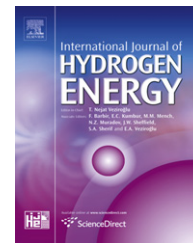


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A novel integrated thermally coupled configuration for methane-steam reforming and hydrogenation of nitrobenzene to aniline

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ABSTRACT

In this work, a novel thermally coupled reactor containing the steam reforming process in the endothermic side and the hydrogenation of nitrobenzene to aniline in the exothermic side has been investigated. In this novel configuration, the conventional steam reforming process has been substituted by the recuperative coupled reactors which contain the steam reforming reactions in the tube side, and the hydrogenation reaction in the shell side. The co-current mode is investigated and the simulation results are compared with corresponding predictions for an industrial fixed-bed steam reformer reactor operated at the same feed conditions. The results show that although synthesis gas productivity is the same as conventional steam reformer reactor, but aniline is also produced as an additional valuable product. Also it does not need to burn at the furnace of steam reformer. The performance of the reactor is numerically investigated for different inlet temperature and molar flow rate of exothermic side. The reactor performance is analyzed based on methane conversion, hydrogen yield and nitrobenzene conversion. The results show that exothermic feed temperature of 1270 K can produce synthesis gas with 26% methane conversion (the same as conventional) and nitrobenzene conversion in the outlet of the reactor is improved to 100%. This new configuration eliminates huge fired furnace with high energy consumption in steam reforming process.

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1. Introduction

Steam reforming of methane or natural gas (SRM) is still the predominant method for producing the hydrogen rich synthesis gas (hydrogen and carbon monoxide) [1–9]. In fact, about 50% of hydrogen demand is satisfied by means of methane-steam reforming [10]. The recent increases in the demand for hydrogen that utilized by many processes such as oil refining, methanol, metallurgy, ammonia, aniline, space transportation, etc., have imposed a strong economic incentive to improve the hydrogen production technology.

The conventional steam reformers (CSRs) have huge fired furnace with high energy consumption and the Ni-based catalyst is packed in vertical tubes. The heat of combustion reaction from burners of furnace is transferred to endothermic steam reforming reaction to produce synthesis gas. Considering the disadvantages of the methane combustion reaction at the furnaces of steam reformers as well as the high rate of energy consumption in the conventional steam reforming process, utilizing the recuperative coupled reactor is suggested in order to solve these issues. Besides, applying this novel configuration may decrease the operational and

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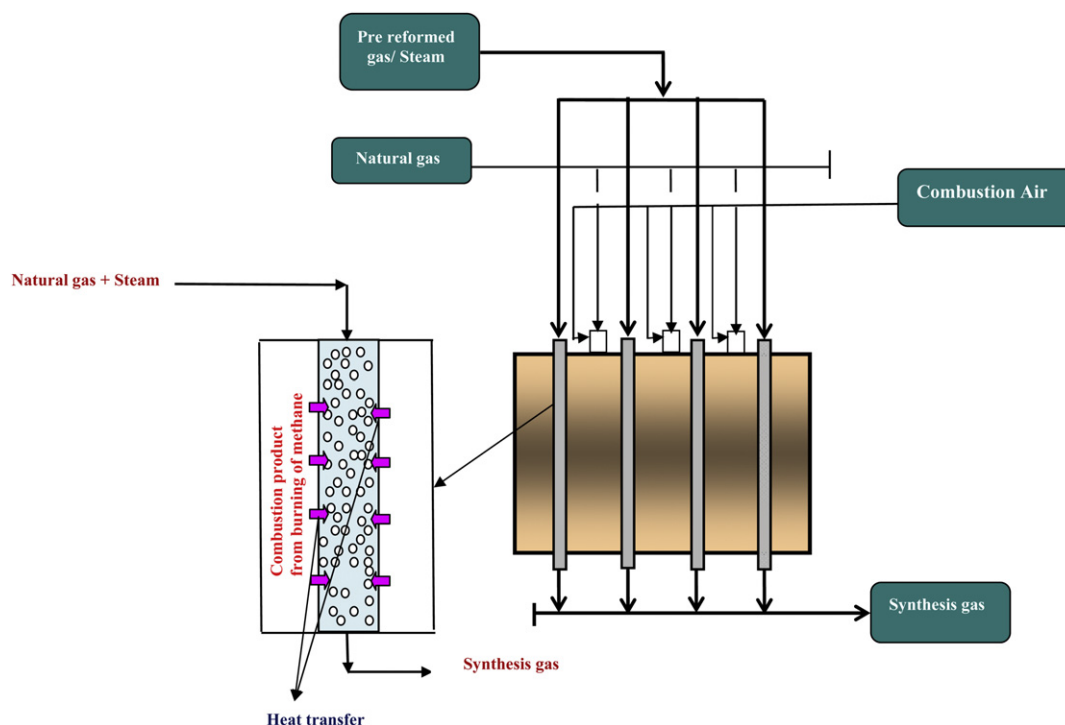


Fig. 1 – Schematic diagram of conventional steam reformer (CSR) reactor configuration [25].

Table 1 – Design specifications and input data of conventional steam reformer (CSR) [25].

Parameter	Value
Tube side	
Feed composition (mol%)	
CO ₂	1.72
CO	0.02
H ₂	5.89
CH ₄	32.59
N ₂	1.52
H ₂ O	58.26
Inlet temperature (°C)	520
Inlet pressure (bar a)	40
Total feed gas flow (kmol/h)	9129.6
Number of tubes (in 4 rows)	184
Inside diameter (mm)	125
Heated length (m)	12
Catalyst volume filled in (total) (m ³)	27.8
Design pressure (bar g)	41
Design temp. (°C)	790
Catalyst shape	10-HOLE rings
Particle size (mm)	19 × 16
Void fraction (–)	0.4
Heat load on tube (100% design case) (kcal/m ² h) (based on tube ID)	68,730
Reformer duty (100% design case) [GJ/h] net	248.2
Shell side	
Combustion air	
Temperature (°C)	330
Pressure (bar a)	1
Flow rate (sm ³ /h)	114,313
Feed gas (fuel)	
Temperature (°C)	34
Pressure (bar a)	3
Flow rate (sm ³ /h)	29,608

capital costs [11]. Also the high thermal energy of the hydrogenation process can be controlled and treated.

Generally, the coupled reactors classify into three main categories: direct coupling, regenerative coupling, and recuperative coupling. Recently, recuperative coupling has gained the interest of many researchers. Itoh and Wu [12] modified the adiabatic coupled reactors by modeling a palladium membrane reactor. Elnashaie et al. [13], and also Moustafa and Elnashaie [14] developed a heterogeneous model for ethyl benzene dehydrogenation process in a membrane catalytic reactor. The dehydrogenation of ethyl benzene occurs in the endothermic side, while the produced hydrogen permeates through the hydrogen perm-selective membrane and provides the exothermic hydrogenation reaction with the necessary reactant. For this case, the mathematical model is developed and the effects of co-current and counter-current flow patterns are investigated. Ramaswamy et al. [15] applied a one-dimensional plug flow model to discuss different behaviors of coupled first order reactions in directly coupled adiabatic reactors. Amin and Yaw [16] analyzed thermodynamic equilibrium of combined carbon dioxide reforming with partial oxidation of methane to syngas. Patel and Sunol [17] simulate a thermally coupled membrane reactor that is composed of three channels for methane-steam reforming. Khademi et al. [18–20] considered methanol synthesis thermally coupled with dehydrogenation of cyclohexane, to prepare pure hydrogen, in a co-current reactor. Ventura and Azevedo [21] developed a numerical model for natural gas steam reforming and coupling with a furnace.

The hydrogenation of nitrobenzene to aniline is an important process in industry which is highly exothermic [22]. The valuable product of this reaction is produced in large scale. Therefore, any improvement in the production rate of aniline attracts attention in industry. Aniline has been applied in methylene diphenyl diisocyanate (MDI) synthesis and also rubber processing. By using nitrobenzene hydrogenation reaction as the heat source in the exothermic side of the

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