I&EC research
Industrial & Engineering Chemistry Research

Review

Cite This: Ind. Eng. Chem. Res. 2019, 58, 9179−9198

pubs.acs.org/IECR

Moving Bed Reactors: Challenges and Progress of Experimental and Theoretical Studies in a Century of Research

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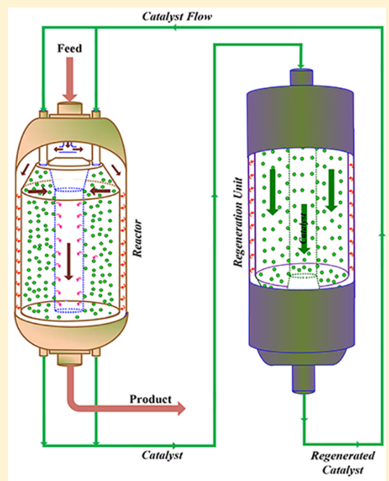
S *Supporting Information*

ABSTRACT: Moving bed reactors (MBRs) have been proposed as a sign of significant progress in the reaction engineering area for performance improving and energy saving. Since their advent in 1890, the MBRs have attracted a wide acceptance in different industries, while they were first developed for the drying industries. The progress that this technology has made during its evolution led to the introduction of these reactors as a pioneer strategy in other industries including petroleum, petrochemical, pyrolysis, and biomass industries. In the traditional reaction systems, the process performance decreases during the operational conditions, while MBRs have obviated this drawback by having an all-around permanent acceptable efficiency. In this context, the present work provides an overview on the evolution of MBRs by investigating the main experimental and theoretical studies. In this way, the experimental studies have typically taken into account operational conditions and production rates of different products, while in the theoretical research, modeling, and simulation of conventional processes, the evaluation of novel configurations and the optimization techniques have been investigated. In the end, some suggestions are proposed to modify the traditional MBRs as helpful ideas for further studies.



1. INTRODUCTION

Chemical Engineering is responsible for the design and operation of processes, which produce a wide range of products in different areas. Accordingly, the entirety of the process from the raw materials until the final products should be considered and evaluated, simultaneously, while the chemical reactors play a key role in all chemical plants.[1,2] In the reactors, multiple reactions occur, and usually different catalysts are required to achieve the desired rates and selectivities. These factors that determine the production rate have a significant impact on the economics of the chemical plants, specifying at the same time the amount of waste products, which can have a major effect on the environment.[1] Thus, the design, scale-up, and performance improvement of chemical reactors have been extensively investigated by many researchers in the past decades.

Table 1. Main Factors in Catalyst Deactivation in Different Chemical Processes

parameters	type	ref
metal impurities	chemical	in 2018, Gavrilovic et al.[35]
thermal instability	thermal	in 2014, Paasikallio et al.; in 2018, Rodriguez et al.[36,37]
carbon deposit	mechanical	in 2018, Rodriguez et al.; in 2014, Argyle et al.; in 2015, Rytter et al.[37−39]
fouling	mechanical	in 2015, Argyle et al.; in 2015, Lang et al.[34,40]
attrition	mechanical	in 2015, Argyle et al.; in 2016, Satyanarayana et al.[34,41]
crushing	mechanical	in 2015, Argyle et al.; in 2016, Satyanarayana et al.[34,41]
loss of metallic-basis	mechanical	in 2018, Braz et al.; in 2018, Megias-Sayago et al.[42,43]
poisoning	chemical	in 2015, Argyle et al.; in 2014, Argyle et al.; in 2015, Lang et al.[34,38,40]

1.1. What Is a Moving Bed Reactor? Moving bed technology (MBT) has been introduced for more than a half century[3−8] and was developed by investigating the steady-state processes[9,3] and modeling analysis through mathematical models.[10−12] The first MBT patent was published in 1890 by

Received: February 28, 2019
Revised: May 5, 2019
Accepted: May 9, 2019
Published: May 9, 2019

DOI: 10.1021/acs.iecr.9b01136
Ind. Eng. Chem. Res. 2019, 58, 9179−9198

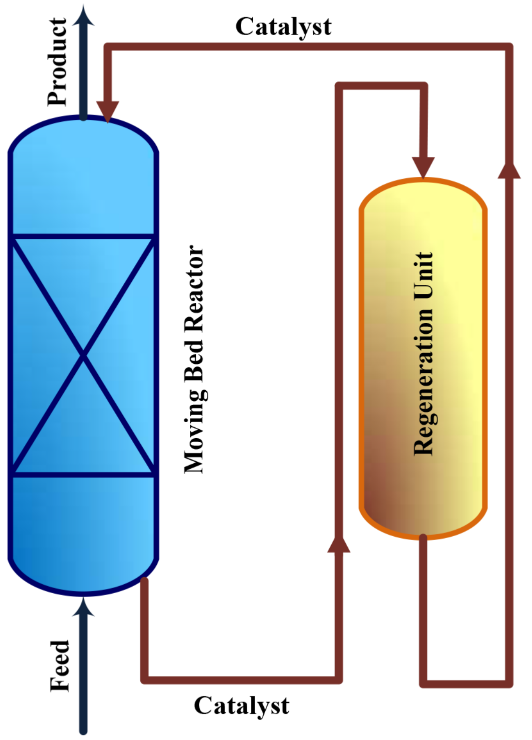


Figure 1. Block diagram of a moving bed reactor connected to a regeneration unit.

Andrews.[13] There was a concern about coating and fouling the gas purifying materials for products or byproducts through the separation process. Consequently, the purifier materials became inert, and no separation occurred after a while. This problem was solved via a separation process by proposing moving bed reactors (MBRs).[13] An MBR is a catalytic reactor in which a layer of catalysts in the form of granules is moved through the reaction bed and regenerated in the regeneration unit, continuously. Since the advent of MBT, it has attracted significant attention in different areas, including the separation of chemical compounds,[14,15] the methane combustion application,[16] the recuperation of petrochemical processes, drying of grains and seeds, and also the organic matter elimination from the effluents,[14] etc. Finally, some new potential applications in bioprocesses have been studied by MBT,[17] especially in biofilm processes for municipal and industrial wastewater treatment.[18−20] On the other hand, the kinetic theory of the granular flows in MBRs has been considered in some engineering systems like the steel production industries from iron ore, some advanced manufacturing products,[21,22] and finally in biomass processes as an ecofriendly energy generation technology.[3] The MBRs have main benefits including low energy consumption and pressure drop, increasing the net profit of the plant by reducing the maintenance costs,[23] improving the process performance,[14] and also enhancing the contact surface of gas−solid systems.[17]

Typically, chemical reactions of different processes[24] take place in the catalytic beds,[25] which can be classified in three main categories[26] including fixed bed, fluidized bed, and moving bed.[27−29] In the petrochemical industries, fixed bed reactors are more common for light petroleum cuts with less than 250 ppm metal impurities treating[29] or hydration and dehydration reactions,[30,31] while the MBR is considered for treating the heavy or light petroleum cuts with more than 250 ppm metal impurities. On the other hand, the rotating bed reactors immobilize the solid phase (catalysts) in a porous chamber, which spins to maximize the axial mixing and convective transport.[32] One of the main drawbacks of rotating reactors is the influence of the shear and/or the bubble formation in the chamber.[33] Through the chemical reactions, on the basis of the scope of the process, different types of materials are produced, some of which may have adverse effects on the catalyst performance. The main agents of the catalyst deactivation[34] are reported in Table 1.

In this way, the deactivated catalysts are required to be regenerated in the regeneration unit to operate as fresh catalysts in the chemical reactors.[44] Specifically, the moving bed technology (MBT) is considered in the chemical and petrochemical industries as a continuous catalyst regeneration (CCR) unit[44−46] to remove the coke deposited on the catalysts surfaces and regenerate them. In the CCR technology, the deactivated catalysts are withdrawn at the bottom of the reactor and introduced to the regeneration unit, which is connected to the top of the reactor. In the regeneration unit, the catalysts are regenerated by moving to the bottom of the unit in the gaseous media. Thereafter, they are introduced to the process as fresh catalysts. This procedure is continued, consistently, with a low pressure drop and energy consumption.[45] Thus, the MBRs have the advantages of higher equilibrium yield[47] and performance[48] as compared to other processes.

In addition to the chemical and petrochemical industries, MBT has attracted much attention in other industries including bioethanol production,[49] hydrogen production,[50] drying,[51] gas separation,[52] and membrane bioreactors,[53] etc. It is worth mentioning that the application of the MBT is at the developing step in these industries, while in the chemical and petrochemical processes it is a mature, reliable, and efficient strategy to control and manage the coke formation and pressure drop.[54] On the other hand, CCR technology is applied in many processes such as biomass[36] and Fischer−Tropsch,[39] as a vital factor in having a high production rate at constant operational conditions.[55] In this way, to maintain the yield and efficiency of the process as high as possible, which is directly influenced by the catalyst activity, it is required to regenerate the deactivated catalysts, continuously.[30] The simple schematic of CCR by using MBT is presented in Figure 1.

It is worth mentioning that, in the literature, there are two types of moving bed reactors including the moving bed biofilm reactor[56−61] and chemical moving bed reactor.[16,62−65] The moving bed biofilm reactor is employed for municipal and industrial wastewater treatment.[60,66] This technology can improve the performance of the process by reducing the size of reactors compared with the conventional ones.[67,68] On the other hand, it should be considered that the simulated moving bed reactor (SMBR) technology[69−73] is a multifunctional reactor in which the chemical and separation reactions take place simultaneously, in a fixed bed reactor for adsorption processes.[74,75]

1.2. Objective.

In this study, the concept of the chemical moving bed reactors has been extensively investigated and reviewed from the advent of this technology until now; this, to the best of our knowledge, is the first review paper in this area. To this goal, all studies are classified in two main classes including (i) experimental studies and (ii) theoretical studies. Then, the accomplished works are defined as subsubjects in these areas.

DOI: 10.1021/acs.iecr.9b01136
Ind. Eng. Chem. Res. 2019, 58, 9179−9198

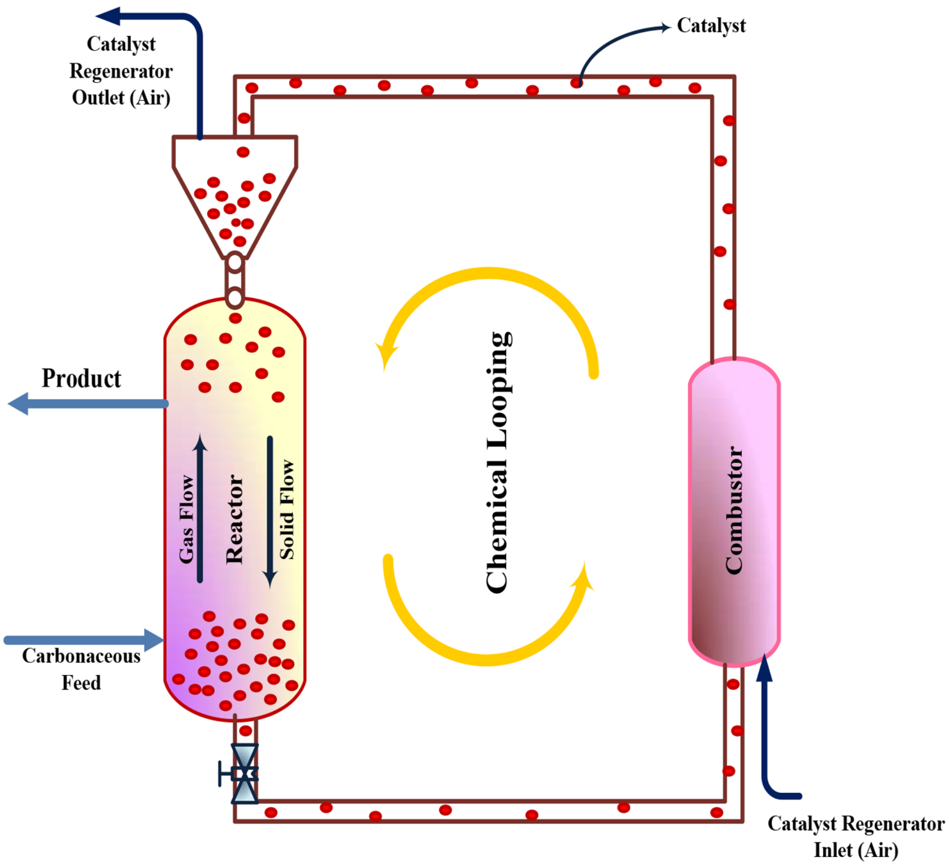


Figure 2. Schematic of chemical looping combustion.

Table 2. Different Types of Chemical Looping Processes

technologies	feed	product	ref
chemical looping gasification	coal	$H_2/CO/H_2S/COS$/liquid product	in 2010, Li et al.; in 2014, Luo et al.[82,83]
chemical looping combustion	gaseous fuel	H_2O/CO_2/heat	in 2012, Adanez et al.; in 2014, Chiu et al.; in 2015, Zeng et al.; in 2013, Hong et al.[84−87]
	solid fuels	H_2O/CO_2/heat	in 2012, Adanez et al.; in 2014, Chiu et al.; in 2015, Zeng et al.; in 2013, Hong et al.; in 2015, Wu et al.[84,88]
chemical looping reforming	hydrocarbons	$H_2/CO/CO_2$/heat	in 2012, Adanez et al.; in 2014, Dou et al.; in 2012, Kang et al.[84,89,90]
syngas chemical looping	methane	H_2O/CO_2/ H_2/heat	in 2013, Tong et al.[91]

2. EXPERIMENTAL STUDIES

2.1. Chemical Looping Combustion. The increasing rate of fossil fuels consumption has contributed to the production of a huge volume of CO_2, which adversely affects the environment and, consequently, the public health.[76] In conventional power plants, there is not too much consideration for CO_2 capture,[77,78] but in recent years, many researchers have focused on developing novel configurations to improve the performance of available processes, based on the carbon capture and storage (CCS) strategy, to modify the energy consumption and reduce the CO_2 emissions, simultaneously.[79] Chemical looping (CL) is a cyclic process in which the CO_2 is separated with minimum consumption of the energy as well as high energy conversion in the process.[76] Figure 2 shows a schematic of the chemical looping process integrated with MBT. Actually, in this process, the main products and carbon dioxide are produced in two separated reactors, which reduces the cost of CO_2 separation and the purity of the obtained products.[80] Furthermore, the integrated moving bed configuration regenerates the deactivated catalysts, continuously.[81] Table 2 illustrates the different types of the chemical looping technologies, the carbonaceous feed, and different products of each chemical looping process.

The employed catalyst in this process is actually a chemical intermediate, which carries the required oxygen for the reactions.[92] The oxygen carrier is reduced in the reactor by the carbonaceous feed to produce the main product and heat; after that, it is oxidized in the combustor. The loop is cycled continuously by using the MBT with a steady-state method.[92] The oxygen carrier should possess some essential features including being low cost and ecofriendly and having a low tendency to agglomeration, high reactivity with the carbonaceous feed and oxygen, the ability to convert the carbonaceous feed to the products, low attrition, as well as a negligible fragmentation tendency.[93]

Recently, scholars have developed the moving bed reactor concept integrated with chemical looping technology on the

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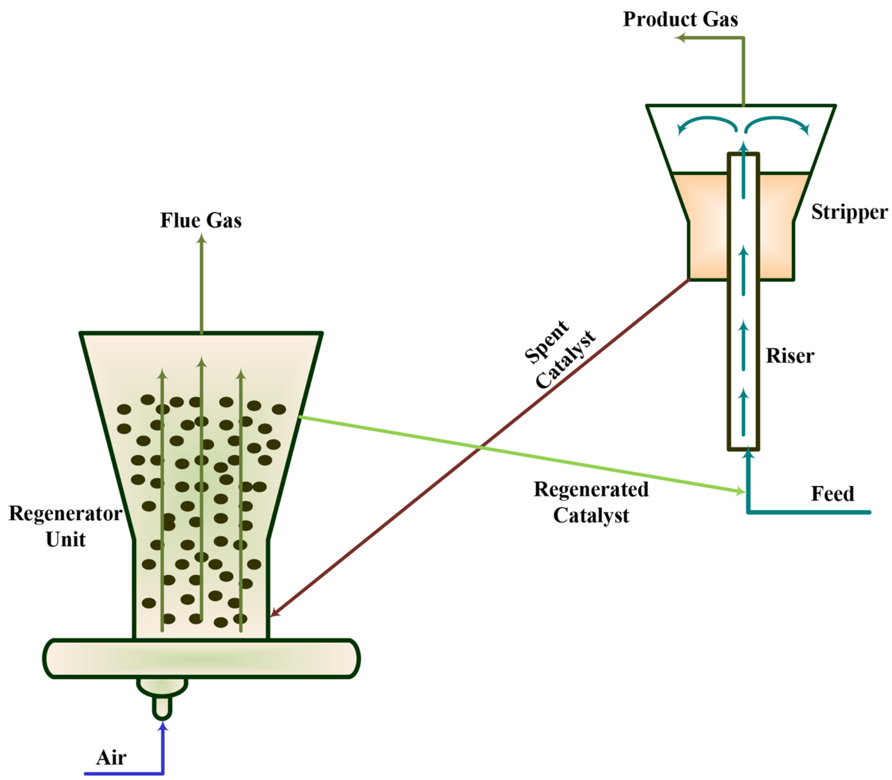

Figure 3. Schematic of the fluid catalytic cracking (FCC) process.

research and pilot scales,[94,95] to produce H_2 from methane[94] and heavy liquid hydrocarbons as carbonaceous feeds.[95] They have also focused on the effective parameters including the type of the oxygen carrier,[94,96] the gas velocity in the moving bed configuration, and the type of the carbonaceous feed,[95] by experimental studies[94,95] as well as mathematical modeling.[97] Furthermore, the study on the chemical looping combustion of the biomass integrated with MBT is ongoing by many researchers.[88,98−100] It can be concluded from these studies that the CL technology integrated with MBT has high energy conversion potential and CO_2 capture ability, which can be a promising concept in different areas.[83,86,89,94,97,101−104]

2.2. Fluid Catalytic Cracking. From around 70 years ago, fluid catalytic cracking (FCC) has been known as one of the key processes to obtain favorable fuels and different required products from the heavier fractions of the crude oil.[105] In this way, the heavy oil, which is not as valuable as lighter cuts (middle distillates, diesel, gasoline, and even light olefins such as propylene), is processed and contributes as a significant commodity in the fuels market.[106] In fact, the FCC process includes a fluidized reactor with a moving bed, in which the applied catalysts are continuously regenerated.[107] The simple schematic of this process is illustrated in Figure 3.

One of the most noticeable experimental works in this area has been devoted to developing more efficient FCC catalysts. From 1960, employing different zeolites has been considered as a basic part of FCC studies,[108] but the potential substitution for zeolites,[109] increasing catalyst performance,[110,111] catalyst modifications,[112−114] and improving regeneration process[115] have been extensively investigated in the past decades. On the other hand, the modification of the operational conditions of the FCC process,[116] pilot-scale experiments,[117] fault-detection of the industrial FCC units,[118] etc. are the other favorable topics in the FCC field, which have been considered so far.

In addition, the combination of the FCC process with chemical looping combustion to reduce the CO_2 emission through the process,[119] the post-treatment of FCC products,[120−123] biofeed synergistic effects on the heavy petrochemical feeds,[124,125] simultaneous removal of catalyst particles from the FCC process,[126] etc. are some of the other main studies, which have been considered to improve the performance and efficiency of this process. More details in this area can be found in the recent review papers, which have been published in Vogt and Weckhuysen,[108] Otterstedt et al.,[127] and Akah.[128]

2.3. Pyrolysis. Pyrolysis is a process which employs the energy to decompose worthless materials to useful products.[80,81,129−132] Recently, the integration of the pyrolysis process with MBR technology has contributed to a more efficient process because of the heat transfer improvement[133,134] as well as the continuous operational condition.[134] To this goal, different heat carriers have been considered as mediums, which enhance the heat transfer rate and the process efficiency.[135] In this way, sands,[135,136] gaseous compounds like nitrogen,[137] etc. have been considered as heating media. Also in some cases, a special MBR has been employed in which the bed is moved mechanically by a belt[138] or a screw-type shaft[134,139] without the heating media.[140,141] Figure 4 shows two common types of MBR configurations in pyrolysis processes, schematically.

Lastly, the recycling of tires and plastics and their conversion into valuable fuels (e.g., fuel gas[135]) have been considered extensively, due to the significant environmental concerns in recent years.[142−144] It is worth mentioning that the economic progress[135] and simplified processes[142] have been developed in the pyrolysis studies, so far. However, more studies are required to be conducted in this area to achieve a lucrative technology.

DOI: 10.1021/acs.iecr.9b01136
Ind. Eng. Chem. Res. 2019, 58, 9179−9198

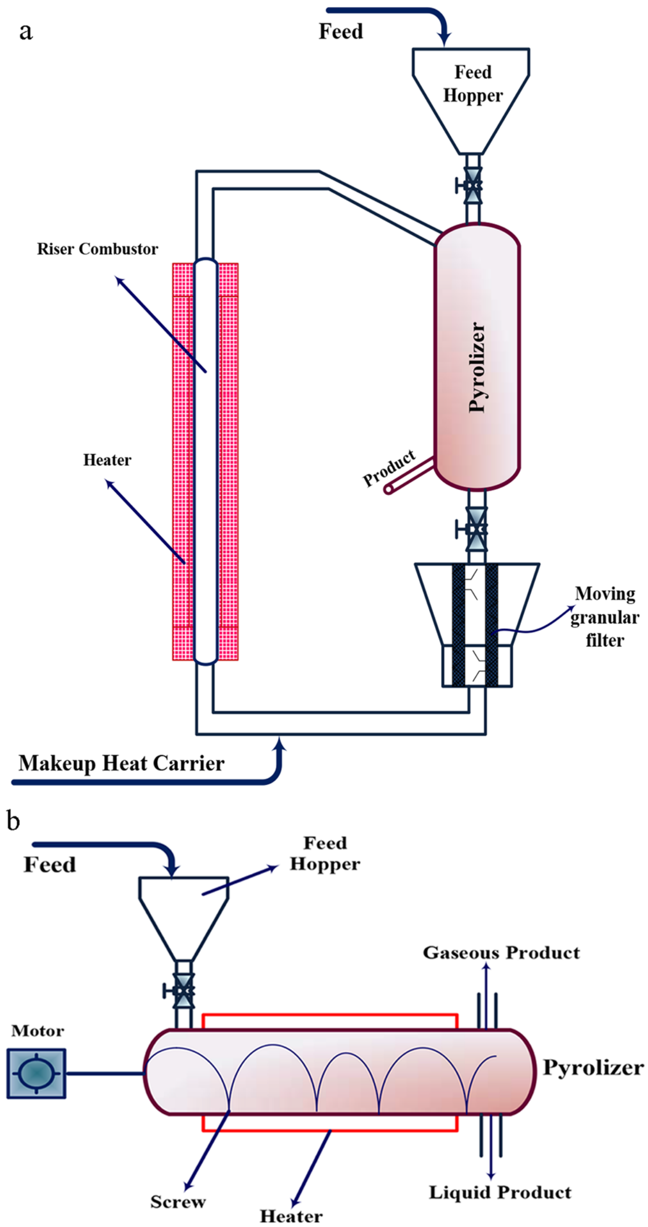

Figure 4. Schematic of the pyrolysis process with a moving bed configuration: (a) with a heating carrier and (b) without a heating carrier.

The pyrolysis of the oil shale has been recently studied in an innovative MBR.[145,146] On the basis of the traditional oil shale pyrolysis system, the heat carrier passes through the solid oil shale to supply the required energy of the pyrolysis process. Hence, the gaseous products are blended with the heat carrier and make a homogeneous gaseous mixture. The separation of the gaseous mixture is a high-cost process and requires a large volume of the heat carrier, which adversely affects the purity of the pyrolysis products. On the other hand, this process is only able to pyrolyze the oil shale particles which have diameters with more than 15 mm[145] and constitute around 15−20% of the total oil shale sample. These drawbacks of the traditional pyrolysis processes have forced researchers to develop an innovative strategy to overcome these limitations. In this way, the moving bed technology with a solid heat carrier (hot shale ash) has emerged due to the greater adaptability for treating the small size of the oil shale, higher oil shale yield, and the gas separation system in a simple way.[145,147]

Upon review, the recent studies on the pyrolysis process integrated with MBT support the idea that many researchers have employed this technology for converting biomass to syngas.[148−152] The integrated pyrolysis technology with the moving bed configuration has eliminated the drying step, which is one of the main steps to obtain useful energy from biomass.[151] The effects of parameters including temperature,[148,150] type of the reactor,[149] the heat carrier type,[151] etc. have been investigated so far, while studies are ongoing in this area. Some of the main research about this process is presented in Table 3.

3. THEORETICAL STUDIES

In this chapter, a comprehensive review on the theoretical studies on the MBRs is presented. In this way, all main modeling studies on MBT have been investigated, while some of these studies are of mathematical modeling or process simulation on the available existing power plants, which have been accomplished on the basis of the industrial data. The other ones are hypothetical research, which have been proposed to improve the performance of the conventional MBRs, and these studies are in the developing step.

3.1. Modeled and Optimized Processes. Mathematical modeling and optimization of MBRs have been considered starting from around 40 years ago.[153,154] Some modeling methods such as computational fluid dynamics (CFD) studies have just been applied to optimize the process.[103,155,156] In the first studies, the composition and the temperature profile through the MBRs as well as the composition and the temperature profile at the effluent were considered as main factors in the MBT.[153,154] They concluded that the analysis of the reactor effluent composition is not sufficient to validate the considered models; thus, the data of the moving bed is also required.[153,157] Gradually, the modeling of the MBRs was developed, and more effective parameters were investigated including cocurrent/countercurrent configurations,[158] the application of nanocatalysts,[12] different types of the feed,[159] radial flow pattern,[160] the catalyst deactivation,[161] the moving bed for the grain drying process,[162] and the CVD coating of the fine particles.[163] The process modeling has been conducted by considering the mass and energy balances,[164] kinetics,[165] the equilibrium,[90] and the physical properties.[90] The MBR configuration for three reactors is schematically demonstrated in Figure 5. It is worth mentioning that the number of reactors is changeable, and based on the type of the process, it can be one or more. Also, the flow patterns in the MBRs can be cocurrent[160,152] or countercurrent,[159] which are demonstrated in Figure 6.

The modeling of MBRs has also been considered in the chemical looping process.[90,97] The methane conversion in a countercurrent moving bed reactor was investigated by Ostace et al.[97] and Kang et al.[90] The mass, energy, and pressure drop balances were applied to model the MBRs by assumptions of the radial[97] and plug[90] flow patterns. On the other hand, the mathematical modeling and the optimization of MBRs integrated with chemical looping technology have been developed through the years. However, this new technology requires deeper studies to be known as a fully developed process.[86,166−168] The modeling of the biomass conversion to the valuable energy in the MBRs has also attracted much attention in recent years.[97,169,170]

DOI: 10.1021/acs.iecr.9b01136
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Table 3. Main Pyrolysis Studies with Moving Bed Technology

heat carrier	kind of reactor	feed	products	ref
sand	screw conveyor (moving bed)	waste plastic	fuel gas	in 2006, Kodera et al.[135]
nitrogen	moving bed	Shenmu bituminous coal	gaseous hydrocarbons and H_2	in 2015, Liang et al.[137]
steam	belt conveyor (cross-flow moving bed)	cellulose	char	in 2006, Yamaguchi et al.[138]
nitrogen	moving bed	waste tire	BTX fraction, short aliphatic chain, and limonene	in 2008, Aylon et al.[134]
nitrogen	moving bed	waste tire	gaseous products and solid residue	in 2010, Aylon et al.[133]
nitrogen	screw conveyor (moving bed)	waste tire	low-molecular-weight products, liquids, or gases	in 2010, Aylon et al.[142]
nitrogen	screw conveyor (moving bed)	inhomogeneous tire chips	gaseous products, pyrolytic oil, and char	in 2017, Rudniak and Machniewski[144]
nitrogen	moving bed	rapeseed stalk	hydrocarbons, H_2, and oil	in 2017, Gao et al.[136]
shale ash	moving bed with internals	oil shale	volatile products	in 2015, Lai et al.; in 2016, Lai et al.[145,146]
nitrogen	screw conveyor (moving bed)	sewage sludge	syngas	in 2018, Zhu et al.[149]
nitrogen	screw conveyor (moving bed)	wet sewage sludge with sawdust	syngas	in 2018, Yang et al.[150]
nitrogen	moving bed	biomass	gaseous products, bio-oil, and biochar	in 2018, Zhang et al.[148]

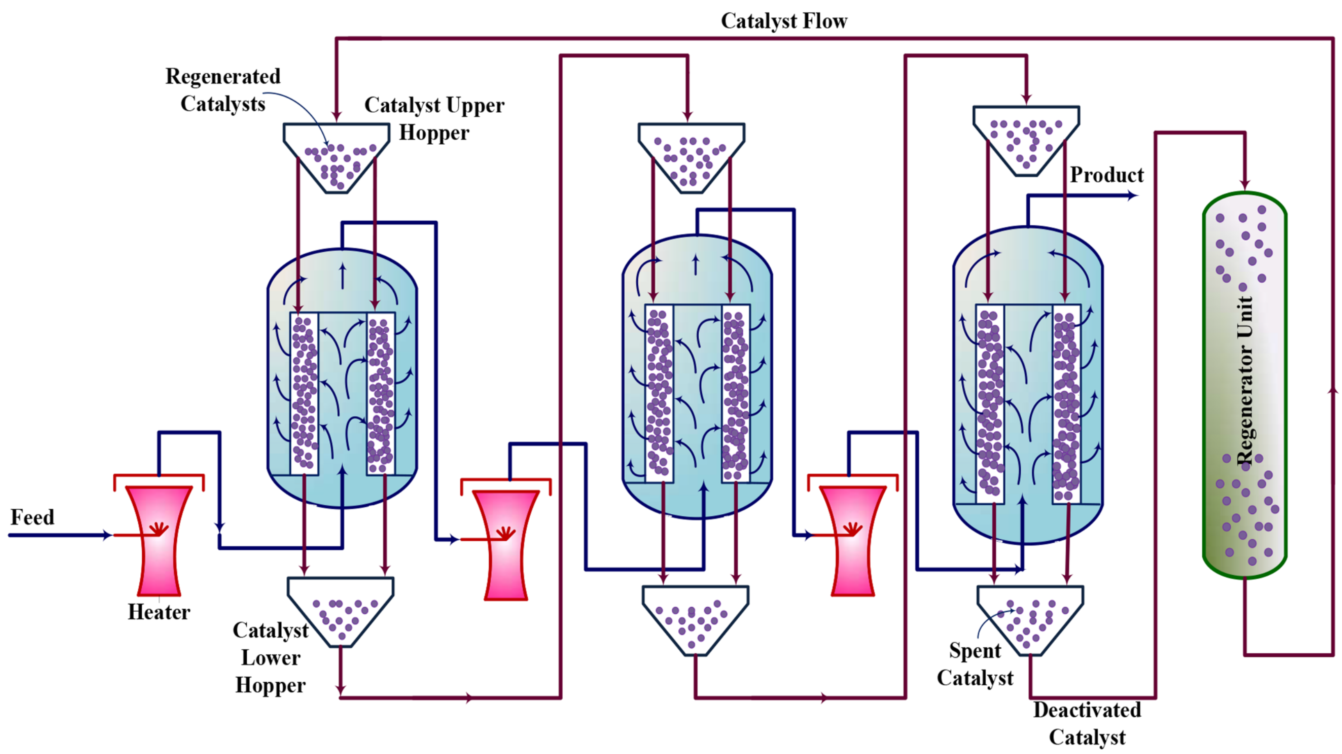

Figure 5. Schematic of a moving bed reactor configuration.

3.2. Gas–Solid Reactors. The main purpose of the gas–solid moving bed reactors is related to some physical or chemical transformations in the moving solids; in the conventional MBRs the moving solid phase operates as a catalyst to accelerate the reaction rate in the gas phase.[127,128] The gas–solid reactors can be categorized in two different regimes: (i) the axial flow regime (cocurrent and countercurrent)[129] and (ii) the cross-flow pattern.[171,174]

The limitations of the gas–solid heat exchanging process convinced researchers to find an efficient strategy to enhance the heat transfer rate in this process.[175] In this way, the gas–solid interaction in the MBRs was proposed to obviate the existing problem through the heat exchanging process.[176] In the axial flow gas–solid moving bed configuration, the advantages of the countercurrent flow regime were applied through the direct reduction of the iron ore to the sponge iron by a reducing gas. In this process, MIDREX technology was propounded[172] (as shown in Figure 7), in which the iron ores come downward while the reducing gas goes upward;[174] meanwhile, the chemical transformation occurs.[177]

Several applications have been introduced for the cross-flow moving bed reactors such as dryers,[162] the moving bed granular filter,[178−181] and catalyst regeneration. Figure 8 shows the drying process for the cross-flow moving bed technology, schematically. After the successful application of the gas–solid moving bed in the MIDREX, researchers tried to employ gas–solid moving bed technology as a chemical reaction bed.[173,176,182]

3.3. Fluid Catalytic Cracking. From the advent of the FCC process, many researchers have considered the theoretical

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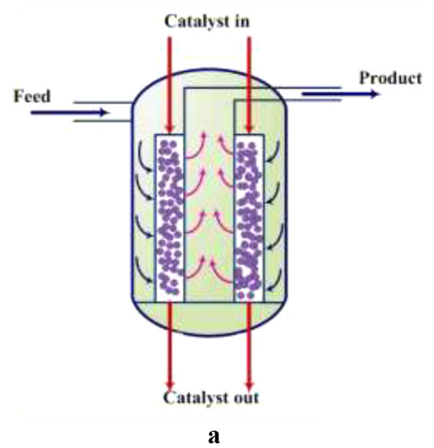

Figure 6. Flow types in the modeling of a moving bed: (a) concurrent and (b) countercurrent.

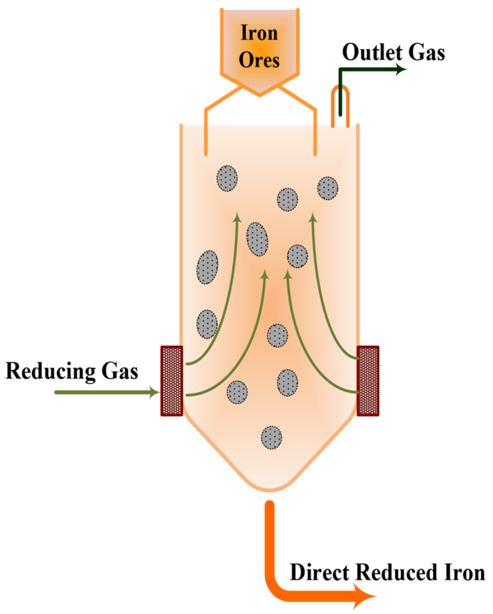

Figure 7. MIDREX technology to reduce iron ores to sponge iron in a moving bed reactor.

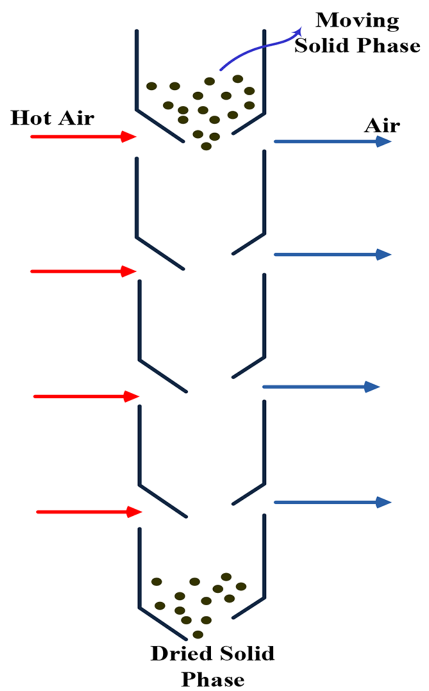

Figure 8. Schematic of a cross-flow moving bed dryer.

studies of this process by focusing on the mathematical modeling, process simulation, and the optimization of the key parameters. In fact, the FCC is a complex process which requires a precise and detailed description of models to cover the interaction of the model inputs and control the model outputs.[183] In this context, various modeling procedures with different assumptions have been implemented. The lumped-kinetic assumption and proposing new methods with different lumped assumptions,[184,185] a higher level of the lumped assumption,[186,187] as well as a comparison of the different models[184] are the most studied subjects in theoretical studies of the FCC process. The evaluation of the nonlinearity in this process and its effect on the model outputs was one of the first considered subjects in this area.[188] Also, the mathematical modeling of the in-operation FCC plant was one of the other primary works.[189] In addition, researchers have studied the hydrodynamics of the FCC process[190,191] (intensifying the catalysts regeneration with baffles in the regenerator,[192] the feed injection patterns and their effects on the FCC process performance,[186] etc.) and the hydrodynamic optimization,[193] which led to the economic improvement in the refineries.

In recent years, the modification of the performance of the available FCC units has also attracted much attention. Thus, the optimal design of the FCC process for increasing the productivity, as one of the most important issues, has been considered.[194] In this way, the combination of different processes[195] and different materials as the modifiers[196,197] through the FCC has been investigated so far.

3.4. Naphtha Processes. The catalytic naphtha reforming is the main industrial process to obtain high-octane gasoline and aromatic hydrocarbons.[198−200] UOP and Axens are two major licenses which represent the commercial reforming processes. According to UOP and Axens technologies, the feed should be hydrotreated to decrease the required catalysts for the reforming process.[201] UOP technology,[45] which employs stacked reactors, was introduced for the first time in 1971 (illustrated in Figure 9), while on the basis of Axens technology the MBRs are designed in a side by side configuration (as shown in Figure 7).

Catalytic naphtha reforming is a basic technology to produce high-octane reformate for gasoline blending.[202] In this process,

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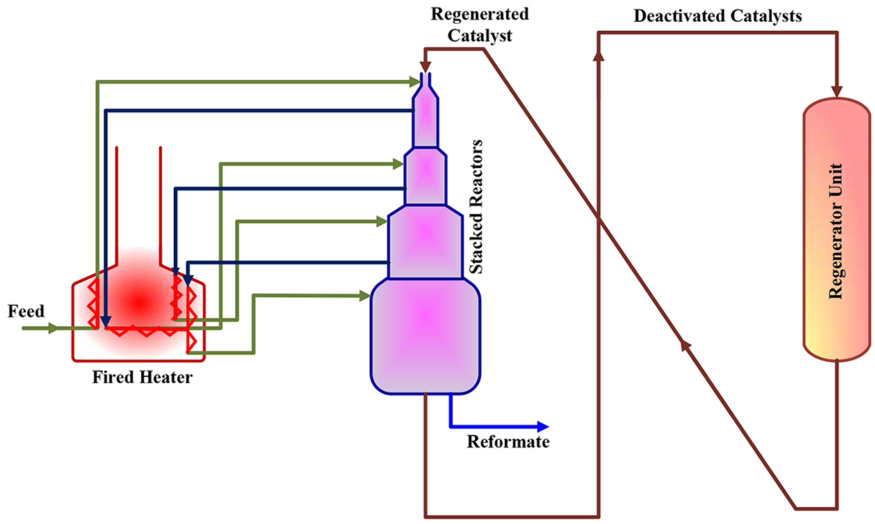

Figure 9. UOP stacked reactor for the naphtha reforming process.

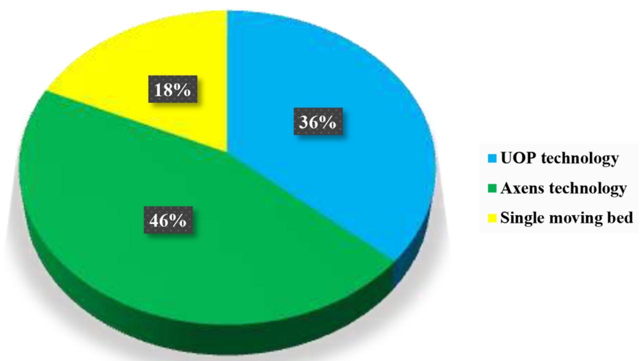

Figure 10. Reactor technology as studied by various researchers.

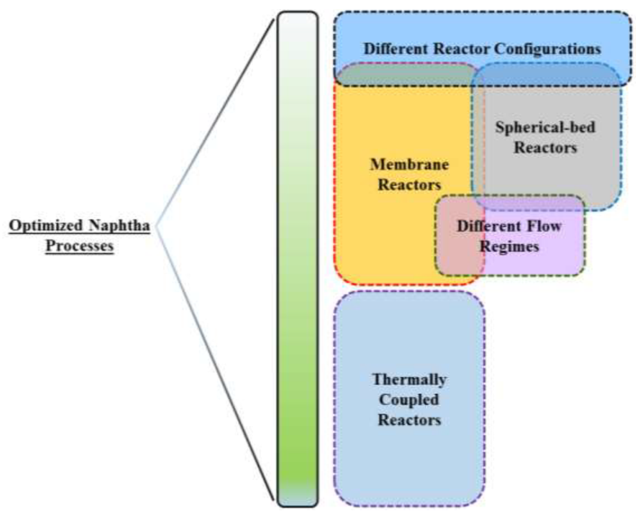

Figure 11. Different methods to improve the performance of the naphtha reforming process.

many reactions take place such as dehydrogenation,[203] dehydroisomerization,[203] aromatization,[204] isomerization,[203] dehydrocyclization,[203,205] hydroisomerization,[203,205] hydrocracking,[203,206] and dealkylation.[204] These reactions, which

Table 4. Exothermic Reactions in Different Studies

entry	exothermic reaction	ref
1	hydrogenation of nitrobenzene to aniline	in 2013, Iranshahi et al.[251]
2	hydrogenation of nitrobenzene to aniline	in 2013, Jafari et al.[252]
3	oxidation of sulfur dioxide	in 2014, Karimi et al.[253]
4	hydrodealkylation of toluene	in 2017, Iranshahi et al.[254]
5	hydrogenation of nitrobenzene to aniline	in 2016, Karimi et al.[245]
6	oxidation of sulfur dioxide to SO_3	in 2017, Saeedi and Iranshahi[255]
7	oxidation of sulfur dioxide to SO_3	in 2018, Iranshahi et al.[256]

occur simultaneously in the reaction bed, are influenced by different parameters.

Naphtha reforming, based on the regeneration cycles, is classified in three main processes[207] including semiregenerative,[208,209] cyclic,[210,211] and continuous regeneration.[212,213] From the 1970s, when the catalytic continuous regeneration (CCR) process by MBR technology was introduced to the industry, it has attracted worldwide attention, and until now, it has been one of the well-known processes in the petroleum and petrochemical industries.[201] In the next sections, the conventional and optimized naphtha configurations (thermally coupled reactors and membrane reactors) are reviewed in detail.

3.4.1. Conventional Naphtha Process. 3.4.1.1. Catalyst Deactivation. Many studies have been devoted to investigating the catalyst deactivation of the naphtha process. The catalyst deactivation decreases the catalyst active sites, and it contributes to the reduction of the catalyst performance and, obviously, the production rate. The catalyst deactivation occurs due to several factors such as poisoning,[214] sintering,[214] feed impurities, unfavorable byproducts,[214] and coke deposition. It is worth mentioning that there are two kinds of deactivation procedures including reversible and irreversible. The reversible deactivation occurs due to the leaching of some catalysts' active components from the catalyst structure[215] or H_2S deposition on the catalyst surface.[216] In these cases, the deactivated catalysts can be regenerated by injection of some special additives to the reaction media. On the other hand, metal functions including arsenic, lead, etc. have a permanent deactivation impact on the catalyst performance.[216] For the irreversible deactivation, the catalysts

DOI: 10.1021/acs.iecr.9b01136
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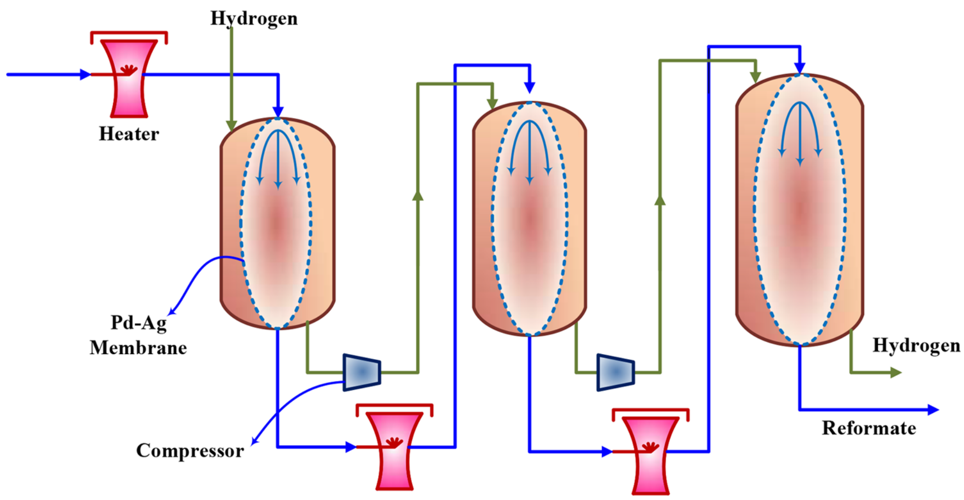

Figure 12. Simple schematic of the membrane catalytic naphtha process.

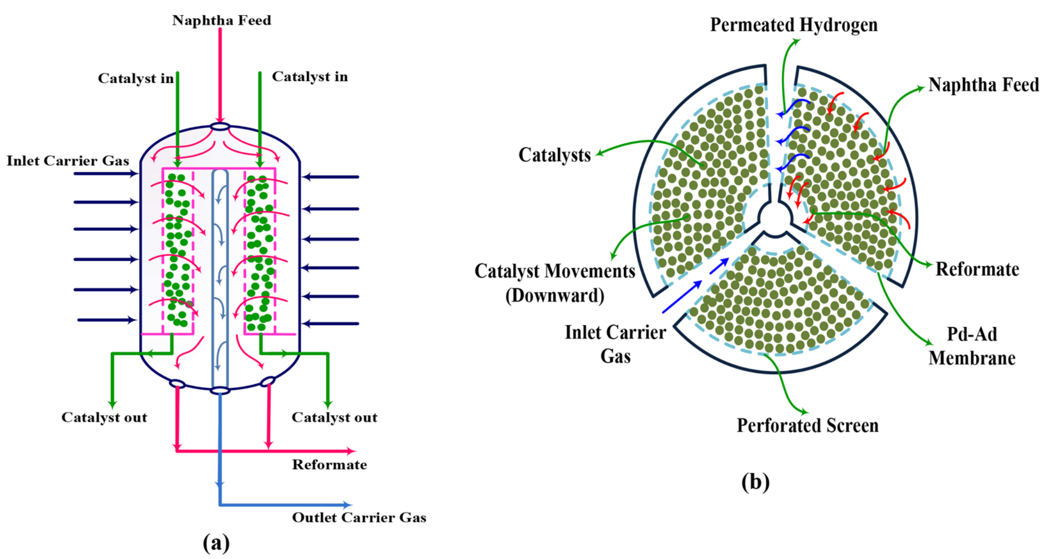

Figure 13. Simplified schematic of a (a) radial and (b) top view of a membrane reactor of CCR naphtha reforming.

usually should be regenerated in a separated unit or withdrawn and replaced with fresh catalysts.[215] Sie[215] studied the relation of the catalyst deactivation rate and process parameters such as the reactor configuration and the reactor type. He reported that the catalyst performance and the deactivation rate are significantly influenced by the process parameters.

Baghalha et al. studied the impact of the coke deposition on the commercial Pt−Re/γ-Al_2O_3 catalyst of the MBR naphtha process.[217] They observed that the coke deposited on the micropores and mesopores of the fresh and reused catalysts at a constant layer thickness (1 nm). Furthermore, they reported the coke deposition rate as the slowest step and the controlling one in comparison with the high rate of the coke ingredient diffusion into the pores. The coke formation and its effects on the catalyst performance at three levels of the active site, the particle, and the reactor were modeled by Froment.[214] He concluded that the experimental deactivation models are not robust enough to present the most efficient way to choose the best reactor type; thus, the economic aspects of the catalyst deactivation should be considered in the modeling of the catalyst deactivation phenomena. In another study,[218] Froment proposed the injection of H_2 to the catalytic reforming of the naphtha reactors to obviate the rapid deactivation of the Pt−Re/alumina. The impact of the chlorine content on the activity of Pt−Re/Al_2O_3 in MBRs was investigated by Verderone et al.[219] They reported a reduction in the catalyst activity by increasing the chlorine content in the feed.

Temperature and H_2 partial pressure are the other factors, which have significant effects on the catalyst deactivation. The optimum inlet temperature for the maximum performance of the catalyst was identified by Taskar and Riggs.[208] The lower temperature profile in the reactors usually contributes to a lower rate of the catalyst deactivation.[220] There are many deactivation models for the naphtha process such as Levenspiel's deactivation kinetic model (LDKM),[221] deactivation models with residual activity (DMRA),[222] etc. Monzon et al.[221] proposed a mathematical relation between the LDKM factors and the DMRA parameters. They reported an equation, which models the Pt/Al_2O_3 reforming catalyst deactivation, by fouling through the methyl cyclohexane dehydrogenation. In some cases,[223,224] the weighted average inlet temperature (WAIT) was described as the most important parameter to compensate the catalyst

DOI: 10.1021/acs.iecr.9b01136
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deactivation, because of the positive impact of the WAIT on the aromatic yield.[224]

3.4.1.2. Kinetics. Many reactions are involved in the naphtha reforming process through the moving bed reactors, which make the kinetics of this process so complicated.[225] The first reaction kinetics were reported in 1959 by Smith[226] and Krane et al.,[227] in which all reactions were considered in the lumped groups. After that, a lot of research improved the naphtha kinetics, in which some pseudocomponents were considered in the network kinetic models.[228−232] In such a context, the main reactions, corresponding feeds, and products, which were considered in these studies, are summarized in Table S2 (Supporting Information).

As previously mentioned, the reaction kinetics of the catalytic reforming are so intricate; thus, all naphtha reactions are typically categorized in three main classes including paraffins, naphthenes, and aromatics reactions, and it is assumed in each category that the same reactions with similar properties take place.[233] Sotelo-Boyas and Froment proposed a kinetic model for the naphtha process on the Pt−Sn/Al_2O_3 catalyst.[234] The model had an acceptable agreement with experimental values, but on the other hand, it contained a multitude of parameters. The pseudo-homogeneous and heterogeneous reactors were modeled in a radial flow regime, and the predicted yields were in an excellent agreement with the experimental data.[234]

3.4.1.3. Process Modeling and Optimization. Modeling and simulation of the catalytic naphtha reforming is a valuable task to evaluate the differential temperature through the reactors,[208] products yield,[235] RON,[235] reformate percentage,[235] feed heater duty,[208] process optimization,[208] as well as the catalyst activity and lifetime.[208] Formerly, the naphtha process was considered in the fixed-bed reactors for process modeling and simulation,[236] but in recent years, on the basis of the requirements of the continuous catalyst regeneration, many researchers have focused on the MBRs modeling, according to the UOP and Axens licenses.[237] In this way, the reactor configurations,[238,239] reaction kinetics,[233] and catalyst deactivation models[165] were considered to determine the process parameters. The main studies in this area have been summarized in Table S3 (Supporting Information). Also, the distribution of the reactor configurations is depicted in Figure 10.

3.4.1.4. Operational Conditions Effects. There are different parameters which affect the naphtha reforming process such as the pressure, temperature, feed composition, hydrogen/hydrocarbon ratio, and the space velocity.[240−242] The quality of the naphtha reforming process is evaluated by investigating the variations in the reformate yield, the coke deposition, the octane number, etc.[240] Also, the H_2/HC ratio has a direct impact on the coke deposition on the catalysts' surfaces and, consequently, the catalyst lifetime; hence, the H_2/HC ratio affects the aromatics production yield, indirectly.[228] On the other hand, many studies have considered the effect of the feed inlet temperature on the product yield.[233,243]

Hongjun et al.[244] studied the naphtha reforming process in the moving bed reactors in the series. They considered two different operational conditions for the feed composition, feedstock flow rate, and H_2/HC ratio. They reported that the higher inlet temperature of reactors leads to the higher rate of the coke deposition and the enhancement of the aromatic yield with increasing the space time. Karimi et al.[245] studied the naphtha reforming process through the MBRs, considering the temperature and H_2/HC ratio as the most effective parameters on the process. The reaction temperature was also introduced as one of the most important parameters affecting the yield of the reforming process. The catalytic naphtha reforming is a highly endothermic process; hence, the process efficiency decreases as the feed goes through the reactors. They considered the hydrogen as a product through the dehydrogenation of naphthenes and the dehydrocyclization of the paraffins, as a reactant in the hydrocracking reactions of the paraffins and naphthenes, and in the hydrodealkylation of the aromatics. According to the fact that the H_2/HC ratio has a significant impact on the coke deposition on the catalysts, it should be at an acceptable level to maintain the catalysts lifetime as high as possible.[245] Also, in another study, Hu et al.[223] introduced the WAIT (weighted average inlet temperature), H_2/HC, and the reaction pressure as key parameters on the performance of the naphtha reforming process.

3.4.2. Optimized Naphtha Configurations. Researchers, continuously, have been looking for innovative ways to improve the efficiency of the reforming process. In this way, enhancing the reformate octane number using thermally coupled reactors,[246,247] membrane reactors,[225] spherical reactors,[248] different reactor configurations,[225] and diverse flow regimes has been investigated, so far. These theoretical methods have been applied together sometimes, which is shown in Figure 11. The designed figure shows the share of each method schematically, by the size of each box.

The refining process is an industry with a high level of energy consumption.[249] Thus, energy optimization has become an important issue, which directly affects the refinery economics.[250] Babaqi et al.[250] studied the heat integration retrofit of the CCR process. They developed a method by pinch analysis technique to save the maximum possible energy. They reduced the utility loads by around 32%, which equals a 4.5% reduction in the total cost index.

3.4.2.1. Thermally Coupled Reactors. The recuperative coupled reactor is one of the other pioneer strategies to reduce the operational and capital cost of power plants which has attracted much consideration in recent years.[246] In this way, the reactors are designed and constructed in a way that an exothermic reaction supplies the required energy for the naphtha process.[251] For example, the huge released energy through the hydrogenation process can be considered as a heat source of endothermic reactions.[251] The main reactions, which have been studied as a source of energy in the naphtha process, are reported in Table 4. It can be found that around 61.5% of the total studies, which were performed in this area, have been devoted to the hydrogenation of the nitrobenzene to aniline, 23.1% to the oxidation of the sulfur dioxide to SO_3, and 15.4% to the hydrodealkylation of the toluene. Furthermore, studies are ongoing to find new exothermic processes which are more applicable and efficient for the naphtha process.

3.4.2.2. Membrane Reactors. Khosravanipour Mostafazadeh and Rahimpour[220] first introduced the concept of membrane reactors for catalytic naphtha reforming in 2008. Figure 12 represents a simple schematic of the membrane catalytic naphtha reforming process. In this way, the hydrogen from the shell side penetrates through the membrane to the tube side, where the reactants flow in the reaction beds. Thermodynamically, the reforming reaction is shifted toward the production side, which increases the production rate.[220]

They reported that a Pd−Ag membrane can enhance the aromatic production rate in the catalytic reforming process. It is worth mentioning that their proposed model was accomplished in the fixed-bed reactors. Recently, Iranshahi et al. proposed the

DOI: 10.1021/acs.iecr.9b01136
Ind. Eng. Chem. Res. 2019, 58, 9179−9198

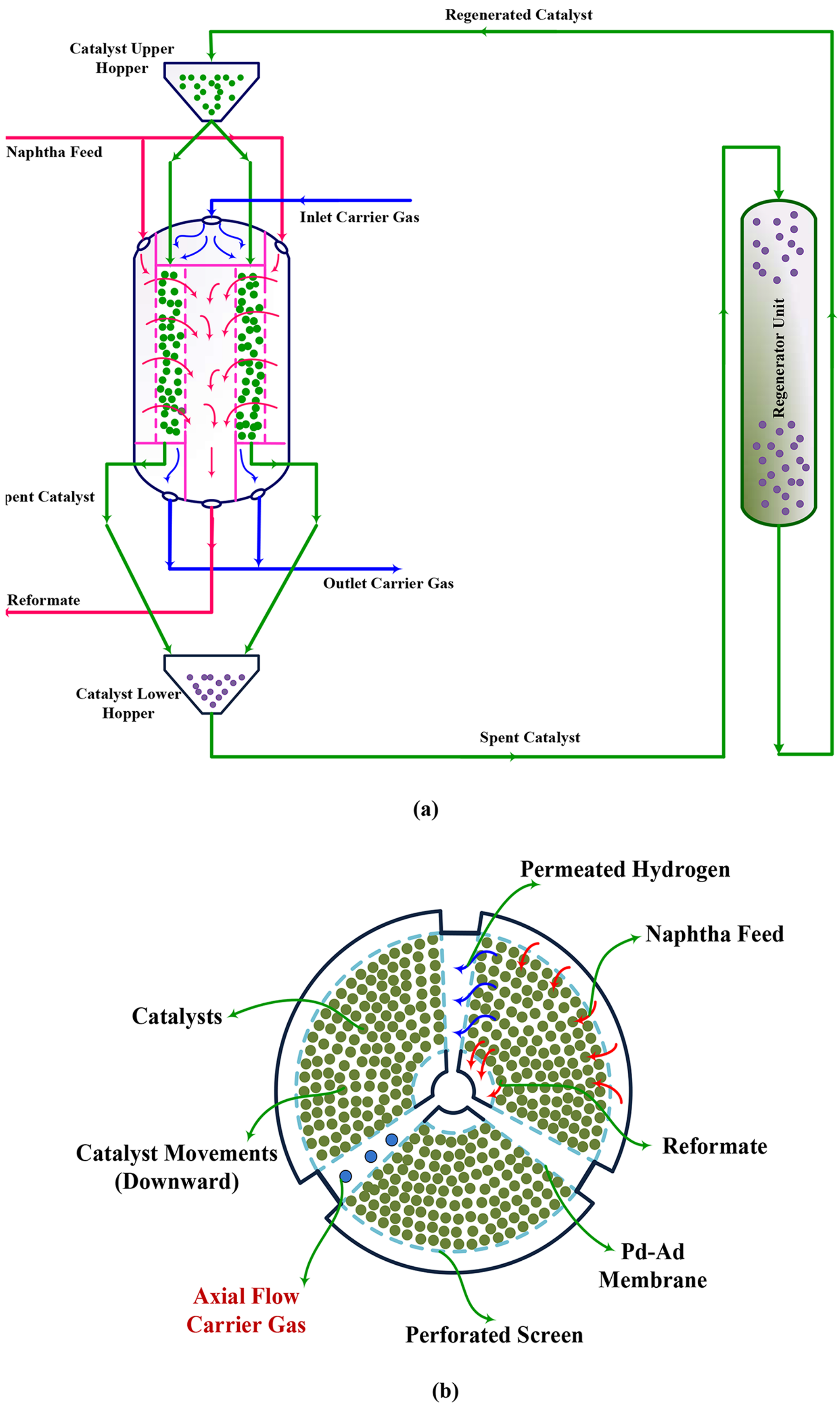

Figure 14. Radial flow regime for reacting materials and axial flow regime for carrier gas in catalytic naphtha reforming: (a) front view and (b) top view.

naphtha process in MBRs by considering a two-dimensional model (in the radial and axial directions).[257] They modified the parameters of membrane reactors with the CCR concept, in which the advantages of MBT were applied. Figure 13 shows a membrane reactor for CCR naphtha reforming with a radial flow pattern in the front and top views, schematically. Saeedi and

DOI: 10.1021/acs.iecr.9b01136
Ind. Eng. Chem. Res. 2019, 58, 9179−9198

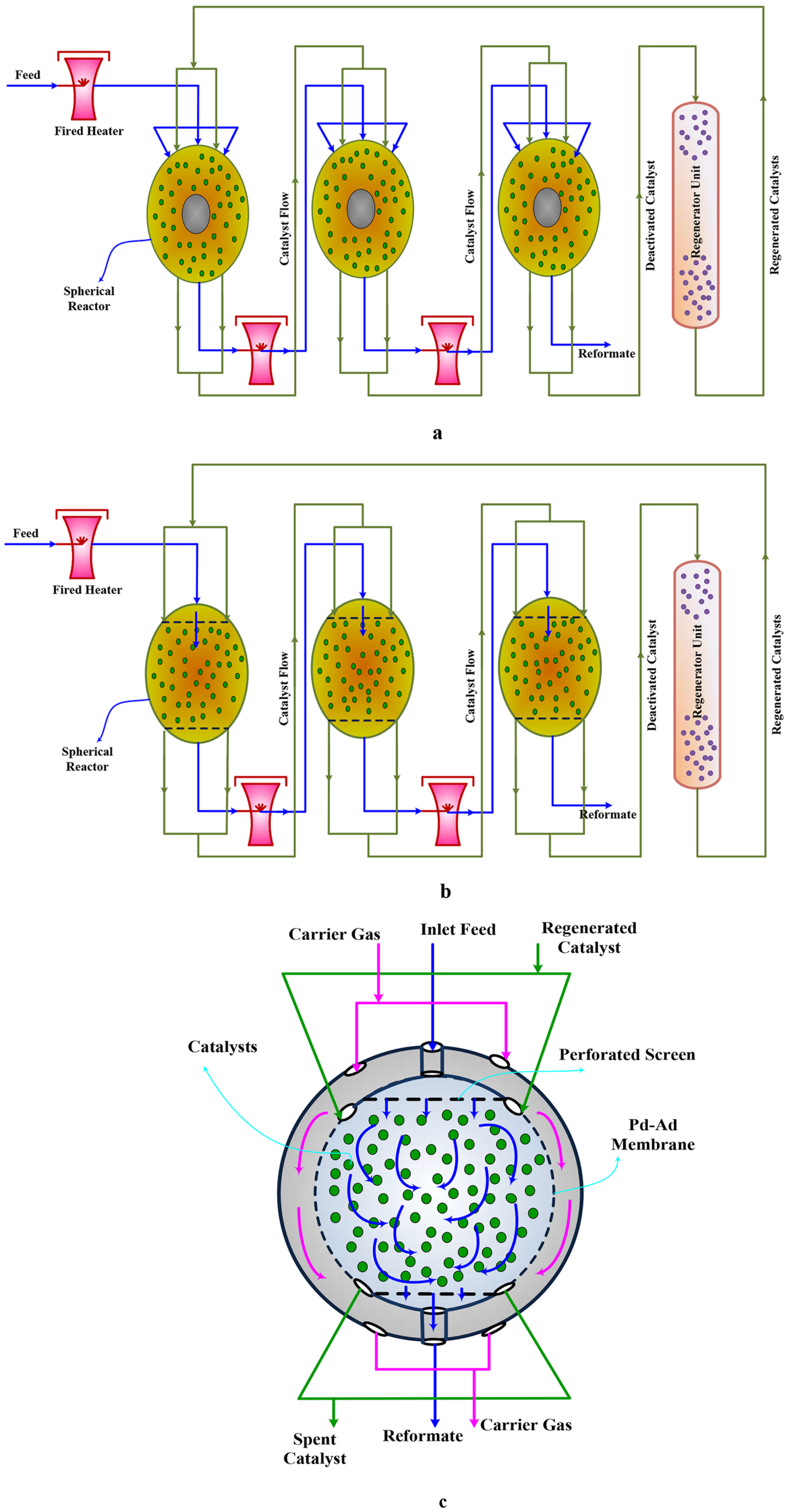

Figure 15. Simple schematic of three spherical MBRs with (a) radial and (b) axial patterns and also with (c) the membrane concept.

Iranshahi performed another study and compared their results with industrial plant data in 2016.[258] They reported a good agreement between the model outputs and industrial values. They also observed that employing the membrane concept on the MBRs enhances the octane number of the gasoline and hydrogen production rate, significantly.

4. DISCUSSION AND SUGGESTIONS

The waste management, environmental challenges, and limitations of fossils fuels have been introduced as main concerns of mankind in recent years.[259−261] On the other hand, the increasing rate of the worldwide energy consumption has intensified the situation and forced researchers to find new sources of energy and improve the quality of available ones. In this way, the MBRs by their widespread applications in chemical looping combustion, pyrolysis technology, fluid catalytic cracking, and the naphtha process have a key role in these industries. The MBR is a valuable technology which can also be coupled with other new processes to develop a highly efficient strategy.[225]

Until around 1960, a high rate of the catalyst deactivation was one of the major restrictions of catalytic reforming, while the advent of MBRs has had a significant effect on this process by developing high-octane reformate and high-quality hydrogen.[262] Then, the development was pursed by a combination of membrane reactors and conventional ones to enhance the process performance. In this way, the radial and axial flow regimes for reactive compounds and the axial flow regime for the carrier gas were proposed to overcome the high pressure drop in the catalytic packed-bed reactors.[248,263] For future studies, the packed-bed reactors can be replaced by the MBRs, which is illustrated in Figure 14.

In addition, the idea of employing spherical MBRs instead of conventional ones (tubular MBRs), on the basis of their lower pressure drop through the catalytic bed, can be an excellent strategy in this area for future studies. A schematic diagram of this proposed model for three MBRs in the catalytic reforming process is presented in Figure 15a. On the other hand, the effects of different flow patterns (axial and radial, Figure 15a,b) on the production rate, hydrogen purity, and RON can be investigated as theoretical or experimental studies. Also, upon consideration, the membrane concepts on the spherical MBRs can improve the process performance,[263] notably (Figure 15c).

A combination of spherical MBRs with membrane tubular reactors to develop an efficient configuration for producing high-octane reformate through the CCR process is also an attractive strategy, which can be studied in detail in future works.

Finally, by considering the significance of the process integration (PI) strategy,[247] merging thermally coupled reactors with proposed spherical/membrane and spherical/radial flow MBRs can be alluring ideas for MBT.

These suggestions show that the innovation in the reactor design and novel configurations can improve the performance of the conventional MBRs, which can be considered for future studies.

5. CONCLUSIONS

The moving bed technology was first introduced as a steady-state operation, in the drying process, and then for the catalyst regeneration in the chemical reactions. Today, on the basis of the high efficiency of MBRs, this technology has been widely employed in different industries. In this study, the gathering and discussion all main studies about MBT from the advent of this technology until now were attempted. Through this review, all studies in MBT were categorized in two main classes: experimental and theoretical studies. In this way, the gas—solid moving bed technology as an important and practical application of MBT was indicated. Also, the importance of MBT in the chemical looping combustion, the pyrolysis industry, and the fluid catalytic cracking process, as three of the leading technologies, was discussed in details. In addition, the naphtha reforming process was investigated in two categories: conventional naphtha processes and optimized processes. While the MBT in the catalytic reforming process has been extensively studied in the literature, and it has contributed to some confusing issues, in this work, the classification and merging of the main studies were attempted to aid researchers to have a better grasp about these concepts.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.iecr.9b01136.

Different types of reactions in the lumped formulation of naphtha catalytic reforming kinetic models; and reactor type, catalyst nature, and kinetics in different catalytic naphtha reforming processes (PDF)

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Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was financially supported by Project POCI-01-0145-FEDER-006984—Associate Laboratory LSRE-LCM funded by FEDER through COMPETE2020—Programa Operacional Competitividade e Internacionalização (POCI), and by national funds through FCT—Fundação para a Ciência e a Tecnologia. M.K. also acknowledges the research grant awarded under Project SFRH/BD/140550/2018 by Portugal Foundation of Science and Technology (FCT).

■ NOMENCLATURE

MBR = moving bed reactor
MBT = moving bed technology
CCR = continuous catalyst regeneration
SMBR = simulated moving bed reactor
CCS = carbon capture and storage
CL = chemical looping
CFD = computational fluid dynamics
LDKM = Levenspiel's deactivation kinetic model
WAIT = weighted average inlet temperature

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