Oxidative Coupling of Methane: A Comparison of Different Reactor Configurations

Aitor Cruellas, Tommaso Melchiori, Fausto Gallucci, and Martin van Sint Annaland*

A quantitative comparison of the performance of the most common reactor configurations proposed for the oxidative coupling of methane (OCM) is made on the basis of numerical calculations with phenomenological reactor models. The configurations that are analyzed can be divided into two main categories, namely, packed-bed reactors (including conventional packed beds with external cooling, packed-bed membrane reactors, and adiabatic packed beds with post cracking) and fluidized bed reactors (bubbling fluidized bed reactor, circulating fluidized bed reactor, and fluidized bed membrane reactor). The challenges in both configuration types, mainly the heat management in the case of the packed-bed reactors and the low C\textsubscript{2+} yields obtained in fluidized bed reactors, are evaluated and quantified. To ensure a fair comparison, La\textsubscript{2}O\textsubscript{3}/CaO is chosen as the OCM catalyst for all the considered cases, mainly in view of the availability of a comprehensive kinetics model. The results show that, with conventional configurations, it is not possible to achieve high C\textsubscript{2+} yields that are needed to make the process economically viable. However, the results also indicate that the C\textsubscript{2+} yield can be significantly improved by feeding the oxygen distributively along the reactor axial length.

1. Introduction

The increasing oil price compared with the price for natural gas makes interesting to use natural gas as feedstock for the production of hydrocarbons, which is why the production of hydrocarbons from natural gas has become an important research field in the last decades. The two main options for carrying out this conversion: 1) the indirect route through syngas production and Fischer–Tropsch synthesis, which is already implemented in industry and involves many steps and 2) the direct route via the oxidative coupling of methane (OCM).

OCM has been investigated since the 1980s\textsuperscript{[1]} and many papers/patents related to this process can be found in the literature.\textsuperscript{[2]} Although this process is performed in just one step, an industrial scale application has never been reached because of the parallel and consecutive undesired reactions that limit the attainable C\textsubscript{2} yield. The primary reactions of OCM are the following:

\begin{align*}
2\text{CH}_4 + \text{O}_2 &\rightarrow \text{C}_2\text{H}_4 + 2\text{H}_2\text{O} \\
\text{CH}_4 + 2\text{O}_2 &\rightarrow \text{CO}_2 + 2\text{H}_2\text{O} \\
\text{CH}_4 + \text{O}_2 &\rightarrow \text{CO} + \text{H}_2\text{O} + \text{H}_2
\end{align*}

Combustion and other consecutive C\textsubscript{2} reactions limit the production of higher hydrocarbons (C\textsubscript{2+}), which thus result in low C\textsubscript{2+} yields. With conventional technologies, in which oxygen and methane are premixed, the maximum yield that has been reported with a stable catalyst was always below 25%\textsuperscript{[2]} However, this yield needs to be improved to make OCM economically feasible for exploitation at industrial scales.\textsuperscript{[3]}

Many different OCM reactor configurations have been proposed over the last decades.\textsuperscript{[2]} The most often proposed configurations, together with some of the most promising novel reactor concepts, are simulated in this work to quantify and compare their performance on the same footing.

The packed-bed reactor is the first configuration that isn't studied in this work because it is one of the simplest and well-known concepts in the chemical industry. For exothermic reactions, like OCM, multitubular configurations are used to allow sufficient heat exchange surface area between the coolant and the reaction mixture. For OCM, Siluria also proposed an adiabatic packed-bed reactor,\textsuperscript{[4]} where the gas is fed at relatively low temperature, and the produced heat is used to carry out the endothermal ethane dehydrogenation in a second stage.

As proposed by many authors in the literature,\textsuperscript{[5–8]} the integration of oxygen membranes in a packed-bed reactor, resulting in a packed-bed membrane reactor, can enhance the performance of the process. The main positive aspect of this design is that the reaction is distributed along the axial length of the reactor while keeping a low oxygen partial pressure, which favors the desired reactions of the process over the combustion reactions and consequently increases the C\textsubscript{2+} selectivity.\textsuperscript{[5,7,9]} Furthermore, the heat released by the reactions is also distributed, making the heat management and control of the system easier. Therefore, the use of membranes in packed-bed reactors has also been evaluated and compared with the conventional packed-bed reactor.

As an alternative to packed beds, heat management issues can also be mitigated by using fluidized beds, which enhance the heat transfer in the reactor, thus preventing local hot spots in

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the bed. However, the exchange between the different phases in a fluidized bed can also influence the achievable performance. Therefore, three fluidized bed cases are simulated and evaluated: 1) a bubbling fluidized bed with a fluidization velocity moderately higher than the minimum fluidization velocity with clearly discernable bubble and emulsion phases; 2) a circulating fluidized bed system, where the solids are transported upward together with the gas in a riser reactor; and 3) a fluidized bed membrane reactor, where the oxygen is distributed along the axial reactor length of the fluidized bed.

To perform the simulation study, different catalysts for different configurations may have to be considered, where each catalyst has a specific performance in terms of activity and selectivity, and different kinetics.[10] One of the most often used OCM kinetics in the open literature was developed for a La$_2$O$_3$/CaO catalyst[11,12] by Stansch et al.,[11] and its behavior is well-known. In the last years, another catalyst, namely, Mn–Na$_2$WO$_4$/SiO$_2$, is investigated in detail. This catalyst is more selective than the La-based catalyst; although its activity is lower, it could also be a good choice for industrial OCM applications. However, no fully reliable kinetic schemes were found in the literature for this catalyst. Tiemersma et al.[13] reported some parameters for the OCM reactions, but the temperature dependency was not considered, and C$_2$ reforming reactions were not included. Lee et al.[14] proposed a more detailed mechanism, including some radical species and the dependency on the catalyst coverage, but it was found to be inconsistent as to the total mass balance. Lomonosov et al.[15] corrected the form of the reaction rate equations for the OCM reactions but did not report the equations and parameters for the side reactions. Other studies like the one proposed by Daneshpayeh et al.[16] need further investigations before being safely implemented.

Because of these reasons and the fact that a very broad range of conditions and reactor configurations are going to be investigated, the La$_2$O$_3$/CaO catalyst kinetics has been selected to perform the simulations in this work. This kinetic model includes both parallel side reactions (mainly total and partial combustion of methane), consecutive side reactions (combustion and reforming of C$_2$), and also one gas-phase reaction (ethane dehydrogenation) and can be seen in the Supporting Information of this work. However, it should be noted that the selected kinetics model has not been validated under some of the conditions at which it is used in this work. An extension of this study may be possible in the future when more refined kinetics models for the tungsten-based catalyst system become available. However, the main conclusions reported in this work are generally valid for different catalysts.

### 2. Packed-Bed Configurations

Among all the experimental works published on OCM, the performance of conventional catalytic packed-bed reactors is the most widely investigated work. A literature review shows that C$_2$+ yields above 19% have not been reported for the conventional packed-bed reactor concept,[11] unless operated under particular conditions, such as the use of unstable catalysts (Li based) or very high feed dilution,[17] which are considered unsuitable for large-scale applications. What is often neglected in these studies is how the reactor temperature can be controlled when upscaling the reactor. Generally, the small dimensions of the experimental setups and the conditions used (e.g., relatively low conversions) allow performing the reaction under nearly isothermal conditions without problems; but for large-scale reactors, the relatively small available surface area for heat exchange can easily become problematic, as shown by a comparative study by Dautzenberg et al.[18] With a simplified quantitative analysis, it is concluded that the multitubular packed beds are not an advantageous solution to conduct OCM, in comparison with fluidized bed reactors. In this section, a more detailed evaluation of these aspects is presented.

A pseudohomogeneous plug-flow reactor model was implemented to simulate the OCM reaction in multitubular packed beds under nonisothermal conditions. The model includes the component and total mass balances, an energy balance, and the Ergun equation in its differential form to account for the pressure drop over the reactor. It should be noted that the pressure drop over the reactor can be the limiting factor in the reactor design, particularly for relatively long tubes, high gas flow rates, or small catalyst particle sizes. The model equations are summarized in Table 1.

The parameters to calculate the species heat capacities were taken from the NIST database,[19] whereas the viscosity of the gas mixture was calculated using a mixing rule.[20]

The efficiency of the cooling is assumed to be ideal outside the reactor tubes—the external heat transfer coefficient is assumed to be infinite, and the coolant temperature is constant along the axial direction (which is valid if the flow rate or the heat capacity of the coolant is very high). The effect of axial diffusion is neglected (it will be shown in the adiabatic simulations that this

### Table 1. Equations used to simulate the packed-bed reactor.

<table>
<thead>
<tr>
<th>Equation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{dc_1}{dz} = r_c(e_r, T, p)$</td>
<td>Gas phase mass balances:</td>
</tr>
<tr>
<td>$\frac{dc_v}{dz} = \sum r_j$</td>
<td>Total energy balance: $\frac{dc_v}{dz} = -\sum_{j=1}^N H_j(T) r_j - U_0(T - T_r)$</td>
</tr>
<tr>
<td>$\frac{dP}{dz} = -\frac{\nu}{\rho} \left( \frac{1 - \epsilon}{\epsilon} \right) \left( \frac{150 \nu (1 - \epsilon)}{\rho d_e} + 1.75 \nu \right)$</td>
<td>Ergun equation</td>
</tr>
<tr>
<td>$C_p = \sum_{j=1}^N x_j C_{p_j}(T)$</td>
<td>Additional equations</td>
</tr>
<tr>
<td>$\rho = \frac{P}{RT} \sum_{j=1}^N x_j M_{W_j}$</td>
<td>$H_j(T) = H_j(T_0) + \int_{T_0}^T C_{p_j}(T)$</td>
</tr>
</tbody>
</table>
is indeed unimportant). By specifying the inlet conditions as boundary conditions, the model can be solved with a standard ODE solver (MATLAB’s ode15s was used), obtaining the axial profiles of the gas composition, gas velocity, temperature, and pressure inside the reactor tube.

2.1. Conventional Packed Bed with External Cooling

In packed beds, the highly exothermic OCM and combustion reactions result in the formation of hot spots inside the reactor. With proper operating conditions and tube dimensions, the system is able to keep the temperature of the hot spot to a sufficiently low value. Otherwise, a very sharp reaction and temperature front are formed at a single point in the reactor, similar to what occurs in a premixed flame. This phenomenon is related to the exponential dependency of the reaction rates on the temperature. The runaway regime must absolutely be avoided for the OCM process, because at very high temperatures, the selectivity to the desired C$_{2+}$ is lost, which poses risks to the catalyst and reactor integrity. The proposed model is able to simulate and evaluate the conditions for which the controlled or the runaway regime is present in the reactor. In this second case, a characteristic temperature profile is established with two inflection points before the maximum value. A qualitative example of a runaway case is shown in Figure 1.

The formation of hot spots is basically related to how the terms in the energy equation can balance each other. The source term associated to the heat of reaction depends on the values of the reaction rates. This will increase with the gas pressure and with the catalyst concentration inside the bed. The gas composition will also play a role in this. The heat transfer term depends on the efficiency of heat exchange (heat transfer coefficient), on the surface area of the tube, and on the temperature of the cooling fluid, whereas the convection term depends on the gas velocity, so on the gas flow rate. In summary, almost all process variables influence the thermal performance of the reactor.

To evaluate the efficiency of the heat management in tubular reactors and the influence of hot spots on the achievable C$_{2+}$ yield, different simulations were conducted with the presented model. The tube dimensions were initially fixed (4 cm diameter, 1 m length, and 3 mm particles), as well as the inlet conditions for the gas ($T = 800^\circ$C, $P = 2$ bar, Methane + Air co-feed, CH$_4$/O$_2$ ratio = 4). The model was used considering different amounts of catalyst in the reactor and different values of gas hourly space velocity (GHSV). In this case, the GHSV was defined as the ratio between the inlet gas volumetric flow rate, calculated at standard conditions, and the volume occupied by the gas (reactor volume scaled by the bed porosity).

For the cooling system, a constant value for the overall heat transfer coefficient ($U = 300$ W m$^{-2}$ K$^{-1}$), based on Dautzenberg et al.’s study, was selected. Moreover, a constant temperature for the coolant ($T_c = 800$ °C) was assumed. This approximates the situation when the coolant has a very high heat capacity or when a very high flow rate is used. The simulations results are shown in Figure 2.

For the selected geometry and operation conditions, the maximum C$_{2+}$ yield that can be obtained with small catalyst fractions ($10^{-4} - 10^{-2}$) and a GHSV in the range of 100–10000 h$^{-1}$ is about 13%–14%, which is in agreement with most catalytic studies about OCM in packed-bed reactors. The isocoulinear lines of the same C$_{2+}$ yield are straight on logarithmic scale (indicating a small influence of $\Delta P$ or $\Delta T$): as expected, the same yield can be obtained when increasing the gas flow rate and the amount of catalyst at the same time. The yield decreases with an increase in the flow rate because of the reduced gas residence time and for lower catalyst fractions.

If the maximum temperature peak exceeds a certain value (around 50–80 °C), a runaway effect can be observed. This corresponds to a quick drop in the selectivity to C$_{2+}$, and a very large temperature rise ($18$, which can lead even to catalyst deactivation. In Figure 2, the red line represents the simulations where the maximum temperature increase in the reactor is 50 °C. Above this line, the isocoulinear lines are very close to each other because of the runaway regime. The green line represents the GHSV for which a relative pressure drop of 20% occurs over the packed bed. To limit the pressure drop and temperature rise, it is necessary to work at a lower GHSV and catalyst fraction. In this case, the operability area is quite affected by the two constraints, but the maximum theoretical yield calculated for the isothermal and isobaric case seems still achievable (however, at the expense of a much-increased reactor volume).

The results shown in Figure 2 are affected by an important simplification related to the heat transfer efficiency, which was assumed constant. In particular, the overall heat transfer coefficient strongly depends on the superficial gas velocity. For a more precise estimation of the constraints, the following expression for the heat transfer coefficient was used:

$$U = \left(1 - 1.5 \left(\frac{D}{d_p}\right)^{-1.5}\right) \frac{\delta}{d_p} Pr^{1/3} Re^{0.59}$$  \hfill (11)

The calculations were repeated with this tube Nusselt correlation, and the results are shown in Figure 3.

Decreasing the gas velocity has a drastic influence on the thermal performance of the reactor. This poses quite a problem because the best yields are theoretically achievable at low flow rates and higher residence times, or high flow rates with higher catalyst concentrations. The constraint on the pressure drop is the limiting factor to operate in the latter regime. These results demonstrate that the formation of hot spots seriously decreases...
the achievable C₂ yields in packed-bed reactors, in comparison with the ideal isothermal case.

The combination of pressure and temperature effects becomes very critical for the OCM system. Very low yields are obtained when choosing this tube size. Note also that the amount of catalyst that can be used in this case is extremely low, which is consistent with the very high activity of La₂O₃/CaO.

Figure 2. Predicted C₂ yields for different GHSVs and amounts of catalyst. Single tube simulations (4 cm diameter, 1 m length, T_n = 800 °C, P_n = 2 bar, Methane + Air cofeed, CH₄/O₂ = 4). Energy balance solved using a constant overall heat transfer coefficient U = 300 W m⁻² K⁻¹. Green line: simulations with ΔP/Pₐ = 20%. Red line: simulations with max(ΔT) = 50 °C.

Figure 3. Predicted C₂ yields for different GHSVs and amounts of catalyst. Single tube simulations (4 cm diameter, 1 m length, T_n = 800 °C, P_n = 2 bar, Methane + Air cofeed, CH₄/O₂ = 4). Energy balance solved calculating the overall heat transfer coefficient from the Nusselt correlation (Equation (11)). Green line: simulations with ΔP/Pₐ = 20%. Red line: simulations with max(ΔT) = 50 °C.
These results shown in Figure 2 and 3 are valid for the selected tube geometry and process conditions. Therefore, the effect of the tube dimensions (diameter and length) on the achievable yield was investigated. The results of the sensitivity analyses are shown in Table 2.

Increasing the heat exchange surface area has a beneficial effect on the temperature control. This is the reason why a higher yield can be obtained when decreasing the tube diameter and increasing its length, without exceeding the maximum temperature rise imposed by the optimization algorithm. In particular, from these results, it is evident that short tubes cannot be used for OCM. Also, tubes with a relatively large diameter (which is important to apply this process on an industrial scale) are not recommended because it is not possible to control the heat management. Thus, the required small diameter of the tubes may become an important source of cost for the reactor. Another option to enhance the efficiency of the heat exchange is to operate at a higher gas velocity in the tubes, but this is limited by the allowable pressure drop, which increases with the increasing tube length. The optimum solution for this kind of reactors appears to be operating with long tubes, low flow rates, and a very diluted catalyst. The results clearly indicate that a very active catalyst is not helpful for packed-bed reactors because large reactor volumes are required to achieve the desired heat exchange. For the lanthanum-based catalyst, extremely high catalyst dilution ratios must be used to balance the reaction rates with the available surface area needed to extract the produced reaction heat.

### 2.2. Packed-Bed Membrane Reactor

To investigate the performance of the packed-bed membrane reactor (see Figure 4), a configuration has been selected where the catalyst bed was positioned around a membrane tube. Air is fed to the membrane section, which is selective to oxygen, thus allowing to introduce pure oxygen to the catalyst bed in a distributed way along the entire length of the reactor. The catalytic section, where the OCM reaction is performed, is placed inside an external tube filled with a cooling fluid to counterbalance the exothermic behavior of OCM.

The mass and energy balances of this system are solved for the two reactor compartments, namely, the retentate (air side) and permeate (fuel side). A mass transfer term between the two compartments is introduced in the oxygen mass balances. The energy balances also account for a heat transfer term between the air and fuel side because of conduction through the membrane. External cooling is considered through an extra term in the energy balance for the annular region, where—as earlier—the external cooling is considered as ideal (i.e., constant temperature of the coolant and no external heat transfer resistance). The model equations are given in Table 3.

The flux of oxygen through the dense selective membrane is described with the generalized equation by Xu and Thomson.

This equation presents three parameters \((D_o, k_v, \text{ and } k_f)\), which depend on temperature through an Arrhenius-type relation. Table 4 shows the values of the permeability parameters used for this study, which corresponds to a hollow fiber \(\text{Ba}_1_{-\text{x}}\text{Sr}_{\text{x}}\text{CO}_1_{-\text{y}}\text{Fe}_y\text{O}_{3-\text{z}}\) (BSCF) perovskite membrane.

It was assumed that the catalytic bed was undiluted. The high concentration of active catalyst increases the reaction rates so much, that all the oxygen is consumed as soon as it reaches the methane in the fuel section, producing a very large amount of syngas. As a consequence of this and the ethylene reforming, which is accounted for in the Stansch OCM reaction mechanism, the methane conversion is very high (\(\approx81\%\)), whereas the predicted \(C_2\) selectivity in the outlet is quite low (\(\approx28\%\)). Subsequently, a simulation was carried out as earlier (same geometry and operating conditions), but for a packed bed that contained only 10% of active catalyst. For these simulations, 800 °C was selected as the reactor temperature with a total

### Table 2. Sensitivity analysis of OCM in a packed-bed reactor with different dimensions.

<table>
<thead>
<tr>
<th></th>
<th>Base case</th>
<th>Sensitivity 1 [diameter]</th>
<th>Sensitivity 2 [axial length]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tube length [m]</td>
<td>1</td>
<td>1</td>
<td>10</td>
</tr>
<tr>
<td>Tube diameter [m]</td>
<td>0.04</td>
<td>0.02</td>
<td>0.04</td>
</tr>
<tr>
<td>Maximum yield [%]</td>
<td>7</td>
<td>9.5</td>
<td>12</td>
</tr>
<tr>
<td>Inlet temperature [°C]</td>
<td>800</td>
<td>800</td>
<td>800</td>
</tr>
<tr>
<td>Temperature increase [°C]</td>
<td>&lt;50</td>
<td>&lt;50</td>
<td>&lt;50</td>
</tr>
<tr>
<td>Inlet pressure [bar]</td>
<td>2</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>Pressure increase [bar]</td>
<td>(\Delta P/P &gt; 20%)</td>
<td>(\Delta P/P &gt; 20%)</td>
<td>(\Delta P/P &gt; 20%)</td>
</tr>
<tr>
<td>(\text{CH}_4/\text{O}_2) ratio</td>
<td>4</td>
<td>4</td>
<td>4</td>
</tr>
</tbody>
</table>

![Figure 4. Scheme of a packed-bed membrane reactor.](image)
Table 3. Equations used to simulate the packed-bed membrane reactor.

Equations for the fuel side
\[
\frac{d}{dz} \dot{\chi}_i = \eta_i + k_i \chi_i \dot{c}_i^m - \dot{c}_i
\]  
(12)
\[
\frac{d}{dz} \dot{c}_i = \sum_{j=1}^{N} \eta_j + k_j \chi_j \dot{c}_j^m - \dot{c}_j
\]  
(13)
\[
\dot{\omega}_i^f \frac{dT^f}{dz} = -\eta_i \dot{\omega}_i^f \dot{c}_i^m - \dot{c}_i^m
\]  
(14)
\[
\dot{\omega}_i^a \frac{dT^a}{dz} = -\eta_i \dot{\omega}_i^a \dot{c}_i^m - \dot{c}_i^m
\]  
(15)
\[
r_i = \sum_{j=1}^{N} \eta_{ij} \dot{\omega}_{ij}^m (1 - \xi)
\]  
(16)

Equations for the air side
\[
\frac{d}{dz} \dot{\chi}_i = k_i \chi_i \dot{c}_i^s - \dot{c}_i^s
\]  
(17)
\[
\frac{d}{dz} \dot{c}_i^s = -\sum_{j=1}^{N} k_j \chi_j \dot{c}_j^s - \dot{c}_j^s
\]  
(18)
\[
\dot{\omega}_i^a \frac{dT^a}{dz} = \sum_{j=1}^{N} \dot{\omega}_i^a \dot{c}_i^m - T^a \frac{dT^a}{dz}
\]  
(19)

Equations at the membrane interface
\[
k_i^m (\dot{c}_i^m - \dot{c}_j^m) = \dot{J}_i^m
\]  
(20)
\[
-k_i^s (\dot{c}_i^s - \dot{c}_j^s) = \dot{J}_i^s \frac{dT^a}{dz}
\]  
(21)
\[
h^i (T^f - T^a) = U (T^f - T^a)
\]  
(22)
\[
h^a (T^a - T^r) = U (T^a - T^r) \frac{dT^a}{dz}
\]  
(23)

Equation for the oxygen flux through the membrane
\[
\dot{J}_i^m = \frac{D_i k_i (\chi_i^m n - \chi_i^m f) + D_i (\chi_i^s n + \chi_i^s f)}{2 \Delta \chi_i (\chi_i^m n - \chi_i^m f) + \Delta \chi_i (\chi_i^s n + \chi_i^s f)}
\]  
(24)

Heat and mass transfer coefficients
\[
k_i^m = 0.0096 \frac{D_i}{\eta_i} S_{caO}^{0.546} R_{caO}^{0.913}
\]  
(25)
\[
k_i^s = 0.0096 \frac{D_i}{\eta_i} S_{caO}^{0.546} R_{caO}^{0.913}
\]  
(26)
\[
h^i = 0.17 \frac{\rho_i v}{\sqrt{\eta_i}} \left( \frac{P_{caO}}{0.7} \right)^{1/2} \theta_{caO}^{0.913}
\]  
(27)
\[
h^a = 0.025 \frac{\rho_i v}{\sqrt{\eta_i}} \left( \frac{P_{caO}}{0.7} \right) \theta_{caO}^{0.913}
\]  
(28)
\[
U = \frac{1}{\frac{1}{\theta_f} + \frac{1}{\theta_f} + \frac{1}{\theta_r} \ln \left( \frac{\theta_f}{\theta_r} \right)}
\]  
(29)

Table 4. Values of the permeability parameters resulting from the fitting of experimental data \(^{[29]}\)

<table>
<thead>
<tr>
<th>Pre-exponential factor (D_i \text{m}^2 \text{s}^{-1})</th>
<th>Activation energy (kJ \text{ mol}^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>9.823</td>
<td>91.8</td>
</tr>
<tr>
<td>15.36</td>
<td>56.3</td>
</tr>
<tr>
<td>308.5</td>
<td>267.0</td>
</tr>
</tbody>
</table>

pressure of 2 bar, whereas the optimal \(\text{CH}_4/\text{O}_2\) ratio, which was used to run these cases, was found to be 2.6. The results are reported in Figure 5, showing the trends of conversion, selectivity, and yield, and in Figure 6, showing the axial temperature profiles.

An enormous increase in the selectivity to \(\text{C}_2\) can be achieved in the packed-bed membrane reactor (78% at the outlet), while
also the methane conversion is slightly higher (81%). As a result, the C2+ yield is increased to 63%. The amount of produced syngas is much lower in this case, which means that a lower fraction of produced C3 is consumed by consecutive reforming reactions. These results clearly show that the activity of the packed bed, that is, the amount of catalyst, can be used to improve the product selectivity because it affects the OCM reactions and consecutive reforming reactions of C2 differently.

Despite the very high conversion obtained, the hot spot can be well controlled. The average reactor (permeate) temperature is higher in this case with the diluted catalyst bed compared with the simulation with only catalyst particles, and this is related to the decreased extent of endothermic reforming. The outlet temperature rise is only 40 °C, which also means that the combined cooling with the external fluid and with air in the retentate compartment is quite effective.

To directly compare the membrane reactor performance with the cofeed operation in the packed-bed reactor, the same base case presented for the membrane reactor was simulated, but assuming that no permeation through the membrane is possible. As the only other difference, the catalyst was diluted with a factor 10², which is necessary to prevent the runaway regime (higher reaction rates result in a too high hot spot with the consequent loss of all C2+ selectivity). Moreover, the risk associated because of working in the explosion regime has been ignored in these simulations, although it is of course a critical aspect when conducting experimental work under these conditions. The results are summarized in Table 5.

Note that the temperature rise at the hot spot is already 70 °C for strongly diluted cofeed packed-bed reactor configuration, which is quite close to the transition to the runaway regime. The temperature rise is much higher than the temperature increase that was observed for the distributed feed operation, despite the much lower amount of catalyst, and the obtained C2+ yield is quite low, only around 5%. For the distributed O2 feeding cases, both conversion and selectivity are increased when diluting the catalyst bed and increasing the reactor length. This example shows that, even if high reactor volumes are necessary to effectively operate membrane reactors, even higher reactor volumes are required for conventional packed beds, because of the enormous catalyst dilution needed to compensate for the inefficient heat transfer in this configuration.

### Table 5. Comparison of the simulation results for packed beds, with cofeed and distributed oxygen feed with diluted and undiluted catalyst beds.

<table>
<thead>
<tr>
<th></th>
<th>Distributed feed [pure catalyst]</th>
<th>Distributed feed [diluted catalyst]</th>
<th>Cofeed [diluted catalyst]</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH4/O2</td>
<td>2.3</td>
<td>2.6</td>
<td>2.6</td>
</tr>
<tr>
<td>% catalyst in solid</td>
<td>100%</td>
<td>10%</td>
<td>0.007%</td>
</tr>
<tr>
<td>X_C2H4</td>
<td>71%</td>
<td>81%</td>
<td>17%</td>
</tr>
<tr>
<td>X_C2</td>
<td>28%</td>
<td>78%</td>
<td>26%</td>
</tr>
<tr>
<td>Y_C2</td>
<td>20%</td>
<td>63%</td>
<td>5%</td>
</tr>
<tr>
<td>max(ΔT)</td>
<td>34 °C</td>
<td>40 °C</td>
<td>70 °C</td>
</tr>
<tr>
<td>ΔP/P0</td>
<td>8%</td>
<td>7%</td>
<td>9%</td>
</tr>
</tbody>
</table>

### 2.2.1. Analysis of the Reaction Rates

The three presented cases show a different operation of the reaction mechanism. The distributed feed case with pure catalyst has more or less the same C2+ selectivity than in the cofeed operation but with a much higher methane conversion. With distributed feeding, a major part of the selectivity loss is related to CO production, whereas in the cofeed configuration, more CO2 is formed. A large improvement in the selectivity can be noted by diluting the catalyst in the membrane reactor. To gain more insight, the average values of the reaction rates inside the reactor were calculated for all three cases, as shown in Figure 7.

The results show that, in case of distributed O2 feeding, the combustion reactions (both complete and incomplete) of methane and ethylene are relatively unimportant compared with the OCM reactions. This is quite different for the cofeed operation, where total and partial combustions are competitive and represent a major source of loss of carbon selectivity to C2+. In membrane reactors, the only important side reaction involving oxygen is the CO combustion: this is not directly related to a decrease in C2+ selectivity because it is only a consequence of the formation of carbon monoxide through reforming of ethylene, which is the real undesired step to be avoided in this case, whereas in cofeed operation, the CO oxidation is not so important. As indicated earlier, C2 reforming is playing a much more important role when using the undiluted catalyst. The larger CO production also results in a higher O2 consumption for its consecutive oxidation to CO2; this is the reason why with pure catalyst, the methane conversion is 10% lower, even if the amount of permeated oxygen is slightly higher than with diluted catalyst (CH4/O2 is lower). In summary, in membrane reactors, the amount of catalyst in the bed affects the extent of reforming reactions, but not the rate of primary OCM reactions, whose rate is controlled by the oxygen flux through the membrane. For this reason, the catalyst concentration can be tuned to optimize the C2+ selectivity.

In conclusion, the simulations show that the integration of membranes may lead to a large improvement in the performance of a packed-bed reactor for OCM. The two key factors are the extent of the different OCM reaction rates along the reactor, which can be relatively controlled by means of the amount of catalyst present in the bed, and the improvement in the heat management due to the distributed reaction and the additional cooling provided by the heat transfer with air.

**Adiabatic Packed Bed with Post Cracking:** In the previous sections, the problems stemming from hot spot formation in OCM tubular packed beds were addressed including how this can be more easily handled in the distributed feed configurations. Another possibility is to operate the packed bed under adiabatic conditions, which can have a clear advantage in terms of simplifying the reactor geometry (no tube and shell structure, but a single vessel), as well as its operability (no more need to handle a cooling fluid and to optimize the heat exchange).

A simple solution for this problem has been proposed by Siluria.[4] The key aspect of this concept is to operate with a relatively cold feed (≈530 °C). The feed temperature should be high enough to allow reaction activation, but at the same time, the
adiabatic temperature rise must not be so high to cause a complete loss of selectivity (the maximum temperature should not exceed approximately 1000 °C). The OCM system is prone to exceed this limit in adiabatic operation, but one possibility to avoid this is to operate with high CH$_4$/O$_2$ ratios: in this case, the temperature rise is limited by the amount of oxygen present in the system.

In their economic analysis, Siluria proposed to use a two-stage adiabatic reactor, where the first stage consists of a catalytic bed in which the OCM takes place, and the second stage consists of a catalytic bed for the dehydrogenation. Between the two stages, extra ethane is fed, and the high temperature reached by the exothermic reactions in the first section is used to carry out the endothermic conversion of ethane to ethylene. A methane inlet flow rate of 1065 ton h$^{-1}$ has been selected for the simulations of this case, with an initial CH$_4$/O$_2$ ratio of 10 and without nitrogen dilution. The inlet temperature was set to 540 °C and the pressure to 9 bar. Finally, the ethane flow rate for the second stage was 90 ton h$^{-1}$.

For this adiabatic reactor case, the diffusion terms were added to all the conservation equations. This was done particularly to evaluate how much the axial heat dispersion influences the axial temperature profiles when no external cooling is applied to the system.

The mass balances for all the individual chemical species are given by:

$$\frac{dc_i}{dz} = \frac{df_i}{dz} + r_i$$

(30)

where the diffusive fluxes are calculated with the generalized Fick’s law:

$$J_i = -c \sum_{j=1}^{N-1} \left( D_i \frac{dx_j}{dz} \right)$$

(31)

A total mass balance is added to evaluate the gas velocity $v$:

$$\frac{dv}{dz} = \sum_{j=1}^{N} (r_j)$$

(32)

The energy balance reads:

$$cC_p \frac{dT}{dz} = \frac{d}{dz} \left( \lambda \frac{dT}{dz} \right) - \sum_{j=1}^{N} H_j r_j$$

(33)

The gas diffusivities were calculated by the binary diffusion coefficients as a function of temperature and pressure. The conductivity of the bed was calculated from the contribution of the solid and gas phase:

$$\lambda = \nu \lambda_g + (1 - \nu) \lambda_s$$

(34)

where $\lambda_g$ was evaluated as a function of temperature and gas composition.

This model was used to simulate the two reactor sections (OCM and cracking). The Stansch mechanism was applied for both the reactor sections; the only difference is that the catalyst concentration was set to zero in the second section. This automatically sets all the reaction rates to zero, except for the dehydrogenation of ethane to ethylene, which is supposed to be a gas phase reaction and thus does not depend on the concentration of La$_2$O$_3$/CaO. Figure 8 and 9 show the simulation results of the two-stage bed, using the reactor dimensions and operating conditions from Siluria. A catalyst fraction of 40% was assumed to reduce hot spot formation in the bed.

In the first reactor stage, the reaction activates at 650–700 °C, and consequently, a very sharp reaction front is formed, where the temperature rises to about 980 °C. This could be considered as a kind of controlled runaway regime: even if the temperature profile is very pronounced, the temperature cannot exceed 1000 °C. After the oxygen is completely consumed, neither OCM nor combustion reactions can occur anymore. At this
correspondence of these temperatures. An even lower amount of oxygen should be fed to fulfill this condition.

In conclusion, the adiabatic packed-bed reactor is probably the simplest concept that can be devised for OCM. It can be applied for this reaction system, even for very high gas flow rates, but with a relatively low performance in terms of achievable C\textsubscript{2+} yield (lower than with multitubular packed beds with cooling). The main limitation is the high methane dilution needed to control the maximum temperature, which strongly affects the achievable conversion.

3. Fluidized Bed Configurations

Fluidized bed reactor technology can overcome the heat management issues of packed-bed reactors. However, the effect of mass transfer limitations between the different phases of both the reactants and the products on the achievable C\textsubscript{2+} yield needs to be better understood and quantified. In this section, the performance of a fluidized bed in the bubbling fluidization regime, a circulating fluidized bed system, and a fluidized bed membrane reactor for OCM is investigated and compared.

3.1. Bubbling Fluidized Bed

First, a fluidized bed reactor in the bubbling fluidization regime is considered. In a bubbling fluidized bed, assumed to consist of Geldart B-type particles, the gas velocity is typically 3–15 times the minimum fluidization velocity. In the bubbling regime, there is a clear distinction between the bubbles, regions characterized by a relatively high porosity via which most of the gas moves from the bottom feeding section to the top freeboard, and an emulsion phase with a much higher solids holdup, through which the gas percolates with a velocity close to the incipient fluidization velocity. In this work, the bubbling fluidized bed model developed by Kunii and Levenspiel\cite{30} was used (see also a.o. Medrano et al.\cite{31}) and extended to include also the wake phase, which represents the region inside the bubbles (around 15% of the bubble volume) where the solids fraction is considered to be the same as in the emulsion phase. A scheme of the proposed model structure is shown in Figure 10, and a brief description of the model is provided below.

The total gas feed entering the reactor with a specified superficial gas velocity, \( u_\text{g} \), is assumed to be distributed between the bubble phase (including the wake), at velocity \( u_\text{b} \), and the emulsion phase (with a velocity \( u_\text{e} \)). The bubbles are considered to be devoid of solids, whereas the amount of solids contained in the wake of the bubbles is assumed to be constant and equal to 15% of the total bubble volume with a porosity equal to the emulsion phase porosity. Mass transfer is accounted for to describe the gas exchanged between the different phases. The component mass balances and constitutive equations for the hydrodynamics and mass transfer processes are summarized in the Supporting Information.

For the OCM system, methane and oxygen need to be transported from the bubble phase to the wake and emulsion phases containing the catalyst, where they react to form C\textsubscript{2+} and side products. Afterward, these species need to be transferred back to the bubble phase, where they move upward to the freeboard.
The key factor to improve the reactor performance is the enhancement of the bubble–emulsion mass transfer rate while decreasing the reaction rates in the bubble wakes so that the mass transfer processes govern the axial concentration profiles in the bubble plus wake phase. To do this, the amount of catalyst present in the bubble phase including the wake should somehow be limited or its activity decreased. If the conversion in the wake phase is limited, the OCM could take place at low oxygen concentration levels. Moreover, it would be beneficial for the process to entirely feed the oxygen in the wake, thus avoiding fast reactions in the emulsion phase close to the bottom distributor with a relatively high oxygen concentration.

In conclusion, bubbling fluidized beds may be a good solution in terms of thermal control but suffer from a poor performance in terms of achievable yields due to mass transfer limitations, as also reported in the literature.\[32\] The key to improve the performance is to try to use the bubbles as a natural way of distributing oxygen to the regions with high catalyst concentrations, but the main obstacle seems to be the mass transfer rates from the bubble wake to the bulk of the emulsion phase, because in the bubble wake, the amount of oxygen is relatively high while at the same time reactions take place.

In the next section, whether operation in a circulating fluidized bed system could overcome these problems and achieve higher $C_2+$ yields is investigated. Finally, the use of membranes in fluidized bed reactors\[33,34\] is also investigated. In this latter
case, the amount of oxygen is low in all phases at the inlet, and the \( \text{O}_2 \) flux can be regulated by the membrane permeability tuned to the hydrodynamics of the gas–solid system.

### 3.2. Circulating Fluidized Bed

An alternative fluidization technology that could also be used for OCM is the circulating fluidized bed.\(^{35–37} \) In this configuration, gas velocities are much higher than in bubbling fluidized beds so that the solids are transported upward in the main reactor section. At the top, a system (commonly a cyclone) is used to separate the particles, which are transported back to the bottom of the reactor. Inside the bed, the solids are also partially recirculated in the region close to the wall, where the solids naturally move downward when operating at higher solids holdups.

A phenomenological model for circulating beds was proposed by Pugsley and Berruti, who applied it for different reaction systems, including OCM.\(^ {38} \) The model is a core-annulus model, which describes the two reactor regions that are characterized by different fluid dynamic conditions and catalyst concentrations. The model assumes that gas is transported upward in the core section, and it is stagnant in the outer region and accounts for radial gas mass transfer between the two sections. The model equations are summarized in the Supporting Information of this work.

For this concept, the following conditions were selected as a base case. The column height was set to 7 m, and a particle size of 70 \( \mu \)m (Geldart A) was selected with a net solids flux of \( 100 \text{ kg m}^{-2} \text{s}^{-1} \). It was assumed that undiluted catalyst particles are fed to the reactor. For the circulating fluidized bed configuration, it is possible to operate without solids dilution despite the very high catalyst activity because of the very high void fractions (>90%) prevailing in the bed. The \( \text{CH}_4/\text{O}_2 \) feed ratio was set to 4 with a bed temperature of 800 °C and a total pressure of 2 bar. The value of the mass transfer coefficient was fixed at 0.05 m s\(^{-1}\), in agreement with the range of values proposed by the authors.\(^ {38} \) The computed axial mole fraction profiles of methane, oxygen, and ethylene for the core and annulus regions are given in Figure 14.
In the bottom part of the reactor, the gas composition in the core and annulus regions is quite different, indicating the importance of mass transfer limitations. When the oxygen concentration in the core decreases, the compositions in the two regions get closer due to the reduced reaction rates, and the kinetic resistance becomes comparable with the mass transfer resistance. In the circulating fluidized bed reactor, the core region acts as a kind of distributed oxygen feeding to the annulus, similar to the bubbles in the bubbling fluidized bed reactor, as discussed earlier. At the reactor outlet, almost all the oxygen from the core region has been transferred and consumed. The corresponding methane conversion amounts to 28% with a $C_2$ selectivity of 54%, giving a $C_2$ yield of 15%.

It is important to mention that the $C_2$ selectivity is enhanced in the annulus part as a result of reduced methane total and partial combustion reactions, resulting from the relatively low oxygen concentration levels. However, as was also the case in the bubbling fluidized bed reactor, the reaction kinetics predominates over the mass transfer rates at the entrance of the reactor, causing that most of the reaction takes place in the core where a relatively high oxygen concentration is still present. Reduced kinetics or improved core-annulus mass transfer rates are required to improve the reactor performance, limiting the reactions in the core and distributing the oxygen of the core more evenly to the annulus, where the reactions could occur at lower oxygen concentrations.

3.3. Fluidized Bed Membrane Reactor

An interesting possibility to increase the overall yield of the process is to integrate oxygen membranes into a fluidized bed reactor. The equations used to simulate the fluidized bed membrane reactor are given in the Supporting Information of this article, whereas the hydrodynamics and mass transfer expressions have been selected to be the same as in the case of the bubbling fluidized bed reactor. Similar to the case of the packed-bed membrane reactor, the membranes distribute the oxygen feed along the axial length of the reactor, keeping the oxygen partial pressure low and consequently selectively enhancing the desired reactions. Also for the fluidized bed membrane reactor, mass transfer between the bubble and emulsion phases influences the maximum yield that can be achieved. Mass transfer resistances could help protecting $C_2$, formed in the emulsion phase and migrated to the bubble phase, from undesired consecutive reactions (reforming and combustion) because of the very low catalyst concentration in the bubbles. In contrast, mass transfer can also have a detrimental effect, because some methane and also oxygen present in the bubbles may remain unreacted because of the low catalyst concentration in the bubble phase, leading to a much reduced conversion of the reactants.

For the simulations, the same conditions have been selected as before for the bubbling fluidized bed reactor simulations. The amount of oxygen has been varied by changing the number of membranes in the reactor, whereas the catalyst dilution was fixed at 10%. The membrane characteristics have been chosen identical to those in the packed-bed membrane reactor simulations, with a membrane diameter of 0.01 m. The membrane length was equal to the reactor length to maximize the oxygen distribution.

In Figure 15, the simulation results for different $\text{CH}_4/\text{O}_2$ ratios have been plotted.

![Figure 15](https://www.entechnol.de)

The figure shows that an optimal $\text{CH}_4/\text{O}_2$ ratio is found at around 1.5, corresponding to 50 membranes and resulting in a $C_2$ yield of 54%. The performance is significantly increased when compared with the other fluidized bed configurations. With this configuration, indeed a good distribution of the oxygen feed is achieved along the bed, keeping a low oxygen partial pressure in both phases, particularly also in the wake phase, where most of the reactions take place.

The overall process performance with a $C_2$ yield well above 30%–35%; the minimum value required to make the process economically viable, is however lower than the $C_2$ yield that can be obtained with the packed-bed membrane reactor. To better elucidate the differences in the performance of the two membrane reactor configurations, the separate performance of the bubble (plus wake) and emulsion phases for the optimal $\text{CH}_4/\text{O}_2$ ratio is shown in Figure 16.

Because of the higher amount of catalyst in the emulsion phase, the methane conversion is much higher, reaching almost 100% at the outlet, whereas in the bubble phase including the wake is much lower (about 65% at the outlet). The $C_2$ selectivity is higher in the bubble phase because of reduced consecutive $C_2$ reactions in the bubble phase, corresponding to the lower amount of catalyst. However, the contribution of the bubble (including wake) phase to the overall performance of the OCM process is higher than the emulsion phase because of the higher superficial gas velocity in the bubble phase.

The differences in reaction rates in both phases result in a difference in required reactor length for optimal $C_2$ yield, necessitating finding a trade-off for the maximum overall yield and consequently losing efficiency in at least one of the two phases. The final consequence of this efficiency loss is a lower overall yield than for the comparable case of a packed-bed membrane reactor, where just one compartment (phase) is considered.

To confirm this effect, the mass transfer coefficients between the different phases have been increased by 100 times for the optimal fluidized bed membrane reactor case, and the simulation results are shown in Figure 17.

Because of the enhancement of the mass transfer between phases, the profiles of both emulsion and bubble phase are almost the same, although a small difference is still discernable because of the faster reaction rates in the emulsion phase. Most
importantly, a very strong increase in the overall performance of the process is observed, where the \( \text{C}_2 \) yield can even be increased to values well above 70% and even surpassing the performance of the packed-bed membrane reactor. By increasing the mass transfer, the methane present in the bubbles is reaching the emulsion phase easily, thus enhancing the methane conversion. In addition, part of the \( \text{C}_2 \) formed in the emulsion and wake phase can migrate back to the bubbles, which are free of catalyst, avoiding consecutive catalytic reactions. As a consequence, the positive effects of the use of fluidized beds are enhanced, whereas the negative effects are minimized by increasing the mass transfer rates between the phases.

In summary, membrane fluidized bed reactors could indeed be a solution to improve the \( \text{C}_2 \) yield, and with simulations, it has been demonstrated that this reactor concept can outperform conventional reactor configurations. The performance of the fluidized bed membrane reactor is lower than the packed-bed membrane reactor, unless the mass transfer rates between the phases can be enhanced. However, fluidized beds provide a much easier temperature control, which is shown to be crucial for the considered reaction system.

### 4. Conclusions

In this work, different reactor concepts for OCM have been compared in detail with numerical simulations. With packed-bed reactor configurations, a \( \text{C}_2 \) yield of around 20% can maximally be achieved, even when assuming isothermal operation, which is not easily accomplished in packed-bed reactor configurations. Cofeeding of all reactants causes almost all conversion to take place close to the reactor inlet, leading to a very large local heat release in that region because of the strong exothermicity of the OCM reaction system, with drastic consequences for the process performance. The optimization of several design and operating conditions, such as dilution of the catalyst with inert material and/or increase of the gas velocity, can solve the heat management issues, however at the expense of a strong decrease in the OCM performance.

Subsequently, a packed-bed membrane reactor was simulated to feed the oxygen in a distributed manner along all the reactor length, thereby maintaining low oxygen partial pressures and consequently favoring the desired reactions, which also helps improving the reactor heat management and control of hot spot formation. With a packed-bed membrane reactor diluting the catalyst ten times with inert material to minimize the effect of the \( \text{C}_2 \) reforming reactions, the \( \text{C}_2 \) yield can be increased up to 60%, making the OCM process economically viable and competitive with other technologies.

Another option to solve the heat management problem is the deployment of a fluidized bed reactor technology, with which the heat transfer is enhanced, facilitating easier thermal control of the reactor. However, the performance of both the bubbling and circulating fluidized bed configurations is rather poor,
achieving \( \text{C}_2 \) yields of only around 12%. Again, the integration of membranes in this configuration can considerably improve the reactor performance, reaching values that—although lower than for the packed-bed membrane reactor—are sufficiently high to render this process competitive with other ethylene technologies. It has been demonstrated that the mass transfer rates between the phases need to be improved to further increase the overall performance.

Although the results obtained in this work of both the packed-bed and fluidized-bed membrane reactor concepts are good enough to satisfy the requirements for industrial exploitation of the OCM process, two important aspects that can significantly affect the computation results need to be further investigated. From one side, the reaction kinetics used for the simulations in this work was not developed for use in a membrane reactor, and some \( \text{C}_2 \) consecutive reactions that can take place have not been accounted for in the used kinetics model. In a membrane reactor, where the desired products are formed throughout the reactor and where oxygen is fed along the entire reactor, \( \text{C}_2 \) consecutive reactions may become more important, especially toward the end of the reactor, thus decreasing the overall performance of the process. Unfortunately, so far no comprehensive reaction kinetics has been found in the literature (suited for application in a membrane reactor model) to reliably quantify the extent of the decrease in performance.

In addition, it should be considered that the \( \text{La}_2\text{O}_3/\text{CaO} \) catalyst (selected for all the simulations) is not able to completely direct the reaction toward the desired products. As a consequence, the overall yield obtained from OCM will always be influenced by the particular characteristics of the catalyst chosen, mainly activity and selectivity, even though the reactor configuration used could help maximizing the yield toward the desired products.

Another important assumption used in this work is related to the selected 1D reactor configuration. In membrane reactors, lateral concentration profiles of methane, oxygen, and \( \text{C}_2 \) species can significantly affect the overall reactor performance. The increased oxygen and \( \text{C}_2 \) concentrations near the membrane surface relative to the methane concentration decrease the \( \text{C}_2 \) yield, and this becomes even more important at higher methane conversion. Future work should focus on the influence of radial concentration profiles on the overall reactor performance.

Finally, it should be mentioned here that the membrane reactor technology for the specific OCM application is far from a feasible application to bigger scale systems. Hence, additional efforts should be put on for the development of OCM membrane reactors to show, first at lab-scale, that the results achieved in the simulations can be reproduced in long-term experiments.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.
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