

Engineering model of an electrically heated steam methane reforming reactor[#]

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ABSTRACT

Electrification of steam methane reforming is an interesting approach towards the EU carbon neutrality target. This paper presents a dynamic continuous-stirred-tank-reactors (CSTRs) model for simulating an electrically heated washcoat tube that represents a steam methane reforming reactor. Results from the engineering model are presented, such as temperature, gas molar fraction, CH₄ conversion, and effects of dynamic operation. The model is validated by a previously published CFD model, which has been validated by experimental data. The engineering model provides much faster results than the CFD model and can accurately re-produce the CH₄ conversion and the temperature profile in the reactor.

Keywords: Electrification, Steam reforming, Catalysis, Methane

NOMENCLATURE

$A_{t,in}$	Inner surface area of the coated catalyst channel [m ²]
$A_{t,out}$	Outer surface area of the coated catalyst channel [m ²]
$C_{p,i}$	Gas heat capacity [$\frac{J}{mol \cdot K}$]
$C_{p,tube}$	Tube heat capacity [$\frac{J}{kg \cdot K}$]
d	Inner diameter of the tube without coat [m]
D	Outer diameter of the tube without coat [m]
$F_{i,n}$	Molar flow rate of component i out of the n 'th reactor [mol/s]
h_T	Heat transfer coefficient [W/m ² K]
ΔH_i	Heat of reaction [kJ/mol]
i	Number of species
L	Length of the tube channel [m]
m_{cat}	Mass of catalyst in the n th reactor [kg]
m_{CSTR}	Mass of each CSTR reactor [kg]
n	Index of reactor

$N_{i,n}$	Number of moles of component i in the n th reactor [mol]
P	Pressure [Pa]
P_E	Input power [W]
R_j	Reaction rate of reaction j [$\frac{mol}{(kg \text{ cat}) \cdot s}$]
$R(P_{i,n,s})$	Reaction rate of species i in the n th reactor [$\frac{mol}{(kg \text{ cat}) \cdot s}$]
T_n	Gas temperature in reactor n [K]
$T_{wall,n}$	Tube wall temperature of reactor n [K]
T_{ref}	Reference temperature [K]
<i>Abbreviations</i>	
CSTR	Continuous stirred-tank reactor
WGS	Water gas shift
<i>Greek</i>	
δ_{cat}	Thickness of the catalyst coat [μm]

1. INTRODUCTION

Steam methane reforming is a commonly used approach in commercial syngas production [1]. An electrically heated steam reforming reactor allows a compact reactor design with low pressure drop [2], which is ideal for industrial hydrogen production. This paper describes a CSTR in series model that is used to simulate an electrically heated tube with a catalytic coating. To the authors knowledge there are no similar engineering models for electrically heated steam reforming reactors in the literature.

The Ni-based catalyst is coated as a thin layer (100 μm) on the inside wall of the tube, and the reactions only takes place here. The gases flow into the reactor with a volumetric flow rate of 200 NL/h under a pressure of 26 bar and a temperature of 473 K. The total length of the tube is 1 meter, and its inner diameter is 5 mm. For the simulations, the tube is divided into 10 CSTRs in series which was found by a sensitivity analysis to be sufficient.

2. MATHEMATICAL MODEL

The CSTR in series model assumes the coated tube can be divided into many small reactors, the input in each reactor is the output from the previous. The concept is shown in Fig. 1. The transient species and energy balances for the gas phase and solid wall are written in Equation (1), (2) and (3). The reaction kinetics are from Xu and Froment [2]. The heat and mass transfer between the wall the gases are calculated based on the heat and mass transfer coefficients.

$$\frac{dN_{i,n}}{dt} = F_{i,n-1} - F_{i,n} + m_{cat}R(P_{i,n,s}) \quad (1)$$

$$\sum_i N_{i,n} C p_i(T_n) \frac{dT_n}{dt} = \sum_i F_{i,n-1} \int_{T_{ref}}^{T_{n-1}} C p_i(T') dT' - \sum_i F_{i,n} \int_{T_{ref}}^{T_n} C p_i(T') dT' + h_T A_{t,in} (T_{wall,n} - T_n) \quad (2)$$

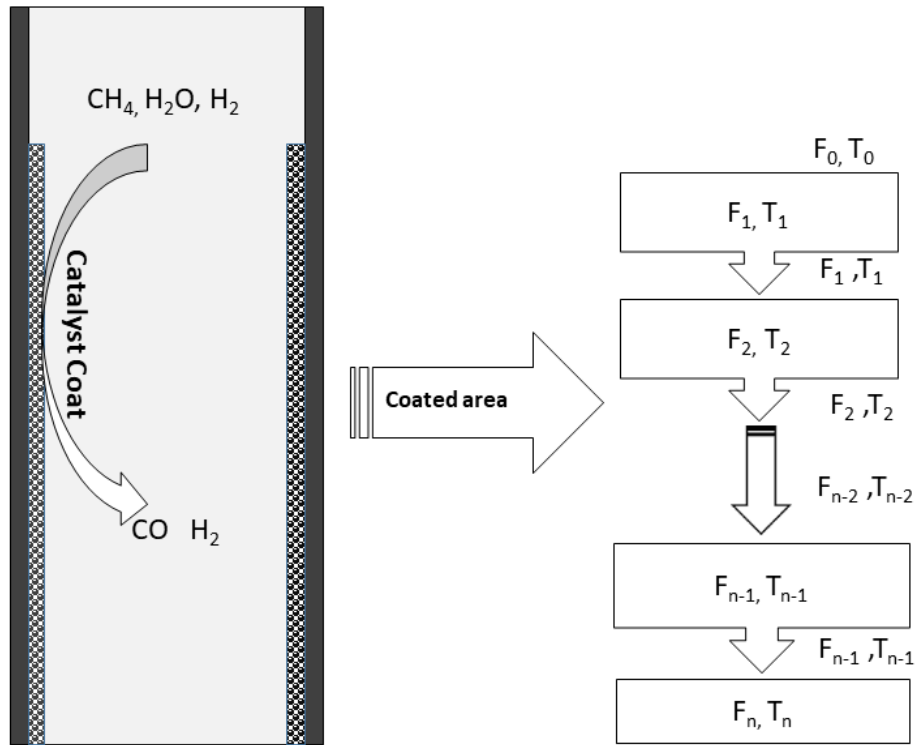


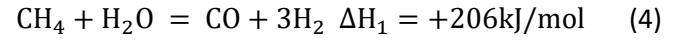
Fig. 1 Structure of the tube reactor and the concept of CSTRs in series

3.1 Detailed simulation results under various powers

In this study, the coated area of the tube is divided into 10 CSTRs, and a total time of 500 seconds of

$$(C p_{tube} m_{CSTR} + C p_{cat} m_{cat}) \frac{dT_{wall,n}}{dt} = h_{T,n} A_{t,in} (T_{g,n} - T_{wall,n}) + \sum_j \Delta H_j(T_n) R_{j,n} m_{cat} + P_E \quad (3)$$

The reactions that happen on the catalyst surface are steam reforming (4) and water-gas shift (5) reactions:



3. SIMULATION RESULTS

The engineering model can predict the temperature and gas concentrations in each CSTR under different operating conditions. In this paper, the main results will be presented under the power of 188 W. For the model validation, the methane conversion and outlet gas temperature under a series of input powers are compared with the results from a detailed CFD model [3].

Table 1 Tube geometry and Initial conditions for the simulation

Flow rate [NL/h]	Geometry				Feed molar fractions					
	L[m]	D[mm]	d[mm]	δ_{cat} [μm]	CH ₄	H ₂ O	H ₂	CO	CO ₂	N ₂
150-300	1	6	5	100	0.32	0.655	0.025	0	0	0

Fig. 2 shows the gas temperature and wall temperature at the outlet of each reactor when the external heating source is 188 W. Reactor 1 is the first reactor in the tube, and reactor 10 is the last reactor in the tube. As is shown in Figure 2, the temperatures increase towards the outlet of the tube. In the first few reactors, the temperature increase is steeper because the wall temperature is lower in the first few reactors, and therefore the reaction rate is small, and the energy that passes from the wall is mainly used to heat up the gas. When the temperature reaches the around 900 K, the reaction rate becomes significant which means that a large fraction of energy is used for the endothermic reaction, and the temperature rise becomes slower.

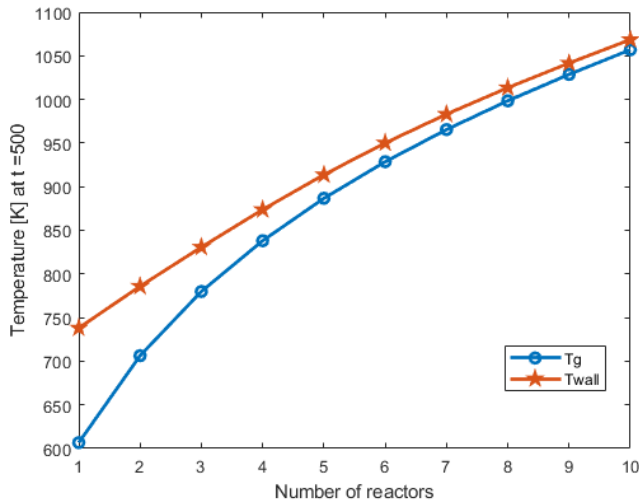


Fig. 2 Gas temperature at the outlet of each reactor at steady state

Fig. 3 shows the gas molar fractions at the outlet the tube. The tube is initially filled with N₂, as is shown in Fig. 3a. When the feed gas flows in, the N₂ is quickly flushed out. As for the other species, especially for CH₄ and H₂O (Fig. 3 b and c), the molar fraction of these two species reaches the feed ratio immediately after N₂ is removed because the temperature of the tube is still too low for any appreciable reaction to happen. After around 60 seconds, both molar fractions of the two species start to decrease, indicating the steam reforming reactions are taking place. This can also be seen from the product side (Fig. 3d and e), where both molar fractions of H₂ and CO start to increase. The molar fraction of CO₂ (Fig. 3f) is much lower than any other species during the whole process. After a certain time, the molar fraction of CO₂ decreases, because the tube reaches a higher temperature and the WGS reaction shift backwards.

Fig. 4 shows both the gas temperature profile and the CH₄ concentration profile for various input powers after the reactor reached steady state. The temperature at each location increases with increasing power. Consequently, the methane concentration decreases with increasing power and along the tube, due to higher temperature. The increasing slope in the temperature profile towards the outlet at a power of 300.8 W indicates near complete methane conversion decreasing the heat consumed by the endothermic reaction. Therefore, the model can be used to indicate safe operating conditions for the reactor, without melting.

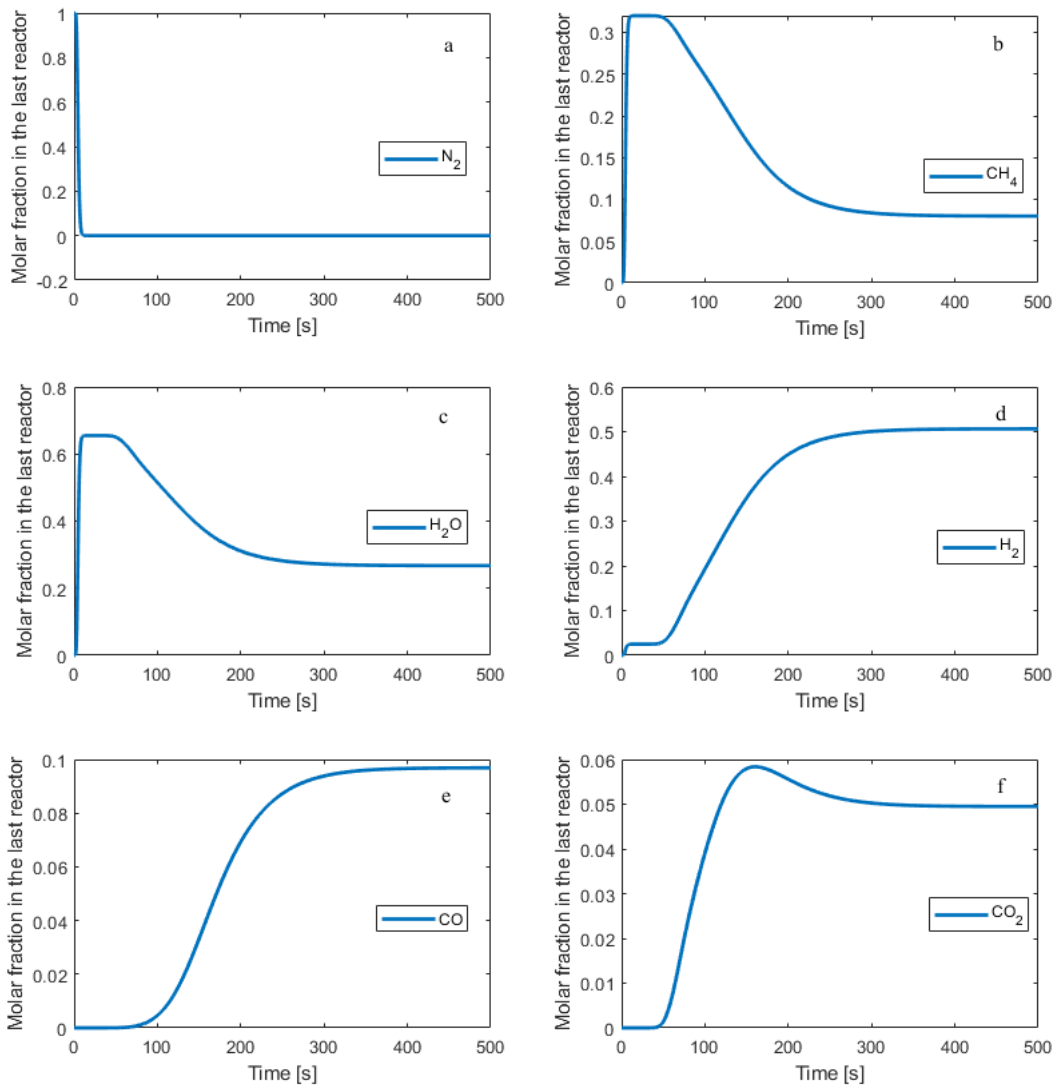


Fig. 3 Gas molar fraction in the last reactor

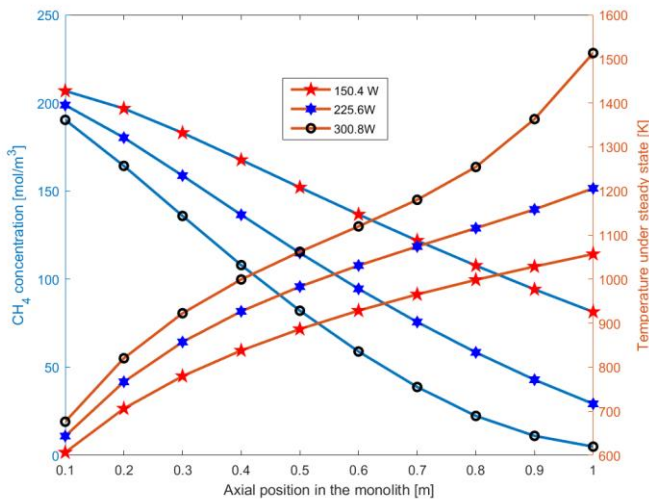


Fig. 4 Temperature and CH_4 concentrations under the power of 150.4, 225.6, 300.8 W, corresponding to a certain axial position, under steady state

3.2 Model validation against the CFD model

Wisnmann et al [3] described a CFD model developed by COMSOL, where the calculation of electric currents, thermal energy, fluid dynamics, mass transport and reaction kinetics are included. In the CFD model, the methane conversion is calculated under a series of power inputs, namely, 75.2, 112.8, 150.4, 188.0, 225.6 and 263.2 W. Fig. 5 and Fig. 6 compares outlet gas temperature and methane conversion, two key parameters, that are calculated from the engineering model presented in this paper and the validated CFD model [3].

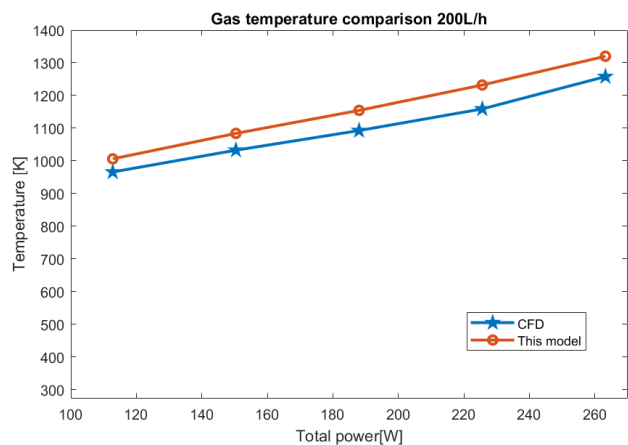


Fig. 5 Comparison of gas temperatures from engineering and CFD model.

At an inlet flowrate of 200 NL/h, the engineering model predict a slightly higher gas temperature than the CFD model. As for the methane conversion, the engineering model can predict almost the same conversion with the CFD model.

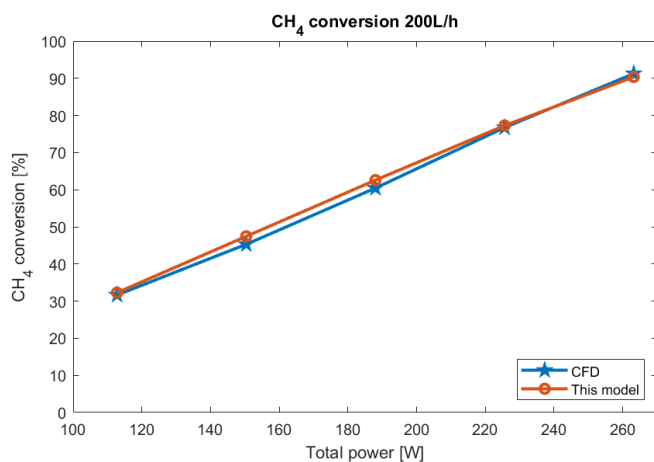


Fig. 6 Comparison of methane conversion from the engineering and CFD model.

4. CONCLUSIONS

This paper presents a dynamic engineering model of an electrically heated steam methane reforming reactor. The model predicts the axial profiles of gas and wall temperature and gas molar fractions. The validation of gas temperature and the methane conversion against a CFD model shows the engineering model can predict relatively accurate results. The engineering model can be a central tool for scaling up electrically heated chemical reactors.

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