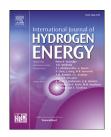
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Comparison of conventional and spherical reactor for the industrial auto-thermal reforming of methane to maximize synthesis gas and minimize CO₂

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ABSTRACT

Auto-thermal reforming (ATR), a combination of exothermic partial oxidation and endothermic steam reforming of methane, is an important process to produce syngas for petrochemical industries. In a commercial ATR unit, tubular fixed bed reactors are typically used. Pressure drop across the tube, high manufacturing costs, and low production capacity are some disadvantages of these reactors. The main propose of this study is to offer an optimized radial flow, spherical packed bed reactor as a promising alternative for overcoming the drawbacks of conventional tubular reactors. In the current research, a one dimensional pseudo-homogeneous model based on mass, energy, and momentum balances is applied to simulate the performance of packed-bed reactors for the production of syngas in both tubular and spherical reactors. In the optimization section, the proposed work explores optimal values of various decision variables that simultaneously maximize outlet molar flow rate of H₂, CO and minimize molar flow rate of CO₂ from novel spherical reactor. The multi-objective model is transformed to a single objective optimization problem by weighted sum method and the single optimum point is found by using genetic algorithm. The optimization results show that the pressure drop in the spherical reactor is negligible in comparison to that of the conventional tubular reactor. Therefore, it is inferred that the spherical reactor can operate with much higher feed flow rate, more catalyst loading, and smaller catalyst particles.

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Introduction

Due to the high demand of synthesis gas (a mixture of H_2 and CO), various catalytic reforming processes attract increasing attention during recent years. Synthesis gas is applied primarily for different industrial products like methanol, Fischer-Tropsch (FT) [1], ammonia/urea or as a reducing agent in the steel industry [2].

Currently, steam reforming, catalytic and non-catalytic partial oxidation, and auto-thermal reforming of natural gas are three major thermochemical reforming methods which are available to produce synthesis gas. Steam reforming is been applied in various industries for synthesis gas production [3]. However, the significant external energy requirement (due to the strong endothermic reforming reactions) and the high initial investment costs (steam reforming section of each industry is responsible for 50-75% of the capital costs) can be referred to as the main drawbacks of this technology [4]. On the other hand, catalytic partial oxidation is generally characterized by very high reaction rates that do not require external energy supplies. Drawbacks of this technology are the high temperatures attained and, especially, the possible hot spots [5] both of which pose high demand to the catalyst and reactor materials and reduce its lifetime. It is worth to mention that, thermal or non-Catalytic Partial oxidation is an exothermic reaction and is usually performed without the use of a catalyst or an external heat source, thus it does not have catalytic restrictions of catalytic partial oxidation in high temperatures. The low hydrogen yield and the production of soot are, however, the major drawbacks of this process [6].

To circumvent these problems, auto-thermal reforming is proposed as a solution in which the thermal effects of steam reforming and partial oxidation reactions are mixed by feeding the fuel, water, and air together into the reactor. The two processes of steam reforming and partial oxidation occur simultaneously in the presence of catalyst in the reactor. The thermal energy produced by partial oxidation is absorbed by the endothermic steam reforming reactions; hence, the overall process is thermally neutral [7,8].

Several research efforts have been made to investigate the catalytic auto-thermal reforming and enhance the production of the desired products [9]. Dias et al. studied the effect of introducing small amount of platinum, palladium and iridium (<0.3% by weight) into Ni/ γ -Al₂O₃ catalysts for the autothermal reforming of CH4. Experimental results of this study indicate that these noble metals increase methane conversion during auto-thermal reforming [10]. Halabi et al. have modeled with a 1-D heterogeneous model the auto-thermal reforming of methane in a fixed bed reformer. They have investigated the process performance under dynamic and steady state conditions with respect to key operational parameters such as: feed gas temperature, oxygen/carbon and steam/carbon ratios, gas hourly space velocity and feed contaminations [11]. Simeone et al. have investigated impact of water addition and stoichiometry variations on temperature profiles in an auto-thermal CH4 reforming fixed bed reactor with Ni catalyst. Furthermore, based on CH4 conversion, H2 production, catalyst deactivation and coke formation a criterion to define favorable reactor operating conditions was

proposed [12]. Zahedi et al. presented a one-dimensional model for heterogeneous auto-thermal reforming of methane over Ni/Mg-Al₂O₄ in a fixed bed reactor. In this study, effects of the suitable feed temperature and pressure, and also feed composition have been investigated [13]. Behroozsarand et al. have investigated multi-objective optimization of industrial auto-thermal reformer for synthesis gas production by applying non-sorting genetic algorithm II. In this study, an optimization method was employed for maximizing of CH₄ conversion and CO selectivity and minimizing of CO₂ makeup flow [14]. Scognamiglio et al. have developed a 1-D heterogeneous model of auto-thermal reforming fixed bed reactor with Rh catalyst and then validated on the basis of experimental results. They have finally concluded that their proposed model was accurate in all the investigated operating conditions and that can be safely applied to design autothermal reforming processes with Rh catalyst [15].

All of the previous investigations were carried out for tubular packed bed reactors. This type of reactor causes high pressure losses. For this type of reactors in process plants, a remarkable amount of energy is mainly dedicated to minimize the pressure drop [16]. Particularly, in gas-phase reactions, the concentrations of reactants are directly affected by changes in total pressure. Indeed, the reaction rates and the degree of conversions are strongly reduced due to the pressure drop. Therefore, development of a number of alternatives for tubular packed bed reactors seems to be necessary [17].

An interesting practical idea for industrial units is the use of spherical reactors. Spherical reactors could effectively overcome the pressure drop problems [18]. Balakotaiah et al. have investigated the effect of flow direction on conversion in isothermal radial flow fixed-bed reactors. Their research showed that the outward flow direction is the preferred one for any reaction with a convex rate expression and a positive change in the number of moles; on the other hand, the inward flow direction is a better choice for any reaction with a concave rate expression and a negative change in the number of moles [19]. Viecco et al. have developed spherical reverse flow reactors. In their suggestion the gas feed enters throughout one hemisphere and leaves through the other one. It was determined that for reversible reactions the spherical reactors present a sufficient increase in conversion due to the lower temperature achieved at the center of the reactor. Furthermore, it was concluded that in this configuration the hot zone could not be occurred [20]. Rahimpour et al. have presented a novel radial-flow spherical bed reactor for methanol synthesis process. The results indicate that for this case study, the two-stage spherical reactor with similar conventional reactor specifications and operating conditions performs better than other alternatives such as single-stage spherical, three-stage spherical and conventional tubular reactors [21]. Farsi et al. have developed a mathematical modeling and optimization procedure for spherical reactor configurations to produce dimethyl ether (DME) from methanol dehydration. Their dynamic model successfully predicted the production rate enhancement for DME in industrial processes, when a series of spherical reactors are applied and thus solving some drawbacks of conventional reactors such as pressure drop [22].

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