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Optimal design of a radial-flow membrane reactor as a novel configuration for continuous catalytic regenerative naphtha reforming process considering a detailed kinetic model

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ABSTRACT

The significance of the catalytic naphtha reforming process in the petroleum refining and petrochemical industry generates continuous evolution of the technology. These improvements would be observed in presenting more efficient reactor setups in order to improve production yield and operating conditions, as well as elucidating better kinetic and deactivation models with higher predicting ability. Both of these items have been considered in this work. An optimized radial-flow moving bed membrane reactor has been proposed as a novel configuration for naphtha reforming process. Optimization has been carried out by differential evolution (DE) method considering 40 decision variables. A detailed kinetic model has also been presented. The proposed kinetic model consists of 32 lumped pseudo-components and 84 reactions. Deactivation rate of catalyst has also been taken into account by considering coke deposition on both acidic and metallic sites. Plant data have been used to validate the modeling results. In order to assess the performance of the proposed configuration, the obtained modeling results have been compared with those of conventional configuration, which shows the superiority of the presented one.
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1. Introduction

Worldwide growing environmental concerns on one hand and increasing demand to gasoline on the other hand have motivated researchers to find new approaches for increasing octane number of gasoline as well as improving the yield of existing gasoline production processes. In this regard, extensive investigations have been accomplished to improve the performance of catalytic naphtha reforming process as one of the most important processes in refining industries. This process has a main role in petroleum refining to obtain high octane number gasoline and in the petrochemical industry to produce aromatics (mostly benzene, toluene, and xylenes). Hydrogen, as a valuable component, and liquefied petroleum gas are also produced in both cases.

1.1. *Continuous catalyst regeneration reformer (CCR)*

Catalytic naphtha reforming units could be categorized according to the catalyst regeneration procedure into three main

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classes, including semi-regenerative catalytic reformer (SRR), cyclic catalytic reformer, and continuous catalyst regeneration reformer (CCR).

SRR is the most commonly used type of the catalytic reforming units [1]. In this process, reactors operate continuously and catalyst activity decreases mainly due to the coke deposition. At the end of the cycle, which occurs approximately once each 6–24 months, all of the catalysts are regenerated in situ during routine catalyst regeneration shutdowns. To maximize the length of time (cycle) between regenerations, SRR units are operated at high pressures, about 1.380–2.070 MPa.

In the cyclic catalytic reformer unit, an extra spare or swing reactor exists and each reactor can be removed from the process flow, regenerated, and then put back into service without shutting down the unit and losing production. It should be mentioned that application of the cyclic catalytic reformer unit for naphtha reforming process is not very common. The operational pressure of cyclic reforming reactors is approximately 1.5 MPa.

The most modern type of the catalytic reformers is CCR, which represents a step change in reforming technology compared to semi-regenerative and cyclic processes. In this unit, the catalyst regenerates continuously in a special regenerator and then is sent to the operating reactors. Due to the continuous regeneration of the catalyst, this process is suited for higher severity operations relative to semi-regenerative fixed-bed reformers [2,3]. The CCR process has enabled ultra-low pressure operations at about 0.345 MPa. The advantages of CCR process against traditional methods are [3,4]:

- Production of higher octane reformate even working with a low feed quality
- Long time working of the process for hydrogen demand
- Using catalyst with less stability but higher selectivity and yield
- Lower required recycle ratio and the lower operational pressure with high yield of hydrogen

1.2. Kinetic model

It is revealed that naphtha is a complex mixture of more than 300 components, mainly hydrocarbons. Considering all of these components and their corresponding reactions in a kinetic model is a complex problem. In order to take into account the main components and reactions, as well as avoiding the complexity of considering large number of components and reactions, “lumped” models have been presented. In this kind of model, the large numbers of chemical components are classified to smaller set of kinetic lumps. The first significant attempt to model a reforming system by this approach has been made by Smith in 1959 [5]. His model consists of three basic components including paraffins, naphthenes, and aromatics (PNA), which undergo four reactions. Krane et al. [6] investigated the presence of various hydrocarbons in the whole naphtha and presented a reaction network of twenty different components, containing hydrocarbons from C_6 to C_{10}. Padmavathi et al. [7] considered 26 lumps of hydrocarbons undergoing 48 reactions. Evolution in the considered number of lumped components and reactions in catalytic naphtha reforming kinetic is presented in Table 1.

1.3. Membrane reactor

Membrane reactors are widely used either as “extractor” in which certain products remove from reaction zone or as “distributer” in which a certain reactant distributes into the reactor [18]. This effective configuration has various advantages such as increasing reaction rate, reducing by product formation, requiring lower energy, and operating under relatively safe conditions. Several researchers have investigated the application of membrane technology in chemical reaction processes. Dittmeyer et al. [19] studied pd-membrane reactors and discussed two different membrane reactor concepts which both rely on supported palladium, on the one hand as a permselective membrane material, and on the other hand as base component of a membrane-type hydrogenation catalyst. Pereira et al. [20] studied ethyl lactate synthesis in a pervaporation membrane reactor. Parvasi et al. [21] analyzed Pd–Ag membrane methanol loop reactors and applied differential evolution (DE) method as a powerful method for optimization of the process.

1.4. Objective

In this work the benefits of the membrane reactor and the continuous catalyst regeneration mode of operation has been combined to improve the performance of the catalytic naphtha reforming unit. The flow pattern in the reactor is considered to be radial-flow due to its advantageous over axial-flow pattern such as lower pressure drop. A reaction network containing 32 pseudo-component and 84 reactions has also been presented. Then for modeling the conventional and membrane reactor, mass and energy balances, thermodynamic and kinetic equations, deactivation model, and auxiliary relations have been applied. The obtained conventional reactor model has been validated by comparing with

Table 1 – Evolution in the considered number of lumped components and number of reactions in catalytic naphtha reforming kinetic.

Number of lumped component	Number of reactions	Year	Investigator	Reference
3	4	1959	Smith	[5]
31	78	1980	Jenkins et al.	[8]
28	81	1987	Froment	[9]
35	36	1997	Taskar et al.	[10]
26	48	1997	Padmavathi et al.	[7]
24	71	2000	Ancheyta-Juarez et al.	[11]
17	17	2004	Hu et al.	[12]
21	51	2004	Hu et al.	[13]
20	31	2006	Weifeng et al.	[14]
18	17	2006	Weifeng et al.	[15]
27	52	2010	Hongjun et al.	[16]
38	86	2012	Wang et al.	[17]

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