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Numerical analysis of catalytic-coated walls of an indirect internal reforming solid oxide fuel cell: Influence of catalyst coating distribution on the reformer efficiency

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ABSTRACT

In this research, the performance evaluation of methane (CH_4) steam reforming in Wall Steam-methane Reformer (WCR), intended to supply hydrogen (H_2) to SOFC, is investigated using homemade code based on 2D numerical modeling. Focus is on the design of the catalyst coating on the walls of reactor. Various designs of nickel-based catalyst coating were closely examined and compared purporting to understand their effect on the WCR efficiency. The WCR designs operate at similar industrial operating conditions and with the same catalyst density. The computations were discussed with respect to the possible improvement on WCR efficiency. Comparing to the worst catalyst design found, the CH_4 conversion rate improving is estimated to 31%, which corresponds to 95.8% more in H_2 production. The obtained results are of practical importance for the design of the catalyst coating and saving fuel energy.

1. Introduction

Faced with the inevitable exhaustion of conventional fossil fuels and the harmful effects of their use on the environment, research activities in clean energies field are constantly increasing in line with rising demand for energy in the world. According to the global status report REN21, the total renewable energies capacity exceeded 2000 GW mark for the first time by achieving a growth of 8.7% at the end of 2016 [1]. If electricity remains the most used energy carrier, hydrogen could also join it in the future as a promising clean energy carrier. H_2 is an attractive chemical element for the energy production field thanks to its high potential energy. Although it is the most abundant element in the universe, H_2 needs to be produced, either from its oxidized form H_2O or carbonated form, e.g. CH_4, to benefit from its energetic aspect. So, H_2 is almost obtained by decomposing water using electrolysis process (either continuously or intermittently) or by reforming hydrocarbons with the use of various processes. The most widely used process in hydrogen production is methane reforming by water vapor, known as Methane Steam Reforming (MSR) [2,3].

In this respect, considerable efforts have been deployed by scientific community aimed at improving the hydrogen production yield and at the same time reducing in fuel consumption and greenhouse gas emissions. Several studies have been conducted to assess "fuel consumption-hydrogen production" relationship by improving the reactors efficiency.

In 1996, Adris et al. [4] reviewed the attempts to improve the MSR reaction in conventional reforming reactor configuration. They demonstrated that conventional steam reformers find severe mass and heat transfer limitations caused by the effectiveness factor of catalysts estimated to less than 5%. They noted also that the MSR process can be enhanced by improving the catalyst used in reactors and optimizing their configuration. Since then, there are many works in the literature about MSR intensification for various applications by reactor configuration optimization and catalyst layout optimization.

There are three types of reformers used in hydrocarbon-based fuel conversion [5]. Their classification depends on the H_2 production stage and the way they are integrated in the system such as Fuel Cells (FC): (1) Direct or Indirect Internal Reforming systems (DIR or IIR) as in [6–8]; (2) External Reforming (ER) or Partial Oxidation (PO_x) systems. By DIR system, H_2 is produced just before entering the fuel cell stacks, while by IIR system H_2 generation is processed separately and then is fed into the FC stacks [9–11]. Concerning ER and PO_x systems, H_2 is produced and stocked to be used in various applications. It has been demonstrated that the use of IIR reduces the expensive cost of ER systems by eliminating external heating to supply the endothermicity of MSR reaction, such for high-temperature SOFC [5,12]. The heat supply

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can be ensured by the dissipated heat from SOFC stacks due to its exothermic nature.

In industry, two major MSR reactor configurations are used to feed FC stacks: tubular reactors [13–18] and plate reactors [19–23]. Considering tubular MSR reactor configuration, Hong et al. [24] conducted a numerical study on the effect of the operating and design parameters on the reactor efficiency. Their physical model was composed of several bayonet reforming tubes supplied by heat using burner placed at the center. They investigated the input conditions, the catalyst layer and the number of reforming tubes used. The results show that the reactor gives a better performance by increasing the number of reforming tubes due to the efficient thermal behavior in the system. On the other hand, the reduction of catalyst layer length is not conductive to the H_2 production. However, they did not describe limitations on the catalyst layer length and number of bayonet tubes. Thus, in such types of reaction, the management of the heat provided to the MSR reaction is of primary importance with the catalyst layers distribution. For operating conditions, the effect of reaction pressure and operative temperature and inlet (H_2O/CH_4) was studied via an orthogonal second order design by Farshad et al. [25]. The considered responses were methane conversion, selectivity of hydrogen, selectivity of carbon monoxide, hydrogen per carbon monoxide ratio, steam per carbon monoxide ratio and coke formation. In general, higher temperatures promote the reaction toward syngas formation and improve the conversion of CH_4 and CO_2. This has been confirmed by Chein et al. [26], who investigated the thermodynamics of biogas dry reforming under different temperatures and pressures. The authors indicate that, introducing a large amount H_2O into the reaction system can eliminate carbon. However, the system must be operated at high temperature to produce high CH_4 and CO_2 conversions. However, higher pressures were not suitable for biogas dry reforming. The same authors (Chein et al.) [26] shows that dry reforming of methane (DRM) is unfavorable at high pressure. CO_2 and CH_4 conversions decrease while the carbon formation increases as the pressure increases. With increasing CO_2/CH_4 ratio in the reaction, CH_4 conversion is enhanced, carbon formation is suppressed, but CO_2 conversion is lower due to the excess CO_2 supply. The simulation results indicated that the introduction of inert gas in the system did not produce a significant effect on DRM performance. Nikoo et al. [27] indicate that when the pressure increased from 1 bar to 30 bar the conversion of CH_4 and yield of H_2 decreased, and the effect of pressure suppressed the effect of temperature on reactant conversion, augmented carbon deposition and decreased CO and H_2 production due to methane decomposition and CO disproportionation reactions. Kim et al. [28] studied experimentally the effect of pressure difference on the CH_4 conversion by studying a membrane reactor comprising palladium composite on porous medium. They confirmed that the methane conversion increases with increasing pressure difference between the shell and the tubes of the membrane reactor. For solar thermochemical reactor applications, Wang et al. [18] conducted a numerical study to analyze the effect of irradiation concentration of H_2 production efficiency. Their results showed that the concentration of irradiation in the inlet of the porous reactor caused an irregularity in temperature distribution, which affect strongly the H_2 production.

In a recent study, Diglio et al. [29] conducted 1D modelling study of sorption-enhanced MSR process in fixed bed reactors. They determined the numbers of reactors to connect to obtain a high H_2 purity. In addition, the overall heat energy in the system was managed so that it becomes energy self-sufficient with CO_2 rate close to zero. Regarding the plate reactor configurations which are often assisted to exothermic reactions such as combustion or Water Gas Shift (WGS) as in Ref. [30]. Seo et al. [31] investigated numerically the heat transfer to the catalyst beds and catalyst reaction in a compact reformer coupled with WGS reactor. The heat generated by combustor which supplies the endothermic MSR reaction was estimated to be 76.1% of the combustion overall energy. They concluded that the improvement of the heat transfer behavior could enhance the CH_4 conversion and thus the reformer efficiency. Ni [23] conducted a numerical investigation on hydrogen production in compact reformer. The author studied the effect of the reformer structure containing a porous matrix and operating parameters such as thermal behavior of reformer and the heat ratio of the reactor performance. Chen et al. [32] studied also the operating conditions of DRM reaction in solar porous reactor, and proposed two correlations for heat transfer coefficient and pressure drop within the porous media. They found that when the inlet velocity of reactants is decreasing, the methane conversion is reduced. However, when the effective thermal conductivity of the porous media is enhanced, the fuel conversion is improved. Patel and Sunol [33] propounded mathematical model of MSR reaction in plate membrane coupled thermally with methane combustion within plate burner. The authors analyzed the effect of operating variables on the reactor performance like inlet fuel concentration and velocity. The simulation results show that using membrane in thermally coupled reactors leads to significant improvement in terms of methane conversion and hydrogen recovery. In addition, by imposing a low reformer inlet velocity and high inlet temperature, the methane conversion could be enhanced. In the same context, Jiwanuruk et al. [34] studied numerically the design of a thermally-coupled monolithic reformer for hydrogen-powered vehicles. Different hydrogen sources and arrangement configurations between combustor and reformer have been investigated. The authors confirm that methane gives the best energy efficiency which could be enhanced by applying a checked arrangement instead of parallel one. The improvement of reactive flow conditions in fuel cells has been the subject of the experimental study conducted by Tseng et al. [35]. They proposed the use of metal foam (580 μm pore size) as a flow distributor in a proton exchange membrane fuel cell in place of conventional flow distribution channels. The results showed that, thanks to the high porosity of the metal foams, the performance of the proton exchange membrane fuel cell can be improved.

In the numerical study of Settar et al. [36], alternative catalytic layers were applied at the walls of plate reformer. Using such catalyst coating arrangement, the methane conversion was significantly improved. CH_4 conversion rate was superior by 46% to that obtained by conventional steam reformer studied experimentally by Lin et al. [37] for same configuration and equivalent catalyst weight. However, the effect of the propounded catalyst arrangement on the plate reformer efficiency has not been fully explored. To further improve the steam reformer efficiency, Settar et al. [36,38] used an inert metal foam matrix. 5% of supplementary CH_4 amount was converted. Diverse intensification ways were used in the literature to improve the conversion process by SMR. Among them, the optimization of catalyst deposition represents an effective mean. De Jong et al. [39] attempted to analyze numerically design of six configurations of reformers from the thermal and kinetics perspectives. 11.2% of hydrogen production enhancement is achieved by managing the heat supply. Vigneault and Grace [40] made experiment measurement and numerical calculations on multichannel reactor designed to produce hydrogen. The reformer designed is composed of five micro reformers and coupled to micro combustors to provide heat. The CH_4 conversion rate attained is 87%. In the same underlying ideas, the work herein aims at comparing different catalyst coating arrangement within a parallel-plate Wall-Coated Reformer (WCR) and investigates their effect on the methane conversion and hydrogen yield. Six catalyst patterning within WCR at industrial operating conditions are compared while keeping the same catalyst weight. Comparing to the studies cited above, the conversion rate enhancement reached by the present work, which is about 28.71%, is significant when the gap among discrete layers is high until certain length.

2. Kinetic model

The kinetic model involved in the formation of H_2 by MSR reaction allows the evaluation of the gas mixture composition at a given point of the reactor. The reaction mechanism of MSR is complex. Eleven