Hydrodynamic Characteristics of an Internal Recycle Berty Catalytic Reactor in Batch/Continuous or Packed/Fluidized Bed Modes

Mengmeng Cui, Shekhar R. Kulkarni, Stefan Wagner, Claudia Berger-Karin, Anton Nagy, and Pedro Castaño*

1. INTRODUCTION

The proper design of laboratory-scale reactors for catalyst screening has always been a permanent goal in reaction engineering. The ultimate frontier of these instruments is the control or depletion of temperature and concentration gradients to avoid mass and heat transfer effects on kinetics, respectively. In this case, the “intrinsic kinetic regime” can be attained, meaning that the chemical kinetics unperturbed by transport limitations can be measured. The development of internally recycled reactors boomed in the 1960s and was reviewed as early as 1972 by Bennett et al. The main objective of recirculation is to remove the heat and mass gradients throughout a reactor and approach perfect mixing. Berty et al. significantly contributed to the field by incorporating a stationary catalyst basket, so such reactor subtypes were called “Berty reactors”. The so-called Berty reactor was first manufactured at Union Carbide Corp. with an impeller at its bottom, where gas is axially forced from the impeller through a fixed bed to create an internal recycle flow. Other representative internally recycled reactors include the Carberry and the Robinson–Mahoney [also known as stationary catalytic basket stirred tank reactor (SCBSTR)] reactors. The Robinson–Mahoney reactor is similar to the rotating packed bed reactor (RPB) of Ramshaw and Mallinson but with a different catalyst basket shape. All of these designs including the variants in batch/continuous or packed/fluidized bed modes are shown in Figure 1.

Internally recycled reactors have been widely used to investigate reaction kinetics and optimize the multiphase homogeneous or heterogeneous catalytic reactions with gas–solid, liquid–solid, and gas–liquid–solid interactions. The main difference between rotating and stationary catalyst basket reactors is the flow direction of the gas penetrating the

Received: October 5, 2021
Revised: November 30, 2021
Accepted: December 1, 2021
catalyst particles. While gas is axially forced through the stationary basket, the actual contact of gas with the catalyst is different for the leading face and backside in rotating bed reactors. Berty described that the filling level in a catalyst basket can be varied to lower degrees, so the possible transition from the packed to fluidized bed mode can be achieved with the momentum obtained from the rotating impeller. Thus, Berty reactors offer one important advantage over their internally recycled reactor competitors, meaning that fluidized bed catalysts can also be tested by reproducing the conditions in commercial fluidized bed reactors. Figure 1 shows the typical Berty configurations of impeller positions (top or bottom) in the batch and continuous modes, where the freeboard and internal recycling can be different.

The group and company of de Lasa et al. achieved great success by studying and commercializing fluidized bed Berty reactors, the so-called CREC riser simulator. This system requires an impeller to be placed on the top of the reactor and to have a very high rotating speed to enable the fluidization of the catalyst bed. The design resembles the first homemade recycle reactor manufactured by Union Carbide Corp. in 1965. Both experimental and simulation approaches proved good mixing and particle fluidizations under certain conditions, for which the reactor was employed in the catalyst testing and kinetic modeling of catalytic reactions of high industrial interests, such as fluid catalyst cracking (FCC), oxidative coupling of methane (OCM), and steam reforming in the batch mode.

The more common application of Berty reactors is in continuous flow experiments, where reactors generally exhibit perfect mixing or behave as a continuously stirred tank reactor (CSTR). Additionally, applications in batch operations have been described through the rapid removal of the products. Approaching perfect mixing eliminates the gradients of heat and mass across the reactor and enhances turbulence in the system, which typically reduces the impact of mass and heat transfer limitations. Consequently, the flow hydrodynamics and mixing patterns in Berty reactors across batch/continuous or packed/fluidized bed modes under different operating conditions are worth studying for system enhancements.

A perfect candidate for planning such detailed studies appears to be the Integrated Lab Solutions (ILS) Berty reactor, which is based on a design patented by the University of Erlangen, Germany. This reactor can provide an ideal internal circulation for the perfect mixing of gas/vapor with packed bed particles in the continuous mode, for which industrial, heterogeneously catalyzed reactions can be simulated from the laboratory scale with a small amount of catalysts. The adoption of a magnetic stirrer in this reactor avoids leakages and extends the lifetime of the reactor's equipment. Simultaneously, the reactor allows catalyst screening at high temperatures (up to 900 °C) and rotation rates (up to 10 000 rpm) for certain challenging chemistries (particularly close to ambient pressures) and enables controlled contact times between reactants and catalysts. Such reactions can potentially be catalytic cracking, the valorization of bio-oil, steam reforming of hydrocarbon mixtures, and OCM.

Works on the hydrodynamics of Berty reactors are scarce. However, some of those works include experimental research on the performance estimations of Berty reactor impellers, the degree of mixing based on residence time distribution (RTD) calculations, the pressure drop across the bed under various physical parameters and volumes of catalyst particles, and a recent computational fluid dynamics (CFD) simulation of a concentration homogeneity assessment, which indicated the resemblance of ideal mixing behaviors at impeller frequencies higher than 6000 rpm for continuous operations. Although it was only a primary simulation without experimental validation, the conclusion is quite inspiring for the further thorough analysis and evaluation of Berty reactors, which are vital for exploiting suitable application potentials under different operational conditions, resulting in improved performance.
The fluid dynamic research on internal recycle reactors has been focused on geometries for SCBSTR and RPB. To study the blade/baffle effect on flow patterns, a CFD hydrodynamic investigation was validated with experimental data that was obtained using the particle image velocimetry (PIV) technology of an SCBSTR. The liquid flow field in an SCBSTR was modeled using CFD simulations, which showed the maximal velocity through the central zone of the vessel and the nonuniformity of mass transfer. A two-dimensional CFD framework was built to investigate the rotating speed and flow velocity effects on the micromixing performance of an RPB. The flow pattern in the RPB was investigated using a simplified section representing the whole geometry, which was capable of reproducing the main flow field characteristics. A three-dimensional (3D) simulation on the gas–liquid flow in an RPB was realized and verified through good agreements with experimental data, where the misdistribution phenomenon could be counteracted by increasing the rotating speed. A new porous media model was adopted for the Eulerian simulation of RPBs, and the predicted holdup distribution agreed well with experimental data and was further applied to the modeling of CO₂ absorption by coupling a novel liquid generation–elimination model. A 3D steady-state gas flow simulation in an RPB with randomly arranged spherical packing was validated to study the rotational speed and flow rate effects on flow behaviors, from which a semiempirical correlation was used to predict the dry pressure drop of the packing zone.

All of the above cases prove the efficiency and reliability of CFD simulations for better understanding the fundamental theories and practical applications of the rotating systems with packed/fluidized beds. In these cases, it is normal to assume the bed to behave as porous media and simplify the geometry into a slice model, significantly reducing computing demands and improving mesh quality. Although different turbulence models have been utilized, Boussinesq’s approximation of the k−ε and k−ω models effectively manifested the gradual transition from laminar flow regimes to turbulent flow regimes with relatively low computing demands and reasonable accuracy and stability.

In this work, we developed a computational strategy for understanding the hydrodynamics of an ILS Berty reactor using a slice model to represent the whole geometry. Using the species transport model, the multiple reference frame (MRF), and the SST k−ω turbulence model, and ANSYS Fluent, we performed a three-dimensional simulation, which was validated with experiments in the continuous mode. The verified simulation approach was further employed to study the hydrodynamic behaviors in the batch mode in terms of the concentration profiles and pressure and velocity distributions in the reactor by evaluating the effects of the transient injection, porosities, and rotation rates. On this basis, we proposed a set of predictive correlations for assessing the catalyst fluidization and contacting time under different porosities.

2. METHODS

2.1. Experimental Setup

As shown in Figure 2a, the ILS Berty reactor can be split into bottom (vessel) and top (lid) parts. The vessel contains threads, into which studs are screwed. The lid is placed onto these studs, much like a flange coupling, and tightened with nuts screwed onto the studs. The vessel contains the main reaction chamber, where a cylindrical basket acts as a separating wall between the center, where the catalyst basket is placed, and the outer fluid volume, where the recycle flow passes through. The impeller is located on top of the basket. The main inlet is located at the bottom of the vessel, and the outlet is located at the sidewall, slightly lower than the impeller. The lid contains the driveshaft, to which the impeller is connected via a bolt. It is noteworthy that a purge stream of inert gas enters the reactor between the driveshaft and the driveshaft housing and acts as a second inlet so as to prevent corrosive gases (like hydrogen and water) from reaching the driveshaft and damaging its components.

We conducted hydrodynamic testing experiments on the ILS Berty reactor in the continuous mode at the laboratories of the University of Erlangen by connecting the inlet and outlet to a three-way ball valve and a digital WIKA S10 pressure transmitter, and the experimental conditions are shown in Table 1. The probes are located directly below and above the catalyst basket, respectively. The basket is filled to the maximum capacity of the quartz particles to diminish the impacts of the porosity settings of the CFD simulation. The impeller is rotated to radially accelerate the fluid so as to create a recycle flow from the inlet to the outlet, which is therefore formed from the top along with the annular side of the reactor to the bottom. There, it is sucked into the catalyst bed, in which the catalytic particles are kept between two screens located at the top and bottom of the bed (where the Hastelloy mesh is adopted in this work), and finally reaches the impeller again. Each experiment is repeated three times to obtain the average values with standard deviations.
Table 1. Experimental Conditions in the Continuous Mode

<table>
<thead>
<tr>
<th>feed</th>
<th>flow rate (mL min(^{-1}))</th>
<th>temperature (°C)</th>
<th>pressure (bar g)</th>
<th>particle diameter (mm)</th>
<th>porosity</th>
<th>Hastelloy mesh bed</th>
<th>rotation rate (min(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>N(_2)</td>
<td>50</td>
<td>175</td>
<td>4</td>
<td>1.3</td>
<td>0.7</td>
<td>0.4</td>
<td>3500–10000</td>
</tr>
</tbody>
</table>

2.2. Simulation Model

2.2.1. Mathematical Model. ANSYS Fluent was used for the 3D, transient, and isothermal simulations in the ILS Berty reactor by adopting the multiple reference frame (MRF) model\(^{31}\) for the rotating domain based on a 60° slice model (1/6th of the reactor)\(^{52,64}\) that was established and meshed by the ANSYS Workbench\(^{32}\) for which rotationally periodic boundaries were adopted to mimic the whole reactor. The catalyst bed was assumed as a homogeneous porous zone\(^{57,63}\) where the permeability and inertial loss coefficient were calculated by the Gidaspow equation\(^{64,65}\) with uniform particle diameters and assigned porosity. With the porous zone setting of the catalytic bed, the calculation is simplified to a single gas phase. To capture the flow transition from laminar to turbulent regimes, the SST k–ω turbulent model with low-Reynold corrections\(^{63,66}\) was selected. With the species transport model, the gas concentration distributions could be monitored.

The basic governing equations\(^{61}\) for the continuity and momentum balances are shown in eqs 1 and 2, respectively.

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = S_m
\]  

(1)

where \(\rho\) is the density, \(\mathbf{u}\) is the velocity, \(t\) represents the time, and \(S_m\) denotes the other source items.

\[
\frac{\partial (\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \mathbf{u}) = -\nabla p + \nabla \cdot (\nabla \cdot \mathbf{F}) + \rho \mathbf{g} + \mathbf{F}
\]  

(2)

where \(p\) denotes the pressure, \(\mathbf{r}\) denotes the stress tensor, \(g\) is the gravitational acceleration, and \(F\) denotes the external body forces, which includes the porous media source in this work. As the porous media is modeled by the addition of a momentum source term to the standard fluid flow equations, it is shown in eq 3 in a homogeneous form, which is composed of two parts: a viscous loss term (Darcey, the first term on the right-hand side of eq 3), and an inertial loss term (the second term on the right-hand side of eq 3).

During modeling laminar flow through the porous media, the second term in eq 3 can be dropped, with the laminar zone frame activated in the fluid dialog box, which means the turbulent model (SST k–ω model in this work) can be turned off for this specific zone. In this work, we considered the turbulence through the bed by keeping the turbulent model and adding the inertial loss term, while the flow though the Hastelloy meshes is regarded as laminar.

\[
F_l = -\left(\frac{\mu}{\alpha} \mathbf{v}_i + C_3 \frac{1}{2} \rho \mathbf{v}_i \right)
\]  

(3)

where \(\mu\) is the viscosity and \(\alpha\) and \(C_3\) denote the permeability and inertial resistance factor, respectively, which can be calculated using the Gidaspow model\(^{64,65}\) through eqs 4 and 5.

\[
K_{gi} = 150 \frac{\epsilon_i(1 - \epsilon_i)}{\epsilon_i d_i^2} + 1.75 \frac{\rho \epsilon_i \mathbf{v}_i \mathbf{v}_i - \mathbf{v}_i^2}{d_i} \text{ if } \epsilon_i \leq 0.8
\]  

(4)

\[
K_{gi} = 3 \frac{C_3 \rho \epsilon_i \mathbf{v}_i \mathbf{v}_i - \mathbf{v}_i^2}{d_i^2} \text{ if } \epsilon_i > 0.8
\]  

(5)

where \(K\) is the interphase momentum exchange coefficient, \(\epsilon\) is the volume fraction, and \(d\) is the particle diameter or equivalent particle diameter for nonspheres. Also, the subscripts \(s\) and \(g\) represent the solid phase and gas phase, respectively, and \(C_3\) is the drag coefficient.

With the MRF approach, the calculation domain was divided into subdomains of the stationary and rotating zones, where the rotational fluid velocities could be transformed from the stationary frame to the moving frame by eqs 6–8.

\[
\mathbf{v}_r = \mathbf{v}_s - \mathbf{u}_i
\]  

(6)

\[
\mathbf{u}_i = \omega_i \times \mathbf{r}
\]  

(7)

\[
\omega_i = \omega_i \hat{a}
\]  

(8)

where \(\mathbf{v}_r\) is the relative velocity viewed from the moving frame, \(\mathbf{v}_s\) is the absolute velocity viewed from the stationary frame, \(\mathbf{u}_i\) is the velocity of the moving frame relative to the inertial reference frame, and \(\omega_i\) is the angular velocity with/without the direction vector \(\hat{a}\).

As one of the Reynolds-averaged Navier–Stokes turbulence models, which simplify the turbulence problem by two additional transport equations, the SST k–ω turbulent model\(^{63,66}\) calculated the turbulence kinetic energy \((k)\) and specific dissipation rate \((\omega)\) using eqs 9 and 10.

\[
\frac{\partial (\rho k)}{\partial t} + \nabla \cdot (\rho \mathbf{u} k) = \nabla \cdot \left( \frac{\mu}{\sigma_k} \nabla k \right) + G_k - Y_k + S_k + G_b
\]  

(9)

\[
\frac{\partial (\rho \omega)}{\partial t} + \nabla \cdot (\rho \mathbf{u} \omega) = \nabla \cdot \left( \frac{\mu}{\sigma_\omega} \nabla \omega \right) + G_\omega - Y_\omega + S_\omega + G_{\omega b}
\]  

(10)

where \(t\) denotes the time, \(\Gamma_1\) and \(\Gamma_\omega\) stand for the effective diffusivities of \(k\) and \(\omega\), respectively, \(G_k\) and \(G_\omega\) denote the generation of \(k\) and \(\omega\) due to the mean velocity gradients, respectively, \(Y_k\) and \(Y_\omega\) denote the dissipations of \(k\) and \(\omega\) due to turbulence, respectively, and \(C_\omega\) and \(G_{\omega b}\) account for the buoyancy terms.

The local mass fraction of each species, \(Y_s\), was solved by the convection–diffusion equation for the species using the species transport equation (eq 11).

\[
\frac{\partial (\rho Y_s)}{\partial t} + \nabla \cdot (\rho \mathbf{u} Y_s) = -\nabla \cdot \left( \frac{\epsilon}{\sigma_s} \nabla Y_s \right) + S_s
\]  

(11)

where \(J\) denotes the diffusion flux of the species.

2.2.2. Mesh Model. A 60° slice model (Figure 2b) was prepared using the ANSYS Workbench\(^{32}\) to represent the ILS Berty reactor by the periodic boundary settings of rotational symmetry, where the stationary recirculation part was divided into several zones with shared interiors for fluid exchange to better control the mesh quality. The interfaces were defined to simulate the velocity transform between the rotating and stationary zones.

Hexahedron meshes were generated for the fluid domains across the catalyst bed and Hastelloy meshes, while the patch confirming method of tetrahedrons was used for the other zones. By adjusting the element size, different mesh models with a maximum skewness within 0.95 were obtained to perform a mesh independence check. Steady-state simulations were conducted by three mesh models with mesh sizes of 111 363, 149 854 (Figure 2c), and 229 036, respectively. The inlet velocity was calculated from the feed flow rate, while the outlet pressure was set to 0 with the operating conditions shown in Table 2. With the porous zone assumptions for the Hastelloy meshes and the catalyst basket, the permeability and inertial resistance factor of the porous zones were determined accordingly. By comparing the velocity and pressure profiles along the central axis (marked by an arrow in Figure 2b), it is observed that the results of the model with 111 363 meshes deviate from the other two models, while the mesh model with the element size of 149 854 shows similar velocity and pressure distribution results to the model with 229 036 meshes with much fewer computation efforts needed. Thus, the 149 854 meshes model was selected for further batch/continuous or packed/fluidized bed...
simulations in consideration of the calculation efficiency and precision. For simplification, the locations of data points, surfaces, and volumes in this work adhere to the coordinate system shown in Figure 2b.

2.3. Simulation Procedures

2.3.1. Continuous Mode. Based on the same experimental operating conditions shown in Table 1 and the simulation strategy proposed in Section 2.2, the hydrodynamic behavior of the ILS Berty reactor under the continuous mode was studied with the settings shown in Table 3. The inlet velocity was calculated from the feed flow rate shown in Table 1, while the outlet was set as an outflow boundary for fast convergence since the mass balance between the inlet and outlet was difficult to reach as a result of the circulation under the pressure outlet boundary setting. In this regard, a transient single-phase simulation with porous zone simplifications for the bed and Hastelloy mesh was performed with the other settings in the default mode.

2.3.2. Batch Mode. To study the hydrodynamic performance of the ILS Berty reactor in the batch mode, we conducted simulations with the boundary conditions shown in Table 4 as a case study for crude to chemical applications. One of the key differences from the continuous mode is the boundary conditions: for batch mode, the outlet was switched to “wall boundary”.

To keep consistency with experiments, the simulation procedures were as follows:

(1) Initialization of calculation: inert gas patched to the entire domain
(2) First round simulation with the impeller at the desired rotation rate until stable hydrodynamics were achieved (10 s)
(3) Pulse injection of the feed mixture at a certain injection time
(4) Second round of simulations with the dynamic steady state.

The physical properties of the feed and the inert gas were simulated from the Aspen HYSYS database, where the mixture of the Arabian light petroleum assay and water was regarded as the feed, while argon was used as the inert gas. For simplification, the single gas phase of the isothermal condition was considered, where the liquid feed at the ambient condition was converted into the vapor phase for the simulation conditions (Table 4) by mass balance to obtain the physical properties shown in Table 5. Since no reactions were involved in this work, monitoring of the Arabian light concentration was done by studying the species transport phenomenon in the reactor, so it was named as tracer for the subsequent content.

2.4. Impeller Relationship

According to the previous experimental investigations, the pressure drop occurring over the catalyst basket was directly correlated with the pressure generated by the impeller, which is a function of the rotation speed and fluid density. Thus, the impeller relationship of the Berty reactor could be expressed as a semipirical expression in eq 12, offering a quantitative way for describing the hydrodynamic behaviors using the monitored/measured parameters, such as the fluid density, pressure, and impeller rotation rate. Through the impeller relationship, we not only verified the modeling strategy through the comparisons between the simulated and experimental results but also better controlled the system.

\[
\frac{\Delta p}{p} = f(\eta^2) = B + A\eta^2
\]  

(12)

where \(\Delta p\) is the pressure drop obtained by either experiments or simulations, \(p\) is the fluid density, \(\eta\) is the rotation rate, \(A\) is the slope constant, which is named as the impeller constant of the Berty reactor, and \(B\) is the intercept constant.

2.5. Mass Balance Correction

With the settings of constant properties, one of the limitations for the batch mode simulation lies in the total mass loss during the injection of feed mixture, the density of which is less than argon at the gas phase. The volume-weighted mixing law leads to a lower mixture density after the feed injection, resulting in a mass decrease with the fixed reactor volume. Although the pressure increase and density change during the injection can be captured by the ideal gas law for compressible flow, the petroleum assay of Arabian light will be regarded as a single component by calculating the density with a certain molecular weight, which is not true. Thus, we adopted the specific properties (Table 5) at the reaction condition for investigating the fluid dynamic characteristics. By combining the mass of the initial Ar with the injected feed mixture, the final concentration of each component can be calculated. In this way, the physical properties of the mixture can be corrected under the mass balance calculation. Based on the impeller relationship expressed in Section 2.4, the pressure drop could be amended accordingly.

---

**Table 2. Simulation Conditions and Settings in Mesh Independence Check**

<table>
<thead>
<tr>
<th>conditions</th>
<th>boundary and zone settings</th>
</tr>
</thead>
<tbody>
<tr>
<td>feed</td>
<td>N&lt;sub&gt;2&lt;/sub&gt; inlet velocity inlet</td>
</tr>
<tr>
<td>flow rate (mL min&lt;sup&gt;−1&lt;/sup&gt;)</td>
<td>50 outlet pressure outlet</td>
</tr>
<tr>
<td>temperature (°C)</td>
<td>175 turbulent model SST k→ω</td>
</tr>
<tr>
<td>pressure (bar g)</td>
<td>4 energy model no</td>
</tr>
<tr>
<td>particle diameter (μm)</td>
<td>1.3 species transport no</td>
</tr>
<tr>
<td>Hastelloy mesh porosity</td>
<td>0.7 transient state no</td>
</tr>
<tr>
<td>bed porosity</td>
<td>0.4 multiphase no</td>
</tr>
<tr>
<td>rotation rate (min&lt;sup&gt;−1&lt;/sup&gt;)</td>
<td>5000</td>
</tr>
</tbody>
</table>

---

**Table 4. Simulation Conditions and Settings in the Batch Mode**

<table>
<thead>
<tr>
<th>conditions</th>
<th>boundary and zone settings</th>
</tr>
</thead>
<tbody>
<tr>
<td>feed volume (μL)</td>
<td>200 inlet velocity inlet</td>
</tr>
<tr>
<td>temperature (°C)</td>
<td>700 outlet NA, change to wall</td>
</tr>
<tr>
<td>pressure (bar)</td>
<td>1 turbulent model SST k→ω</td>
</tr>
<tr>
<td>particle diameter (μm)</td>
<td>66 energy model no</td>
</tr>
<tr>
<td>particle density (kg m&lt;sup&gt;−3&lt;/sup&gt;)</td>
<td>1500 species transport yes</td>
</tr>
<tr>
<td>Hastelloy mesh porosity</td>
<td>0.7 mixture density volume-weighted mixing law</td>
</tr>
<tr>
<td>bed porosity</td>
<td>[0.47, 0.8] transient state no</td>
</tr>
<tr>
<td>rotation rate (min&lt;sup&gt;−1&lt;/sup&gt;)</td>
<td>4000–10000 multiphase no</td>
</tr>
<tr>
<td>injection time (s)</td>
<td>0.5</td>
</tr>
</tbody>
</table>

"The regime from the packed bed to turbulent fluidization was assumed. With the velocity inlet setting for the batch mode, the injection time was relevant." Batch operation. Isothermal.

---

**Table 3. Simulation Conditions and Settings in the Continuous Mode**

<table>
<thead>
<tr>
<th>inlet</th>
<th>outlet</th>
<th>turbulent model</th>
<th>energy model</th>
<th>species transport</th>
<th>transient state</th>
<th>multiphase</th>
</tr>
</thead>
<tbody>
<tr>
<td>velocity inlet&lt;sup&gt;a&lt;/sup&gt;</td>
<td>outflow&lt;sup&gt;a&lt;/sup&gt;</td>
<td>SST k→ω</td>
<td>no&lt;sup&gt;b&lt;/sup&gt;</td>
<td>no&lt;sup&gt;d&lt;/sup&gt;</td>
<td>yes</td>
<td>no</td>
</tr>
</tbody>
</table>

<sup>a</sup>Calculated from Table 1. <sup>b</sup>For mass balance. <sup>c</sup>Isothermal. <sup>d</sup>Single species.
Table 5. Physical Properties of the Feed and the Inert Gas from the HYSYS Database

<table>
<thead>
<tr>
<th>gas</th>
<th>density (kg m$^{-3}$)</th>
<th>heat capacity (kJ kg$^{-1}$ K$^{-1}$)</th>
<th>thermal conductivity (W m$^{-1}$ K$^{-1}$)</th>
<th>viscosity (cP)</th>
<th>molecular weight (g mol$^{-1}$)</th>
<th>note</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arabian light</td>
<td>2.61</td>
<td>3.32</td>
<td>$6.39 \times 10^{-2}$</td>
<td>$4.96 \times 10^{-3}$</td>
<td>209.9</td>
<td>feed mixture</td>
</tr>
<tr>
<td>water-vapor</td>
<td>0.223</td>
<td>2.28</td>
<td>$9.42 \times 10^{-2}$</td>
<td>$3.65 \times 10^{-2}$</td>
<td>18.02</td>
<td></td>
</tr>
<tr>
<td>feed mixture</td>
<td>0.410</td>
<td>2.795</td>
<td>$8.11 \times 10^{-2}$</td>
<td>$1.67 \times 10^{-2}$</td>
<td>33.18</td>
<td></td>
</tr>
<tr>
<td>argon</td>
<td>0.494</td>
<td>0.521</td>
<td>$4.17 \times 10^{-2}$</td>
<td>$5.70 \times 10^{-2}$</td>
<td>39.95</td>
<td>inert gas</td>
</tr>
</tbody>
</table>

3. RESULTS

3.1. Hydrodynamics in the Continuous Mode

3.1.1. Experimental Validation. We conducted the single-phase CFD simulation of the ILS Berty reactor based on the continuous operating conditions shown in Tables 1 and 3. With the semiempirical expression shown in eq 12, the hydrodynamic performance could be obtained at the macroscopic scale through both experimental and simulation approaches, resulting in convenience for the validation of the complex physics. The experimental relationship between the ratios of the pressure drop to gas density under various rotation rates is illustrated in Figure 3, which also includes the simulated results under identical conditions based on the strategy detailed in Section 2.

The impeller constant from the simulation ($1.543 \times 10^{-6}$ Pa min$^2$ m$^3$ kg$^{-1}$) is in close agreement with that obtained from the experiments ($1.556 \times 10^{-6}$ Pa min$^2$ m$^3$ kg$^{-1}$) regardless of the nonhomogeneous porosity of the particles in the experiments, which verifies the reliability of the simulation approach in terms of both the chosen sectional geometry and the numerical framework of the computational model. Consequently, this modeling strategy can be further used in studying hydrodynamics under various operational conditions and other procedures as in the batch mode.

3.1.2. Flow Behaviors in Continuous Packed Bed Operations. By taking one representative surface from the entire revolving geometry, the pressure and velocity contours of the fluid domain under the rotation rates of 5000, 6000, 8000, and 10 000 rpm at the dynamic steady state in the continuous packed bed mode were reported, as shown in Figure 4. To better display the pressure distribution, the relative static pressure to the operating condition was demonstrated.

Figure 4 shows that with an increase in rotation rate, the pressure in the fluid domain accordingly increased. Similar pressure distributions were observed for all of the conditions with the lowest pressure at the outlet and the major pressure drops across the porous zone of the catalyst bed and the Hastelloy mesh in the stationary domain. Consequently, the outlet pressure needed to be well adjusted for product collection during the experimental operations. Although the major pressure drop occurred at the porous zone, the contours line was not horizontally distributed as a result of the continuous flow. Fortunately, with the regular spacing between the contiguos contour lines, the simulation assumption for the uniform porosity setting of the packed beds still made sense, while a further simulation strategy needs to be developed for nonpacked beds in the continuous operating modes. As shown in Figure 4, the impeller center and a sharp corner of the blades witnessed a lower pressure in the rotating zone than in the rest of the reactor, for which fluid can easily be stored with low velocity, indicating potential optimization directions for the impeller geometry.

There is no doubt that the velocity increased with the rotation rate because of the increased momentum received from the impeller. However, the rotation rate had no obvious effects on the velocity distribution of the fluid domain, as shown in Figure 4. The spinning velocity of the impeller was well transferred to the adjacent stationary zone, which attenuated along the circulation zone to flow through the catalyst bed from the bottom, where the velocity was kept relatively low. Regardless of the increase in the rotation rate, the fluid velocity across the bed was extremely low compared with the other zones, demonstrating a relatively high loss of momentum in the current design.

According to the velocity contours, the velocity in the bed was almost uniform across the catalytic bed, which was further studied by comparing the area-averaged velocities of the bottom and top surfaces with the mass-averaged velocity of the entire catalytic bed zone, as shown in Figure 5. Also, Figure 5 shows that similar velocities at the surfaces were obtained as the mass-averaged bed velocity under each rotation rate, from which the uniform bed velocity reflected a possible ideal mixing behavior in the reaction zone. An important hydrodynamic behavior of the ILS Berty reactor in the continuous mode was observed with regard to the linear bed velocity and the rotation rate, indicating that the momentum generated by the impeller is directly applied to the bed under continuous operating circumstances at the dynamic steady state since the rotating linear velocity of the impeller was also in a linear relationship with the rotation rate.

3.2. Hydrodynamics in the Batch Mode

With the validated simulation approach, we investigated the hydrodynamic performance of the ILS Berty reactor with packed/fluidized beds in the batch mode under various...
operating conditions (Tables 4 and 5), for which the simulation procedures can be found in Section 2.3.2.

3.2.1. Transient Injection in Packed Bed Operations.

One of the key characteristic differences between the continuous and batch modes is the fluid disturbance caused by the feed injection. With a constant feed volume, the injection velocity was determined by the injecting time, which could be controlled during the experiments using the ILS injecting system. In consideration of the pressure increase caused by feed injection and reaction, a pulse injection of 0.5 s was adopted for studying the transient phenomenon. The tracer fraction contours of the fluid domain after pulse injection under the rotation rates of 4000, 6000, 8000, and 10 000 rpm are displayed in Figure 6 by taking one representative surface from the entire revolving geometry. A similar concentration distribution was observed for different rotation rates, with the fact that the entire reactor is almost filled with the tracer right after the injection except for the catalytic bed. With the porous media setting of the bed, it is easier for the tracer to flow along the opposite direction of the recycle flow than through the bed. To have a better understanding of this transient injection, the tracer fractions at the bottom surface, top surface, and entire bed were plotted, as shown in Figure 7, where the impeller rotation rates were kept at 4000, 6000, 8000, and 10 000 rpm with the fully packed catalyst in the bed.

Figure 7 displays the F curves, or area-averaged oil fraction (normalized) over time, for the different cases studied before. As seen in these results, the same trends were observed for different rotation rates for the bottom and top surfaces of the catalyst bed. A sharp peak existed at the bottom right after the

Figure 4. Pressure and velocity contours at the dynamic steady state for the continuous packed bed operations: (a and e) rotation rate, 5000 min$^{-1}$; (b and f) rotation rate, 6000 min$^{-1}$; (c and g) rotation rate, 8000 min$^{-1}$; and (d and h) rotation rate, 10 000 min$^{-1}$. All of the contours are displayed at the y = 0 surface, and the simulation conditions are shown in Tables 1 and 3.

Figure 5. Averaged velocity at the bottom and top surfaces along the Z direction of the bed and the entire volume of the bed for continuous packed bed operations under different rotation rates. The simulation conditions are shown in Tables 1 and 3.
feed injection, where the tracer arrived at the catalyst bed almost instantaneously, but the movement was retarded by the packed catalysts; thus, the tracer flows toward the other directions, leading to a short decrease of the tracer fraction at the bottom surface. With the recycle effects of the impeller, the system gradually reached the dynamic steady state with the following increase in the tracer fraction. In this regard, the tracer could be monitored at the top just after the injection, while the increase to the final state was a combined result of the recycle flow generated by the impeller and the fluid flow through the bed with a gradual increase in the tracer fraction. The dynamic steadiness of the system is a tradeoff of the injected disturbance, the rotating momentum, with the impeded fluid flow through the catalytic bed. In addition, the profiles of the $F$ curves show that the reactor approaches perfect mixing conditions or, in other words, the residence time distribution (RTD) of the tracer is like a CSTR.

Figure 7 shows that it took almost 45 s for the system to reach another dynamic steady state after the injection even at a relatively low rotation rate of 4000 rpm, less than the single-round contact time (calculated in Section 3.2.2) between the tracer and the catalysts. The system witnessed a short unstable period in the reactor inlet (bottom the bed) at the beginning of the pulse injection ($t < 10$ s). However, this instability quickly led to a perfect back-mixing behavior, and the stabilization of the tracer concentration across the bed, which occurs after $\sim 40$ s at 4000 rpm and after $\sim 30$ s at 10 000 rpm.

3.2.2. Flow Behaviors in Batch Packed Bed Operations. By taking one representative surface from the entire revolving geometry, the pressure and velocity contours of the fluid domain under the rotation rates of 4000, 6000, 8000, and 10 000 rpm at the dynamic steady state with the packed bed in the batch mode were demonstrated, as shown in Figure 8. To better display the pressure distribution, the relative static pressure to the operating condition was shown. Figure 8 shows analogous hydrodynamic behaviors for the augmented rotating speeds with increasing pressures. The low-pressure part in the rotating zone decreased accordingly, indicating the reduction of the dead volume in the spinning part, as verified by Figure 8. Since the stationary part is of more concern for this study, Figure 8 shows that the major pressure drop took place at the porous zones across the Hastelloy meshes and the catalyst bed in between, where the lowest pressure was located at the top of the Hastelloy meshes, demonstrating the necessity for a vacuum product collector at the outlet to prevent further flow to the rotating zone. Unlike the continuous mode, the contours lines were almost horizontally distributed across the porous zone with regular spacing, indicating that the simulation assumption of the uniform porosity setting was quite reasonable for the batch mode, so the simulation strategy can be used for investigations under different porosity conditions in batch operations.

Figure 8 demonstrates that the velocity increased with an increase in the rotation rate as a result of an increase in the momentum received from the impeller, but the rotation rate
had no obvious effects on the velocity distribution of the fluid domain, especially for the stationary part, where the velocity across the bed was relatively low, even in most of the stationary zone compared with the rotating linear velocity generated by the impeller, showing a dramatic momentum loss in the circulation part without any velocity contour lines.

The area-averaged velocities of the bottom and top surfaces and the mass-averaged velocity of the entire catalyst bed zone under the batch packed bed mode are shown in Figure 9. The same phenomena as the continuous mode of the uniform bed velocity under each rotation rate appeared, which reflected the ideal mixing behavior in the catalyst bed for both conditions. However, the bed velocity was in a quadratic polynomial relationship with the rotation rate.

3.2.3. Effects of the Operating Parameters. Under the operating conditions exhibited in Table 4, the minimum fluidization velocity was 0.24 cm s\(^{-1}\) with the simulated mixture as the stripping gas,\(^{67}\) which is higher than the fluid velocities in the catalyst bed, as shown in Figure 9. However, fluidization may occur with a smaller catalyst mass of higher porosities in the catalytic packed bed. As concluded from Figure 8, the uniform horizontally distributed pressure and velocity across the porous zone under the batch mode allowed further hydrodynamic investigations on the effect of operating conditions and porosities, for which the simulations were performed based on the same conditions shown in Tables 4 and 5.

The average velocities in the bed are shown in Figure 10, from which the same quadratic polynomial relationships were discovered. The bed velocities under the rotation rate of 4000–10 000 for the bed with the porosity of 0.8, as well as

Figure 8. Pressure and velocity contours at the dynamic steady state for the batch packed bed operations: (a and e) rotation rate, 4000 min\(^{-1}\); (b and f) rotation rate, 6000 min\(^{-1}\); (c and g) rotation rate, 8000 min\(^{-1}\); and (d and h) rotation rate, 10 000 min\(^{-1}\). All of the contours are displayed at the \(y = 0\) surface, and the simulation conditions are shown in Tables 4 and 5.

Figure 9. Averaged velocity at the bottom and top surfaces along the \(Z\) direction of the bed and the entire volume of the bed for the batch packed bed operations under different rotation rates with the simulation conditions in Tables 4 and 5.
under various operating porosities are almost identical, which agrees with the experimental conclusion that the generated pressure drop is independent of the catalyst quantities, suggesting fewer catalysts and a higher rotation rate for fluidization applications. A nominal difference appears between Figure 11 and the validation case in Figure 3, demonstrating that the reactor hydrodynamic characteristics are also related to the particle properties. As the ratio of the pressure drop to mixture density could be estimated by the rotation rates, a larger inert density is recommended for severe fluidization with higher pressure drops and fluid velocities. In such conditions, argon generates a larger velocity in the catalytic bed than nitrogen with a higher density. In addition to that, the minimum fluidization velocity with the stripping gas of argon is less than nitrogen as a result of the higher viscosity under the same operating conditions. In this regard, argon is better than nitrogen for fluidization in the Berty reactor.

With the top impeller, the hydrodynamic performance of the ILS Berty reactor could be estimated by the impeller relationships for both the packed and fluidized beds, where the pressure drop is mainly relevant to the physical properties of the feed gas and catalyst particles. Also, the rotation rates differ from the typical pressure drop profiles of the packed (linear with velocity) and fluidized beds (invariant after minimum fluidization velocity) in the riser without continuously mixed momentum generated by the impeller. Since most of the pressure drop between the monitored experimental points took place at the porous zones, where the gas–particle interaction in the bed is described by the Gidaspow equation in eqs 4 and 5 and the pressure drop through the Hastelloy meshes can be calculated by eq 3, the most significant hydrodynamic characteristics in the ILS Berty reactor can be assessed by eqs 13–15.

\[
\frac{\Delta P_{\text{total}}}{\rho_g} = -5.738 + 1.758 \times 10^{-6} \eta^2 
\]

\[
\frac{\Delta P_{\text{basket}}}{H_{\text{basket}}} = 150 \left( 1 - \varepsilon_g \right)^2 \frac{\mu \nu \rho_g}{\varepsilon_g \nu_s} + 1.75 \frac{(1 - \varepsilon_g) \rho \nu^2}{\varepsilon_g \rho_s} 
\]

\[
\frac{\Delta P_{\text{mesh}}}{H_{\text{mesh}}} = \frac{\mu_k}{\alpha \rho_g} 
\]

By substituting the physical parameters with the data from Table 4, the predicted bed velocity could be obtained as a simplification of the porosity and rotation rate by summing the monitored pressure drop contributions of eqs 13–15. In this regard, the estimated bed velocity and contacting time profile were illustrated, as shown in Figure 12, for different porosities, which agrees well with the simulated results obtained with the maximum absolute relative deviations within 10%, thus verifying the reliability of representing the complex hydrodynamics of the ILS Berty reactor through simple equations. This way, we could obtain the bed velocity in the catalytic bed without running any experiments, evaluate the prospects of fluidization (given certain particles), and enhance the reactor performance by adjusting the operating conditions, such as the rotation rates and particle porosities. Although the obtained results in the section are based on the physical properties in Table 4, the methodology can be applied to other cases with different feed and particle properties.
4. DISCUSSION

By comparing the impeller relationship between the experiments and simulations, the simulation strategy was validated in the continuous packed bed mode, from which the hydrodynamic behaviors of the pressure and velocity distributions were obtained. As a result of the continuous flow, the pressure contours line was not horizontally distributed, which limits the application of this simulation strategy in fluidization investigations, where the assumption of uniform porosities by regarding catalyst beds as porous media is not accurate enough. With the lowest pressure at the outlet, which is suggested to be well adjusted and maintained during experiments, and the relatively low catalyst bed velocity caused by the momentum loss in the circulating zone, the uniform velocities were obtained for the catalyst bed, which is in a linear relationship with the rotation rate. Further geometry modifications should be conducted to better utilize the momentum generated by the impeller.

With the almost horizontally distributed contours lines across the porous zone with regular spacing, the simulation assumption of the uniform porosity setting is quite reasonable for the batch mode, so the hydrodynamic behaviors of both the packed and fluidized beds were studied to propose predictive equations for evaluating the bed velocity. With the lowest pressure located at the top of the Hastelloy meshes, a vacuum product collector at the outlet is suggested to prevent further flow to the rotating zone. A dramatic momentum loss in the circulation part was also observed, as the velocity across the catalyst bed even in most of the stationary zone was relatively low compared with the rotating linear velocity generated by the impeller. A uniform bed velocity under different rotation rates appeared with the quadratic polynomial relationship with the rotation rate, indicating an almost perfect mixing in the catalyst bed. These results together with our residence time distribution (RTD) simulations led to conclude that the reactor approaches perfect mixing behavior, and it is free of gradients across the bed after 30–40 s, depending on the rotating speed. Thus, the system can be well operated for the batch packed or fluidized bed mode through the simple control of the rotation rate and solid loading.

Our work adopted the isothermal and nonreactive assumption, so the gradient in the reactor lies in velocity and pressure. Based on our RTD results, the contacting pattern of the reactor was defined as “perfectly mixed”. This pattern eases the interpretation of hydrodynamic behavior and makes this reactor attractive for kinetic studies, particularly in “fast” reactions occurring in the second scale. The relatively high linear velocities attainable in the reactor ensure a more turbulent regime in the system, and thus, for a given kinetic application, it can decrease the mass and heat transfer resistances.

The continuous mode enables the reactive testing to be conducted in conditions where the reactor-scale transport limitations are minimal. During the batch mode, due to the high internal recycle flow, the external mass and heat transport limitations can be diminished. For any prospective kinetic application, a more thorough study is needed to guarantee the intrinsic kinetic operation.

From an application point of view, the ILS Berty reactor finds a niche in areas involving fast chemistries that are more prone to suffer from heat or mass transfer limitations in conventional chemical reactors. From the case study of the
crude to chemical application in the batch mode in Section 3.2, the single-round contact time at a porosity of 0.8 between the catalysts (~3.75 g) and feed (200 µL) in the bed was about 3 s at a rotation rate of 5500 rpm, which can reproduce the conditions of industrial riser reactors \(^3\) for crude oil or vacuum gas oil catalytic cracking. In fact, most of the viable reactions involving methane, which are challenging, can be potential candidates for the ILS Berty reactor due to their combination of high temperatures and relatively short contact times by adjusting the rotation rates and catalyst weights. Another assumption for the simulation was the homogeneous porous media setting of the bed. This regime can be another operation pattern of the so-called “packed fluidization” for this reactor with smaller particles in the interstices of a bed of larger stationary particles. The benefit of this strategy is that the fluidization is more homogeneous, slug-free, and with lower operating costs. With the model established in this work, the application of the packed fluidization mode of the Berty reactor can also be investigated by several layers of porous media setting for the bed with different particle sizes, permeabilities, and inertial resistance factors by simulations after validating with the visualization experiments.

5. CONCLUSIONS

In this work, to study the hydrodynamic characteristics of an ILS Berty reactor, the CFD approach was adopted using a slice model to represent a three-dimensional symmetric geometry with porous zone settings for the catalyst bed, coupled with the species transport model, multiple reference frame, and SST \(k−\omega\) turbulence model. The effectiveness of the computational strategy was verified by comparing the impeller relationship with the experimental results. Then, the model was applied to the continuous packed and batch packed/fluidized bed modes.

The hydrodynamic behaviors in both the continuous and batch modes indicated a uniform ideal mixing behavior in the catalytic bed, while the relationship of the velocity with the rotation rate was different: linear for continuous flow and quadratic polynomial during batch operations. This is convenient for mathematical modeling and provides an easier interpretation of the hydrodynamic behavior.

For batch operations, by analyzing the transient injection under various rotation rates, the system turned to be stable in the initial 30–40 s, depending on the rotating speed. An initial instability occurs in the reactor inlet, but this effect turns out to be negligible considering its null effects on the catalytic bed. The residence time distributions indicate that the reactor behavior approaches the perfectly mixed regime.

For continuous operations, internally circulating beds can increase the linear velocities at the same overall flow rates compared with non-circulating one-through reactors, leading to a more turbulent regime across the catalytic bed and the minimization of transport resistances.

The effects of the porosities and rotation rates on the bed velocity and pressure drop were investigated, from which particle fluidizations appeared under certain conditions. The major pressure drop of the stationary part took place at the porous zones across the Hastelloy meshes and the catalyst bed in between, which is independent of the catalyst quantities validated by the experimental observations and different from the other packed/fluidized bed behaviors without continuous mixing. In this regard, a correlation for predicting hydrodynamic behaviors was proposed to calculate the bed velocity and assess the contacting time under different porosities, thus simplifying the fluidization operations and performance enhancements. However, a momentum loss was observed for the stationary recirculation zone for both the continuous and batch modes, for which further geometry modification and optimization are required.

■ AUTHOR INFORMATION

Corresponding Author

Pedro Castaño — Multiscale Reaction Engineering, KAUST Catalysis Center (KCC), King Abdullah University of Science and Technology (KAUST), 23955-6900 Thuwal, Saudi Arabia; orcid.org/0000-0002-6454-9321; Email: pedro.castaño@kaust.edu.sa

Authors

Mengmeng Cui — Multiscale Reaction Engineering, KAUST Catalysis Center (KCC), King Abdullah University of Science and Technology (KAUST), 23955-6900 Thuwal, Saudi Arabia; School of Petroleum and Natural Gas Engineering, Southwest Petroleum University, 610500 Chengdu, China
Shekhar R. Kulkarni — Multiscale Reaction Engineering, KAUST Catalysis Center (KCC), King Abdullah University of Science and Technology (KAUST), 23955-6900 Thuwal, Saudi Arabia; Email: shekhar.kulkarni@kaust.edu.sa
Stefan Wagner — ILS—Integrated Lab Solutions GmbH, 12489 Berlin, Germany
Claudia Berger-Karin — ILS—Integrated Lab Solutions GmbH, 12489 Berlin, Germany
Anton Nagy — ILS—Integrated Lab Solutions GmbH, 12489 Berlin, Germany

Complete contact information is available at: https://pubs.acs.org/10.1021/acsengineeringau.1c00026

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

Funding for this work was provided by King Abdullah University of Science and Technology (KAUST).

■ NOMENCLATURE

\(C_s\) inertial resistance factor \((m^{-1})\)
\(C_d\) drag coefficient
\(d\) diameter \((m)\)
\(F\) external body forces \((Pa\ m^{-1})\)
\(g\) gravitational acceleration \((m\ s^{-2})\)
\(H\) height \((m)\)
\(k\) turbulence kinetic energy \((m^2\ s^{-2})\)
\(K\) interphase momentum exchange coefficient \((kg\ m^{-3}\ s^{-1})\)
\(p\) pressure \((Pa)\)
\(t\) time \((s)\)
\(v\) velocity \((m\ s^{-1})\)

■ SYMBOLS

\(\alpha\) permeability \((m^2)\)
\(\varepsilon\) volume fraction
\(\eta\) rotation rate \((min^{-1})\)
\(\mu\) viscosity \((Pa\ s)\)
\(\rho\) density \((kg\ m^{-3})\)
\(\omega\) specific dissipation rate
\(\omega_i\) angular velocity \((s^{-1})\)
REFERENCES


