SENSITIVITY ANALYSIS AND OPTIMIZATION OF A MEMBRANE REACTOR FOR HYDROGEN PRODUCTION THROUGH METHANE STEAM REFORMING

ANÁLISE DE SENSIBILIDADE E OTIMIZAÇÃO DE UM REATOR DE MEMBRANA PARA PRODUÇÃO DE HIDROGÊNIO POR REFORMA A VAPOR DO METANO

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Abstract: Hydrogen is one of the most studied sources for clean power generation in the near future. Nowadays, hydrogen is mainly produced through methane steam reforming in packed bed reactors, with a promising alternative to this technology being the implementation of hydrogen-selective membrane reactors. This work compares the isothermal mathematical models of both designs by assessing the effects of multiple design variables on methane conversion, while also providing recommended operating conditions for maximum efficiency of the membrane reactor over the packed bed technology. Additionally, an optimization study is carried by dividing the reactor length in isothermal segments to achieve higher efficiency. Results showed that the membrane technology considerably increases hydrogen production, with temperature being the most influential variable on methane conversion. While the temperature profile optimization provided similar conversions compared to the isothermal models, the membrane reactor’s efficiency was increased, further justifying its implementation.

Keywords: Membrane reactor. Hydrogen. Sensitivity analysis. Optimization. Scilab.

Revista Mundi, Engenharia e Gestão, Paranaguá, PR, v. 5, n. 6, p. 281-01, 281-14, 2020
DOI: 10.21575/25254782rmetag2020vol5n61315
Resumo: O hidrogênio é uma das fontes mais estudadas para a produção de energia limpa no futuro próximo. Atualmente, a maior parte da produção de hidrogênio se dá pela reforma a vapor do metano em reatores de leito fixo, enquanto a implementação de reatores com membranas seletivas ao hidrogênio se apresenta como uma alternativa promissora para esta tecnologia. Este trabalho compara os modelos matemáticos isotérmicos de ambas tecnologias analisando-se os efeitos de múltiplas variáveis de projeto na conversão do metano, além de serem apresentadas as condições operacionais recomendadas para a máxima eficiência do reator de membrana em relação à tecnologia com leito fixo. Ademais, um estudo de otimização é realizado por divisão dos reatores em segmentos isotérmicos para aumento de suas eficiências. Os resultados demonstraram que a tecnologia com membrana aumenta consideravelmente a produção de hidrogênio, sendo a temperatura a variável mais influente na conversão do metano. O estudo de otimização do perfil de temperatura forneceu conversões similares às do modelo isotérmico; não obstante, esta modificação permitiu aumentar a eficiência do reator de membrana, justificando ainda mais sua implementação.

1 INTRODUCTION

Due to the ever-increasing environmental impacts caused by industrial and economic activities, several studies have been made throughout the last decades regarding natural resource management and sustainable energy supply systems. In the context of power generation, electricity production from hydrogen using fuel cell technology presents itself as a promising alternative to the burning of fossil fuels due to the former’s considerably reduced emissions of greenhouse gases (WILBERFORCE et al., 2017).

Regarding sustainable hydrogen production, photo-induced water decomposition can be cited as a viable technology to construct clean energy systems (KUDO; MISEKI, 2009). However, since hydrogen is currently mainly produced from primary energy sources such as coal and fossil fuels (WILBERFORCE et al., 2017), these production processes must be designed for efficient energy and raw materials consumption until technological advancements allow renewable energy to become the main source for hydrogen production.

As of 2011, hydrogen is mainly produced from methane and water vapor (VALLADARES, 2011). In this process, the main reactions are the methane steam reforming (MSR), in which the reactants form carbon monoxide and hydrogen, and the water-gas shift (WGS), in which the carbon monoxide further reacts with water vapor to form carbon dioxide and more hydrogen (BROWN, 2001). While multiple side reactions are also involved, XU and FROMENT (1989)’s study demonstrated that the system can be simplified to the MSR and WGS reactions and the methanation reaction, which are respectively described by Equations (1) to (3).

\[
\begin{align*}
CH_4 + H_2O & \leftrightarrow CO + 3H_2, \\
CO + H_2O & \leftrightarrow CO_2 + H_2, \\
CH_4 + 2H_2O & \leftrightarrow CO_2 + 4H_2.
\end{align*}
\]

Industrial methane reformers usually consist of fixed bed reactors packed with nickel-based alumina catalyst, with external heat being provided due to the endothermic nature of the MSR reaction. Following the previously presented importance of improving hydrogen production processes, a promising technology that has been proposed over the past decades is the implementation of hydrogen-selective membranes inside the MSR reactors (LI et al., 2008), as presented on Figure 1.

In this design, hydrogen is produced in the catalyst-packed reaction zone, while a Pd- or Pd/alloy-based catalytic membrane covers the permeation zone where a sweep gas is employed to create a pressure difference between both sections. The hydrogen flux through the membrane...
decreases its partial pressure in the reaction zone, which in turn pushes the equilibrium-controlled reactions towards an increase in hydrogen production (ROUX, 2011).

Compared to the conventional design, the membrane technology allows for higher methane conversion at the same operating conditions or, most importantly, the same conversion at lower temperatures, which can reduce energy costs and increase catalyst lifetime (AYTURK; KAZANTZIS; MA, 2009). However, since implementing the membrane results in additional costs, the operating conditions must be adjusted to provide a considerably higher raw material conversion and, thus, justify the increase in initial investment.

Several studies have used computer simulation to assess the performance of Pd-based membrane reactors for hydrogen production from methane (AYTURK; KAZANTZIS; MA, 2009; BRUNETTI; BARBIERI; DRIOLI, 2011; CORONEL et al., 2011), while others investigated the use of similar designs, albeit for hydrogen production from thermal decomposition of hydrogen sulfide (ZAMAN; CHAKMA, 1995). This work aims to compare the performance of membrane reactors and packed bed reactors for hydrogen production from methane based on the isothermal mathematical models described in (ROUX, 2011). Sensitivity analyses were carried to assess the methane conversion’s dependence on temperature, pressure, and inlet water/methane ratio. Additionally, an optimization study considering isothermal reactor segments of equal length was used to find temperature profiles that increase the membrane reactor’s efficiency compared to the isothermal models. Similar optimization studies were also carried to find the ideal temperatures at different operating conditions.

2 MATHEMATICAL MODELS

This section presents the isothermal mathematical models described in (ROUX, 2011) for the membrane reactor (MR) and the packed bed reactor (PBR). As previously described, the
system can be simplified to Equations (1) to (3), whose reactions rates are respectively shown on
Equations (4) to (6), where $r$ are the reaction rates in kmol/kgcat. $h, k$ are the reaction constants,
$K$ are the reactions’ equilibrium constants and $P$ are the components’ partial pressures in bar.
The denominator $DEN$ is described by Equation (7), where $K$ are the components’ adsorption
constants on the catalyst’s surface.

$$r_1 = \frac{k_1}{(P_{H_2})^{2.5}} \cdot \frac{(P_{CH_4} \cdot P_{H_2O} - \frac{(PH_2)\cdot P_{CO}}{K_1})}{(DEN)^2},$$  (4)

$$r_2 = \frac{k_2}{P_{H_2}} \cdot \frac{(P_{CO} \cdot P_{H_2O} - \frac{P_{H_2} \cdot P_{CO_2}}{K_2})}{(DEN)^2},$$  (5)

$$r_3 = \frac{k_3}{(P_{H_2})^{3.5}} \cdot \frac{(P_{CH_4} \cdot (P_{H_2O})^2 - \frac{(PH_2)^3 \cdot P_{CO_2}}{K_3})}{DEN^2},$$  (6)

$$DEN = 1 + K_{CO}P_{CO} + K_{H_2}P_{H_2} + K_{CH_4}P_{CH_4} + K_{H_2O} \frac{P_{H_2O}}{P_{H_2}}.$$  (7)

Reaction, equilibrium, and adsorption constants are expressed by Arrhenius models
according to Equations (8) to (10), respectively, where $k_{i0}$, $E_a$ and $\Delta G_i^0$ are reaction $i$’s pre-
exponential factor, activation energy and Gibbs free energy variation, respectively, $K_{aj}$ and
$\Delta H_j$ are component $j$’s pre-exponential factor and adsorption enthalpy, respectively, and $T$ and
$T_{ref}$ are the actual temperature and the reference temperature, respectively.

$$k_i = k_{i0} \cdot \exp \left[ \frac{E_a}{R} \left( \frac{1}{T_{ref}} - \frac{1}{T} \right) \right],$$  (8)

$$K_{aj} = K_{aj0} \cdot \exp \left[ \frac{\Delta H_j}{R} \left( \frac{1}{T_{ref}} - \frac{1}{T} \right) \right],$$  (9)

$$K_{ei} = \exp \left( \frac{\Delta G_i^0}{RT} \right).$$  (10)

ROUX (2011)’s mathematical models for the reactors use a one-dimensional, steady state
analysis on a control volume $dV$ between axial lengths $x$ and $x + dx$. In order to provide a
fair comparison of the results for the MR and PBR technologies, both reactors are calculated
considering the same cross section area $A_c$, which, for the MR, is described as a function of its
inner radius $R_i$, the permeation zone’s outer radius $r_0$ and the membrane thickness $\delta$, according
to Equation (11).

$$A_c = \pi [R_i^2 - (r_0 + \delta)^2].$$  (11)
The PBR system is correctly defined by two differential equations: one for the methane conversion and another for the carbon dioxide conversion, which are obtained from the mass balances on the control volume $dV$. The resulting system is described by Equations (12) and (13) as a function of the dimensionless length, which, in turn, is described by Equation (14), where $\rho_b$ is the catalyst bulk density in kg/m$^3$, $L$ is the reactor length in m, $F_{CH_4}^0$ is the initial molar flow rate of methane in kmol/h and $\eta$ is the effectiveness factor of each reaction.

$$\frac{dX_{CH_4}}{d\xi} = \frac{\rho_b A_c L}{F_{CH_4}^0} (\eta_1 r_1 + \eta_3 r_3),$$  \hspace{1cm} (12)

$$\frac{dX_{CO_2}}{d\xi} = \frac{\rho_b A_c L}{F_{CH_4}^0} (\eta_2 r_2 + \eta_3 r_3),$$  \hspace{1cm} (13)

$$\xi = \frac{x}{L}.$$  \hspace{1cm} (14)

The MR system uses the same differential equations of the PBR, with the addition of a third one for the hydrogen flow ratio, as described in Equation (15), where $P_{H_2}^{ret}$ and $P_{H_2}^{perm}$ are the hydrogen partial pressures in bar in the reaction and permeation zones, respectively, and $Q_{pd}$ is the hydrogen permeability in m$^3$.\mu.m.h/m$^2$.bar$^{0.5}$, whose expression is described in (AYTURK; KAZANTZIS; MA, 2009) considering a pure platinum membrane according to Equation (16).

$$\frac{dY_{H_2}}{d\xi} = \frac{Q_{pd} 2\pi (r_0 + \delta) L}{F_{CH_4}^0 \delta \cdot 22.4} \left[ (P_{H_2}^{ret})^{0.5} - (P_{H_2}^{perm})^{0.5} \right],$$  \hspace{1cm} (15)

$$Q_{pd} = 6322.7 \cdot \exp \left( -\frac{15630}{RT} \right),$$  \hspace{1cm} (16)

The mathematical models use the boundary conditions described in Equation (17), considering that at the reactors’ inlets the methane conversion, carbon dioxide conversion and hydrogen flow ratio are equal to zero, with the last condition being used only in the MR system.

$$At \ \xi = 0 \rightarrow X_{CH_4} = 0, \ \ X_{CO_2} = 0, \ \ Y_{H_2} = 0.$$  \hspace{1cm} (17)

3 METHODOLOGY

This work used the open source software Scilab 6.0.2 to run the aforementioned reactor models. An extensive list of the kinetic constants and thermodynamic properties used in the simulations can be checked in (ROUX, 2011). A set of parameters to be used in the sensitivity analyses and optimization studies (i.e. reactor dimensions and operating conditions) were
proposed by the author and are presented in Table 1.

<table>
<thead>
<tr>
<th>Parameter / Operating condition</th>
<th>Symbol</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reactor length</td>
<td>$L$</td>
<td>7</td>
<td>m</td>
</tr>
<tr>
<td>Reactor inner radius</td>
<td>$R_i$</td>
<td>6.35</td>
<td>cm</td>
</tr>
<tr>
<td>Permeation zone outer radius</td>
<td>$r_0$</td>
<td>1</td>
<td>in</td>
</tr>
<tr>
<td>Membrane thickness</td>
<td>$\delta$</td>
<td>5</td>
<td>$\mu$m</td>
</tr>
<tr>
<td>Permeation zone pressure</td>
<td>$P_p$</td>
<td>1</td>
<td>bar</td>
</tr>
<tr>
<td>Methane inlet</td>
<td>$F_{\text{CH}_4}$</td>
<td>1</td>
<td>kmol/h</td>
</tr>
<tr>
<td>Inlet water/methane ratio sweep</td>
<td>$\theta_{\text{H}_2\text{O}}$</td>
<td>3</td>
<td>–</td>
</tr>
<tr>
<td>Sweep gas/methane ratio sweep</td>
<td>sweep</td>
<td>10</td>
<td>–</td>
</tr>
</tbody>
</table>

Source: (ROUX, 2011).

The actual reactor inlet consists solely of methane and water; however, as pointed out by ROUX (2011), an impossible numerical value can appear at the reaction rates when the initial hydrogen partial pressure is taken as zero, due to its use as a denominator on Equations (4) to (6). Therefore, an initial hydrogen/methane ratio equal to 0.01 was used in the simulation as it was deemed by the author to be a small enough value to be negligible in front of the other flow rates while also ensuring there would be no convergence issues in the mathematical model.

The differential equation systems that describe the reactor models were solved using the “ode” subroutine with the Adams method. An iteration step of 0.001 was used since it provided the highest precision without creating errors in the subroutine.

Two sensitivity analyses were carried using the simulation of the reactor models: the first one for the dependence of methane conversion on temperature and pressure, and the second one for the same conversion’s dependence on temperature and inlet water/methane ratio.

3.1 Optimization Functions

Alongside the sensitivity analyses, optimization studies were carried to obtain the conditions for maximum efficiency of the membrane reactor. In these analyses, the main variable considered for evaluating reactor performance was the methane conversion ($X_{\text{CH}_4}$).

Since no limitations for operating conditions were presented in (ROUX, 2011) (e.g. temperature and pressure limits for the components and the catalyst), the optimization studies in the present work were formulated as unconstrained problems. However, the methane conversion in both reactors increases along with the temperature, reaching maximum values at approximately 550 °C for the MR. Since one of the major drawbacks of the MR technology is the implementation cost, to determine the operating conditions at which the MR has the highest...
efficiency over the PBR technology, this study used the objective function \( \Delta \), as proposed in (ROUX, 2011) according to Equations (18) and (19), where \( X_{CH_4}^{MR} \) and \( X_{CH_4}^{PBR} \) are the methane conversion in the MR and the PBR, respectively. In the sensitivity analyses, the \( \Delta \) function was optimized using the reactor temperature as decision variable \( x \).

\[
\max_x [\Delta], \tag{18}
\]

\[
\Delta(x) = X_{CH_4}^{MR} - X_{CH_4}^{PBR}. \tag{19}
\]

Another series of optimization studies was carried by dividing the reactors into several isothermal segments of equal lengths, in order to find an optimum temperature profile and increase the \( \Delta \) function at different pressure conditions. In these studies, \( x \) was taken as a decision variable vector, where each element refers to the temperature of an isothermal segment. All optimization problems were solved using Scilab’s “fminsearch” subroutine.

4 RESULTS

This section presents the results of the sensitivity analyses and optimization studies of the MR and the PBR’s mathematical models. Since (ROUX, 2011) is mostly comprised of several case studies with few precise, numerical results, a graphical analysis was used to validate the reactor models. The temperature and methane conversion profiles obtained were like the results provided by the original author, showing that the mathematical models were correctly implemented in the Scilab language.

4.1 Sensitivity Analysis

The results of the temperature \( \times \) pressure sensitivity analysis for the MR and the PBR are presented in Figure 2, showing that, while the temperature increases methane conversion for both reactors, the pressure increases conversion in the MR and decreases it in the PBR.

Results for the \( \Delta \) function are presented in Figure 3, demonstrating that its value increases with the total pressure but peaks for temperatures from approximately 500 to 600 °C. This is explained due to the MR achieving near maximum conversion at this range, making further temperature increases to gradually reduce the benefits of its implementation.

These results were also used to determine the temperatures at different pressures to achieve a minimum value of 0.7 for the \( \Delta \) function, which was considered an enough increase in methane conversion to justify implementation of the MR. These recommended ranges are presented in Table 2 alongside the optimum temperature \( T_{opt} \) at each pressure and the
Figure 2: $T \times P$ sensitivity analysis for the MR and the PBR.

Figure 3: $T \times P$ sensitivity analysis for the $\Delta$ function.

Source: The authors (2020).

Figure 4 presents the results of the temperature $\times$ inlet water/methane ratio sensitivity analysis for the MR and the PBR at a pressure of 30 bar.

While for the MR the temperature remained as the main variable for increasing methane conversion, the inlet water/methane ratio was shown to considerably increase conversion for the PBR at higher temperature values.
Table 2: Recommended temperatures from the $T \times P$ sensitivity analysis.

<table>
<thead>
<tr>
<th>$P$ (bar)</th>
<th>Temperature range ($^\circ$C)</th>
<th>$T_{opt}$ ($^\circ$C)</th>
<th>$\Delta_{opt}$</th>
<th>$X_{CH_4}^{MR}$</th>
<th>$X_{CH_4}^{PBR}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>542 - 586</td>
<td>563.44</td>
<td>0.728</td>
<td>0.983</td>
<td>0.255</td>
</tr>
<tr>
<td>20</td>
<td>497 - 629</td>
<td>540.96</td>
<td>0.830</td>
<td>0.994</td>
<td>0.164</td>
</tr>
<tr>
<td>30</td>
<td>480 - 656</td>
<td>526.74</td>
<td>0.872</td>
<td>0.997</td>
<td>0.125</td>
</tr>
<tr>
<td>40</td>
<td>470 - 675</td>
<td>516.77</td>
<td>0.894</td>
<td>0.998</td>
<td>0.104</td>
</tr>
<tr>
<td>50</td>
<td>463 - 691</td>
<td>509.14</td>
<td>0.909</td>
<td>0.998</td>
<td>0.089</td>
</tr>
</tbody>
</table>

Source: The authors (2020).

Figure 4: $T \times \theta_{H_2O}$ sensitivity analysis for the MR and the PBR.

Source: The authors (2020).

Results of the $T \times \theta_{H_2O}$ sensitivity analysis for the $\Delta$ function at the same pressure of 30 bar are presented in Figure 5, showing that the function reaches its maximum values for temperatures from approximately 500 to 700 $^\circ$C. As previously mentioned, the inlet water/methane ratio is a more influential variable on the PBR, which is demonstrated by the maximum values of the $\Delta$ function being observed at lower values of $\theta_{H_2O}$.

The results of this sensitivity analysis were also used to determine the temperatures at different inlet water/methane ratios for a minimum value of 0.7 for the $\Delta$ function at a pressure of 30 bar. The recommended temperature ranges, alongside the optimum temperatures and the corresponding values of the $\Delta$ function and methane conversion on the MR and PBR are presented in Table 3.

The optimization results show that the higher values of the $\Delta$ function are achieved at $\theta_{H_2O}$ values from 2 to 3, which explains the value of 3 suggested by ROUX (2011), previously presented in Table 1.
Figure 5: $T \times \theta_{H_2O}$ sensitivity analysis for the $\Delta$ function

Table 3: Recommended temperatures from the $T \times \theta_{H_2O}$ sensitivity analysis for $P = 30$ bar.

<table>
<thead>
<tr>
<th>$\theta_{H_2O}$</th>
<th>Temperature range (°C)</th>
<th>$T_{opt}$ (°C)</th>
<th>$\Delta_{opt}$</th>
<th>$X_{CH_4}^{MR}$</th>
<th>$X_{CH_4}^{PBR}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>604 - 743</td>
<td>666.00</td>
<td>0.755</td>
<td>0.923</td>
<td>0.168</td>
</tr>
<tr>
<td>2</td>
<td>485 - 695</td>
<td>549.75</td>
<td>0.875</td>
<td>0.988</td>
<td>0.113</td>
</tr>
<tr>
<td>3</td>
<td>480 - 656</td>
<td>526.74</td>
<td>0.872</td>
<td>0.997</td>
<td>0.125</td>
</tr>
<tr>
<td>4</td>
<td>481 - 626</td>
<td>524.85</td>
<td>0.845</td>
<td>0.996</td>
<td>0.151</td>
</tr>
<tr>
<td>5</td>
<td>485 - 603</td>
<td>525.72</td>
<td>0.816</td>
<td>0.994</td>
<td>0.178</td>
</tr>
<tr>
<td>6</td>
<td>490 - 584</td>
<td>527.10</td>
<td>0.787</td>
<td>0.991</td>
<td>0.204</td>
</tr>
</tbody>
</table>

Source: The authors (2020).

4.2 Temperature Profile Optimization

The temperature profile optimization studies were carried for 5 different pressure conditions. The number of reactor segments for each pressure was determined after multiple tests to maximize the $\Delta$ function. The obtained temperature profiles are presented in Figure 6, along with the optimum temperatures for the isothermal models as previously presented in Table 2. It should be noted that pressure drop was not considered in this optimization or in the previous calculations since initial tests by ROUX (2011) showed it has no influence in the reactor systems.

The results for all evaluated pressure conditions shared similar profiles, with high inlet temperatures followed by a gradual reduction until achieving lower outlet temperatures than
obtained for the isothermal model. The number of reactor segments for each pressure, inlet and outlet temperatures, optimum $\Delta$ function values and corresponding methane conversions for the MR and PBR are presented in Table 4.

**Table 4: Temperature profile optimization results.**

<table>
<thead>
<tr>
<th>$P$ (bar)</th>
<th>Reactor segments</th>
<th>Inlet temp. ($^\circ$C)</th>
<th>Outlet temp. ($^\circ$C)</th>
<th>$\Delta_{opt}$</th>
<th>$X_{CH_4}^{MR}$</th>
<th>$X_{CH_4}^{PBR}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>8</td>
<td>1339.14</td>
<td>473.58</td>
<td>0.836</td>
<td>0.977</td>
<td>0.141</td>
</tr>
<tr>
<td>20</td>
<td>8</td>
<td>1153.57</td>
<td>466.07</td>
<td>0.891</td>
<td>0.989</td>
<td>0.099</td>
</tr>
<tr>
<td>30</td>
<td>9</td>
<td>817.26</td>
<td>463.80</td>
<td>0.912</td>
<td>0.994</td>
<td>0.082</td>
</tr>
<tr>
<td>40</td>
<td>9</td>
<td>753.20</td>
<td>462.76</td>
<td>0.924</td>
<td>0.996</td>
<td>0.073</td>
</tr>
<tr>
<td>50</td>
<td>10</td>
<td>720.53</td>
<td>462.18</td>
<td>0.931</td>
<td>0.998</td>
<td>0.067</td>
</tr>
</tbody>
</table>

**Source:** The authors (2020).

Results showed that, while all pressure conditions have similar outlet temperatures, the inlet temperature is considerably reduced at higher pressure values, which is in agreement with the results obtained for the $T \times P$ sensitivity analysis, which demonstrated that the $\Delta$ function is increased at higher pressures.

Compared to the optimum temperatures presented in Table 2, while the optimized temperature profiles did not increase methane conversion in the MR, the $\Delta$ function was maximized due to lower conversion values for the PBR, thus increasing the membrane technology’s efficiency.
5 CONCLUSIONS

This work used Scilab to run the mathematical models presented by ROUX (2011) for a membrane reactor and a packed bed reactor for producing hydrogen through methane steam reforming and water gas shift. Two sensitivity analyses were carried to assess the effects of three process variables on methane conversion, and their results showed that temperature was the most influential parameter. A series of optimization studies was used to find the operating conditions for maximum efficiency of the membrane reactor, while also obtaining recommended temperature ranges at different pressure and inlet water/methane ratio conditions for maintaining high performance. Lastly, the temperature profile optimizations considering isothermal reactor segments provided better results for the objective function than the isothermal models, further justifying the implementation of the membrane technology.

ACKNOWLEDGMENTS

This study was financed in part by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - Brasil (CAPES) - Finance Code 001.

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