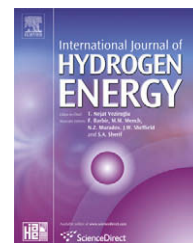


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Partial oxidation of methane in a reverse flow porous media reactor. Water admixing optimization

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ARTICLE INFO

Article history:

Received 1 April 2008

Received in revised form

9 July 2008

Accepted 10 July 2008

Available online 14 September 2008

Keywords:

Partial oxidation

Methane

Hydrogen

Filtration combustion

Numerical simulation

Porous media

Optimization

ABSTRACT

The influence of the addition of steam on methane–air partial oxidation in a reverse flow porous media reactor is investigated numerically. The model is validated via comparison with the experimental data obtained without steam addition. The model of chemical kinetics includes 6- component overall model and GRI 3.0 gas phase methane oxidation kinetics. It is shown that hydrogen concentration in the product gas may be increased by 0.5–1% and the methane-to-hydrogen conversion ratio by 10–15% by means of adding steam to a working mixture. The optimum equivalence ratio remains the same as in the water free case. Steam concentration which maximizes H_2 is in the range of 5–10%; steam concentration which maximizes the conversion ratio is in the range of 20–50%. The role of the thermal insulation of the reactor and of the working gas preheating in this aspect is shown quantitatively.

The results may be utilized for design of porous media reactors and process optimization.

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1. Introduction

Partial oxidation of methane is considered as a perspective technology of hydrogen production [1,2]. It was shown in the works carried out at the Heat and Mass Transfer Institute (Minsk) [3–5], UIC (Chicago) [6–8], University of Erlangen–Nurnberg [9], Texas University (Austin) [10] and some others that partial oxidation of methane in air may be conducted in an excess enthalpy (“superadiabatic”) wave within inert porous media. The concept of the excess enthalpy combustion in porous media (the term “filtration combustion” is sometimes used) was developed in the works by Weinberg [11], Matros [12], Fateev [13] and evolved in [14,15] and other works. Its essence is in the possibility of the thermal process

intensification in the non-stationary thermal wave due to heat recuperation in a gas-porous medium system.

Methane partial oxidation can be carried out in the so-called reverse flow reactor (RFR), where reactions take place in periodically reproduced co-flow propagating thermal wave [16,17] (Fig. 1). The methane-to-hydrogen conversion ratio, determined by the hydrogen molar output to methane molar input ($[H_2]/2[CH_4]_0$) was in the range ~60–70% [3,7]. Note that this technology was explored and tested for various hydrocarbons (including liquids) conversion [4,18] as well as hydrogen sulfide conversion to hydrogen [7,19].

The practical implementation of partial oxidation in RFR demands optimization of operating conditions, investigation of the influence of constructive and scale up factors on the

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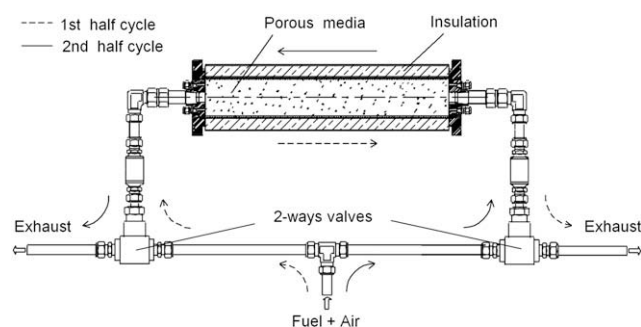


Fig. 1 – Schematic of a reciprocal flow reactor. Arrows correspond to flow direction during the first and the second half of cycle.

product gas cycle composition, methane conversion ratio, temperature fields, etc. Numerical simulation can be effective and in some cases the only reasonable method of the system optimization and design support. The bottleneck problem here is the choice of the adequate model of chemical kinetics. A simple overall model for simulation of partial oxidation of methane in inert porous media was proposed in [20]. The model made it possible to approximate experimental data on partial oxidation of methane and was used to investigate the influence of scale up factors and other parameters on partial oxidation of methane in reverse flow filtration combustion reactor [21]. Among the variable parameters there were the gas flow rate, diameter of porous medium particles, porosity, thermal insulation width and an input pressure.

The influence of the input gas composition on methane conversion is another important aspect of the process optimization. The influence of the enrichment or depletion of oxygen on partial oxidation in inert porous media was theoretically and experimentally explored in [8]. The increase in the hydrogen content in the product gas at the constant fuel-to-oxidizer equivalence ratio ($\Phi = [F/O]/[F/O]_{\text{stoich}}$) was obtained on increase of oxygen. The data on thermal wave velocity and maximum temperature are given for the oxygen content variation from 10 to 35% vol. The data were obtained for the single gas flow rate $u_g = 0.12$ m/s.

Compared to oxygen, water is a cheap and safe component having a potential for improving the methane-to-hydrogen conversion. The influence of water admixing on methane conversion in inert porous media has not been explored consistently as yet. Numerical investigation of this problem as applied to the specific technology seems to be an urgent and important task, especially keeping in mind that numerical investigations of practical filtration combustion systems within the frames of volume-averaged model has become a widely approved and validated practice [5,10,21]. The problem of formulation of an appropriate model of chemical kinetics for the processes is discussed below in Section 3.

This work is a logical continuation of the research [20,21]. The influence of water admixing on partial oxidation of methane in an inert porous medium reverse flow reactor (RFR) is investigated numerically. The appropriate model of chemical kinetics is formulated. The basic parameters of the process such as hydrogen and carbon oxide concentration in

the product gas and maximum temperature of porous media are investigated as functions of the gas mixture flow rate, admixed water content, fuel-to-oxidizer equivalence ratio, working mixture preheating value and the thermal insulation of the reactor. A laboratory scale reactor is under consideration. Recommendations for the optimum processing are formulated. The results of the study can be used for design and optimization of laboratory and industrial scale reactors.

2. Problem statement

Let us consider a RFR reactor consisting of a cylindrical chamber filled with a porous medium (a packed bed of ceramic balls) and a thermal insulation layer. A gas supply system provides constant mass flow rate, gas mixture composition and temperature of the incoming gas and periodical (time period τ_{ctrl}) switch of the flow direction, Fig. 1.

Assume that the radial temperature variation within the reactor chamber is much less than the temperature difference in the insulation layer ($\Delta T_s/\Delta T_{\text{isl}} \ll 1$) and a 1D approximation may be used for reactor simulation.

The geometrical and other parameters of the system correspond to the experimental setup of [4] and are presented in Table 1. The switch period parameter τ_{ctrl} is chosen according to the optimal operation recommendations given in [16] and correlate with the gas flow rate and reactor length, Eq. (10).

Water is added to the working mixture in a gas phase. In practice the temperature of the incoming gas should be raised to avoid liquid condensation if the partial pressure of steam is higher than saturation pressure. For reference, some of the data on saturation pressure are presented: $P_{\text{sat}}(T = 20^\circ\text{C}) = 0.023$ atm;

Table 1 – Parameters of the reactor

Parameter	Dimension	value	Parameter Description
G	Nm^3/h	4	Gas flow rate
D_0	m	0.038	Reactor chamber diameter
L	m	0.38	Reactor length
d_0	m	5×10^{-3}	Packed bed particle diameter
L_{isl}	m	9×10^{-3}	Refractory layer width
β	$\text{W}/\text{m}^3/\text{K}$	2100	Heat loss coefficient
T_0	K	300	Initial temperature of the system
p_0	Pa	1.013×10^5	Pressure at exit of reactor
ε	–	0.6	Emissivity of the packed bed particles
m	–	0.4	Porosity
ρ_s	kg/m^3	2810	Packed bed material density
c_s	$\text{J}/\text{kg}/\text{K}$	794	Packed bed material heat capacity
λ_s	$\text{W}/\text{m}/\text{K}$	0.2	Packed bed material thermal conductivity coefficient
λ_{isl}	$\text{W}/\text{m}/\text{K}$	0.15	Thermal conductivity of the refractory insulation layer
G_m	$\text{kg}/\text{m}^2/\text{s}$	1	Mass flow rate density

Packed bed – Al_2O_3 balls.

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