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Design of a biogas steam reforming reactor: A modelling and experimental approach

Francesco Cipiti*, Orazio Barbera, Nicola Briguglio, Giosuè Giacoppo, Cristina Italiano, Antonio Vita

CNR, Institute for Advanced Energy Technologies “Nicola Giordano”, Via S. Lucia sopra Contesse n. 5, 98126 Messina, Italy

ARTICLE INFO

Article history:

Received 28 September 2015

Received in revised form

9 December 2015

Accepted 14 December 2015

Available online 8 January 2016

Keywords:

Biogas

Steam Reforming

Hydrogen production

Fixed bed reactor

Heterogeneous catalysis

ABSTRACT

This paper covers the research activities performed at the CNR Institute for Advanced Energy Technologies “Nicola Giordano”, aimed at developing and testing a biogas steam reforming reactor. A mathematical model has been developed in order to describe the performance of the above-cited steam reforming reactor (packed bed). To study the effects on reaction performance, a parametric analysis was performed varying operating conditions such as inlet temperature and reagent molar ratio. The model was validated by comparing the calculated data with the experimental data obtained with a proprietary Ni/CeO₂ based catalyst in packet bed micro-scale reactor at different $T = 700\text{--}900\text{ }^{\circ}\text{C}$, $S/C = 1\text{--}5$ and $GHSV = 30,000\text{ h}^{-1}$, showing a good agreement between the experimental and theoretical results.

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Introduction

There is an urgent need in Europe to increase the use of renewable fuels to comply with the EU 20–20–20 objectives [1]. In this scenario biomasses and the derived biofuels represent a source of alternative energy.

The biogas, obtained from a variety of biomasses and processes such as degradation of urban and industrial waste, landfills, co-digestion of zootechnical effluents, agricultural waste and energy crops, can be considered as one of the most known and widespread renewable fuels [2]. At present, biogas is mainly used to produce electricity and heat using cogeneration systems; internal combustion engines (ICE) and gas turbines represent the most used technologies, this type of utilization is strongly influenced by the limits of the

conventional engines: low electrical conversion, high levels of noise, maintenance and pollutant emissions, especially for small biomass gathering terminals [3–6].

More attention has been paid to an alternative use of biogas for hydrogen/syngas production. In this respect, there is an increasing interest for small scale locally distributed hydrogen production units, based on reforming processes, to serve as sources of hydrogen for fuel cells [7]. The production of syngas is important also for the utilization as a raw material for the production of synthetic fuels with low environmental impact (hydrogen, methanol, dimethylether and synthetic gasolines) [8].

The principal components of biogas are CH₄ and CO₂, therefore, the Dry Reforming (DR) process could be the most appropriate to produce hydrogen/syngas, but the severe

* Corresponding author. Tel.: +39 090 624 297; fax: +39 090 624 247.

E-mail address: francesco.cipiti@itae.cnr.it (F. Cipiti).

<http://dx.doi.org/10.1016/j.ijhydene.2015.12.053>

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Table 1 – Reaction equilibrium constants and Arrhenius kinetic parameters.

Reaction, j	Equilibrium constant, K_j	k_{oj} (mol/(kg _{cat} s))	E_j (J/mol)
1	$K_I = \exp\left(\frac{-26,830}{T_s} + 30.114\right)$ (bar ²)	1.7×10^{15} bar ^{0.5}	240,100
2	$K_{II} = K_I K_{III}$ (bar ²)	2.3×10^{14} bar ^{0.5}	243,900
3	$K_{III} = \exp\left(\frac{4,400}{T_s} - 4.036\right)$	5.43×10^5 bar ⁻¹	67,130

$k_j = k_{oj} \times \exp(-E_j/RT)$.
R is gas constant in J/(mol*K).

operating conditions (high reaction temperature and absence of steam) resulting in catalyst deactivation by coke deposition and/or sintering of the metallic phase and support [9]. This means that more efforts must be taken, especially for the development of active and stable catalyst formulations.

Actually, Steam Reforming of Methane (SRM) is the most common chemical process to convert methane into hydrogen. SRM is an endothermic reaction and the ratio of water and methane is 1:1 that is calculated from stoichiometry of SRM, however, excess steam is usually employed to suppress carbon formation (around more than 3 times of stoichiometry). Thus, the Steam Reforming of biogas in heterogeneous catalytic reactors, is expected to become a short-term viable method for the production of hydrogen/syngas [10,11]. Ni-based catalysts were investigated mostly for this reaction and are of potential for industrial application [12–14].

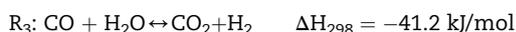
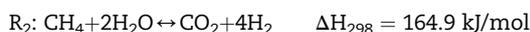
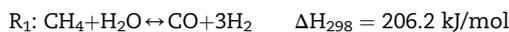
The design of a biogas reactor is the key aspect for the performance and efficiency of a hydrogen generator: weight and volume should be minimized and the heat management system optimized for different operating conditions. In this respect, the development of a biogas steam reforming reactor, has been carried out at the CNR/ITAE Institute. Particularly, a mathematical model has been developed in order to describe, the performance of the above-cited steam reforming reactor (packed bed). The aim of the model is to investigate the performance of the reactor in order to enhance optimization and control of the steam reforming unit. The model plays a key role in overcoming the issues of system design, by evaluating the temperature and the gas concentration profiles in the reactor. The model was validated by comparing the calculated data with the experimental data obtained with a proprietary Ni/CeO₂ based catalyst [14] in packet bed micro-scale reactor at different $T = 700\text{--}900$ °C, $S/C = 1\text{--}5$ and $GHSV = 30,000$ h⁻¹.

Computational methods

Reactor modelling

The simulation of the processes is achieved through the description of transport phenomena by Partial Differential Equations (PDEs), numerically solved using the Finite Element Methods. The model is able to couple heat and mass transport and chemical reactions occurring in the reactor. Energy and mass balances have allowed to define the temperature and the products profiles along the length of the reactor.

According to the literature the following reactions are the prevailing reaction routes [15]:



As the first two reactions are highly endothermic, excessive amounts of heat have to be supplied to the reactor in order to maintain the desired high temperature.

The reactor is simplified and modelled as a non-isothermal plug flow reactor. Assuming a steady-state, the main equations of the model are presented below:

Species mass balance:

$$\frac{dF_i}{dV} = R_i \quad (1)$$

where F_i is the species molar flow rate (mol/s), V represents the reactor volume (m³), and R_i represents the species net reaction rate (mol/(m³ s)).

Reactor energy balance:

$$\sum_i F_i C_{p,i} \frac{dT}{dV} = w_s + Q + Q_{ext} \quad (2)$$

where $C_{p,i}$ is the species molar heat capacity (J/(mol·K)), and Q_{ext} is the heat added to the system per unit volume (J/(m³·s)). Q denotes the heat due to chemical reaction (J/(m³·s)).

$$Q = \sum_j H_j r_j \quad (3)$$

where H_j the heat of reaction (J/mol), and r_j the reaction rate (mol/(m³ s)).

Table 2 – Van't Hoff parameters for species adsorption.

	K_{oi} (bar ⁻¹)	ΔH_i (J/mol)	K_{oi}^C (bar ⁻¹)	ΔH_i^C (J/mol)
CH ₄	6.65×10^{-4}	-38,280		
CO	8.23×10^{-5}	-70,650		
H ₂	6.12×10^{-9}	-82,900		
H ₂ O	1.77×10^5 bar	88,680		

$K_i = K_{oi} \times \exp(-\Delta H_i/RT)$.
R is gas constant in J/(mol*K).

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