



Conversion of hydrocarbon gases to synthesis gas in a reversed-flow filtration combustion reactor



S.O. Dorofeenko*, E.V. Polianczyk

Institute of Problems of Chemical Physics, Russian Academy of Sciences, 1 Academician Semenov Ave., Chernogolovka 142432, Russia

HIGHLIGHTS

- We simulate the methane–oxygen–steam reforming in a novel version of reversed flow reactor.
- Parameters of a process are assessed using approximation of thermodynamic equilibrium.
- Separate supply of reactants provides high temperature for low consumption of oxygen.
- This process promises high energy efficiency of POX conversion.

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ABSTRACT

The conversion of hydrocarbon gases to synthesis gases via partial oxidation in a novel type of reversed-flow porous medium reactor is investigated. The reactor is distinguished by separate supply of the hydrocarbon and the oxidant gas. A calculation scheme is given for the combustion regimes and composition of synthesis gas as controlled by the flowrates of the reactants. The calculation is performed under assumption of thermodynamic equilibrium in the combustion products and zero heat loss through the reactor walls. Calculations for the methane–steam–oxygen/air reaction system show that proposed reactor type provides a possibility to combine a high combustion temperature with low net heat effect and thus, a high chemical efficiency of conversion.

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1. Introduction

Conversion of various hydrocarbon feedstocks ranging from natural gas, petroleum gases to pyrolysis tars into synthesis gas, which can be used, e.g., to produce liquid fuels, involves a reforming process. A variety of the processes such as a steam reforming and catalytic partial oxidation have been developed so far. The main problem related to these processes is that catalysts are prone to sulfur poisoning [1]. Since these processes are highly hydrocarbon feedstock- (i.e. fuel-) consuming, another concern is the need to enhance the energy efficiency of the processes.

The non-catalytic partial oxidation processes performed in the superadiabatic regimes in a porous matrix [2] provide an attractive alternative for hydrocarbons reforming into synthesis gas. Those promise elimination of susceptibility of a catalyst by relying on the high reaction temperature, while providing a substantial heat

recuperation via the gas–solid heat exchange. Thus, these processes provide a possibility to attain a high reaction temperature for a relatively low net heat release (which is the energy expenditure of the process). A possible way to achieve such a superadiabatic process is to perform reforming by partial oxidation in a reverse-flow reactor. The reverse-flow reactor consists of a vessel filled with an inert refractory porous material whose heat capacity accumulates the reaction heat as the hot gas flows through the porous bed. When the porous bed heats to the reactor outlet, the flow direction is reversed and the heat accumulated in the bed is used to preheat the incoming cold gas. This approach to conversion of hydrocarbon fuels has been studied extensively [3–9]. Ideally, this process should provide a highest temperature limited only by the structural strength of the porous bed and the reactor vessel, since higher temperature shifts the chemical equilibria in the favorable direction of formation of hydrogen and carbon monoxide. The high temperature also accelerates the chemical reactions and thus provides a higher throughput of the reforming reactor. This high temperature should be achieved for the lowest possible

* Corresponding author.

E-mail address: dorofeenko@icp.ac.ru (S.O. Dorofeenko).

Nomenclature

Symbols

ϕ	stoichiometric ratio for methane oxidation
A	the reactant supplied through a reactor end
B	the reactant supplied through the center inlet
G	the syngas
H_i	molar enthalpy of the i th component [J mol^{-1}]
K_c	partial pressure of carbon vapor over solid carbon [Pa]
K_i	dissociation constant of the i th gaseous component [Pa, Pa ²]
P	pressure [Pa]
T	temperature [K]
T_0	initial temperature of the supplied gas [K]

T_s	temperature of combustion [K]
y, x, w, m, d, h, n	stoichiometric coefficients
z	the spatial coordinate along the reactor axis [m]

Subscripts

i	species $i = x, w, e, m, d, h, n$
s	related to syngas
0	related to supplied gas

Abbreviations

LHS, RHS left, right hand side of equation

energy cost, which in this case is the net heat release from the chemical reactions in the reactor. Owing to heat recuperation within the filtration combustion wave one can combine high temperature favorable for formation of hydrogen and carbon monoxide and low net heat of reaction (hence, energy expenditure of the process). Addition of water (steam) to the reaction system promises further enhancement of hydrogen yield for lower energy expenditure as the endothermic water reactions with methane and other hydrocarbons would yield hydrogen.

It has been shown previously on a simplified model of a single Arrhenius-type rate controlling reaction that the combustion temperature of a reactive gas filtering through a porous bed grows with the gas flow velocity but levels at a limiting value determined by the kinetic parameters of the rate-controlling reaction [10]. The studies of filtration combustion of ultra-rich methane–oxygen–

steam mixtures in inert porous matrices using more complicated detailed reaction kinetic mechanisms show that the process “chooses” for itself the temperature too low to achieve reactions of water with methane having any noticeable rate [8,11].

2. Formulation of the problem

A new version of a reciprocal flow reactor that provides a possibility to achieve a higher heating of the reaction mixture has been proposed in [12]. Here we briefly describe the gist of the method proposed.

Since the feature that prevents attainment of the desired high temperature in filtration combustion of premixed gases is that the reactive mixture prematurely starts to actively react, we prevent this reaction by simply postponing mixing of the reactants until one of them gets highly preheated. Consider first the conversion process for methane with oxygen and steam. Extension of the approach to other hydrocarbon species is straightforward, as will be shown.

The cyclic reversed flow reactor operates as follows. Consider the established cyclic process. Schematic of the process is presented in Fig. 1.

During the first half-cycle, reactant A , in this case methane–steam mixture, is supplied to the reactor 1 via the three-way valve 7 from the lower inlet 3. As the methane–steam mixture filters through the porous matrix 2, which was preheated during previous cycles, it heats up and reaches the middle part of the reactor at a high temperature, while the part of the porous matrix adjacent to the inlet 3 cools down. The reactant B (oxygen) is supplied via the inlet 8 to the middle part of the reactor. Since we wish to obtain syngas by partial oxidation, the oxygen is supplied substoichiometrically. It reacts with methane with a substantial heat release. At high temperature the water might also enter the reaction with methane yielding some additional hydrogen.¹ The reaction zone in the middle part of the reactor is relatively narrow compared to total dimensions of the reactor. The hot syngas formed in the reaction zone filters through the porous matrix, which was cooled down near the outlet 4 during the previous half-cycle, and lends its sensible heat to heat the porous matrix again. Thus, the syngas G leaves the reactor via the outlet 4 and valve 6 to the withdrawal 9 being substantially cool. When the porous matrix in the upper part of the reactor heats up, the valves 6 and 7 are switched

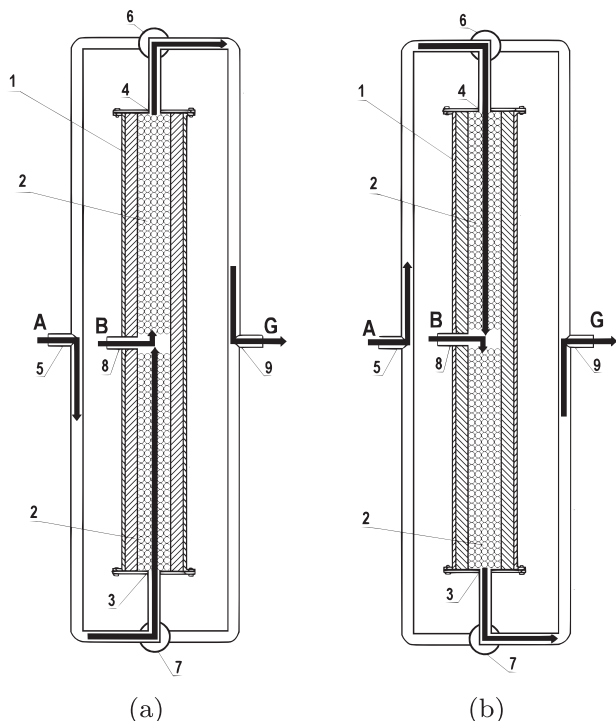


Fig. 1. Schematic of the conversion cycle [12]. (a) first half-cycle; (b) second half-cycle. 1 is reactor vessel, 2 – porous matrix; 3, 4 – inlets/outlets of the reactor, 5 – supply of methane–steam mixture (reactant A); 6, 7 – three-way valves; 8 – supply of oxygen (reactant B); 9 – withdrawal of syngas G .

¹ Water can start reacting with methane even before the heated methane–steam mixture reaches the middle part of the reactor. However this process is kinetically retarded until abundance of free radicals develops in the oxygen reaction zone. For the sake of simplicity so far we disregard this possibility.

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