Study on the FCC Process of a Novel Riser–Downer Coupling Reactor (III): Industrial Trial and CFD Modeling

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Industrial trials detailed in a riser–downer coupling reactor proposed for the fluid catalytic cracking (FCC) process are presented in this paper. The trials are based on the fundamental research of hydrodynamics, mixing behaviors, and laboratory-scale hot experiments described in our previous investigations. Compared with the riser reactor with the same feeds and catalyst, the LPG and propylene yield increased by 8.15 and 4.30 wt %, respectively. The gasoline octane number likewise reached 94.8 with 28 wt % olefin content. However, dry gas is significantly suppressed, and the coke has little change in yield even with the increased catalyst to oil ratio. With some gasoline recycling, the LPG and propylene yield increased by 11.45 and 5.06 wt %, respectively, and the olefin content in gasoline significantly decreased to 22 wt %; the high octane number (95.4) is maintained. The computational fluid dynamics (CFD) coupled with a 6-lump kinetic model is also applied to simulate the FCC process for the industrial trials. The yield of propylene and butylene and the temperature profile along the axis direction demonstrated consistency between the simulation results and the experimental data.

1. Introduction

Since the discovery of the zeolite catalyst, the circulating fluidized bed (CFB) riser has been successfully applied in the petroleum refining process (that is, fluid catalytic cracking or FCC) for many decades and continues to play an important role as a gasoline-producing unit. Among the reactors adopted in FCC, the riser type is the most common. It has the advantages of high gas–solid contact efficiency, high gas/solid flux, and high operating flexibility. Nonetheless, the conventional riser reactor’s inherently nonuniform flow structure and sizable back mixing of solids has put it at a disadvantage. This high-severity operation of risers results in enhanced coke and dry gas formation, limited conversion, and selectivity to liquid products; gasoline is an example. With this in mind, the concept of a “downer” reactor was later proposed in the 1980s,† and extensive studies were conducted on all significant aspects.‡–‡‡ In downers, the gas and solids flow concurrently downward in the same direction as gravity, thus rendering distinguished features such as significantly less back-mixing of both gas and solids as well as the “close to plug flow” pattern§ in the entire reactor. Moreover, some of its hot model experiments in the laboratory scale††–‡‡‡ have demonstrated its high-severity process to enhance gasoline and light olefins production. This improves the selectivity of desired products in comparison with the commercial riser when adopting the same feed and catalyst, for example, increasing yields of propylene and gasoline but reducing dry gas and coke formation. The downer process boasts of high catalyst-to-oil (C/O) ratios and a short contact time as well.

Though downers have demonstrated their prospective use in petroleum refining, there are still certain hindrances that need to be overcome in order to smoothly industrialize this new type of reactor. For instance, a downer design strongly relies on initial phase contact due to the inlet configuration considering the overall ultrashort reaction time (typically less than 1 s)‡‡‡. In addition, highly diluted catalyst flows in downers may exert a negative effect on the transport phenomena in the reactor and consequently on the total conversion; the product distribution is sensitive to the contact efficiency between oil droplets and catalyst particles.‡

The everincreasing demand for light olefins accompanied by growing environmental concerns calls for current FCC techniques‡‡‡directed at maximizing the high added value products of light olefins (C2–C4) all the while achieving higher quality gasoline with less olefins and aromatics content. These new challenges require a high-severity FCC process made up of a reactor operated under high cracking temperature conditions, short contact time, and plug–flow pattern.

In line with the aforementioned impediments, we propose a novel riser–downer coupling reactor (henceforth referred to as “coupling reactor”) with the following mechanism:‡‡‡ First, the feedstock enters the riser reactor to execute the first-stage reactions while taking advantage of the excellent heat and mass transfer in the riser, eventually ensuring overall conversion. Subsequently, the reacting flow smoothly transfers to the downer that acts as the second-stage reactor where the remaining reactions are completed. It is likewise here that less back-mixing is utilized to avoid the potential for overcracking which, in contrast, occurs in a typical riser FCC unit running at high severity. The selectivity of reactions can then be secured. The reactor is notably made to adopt the configuration of a dual cylinder with the annulus operating as a riser and the inner cylinder as a downer, thus ensuring good transition between the riser and the downer. This is best illustrated in a cold experimental apparatus with 5.5 m length downer§§ where, at a position of 0.8 m away from the inlet, the solid fraction shows a pattern with a flat central zone and a dense annular near the wall, which is close to the typical density profile in the fully developed downer. The carefully designed junction can serve as an inlet structure of the downer component to attain uniform

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distribution of solids without increasing the back mixing of particles in the whole coupling reactor. The particle dispersion mechanism has also been investigated,16–18 and the differences in microstructures can explain the characteristic particle mixing behaviors observed in the coupling reactor. Furthermore, the injection points of feed oils can be flexibly adjusted along the riser and downer. As a result, the total residence time and distribution of each phase can be effectively controlled. This leads to large operation flexibility for different cracking purposes when running this proposed reactor prototype in the industry.

In our previous work,19 a two-dimensional dispersion model was developed based on the hydrodynamics and mixing behaviors of the reactor combined with an 11-lump FCC reaction kinetics model to simulate the FCC process occurring in the coupling reactor. The simulation results on the FCC process revealed that in order to obtain the same gasoline yield, the coupling reactor requires a shorter length than the separate downer reactor. Nevertheless, it still possesses much higher gasoline selectivity compared to the separate riser. The simulation also confirms that by adjusting the feed injection positions the coupling reactor can be flexibly operated equal to the various situations of stock and product markets.

At the first industrial demonstration plant, this new coupling reactor with a capacity of 150 000 t/a and built at the Jinan Petroleum Refinery of SINOPEC in China, was successfully tested in two cracking processes, namely, the deep catalytic cracking (DCC) and residue fluid catalytic cracking (RFCC) processes. The results can further greatly encourage the industrial application of coupling reactors, and full commercialization is expected to come soon. In this paper, the RFCC process and results are discussed in detail. It is also worth mentioning, however, that due to its complex hydrodynamics, the experiment does not cover all the detailed aspects. The simulation, particularly by computational fluid dynamics (CFD), provides an easy way of understanding this point.20–22 Thus, for better elucidation of the complex hydrodynamics and kinetics in the RFCC process, the CFD and a 6-lump reaction kinetics model are adopted to simulate the FCC process. These are expected to provide reasonable predictions of reacting flow in a coupling reactor that can contribute to the scale-up procedure and improvement of the reactor design.

2. Industrial Trial and Results

Figure 1 illustrates the original schematic diagram wherein the apparatus has a total height of 41.0 m. The feed used in the experiments is residue oil: its density is 930 kg/m³ (20 °C), viscosity is 16 mm²/s (100 °C), pour point is 40 °C, and carbon residue is 4.04; sulfur and nitrogen content are 4600 and 2500 ppm, respectively, while the concentrations of saturated hydrocarbons, aromatic hydrocarbons, asphaltene, and gum correspond to 60.8, 21.87, 0.81, and 16.52 wt %, respectively. The catalyst used is the MLC-500: its packing density is 0.89 kg/cm³ with an average particle diameter of 70 µm; its catalytic activity is 66.

The regenerated catalyst is made to enter the main riser and then raised by steam or dry gas to the riser–downer conjunction where the gas–solids system changes its flow direction and decelerates before entering the downer. A quick separator is fabricated at the outlet to isolate the deactivated catalyst from the gas products in a very short time. The hot gas is discharged for cooling and further separation, while catalyst particles are stripped of the absorbed gas/oil in the stripper and introduced into the regenerator (composed of a vessel combustor, a dense fluidized bed, and a dilute fluidized bed) for coke removal. The regenerated catalyst is then sent back to the reactor to be reused in the next cycle. In addition, seven groups of high-efficiency atomizing injecting nozzle BMJ are set at the coupling reactor as shown in Figure 2. By injecting the stocks from various nozzles, the reaction time can be adjusted by a wide range without changing the feeding rate or solids flux. Figure 3 shows a photo of the industrial coupling reactor in the Jinan Petroleum Refinery of SINOPEC in China, which was built based on the novel riser–downer coupling reactor concept.

On the basis of the industrial test, operation of the pilot plant is stable and the FCC catalysts are in an excellent fluidized state in the reactor with the maximum catalyst to oil (C/O) ratio reaching 22. Moreover, the riser–downer conjunction exhibited good performance in operation: the mean density reached 300 kg/m³, the gas velocity in the quick separator varied between 20 and 43 m/s, the pressure drop of the reactor is 2–8 kPa, the separation efficiency is above 98%, and the solid fraction in liquid slurry is less than 2 g/mL, which indicates that the quick separator satisfied the separation requirements.

By adopting the same stock (residue oil) and FCC catalyst, the catalyst to oil ratio was approximately 10 and the stock injecting nozzle of the coupling reactor was labeled as E (see Figure 1). Table 1 shows a comparison of the typical product profiles obtained from the coupling reactor and a conventional riser reactor of 380 mm i.d. and 48.0 m in length. It can be observed that in contrast to the riser, the gasoline yield increased...
that of an industrial riser FCC by 1.4 Mt/a, its heat loss and
because this riser is also notable in rigorous environmental protection. It is also notable
number of gasoline achieved 95.4. Hence, these values can be
dramatically decreased to 22%, while the octane
yield decreased by 1.32%. Additionally, the olefin content in
dry gas increased by 11.45% and 5.06%, respectively, and the dry gas
loss of 0.03% liquid yield, the LPG and propylene yield
due to the gasoline’s reduced overcracking. It is generally accepted that suppressing
back-mixing can also restrain secondary reactions in FCC, thus increasing the intermediate products. Therefore, the improved selectivity of the desired products in the coupling reactor can be attributed to adaptation of the downer reactor, which consequently eliminates the back-mixing that is obviously desirable for the cracking process. Such a trend agrees well with the results obtained by Deng et al. from the DCC process in a downer reactor.

It can be concluded that the products in the coupling reactor tend to concentrate on those with middle molecular weight (propylene, liquefied gas, and gasoline), while those with very high (diesel oil) and very low (dry gas) molecular weights are significantly suppressed. The olefin content in gasoline is also lower than those from the riser and mainly due to the gasoline’s reduced overcracking. It is generally accepted that suppressing back-mixing can also restrain secondary reactions in FCC, thus increasing the intermediate products. Therefore, the improved selectivity of the desired products in the coupling reactor can be attributed to adaptation of the downer reactor, which consequently eliminates the back-mixing that is obviously desirable for the cracking process. Such a trend agrees well with the results obtained by Deng et al. from the DCC process in a downer reactor.

Figure 4 illustrates the schematic diagram of an online sampling process. It is a tube that is 2 mm i.d. and 2.0 m in length with a metal net filter on its tip to prevent the catalyst from entering the probe. A rubber seal is installed at the cold end of the sampling opening to prevent the high-temperature poisonous gas from leaking out of the reactor and allowing the sampling probe to move along the radial direction. The sampled gas is then collected by a bag for further gas chromatograph analysis.

The gas sampling results in the G nozzle and inlet of the quick separator are shown in Figure 5. The propylene and butylenes are the main composition of the product, which is about 40% in the gas phase. Meanwhile, based on the trend of the product distribution along the axis direction, the decreasing olefin content in gasoline can result in the increase of olefin in gas products, which agrees well with the results in Table 1. Furthermore, as illustrated in Figure 5b, the product obtained by the quick separator shows that the composition of the propylene increased to over 30%. This is mainly attributed to the cracking of the liquid component, mainly gasoline. Subsequently, the distribution of the product along the axis direction became uniform. The G nozzle is about 6 m down from the injecting nozzle E, where the radial distribution of the gas sampling showed larger variation. This is contrary to the common understanding of the downer with a more uniform profile with the higher solids fraction in the downer being the main reason for the discrepancy. Generally, a downer operates with a much lower solids fraction than a conventional riser; in a coupling reactor, both downer and riser operate with the same higher solids fraction. Indeed, the local solids fraction plays a very important role in the dispersion behavior; with increased solids fraction, particles are likely to be more difficult to disperse, especially in the radial direction. As the reaction proceeds, the molar gas phase changes, leading to obvious changes in gas velocity and thus directly affecting the distribu-

![Figure 3. Photo of the RFCC pilot plant: riser, 380 mm i.d., 40 m high; downer, 480 mm i.d., 15 m high; capacity, 150 000 t/a, Jinan Petroleum Refinery, Shandong Province SINOPEC. Figure 4. Schematic diagram of the sampling device: (1) reactor; (2) metal net; (3) inner tube; (4) outer tube; (5) rubber seal; (6) bag for gas collecting.]

Table 1. Comparison between the Coupling Reactor and Commercial Riser

<table>
<thead>
<tr>
<th></th>
<th>riser</th>
<th>coupling reactor</th>
<th>coupling reactor with gasoline recycling</th>
</tr>
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<tbody>
<tr>
<td>carbon residue</td>
<td>4.56</td>
<td>4.32</td>
<td>4.04</td>
</tr>
<tr>
<td>dry gas</td>
<td>5.10</td>
<td>3.27</td>
<td>-1.83</td>
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<tr>
<td>liquid gas</td>
<td>14.48</td>
<td>22.62</td>
<td>8.14</td>
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<tr>
<td>gasoline</td>
<td>37.97</td>
<td>41.87</td>
<td>3.90</td>
</tr>
<tr>
<td>diesel oil</td>
<td>28.15</td>
<td>17.68</td>
<td>-10.47</td>
</tr>
<tr>
<td>coke</td>
<td>9.03</td>
<td>9.97</td>
<td>0.94</td>
</tr>
<tr>
<td>liquid yield</td>
<td>80.60</td>
<td>82.17</td>
<td>1.57</td>
</tr>
<tr>
<td>propylene</td>
<td>5.30</td>
<td>9.6</td>
<td>4.30</td>
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<tr>
<td>conversion</td>
<td>66.58</td>
<td>77.73</td>
<td>11.15</td>
</tr>
<tr>
<td>olefin content</td>
<td>45</td>
<td>28</td>
<td>22</td>
</tr>
<tr>
<td>octane number</td>
<td>90</td>
<td>94.8</td>
<td>95.4</td>
</tr>
</tbody>
</table>

by 3.9%, the total liquid yield increased by 1.57%, LPG and propylene yield increased by 8.15% and 4.30%, respectively, the dry gas yield decreased by 1.83%, and the coke yield only increased by 0.94%. Meanwhile, the olefin content in gasoline decreased significantly to 28 wt % with the octane number increasing to 94.8.

With a portion of the product gasoline recycling into the coupling reactor, the gasoline yield decreased as opposed to absent gasoline recycling conditions. On the other hand, the decrease of gasoline led to a notable increase of diesel oil, propylene, and liquid gas with slight dry gas increases while the coke formation decreased. In contrast to the riser, with a loss of 0.03% liquid yield, the LPG and propylene yield increased by 11.45% and 5.06%, respectively, and the dry gas yield decreased by 1.32%. Additionally, the olefin content in gasoline dramatically decreased to 22%, while the octane number of gasoline achieved 95.4. Hence, these values can be helpful in rigorous environmental protection. It is also notable that because this riser—downer FCC unit is much smaller than that of an industrial riser FCC by 1.4 Mt/a, its heat loss and
tion of solids velocity and fraction. On the other hand, in the inlet of quick separator, which is the end of coupling reactor, the radial distribution is relatively uniform in the fully developed region of a downer.

It should be noted that in the application of the coupling reactor, hot experiments also indicate the importance of adopting a catalyst with high cracking activity. As stated above, the reason for introducing the coupling reactor in the FCC process is that the downer reactor cannot offer a sufficiently high feed conversion and gasoline yield. Conversely, the riser can achieve this. The lower the catalyst activity needs, the longer the effective riser length. A long riser part in the coupling reactor, however, indicates that the effects of the desirable flow pattern in the downer part are not fully utilized. Alternatively, commercial FCC catalysts for the riser reactor are designed, which optimally perform to avoid low conversion or overcracking. A replacement of the present riser with a downer will thereby decrease the feed conversion, albeit neither C/O ratio increase (due to the limit of pressure balance) nor reaction temperature increases (due to the intensity of thermal cracking) could completely overcome it. The potential solution is to develop a very high activity catalyst for the downer or coupling reactor, which renders it possible to simultaneously achieve high conversion and gasoline yield in a finite reactor length.

The experimental results likewise reveal that the injection locations can be altered to obtain various desired products. For example, if the liquefied petroleum gas (LPG) is favorable, the adopted nozzles should be much closer to the downer reactor than those selected for gasoline production because a shorter residence time is needed. This can be verified by industrial operation (Figure 6). The liquid yield also decreased by adjusting residence time is needed. This can be verified by industrial examples, if the liquefied petroleum gas (LPG) is favorable, the locations can be altered to obtain various desired products. For example, if the liquefied petroleum gas (LPG) is favorable, the locations can be altered to obtain various desired products. For example, if the liquefied petroleum gas (LPG) is favorable, the locations can be altered to obtain various desired products. For example, if the liquefied petroleum gas (LPG) is favorable, the locations can be altered to obtain various desired products.

3. Simulation on the FCC Process in the Coupling Reactor

The industrial trial is costly and information complex, particularly the interaction among the hydrodynamics and kinetics. Nonetheless, the nature of a large reactor is essential, and the previously applied CFD technique is being employed to simulate the industrial unit of the coupling reactor. As a consequence, simulation via CFD coupled with the kinetic model revealed some details of the reactor which can finally assist the scale up of the RFCC process. On the basis of our previous CFD work, a 2-D two-fluid model considering the turbulence nature of the high-velocity flows in the reactor is proposed in this work, while 6-lump reaction kinetics is incorporated in the governing equations to simulate the reacting flows in the industrial unit. The above-mentioned industrial data are available for comparison with the model predictions on reacting flows. Detailed simulations are to be carried out to examine the coupling reactor and the influence of feed positions on the product distribution.

The general form of the governing equations for steady flow in the two-dimension cylindrical coordinate is represented by

\[
\frac{\partial}{\partial x}(\alpha \rho u \varphi) + \frac{\partial}{\partial r}(\alpha \rho u \varphi) = \frac{\partial}{\partial x}\left(\Gamma \frac{\partial \varphi}{\partial x}\right) + \frac{\partial}{\partial r}\left(\Gamma \frac{\partial \varphi}{\partial r}\right) + S_{\varphi i} + S_{\varphi j}
\]

where \(\varphi\) denotes general variables, \(\Gamma\) is a diffusion coefficient, \(S_{\varphi i}\) is the source term, \(S_{\varphi j}\) is the source term due to the interaction between the two phases, and \(i\) and \(j\) denote the gas and solid phases, respectively. For details, please refer to the work by Zheng et al.26

In reality, FCC is quite a complex reaction process involving many species interrelated by complicated chemical reactions. It is thus difficult to study FCC reaction kinetics using ordinary methods. However, many kinds of lump kinetic models have been developed since the 1960s.27–29 In this present work, the 6-lump kinetic models30 are adopted, and the related reaction net is shown in Figure 7.

The 6-lump kinetic model is divided into two parts: the first part is formation of dry gas and coke produced via the free radical mechanism, while the second involves catalytic reactions with two different decay functions for the olefin and paraffin reactions. The latter affects gasoline and LPG yield. Despite its simplicity, the model can predict the yield distribution of...
the main products according to gas sampling in the whole reactor, making it sufficient to study the interaction between hydrodynamics and the chemical reaction. The detailed reaction rate equations and kinetic constants are shown in the study by Liu.30

The flow and reaction model equations are coupled and coded according to the finite-volume solver CFX4.4. The SIMPLEC iterative algorithm is used to relate the velocity, which is similar to the Inter-Phase Slip Algorithm (IPSA) that employs the Partial Elimination Algorithm (PEA) developed by Spalding;31 this is used by the CFX4.4 solver. The operation conditions used in the simulation are the same as the industrial trial stated above. As the injection nozzle is E and the riser—downer conjunction exhibits good performance in operation, we can only simulate the downer for simplicity; the initial condition is set to be a uniform flow field. As the wall, the gas obeys the no-slip condition, whereas the solid phase is allowed to slip along the wall. For details, please refer to the work of Zheng et al.26

Figure 8 shows the axial yield distribution of all lumps and the industrial yield of gasoline, dry gas plus coke, and propylene. With the increase of height, conversion of the residue oil increased. Meanwhile, the gasoline, dry gas, and coke yield rapidly increased, and most of the reaction is completed within a relative distance of 0.4 along the coupling reactor from nozzle E. In this region, the concentration of all lumps rapidly changed and there are large concentration gradients along the directions of the axis. To decrease the negative effect on the reaction yield and selectivity, rapid and uniform mixing of the feed oil and catalyst in the inlet region is required. The riser—downer conjunction adequately solves this requirement. Additionally, the simulation yields agreed considerably with the industrial data, especially in the yield of propylene and dry gas plus coke.

To validate the simulation, the comparison of the main product and temperature distribution is demonstrated in Figure 9. Figure 9a, in particular, shows the radial profile of propylene and butylenes yield as they are considered only in the 6-lump kinetic model. Figure 9b shows the temperature radial distribution. It is noteworthy to add that the simulation results agreed well with the industrial sampling data. In such sampling position, the radial concentration profile showed minimal difference. The wall effect, however, is neglected in the model. Thus, there is minimal error at the center and higher near the wall. The temperature sampling results indicate that the temperature in the radial position slightly changed, which agreed well with the simulation catalyst temperature. The coupled model based on the two fluid models and lump kinetic model can then be applied in such industrial processes with optimal simulation results so provided. Thus, further CFD work should be performed to further understand the details of the reactor and scale up as well as optimize the reactor in the future.

4. Conclusions

The coupling reactor, in which a riser of appropriate length is fabricated in front of the downer, is advantageous in terms of both the riser and the downer. The RFCC process in an industrial coupling reactor shows optimal product distribution. Compared with the riser reactor with the same feeds and catalysts, the total yields of liquid products increased by 1.5 wt %, the propylene yield increased over 5 wt %, and the octane number achieved a high value; this is in the face of reduced olefin content in gasoline. More importantly, the dry gas is significantly suppressed and coke formation fails to increase (even with the increased C/O ratio). With some gasoline recycling, the yield of propylene increases and the olefin content in gasoline decreases, although the gasoline’s octane number increases. Following computational fluid dynamics (CFD) coupled with a 6-lump reaction kinetics model application to
simulate the RFCC process, results revealed high agreement with the experimental data. Therefore, the simulation method is an optimal tool in providing useful information for further scale up of the unit in the future. Moreover, application of the feed nozzle below E exhibited low conversion and can be attributed to the catalyst’s low activity. The most feasible way of improving the process is to develop a catalyst with high activity for the coupling reactor, making possible high conversion and gasoline yield simultaneously in the future.

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