text{C}$, $I=3$ and $P=140 \text{ kPa}$), the methane conversion changed from 31% to 54%. On the other hand, the hydrogen recovery increases from 0.99 to 1.20 for a S/C: ratio varying from 1 to 3, and decreases to 1.17 for S/C equal 4. This may be due to the existence of a radial distribution of the hydrogen concentration [6]. The increase of the steam to methane ratio beyond 3 increases the hydrogen production by favouring the reforming reaction. But, we expect a dilution of the hydrogen in the reaction zone caused by the excess steam which decreased the hydrogen recovery. Therefore, a high steam to methane ratio allowed a better performance in terms of reaction (methane conversion), but a worse one in terms of separation (hydrogen recovery). So, an intermediate value of S/C ratio should be set as a suitable condition between the opposite effects, checking that coke formation is prevented in the reaction zone. It is noteworthy that, methane steam reforming usually proceeded in the presence of an excess of steam to prevent the carbon deposition over the catalyst surface and to enhance the steam reforming [6].

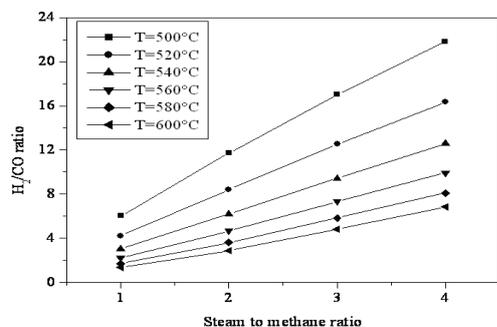


Fig. 4: Effect of steam to methane ratio on H₂/CO ratio (I=3, P=140kPa, P_p=101kPa)

B. Effect of Sweeping Gas

The sweeping gas flow rate has a significant effect on the membrane reactor. Its increase is expected to enhance the methane conversion owing to the decrease of the hydrogen partial pressure on the permeation side. In this study, the effect of sweeping gas was examined via a sweep ratio (I) defined as the ratio of the sweeping flow rate to that of the methane inlet (constant methane inlet flow rate). At fixed temperature, pressure reaction side, pressure permeation side and steam to methane ratio, both conversion of methane and hydrogen recovery increase, but the H₂/CO ratio decreases when sweep ratio and temperature increase. For example (Fig. 5, 6 and 7), at a moderate pressure and temperature (P=140kPa, T=500°C) and steam to methane ratio equal to 3, the methane conversion achieved is only 50%, the hydrogen recovery is 1.2 and a higher H₂/CO ratio (17.01) is obtained. The results showed that the increase of the ratio of the sweeping gas to the inlet methane flow rate from 1 to 4 and at constant permeation pressure (P_p=101kPa) and T=500°C, the conversion of methane boosted from 37% to 53% and the hydrogen recovery decreases from 0.5 to 1.46. According to [30], the improvement in methane conversion results from the higher reaction rate due to hydrogen removal from the reaction zone. This is due to the reduction of hydrogen partial pressure in the permeate side, increasing the driving force for permeation and resulting in the higher rates of hydrogen removal from the reaction zone [20]. The same results were obtained with other conditions (P=200-600kPa, S/C=1-2) and satisfactory results were obtained under elevated pressure and sweep ratio. For example, at T=580°C, steam to methane ratio equal to 3 and sweep ratio equal to 4, the conversion of methane obtained is 97% with a low H₂/CO ratio (2.73). Under the same conditions the hydrogen recovery achieved is 3.72. This may be explained by the favourable effect of the pressure on reaction performances. Furthermore, important results (Table III) were obtained under the following conditions: S/C=3, T=580°C, I=4 and P=400-600kPa. A nearly complete conversion and an important hydrogen recovery were obtained. The H₂/CO ratio is satisfactory. We note that, the optimum ratio varied from 0.7 to 3 [28]. According to [21], the hydrogen flux through a palladium membrane depends on the pressure difference between the reaction side and the permeation side. A larger quantity of sweep gas, that decreases the partial pressure of hydrogen on the permeation side results in the higher hydrogen separation rate and methane conversion. An extremely high methane conversion could be obtained at a

temperature of 580°C. However, this high conversion (94%-97%) could only be achieved at a high sweep ratio (equal to 4) and a pressure varying between 400-600kPa, but the employment of a large quantity of sweep gas is economically undesirable in the industrial process [6, 40], and causes problems of hydrogen separation from the sweep gas (dilution problems) [30].

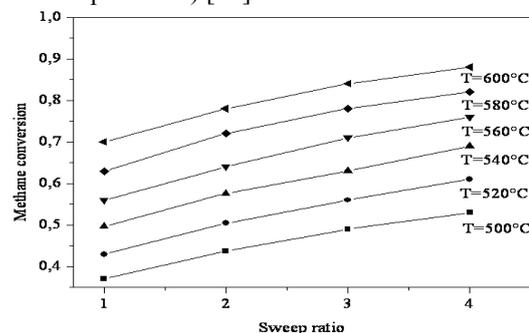


Fig. 5: Effect of sweep ratio on methane conversion (S/C=3, P=140kPa, P_p=101kPa)

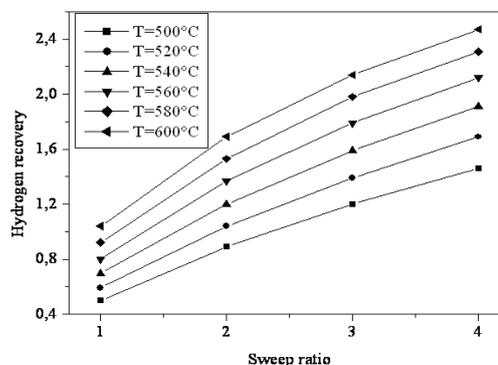


Fig. 6: Effect of sweep ratio on hydrogen recovery (S/C=3, P=140 kPa, P_p=101kPa)

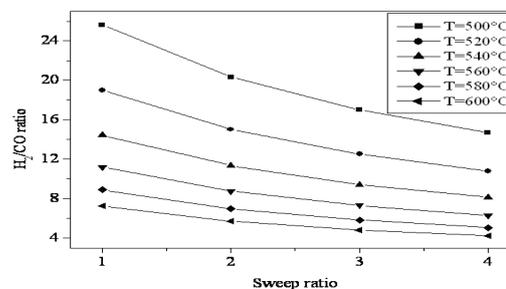


Fig. 7: Effect of sweep ratio on H₂/CO ratio (S/C=3, P=140kPa, P_p=101kPa)

TABLE III: REACTION PERFORMANCE AT: S/C=3, T=580°C, I=4

Reaction pressure side	Methane conversion	Hydrogen recovery	H ₂ /CO ratio
400	0.94	3.48	3.10
500	0.96	3.63	2.87
600	0.97	3.72	2.73

C. Effect of Temperature

The reaction temperature plays an important role in the reactor performance via thermodynamics and kinetics. At fixed pressure, steam to methane ratio and sweep ratio, both conversion and hydrogen recovery increase with the temperature increasing. The H₂/CO ratio also decreases with increasing temperature (Fig. 8 to 11). When the temperature is 500°C, the reforming reaction could not reach a satisfactory conversion level (50%) due to the relative low

methane steam reforming reaction kinetic. We have found that the hydrogen recovery is about 1.20 (at S/C=3, I=3 and P=140kPa). The hydrogen removal through the membrane can only slightly shift the reforming equilibrium. As the temperature increases, the hydrogen permeation rate through the membrane as well as the reaction rate increase resulting in the increase of methane conversion. At temperatures ranging from 520 to 600°C, membrane separation can result in a great improvement in the steam reforming equilibrium. For example, at a total pressure of 140kPa, a reaction temperature of 520°C, and a molar steam to methane ratio of 3, a methane conversion of 56% was achieved. Under the following operating conditions: S/C=3, P=140kPa and T=540°C-600°C, the achieved methane conversion was 63%-84% and the H₂/CO ratio was reduced from 9.45 to 4.82. According to [20], when the temperature increases, the hydrogen rate through the membrane as well as the reaction rate increase resulting in the increase of methane conversion. At 600°C, the conversion in a packed bed membrane reactor is 84%. The results showed that, in the case of a Pd/SS reactor, the temperature could not allow to achieve a high performance. The Pd-based membrane has limitations that have restricted its commercial use. Key limitations include embrittlement and thin films that are free of cracks or pinholes. According to [17, 33], palladium membranes are not suitable for this kind of high temperature reactions, because the formation of carbon filaments and pinholes during the reaction destroys the metallic layer.

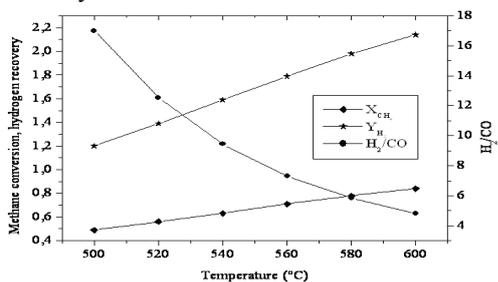


Fig. 8: Effect of temperature on reaction performance (I=3, S/C=3, P=140 kPa, PP=101kPa)

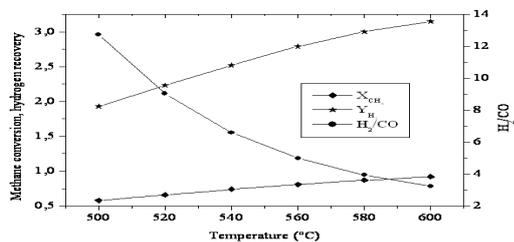


Fig. 9: Effect of temperature on reaction performance (I=3, S/C=3, P=300 kPa, PP=101kPa)

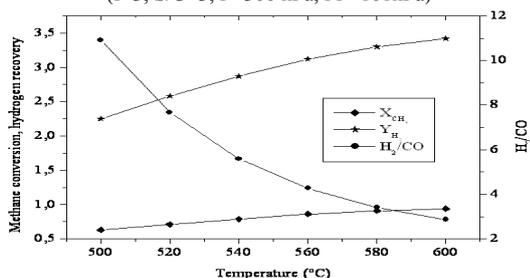


Fig. 10: Effect of temperature on reaction performance (I=3, S/C=3, P=400 kPa, PP=101kPa)

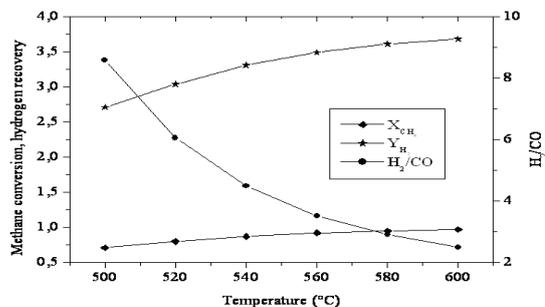


Fig. 11: Effect of temperature on reaction performance (I=3, S/C=3, P=600 kPa, PP=101kPa)

The study of the reaction of methane steam reforming conducted at low temperature (500°C) and under conditions of S/C=3, I=3, P=140kPa showed a great limitation for obtaining high performance in term of conversion. It should be noted that the conversion obtained in the investigated conditions was only 50%. It should be noted also that the value of methane conversion obtained in the fixed bed reactor operating under the same conditions was about 30%. It is shown that in general it is not possible to obtain a satisfactory conversion of methane at a temperature below 500°C. We can explain the inefficiency of methane steam reforming conducted in on Pd-membrane reactor via effectiveness factors whose evolution is shown in Fig. 12.

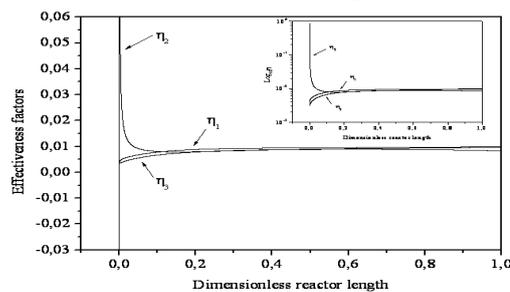


Fig. 12: Evolution of effectiveness factors along the reactor (T=500°C, I=3, S/C=3, P=600 kPa, PP=101kPa)

The effectiveness factor of the Water-Gas-Shift reaction (η_2) shows a behaviour different from that of the reforming reaction (reaction 1) and that of the global reaction (reaction 3). It is to be noted that at the reactor entrance, the effectiveness factor η_2 changes from negative to positive values. This could be explained by the fact that the reaction inside the catalytic particle is reversed from the formation of carbon dioxide and hydrogen back to the formation of carbon monoxide and water. This displacement of the equilibrium is caused by the presence of a small amount of hydrogen and carbon dioxide at the starting of the chemical reaction. After this trend, the effectiveness factor η_2 decreases along the reactor reaching positive values of 0.008 above which the results show continuity and stability with small variations up to a final value of 0.01. The effectiveness factor value of the reforming reaction η_1 and that of the global reaction η_3 increase monotonously along the reactor (final values respectively 0.01 and 0.009), favouring the reaction in the hydrogen production direction. In order to have an idea about the behaviour of the reaction and its composition, the evolution of the partial pressures of the various reaction species along the reactor were determined (Fig. 13); under investigated conditions the plots of pressure for the different

reaction species show a small sharp increase or decrease at the entry of the reactor at about 20% of the reactor length before leveling off. This could be due to the removal of hydrogen via the Pd-membrane. It should be noted that under the same conditions the hydrogen recovery is about 1.2. It is obvious that the various profiles of pressures progress slowly in the direction of hydrogen production.

D. Effect of Reaction Zone Pressure

At fixed temperature, steam to methane ratio and sweep ratio, an increase of methane conversion was predicted when the reaction pressure increased. The hydrogen recovery increases and the H₂/CO ratio decreases when the pressure increases. A nearly complete conversion was obtained under the following conditions: S/C=3, I=3, P=140–600kPa and T=580°C. At these conditions, the conversion of methane changes from 78% up to 95%, the hydrogen recovery changes from 1.98 to 3.61 and the H₂/CO ratio reduces from 5.84 to 2.91 (Fig. 14). The H₂/CO ratio is accepted for P=500kPa and T=600°C (respectively 3.11, 2.91). So, the influence of pressure reaction on performances under the investigated conditions (S/C=3, I=3, T=600°C) are represented in Fig. 15. Increasing the reaction pressure from P=140 to 600kPa resulted in a monotonic methane conversion enhancement corresponding to an increase from 84% to 97%, the hydrogen recovery changes from 2.14 to 3.68 and the H₂/CO value reduced from 4.82 to 2.50. It is noted that the H₂/CO ratio is accepted for P=400kPa, 500kPa and 600kPa (respectively 2.87, 2.64, 2.53). When the pressure increases, both conversion of methane and hydrogen recovery increase. On the other hand, a high reaction pressure also increases the driving force for the products permeation, especially for hydrogen recovery resulting in an enhancement of the methane conversion [41]. When the steam to methane ratio S/C=3, I=4, P=140-600kPa and T=580°C, a nearly complete conversion (Fig. 16) is obtained (82%-97%), the hydrogen recovery changes from 2.31 to 3.72 and the H₂/CO ratio reduced from 5.08 to 2.73. The H₂/CO is accepted for P=400 kPa, 500kPa and 600kPa (respectively 3.10, 2.87, 2.73). The results showed that the reaction pressure has a positive effect on the membrane reactor performance. Increasing the pressure allows a higher rate of hydrogen to be recovered in the permeation zone. Under the following conditions: S/C=3, I=4, P=140-600kPa and T=600°C, the conversion of methane (Fig. 17) changes from 88% up to 98%, the hydrogen recovery from 2.47 to 3.76 and the H₂/CO ratio reduced from 4.23 to 2.38. The H₂/CO is accepted for P=300, 400, 500 and 600kPa (respectively 2.95, 2.66, 2.49, 2.38).

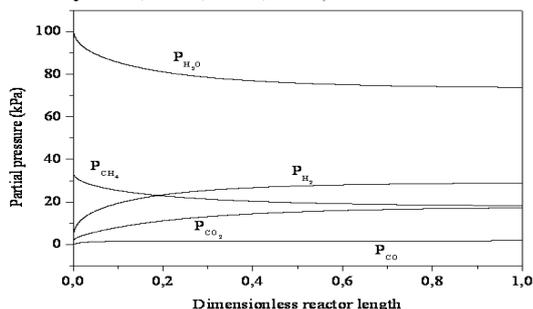


Fig. 13: Component partial pressure profiles along the reactor (T=500°C, I=3, S/C=3, P=600 kPa, PP=101kPa)

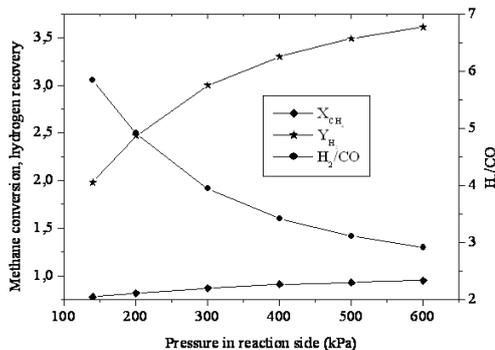


Fig. 14: Effect of pressure on reaction performance (I=3, S/C=3, T=580°C, PP=101kPa)

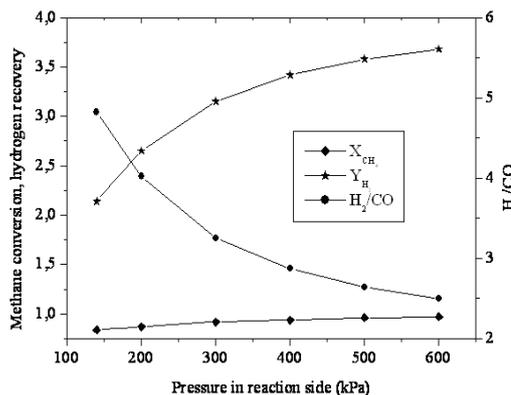


Fig. 15: Effect of pressure on reaction performance (I=3, S/C=3, T=600°C, PP=101kPa)

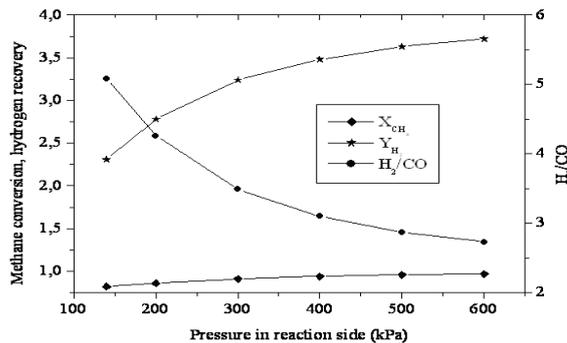


Fig. 16: Effect of pressure on reaction performance (I=4, S/C=3, T=580°C, PP=101kPa)

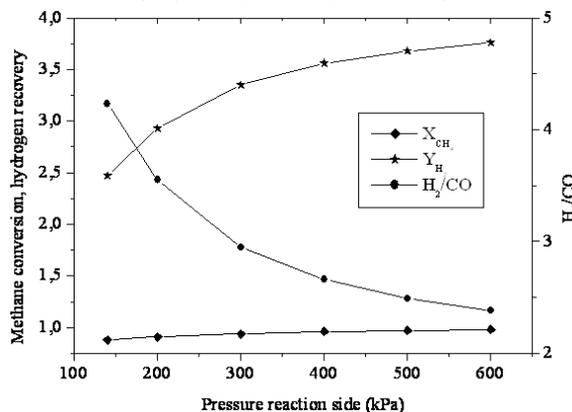


Fig. 17: Effect of pressure on reaction performance (I=4, S/C=3, T=600°C, PP=101kPa)

V. CONCLUSIONS

In the present work, a mathematical model approach for the simulation of a packed bed membrane reactor has been developed. This study allowed concluding that the main

results showed that the reaction of steam methane reforming which was studied in the Pd/SS reactor had some limitations in terms of performance. This performance is characterized by a conversion ranging between 50% and 71%, which was obtained at 500°C, at a pressure ranging from 140 to 600kPa, with a steam to methane ratio of 3 and a sweep gas ratio of 3. Although the reactor operated at a temperature of 500°C, it gave a significant hydrogen recovery. These results showed that a good performance, in terms of conversion and H₂/CO ratio, could not be obtained under the above conditions. Moreover, great values of the H₂/CO ratio were obtained. The conversion of methane can be improved up to 78% using a temperature of $T=580^{\circ}\text{C}$. At this temperature and a reaction pressure of 600kPa, the following performances are possible: a nearly complete conversion of methane (95%), an important hydrogen recovery and a H₂/CO ratio relatively low (2.91). Those performances could also be obtained by increasing the temperature up to 600°C and reducing the pressure from 600kPa to 400kPa. Other results (conversion of methane=92%, hydrogen recovery=3.15 and H₂/CO=3.25) were also obtained under the following conditions: steam to methane ratio=3, sweeping gas flow rate=3, pressure=300kPa and temperature=600°C. This makes an important difference for the hydrogen production under the investigated conditions.

VI. NOMENCLATURE

ΔH	Heat of reaction, kJ/mol
D_e	Effective diffusion coefficient, m ² /s
E_p	Hydrogen permeation activation energy, kJ/mol
F	Molar flow, kmol/h
I	Sweep ratio
K	Equilibrium constant
k	Rate constant
l	Reactor length, m
M	Molar mass
m	Steam to methane molar ratio
P	Partial pressure, kPa
Q	Permeation coefficient of hydrogen, mol/m.s.kPa ^{0.5}
Q_0	Pre-exponential factor, mol/m.s.kPa ^{0.5}
R	Ideal gas constant, 8.314 J/mol.K
r_p	Particle radius, m
r	Reaction rate, kmol/kg _{cat} .h
r_m	Membrane radius, m
T	Temperature, K
W	Catalyst weight, g
X_{CO_2}	Carbon dioxide conversion
X_{CH_4}	Methane conversion
Y_{H_2}	Hydrogen recovery
z	Axial position along the reactor, m
i	Reaction species i
j	Reaction j
p	Permeation side
r	Reaction side
s	Solid
η	Effectiveness factor

ρ	Catalyst density, kg/m ³
δ	Membrane thickness, m
ζ	Radial position dimensionless in solid particle
Ω	Reactor section, m ²
ID	Internal diameter, m
MSR	Methane steam reforming
S/C	Steam to methane ratio

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