Electronic Supplementary Material (ESI) for Reaction Chemistry & Engineering.
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Supplementary Information

Characterization of Single-Phase Flow Hydrodynamics in Berty Reactor using Computational Fluid Dynamics (CFD)

Khunnawat Ountaksinkul, Sirada Sripinun, Panut Bumphenkiattikul, Surapon Bubphacharoen, Arthit Vongachariya, Amnart Jantharasuk, Piyasan Praserthdam, and Suttichai Assabumrungrat*

Experimental Set-up

Table S1: Operating condition of the gas-chromatography.

Gas Chromatography	**Agilent 7820A**
Detector	TCD
Capillary column	Fuse Silica
Carrier gas	Nitrogen
Carrier gas flow rate (mL/min)	1.5
Injector temperature (K)	423.15
Detector temperature (K)	523.15
Initial column temperature (K)	333.15
Initial holding time (min)	1
Secondary column temperature (K)	453.15
Secondary holding time (min)	180
Programmed rate (K /min)	288.15
Analyzed gas	Helium

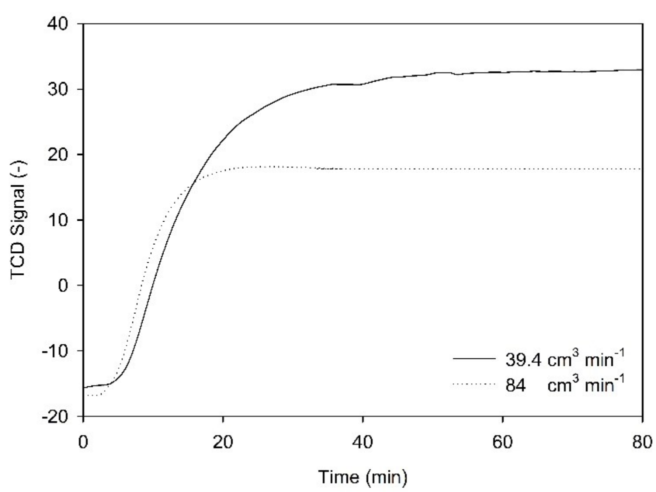
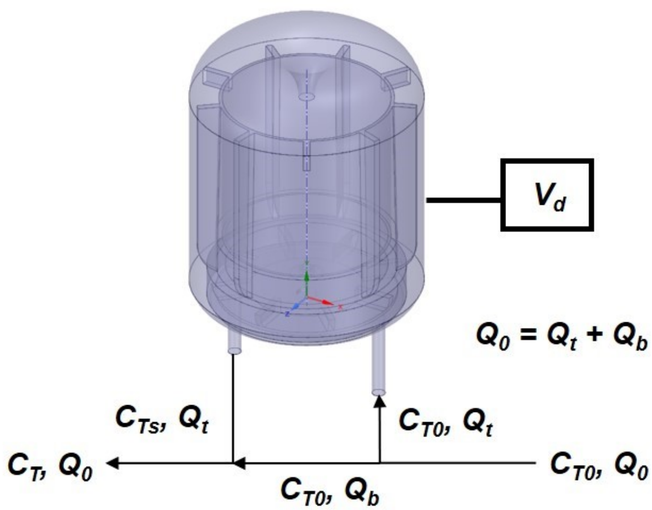


Figure S1: Raw data of TCD signals under the temperature 35 °C, atmospheric pressure, the total flow rates of 39.4 and 84 cm^3 min^{-1}, and impeller speed of 1896 rpm.

Figure S1 shows the TCD signals obtained from the gas chromatography (Agilent 7820A) under the temperature 35 °C, atmospheric pressure, the total flow rates of 39.4 and 84 cm^3 min^{-1}, and impeller s of 1896 rpm. At time = 0 min, it represents the point that He tracer enters the Berty reactor. After tracer experiments, the TCD signals were normalized to provide the F-curves as shown in Figure S4 (experimental data without deconvolution).



Non-ideal CSTR Reactor Model with Dead Volume and Bypassing

Figure S2: Real Berty reactor modelled by a single CSTR with dead volume and short-circuiting.[1]

The non-ideal CSTR model was generated with the combination of active volume (V_t) and dead volume (V_d) for step-input tracer experiment. The balance in non-stationary regime of tracer in the volume of reactor (V_t) was written as.

$$Q_t \cdot C_{T0} - Q_t \cdot C_{Ts} = V_t \frac{dC_{Ts}}{dt} \quad (S1)$$

where Q_t is the volumetric flow rate of non-bypassing flow (m^3 s^{-1}). C_{T0} is the steady input tracer concentration (mol m^{-3}) and C_{Ts} is the tracer concentration without the effect of bypassing (mol m^{-3}). Note that the boundary conditions for positive step-input can be defined as:

$$t < 0 \rightarrow C_T = 0 \quad (S2)$$

$$t \geq 0 \rightarrow C_T = C_{T0} \quad (S3)$$

where t is time (s). C_T is the exit concentration (mol m^{-3}). Then, the tracer balance at the point of union of both streams can be given below:

$$C_T = \frac{Q_b \cdot C_{T0} + Q_t \cdot C_{Ts}}{Q_0} \quad (S4)$$

where Q_b is the bypassing volumetric flow rate (m^3 s^{-1}). Q_0 is the total volumetric flow rate (m^3 s^{-1}). If we define:

$$\alpha = \frac{V_t}{V},\ \beta = \frac{Q_b}{Q_0},\ \text{and}\ \bar{t} = \frac{V}{Q_0} \quad (S5)$$

Alpha (α) is a fraction of non-stationary regime (V_t) to total volume (V), whereas Beta (β) is a fraction of short-circuiting or bypassing (Q_b) to total volumetric flow rate (Q_0). Last, the average residence time ($\bar{t}$) was calculated by total volume (V) divided by total volumetric flow rate (Q_0). Integrating and replacing in the expression of tracer balance in the reactor, the equation of tracer concentration leaving the system can be obtained as shown in Equation S6.

$$F(t) = \left[\frac{C_T}{C_{T0}}\right] = 1 - (1 - \beta) \cdot exp\left[-\frac{1 - \beta}{\alpha}\left(\frac{t}{\bar{t}}\right)\right] \quad (S6)$$

The E-curve of this system can be derived from “F-curve” and written in Equation S7. For ideal behavior, the α and β are equal to 1 and 0, respectively. In this research, Equation S7 was used with the convolution integral method to validate the experimental data. Moreover, this equation was also used together with CFD modeling for parameter estimation to determine the dead volume and bypassing in the Berty reactor.

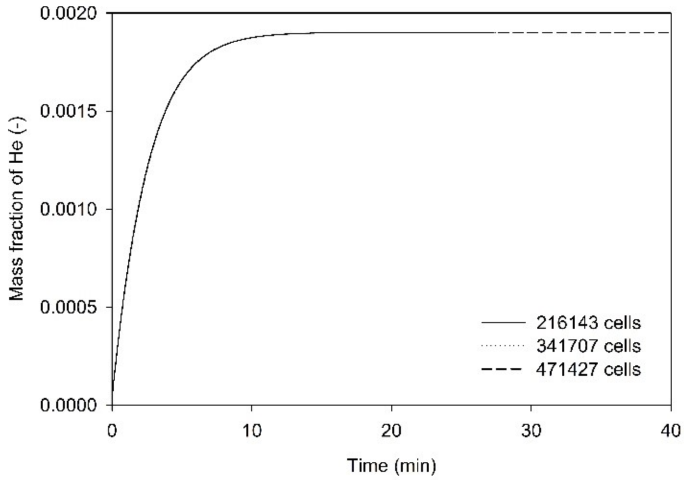
$$E(t) = \frac{(1 - \beta)^2}{\alpha \bar{t}} \cdot exp\left[-\frac{1 - \beta}{\alpha}\left(\frac{t}{\bar{t}}\right)\right] \quad (S7)$$

Vessel Dispersion Number

According to Levenspiel (1999), the theory of the axial dispersion model has been widely used to describe the non-ideal flow behavior inside the reactor.[2] D/uL, called "vessel dispersion number", refers to the flow characteristics of the reactor as explained below.[2]

If D/uL is close to 0	low dispersion, then plug flow
If D/uL is near ∞	large dispersion, then mixed flow

where D is dispersion coefficient depending on molecular diffusion.
u is velocity.
L is characteristic length.



Mesh Generation

Figure S3: The mesh independence study under the temperature of 35 °C, atmospheric pressure, impeller speed of 1896 rpm, and total flow rate of 150 cm^3 min^{-1} at 216,143, 341,707, and 471,427 mesh cells.

To investigate the mesh quality in CFD simulations, the mesh cells for the Berty reactor model were varied in this research (216,143, 341,707, and 471,427 mesh cells). Figure S3 shows that the mass fraction curves of He in three mesh cells are not different. Therefore, the mesh cell at 471,427 cells was used to represent the Berty reactor CFD model in this research. Moreover, the minimum orthogonal quality in this case, is equal to 0.3, which provides a good orthogonal quality mesh.

Results and Discussion

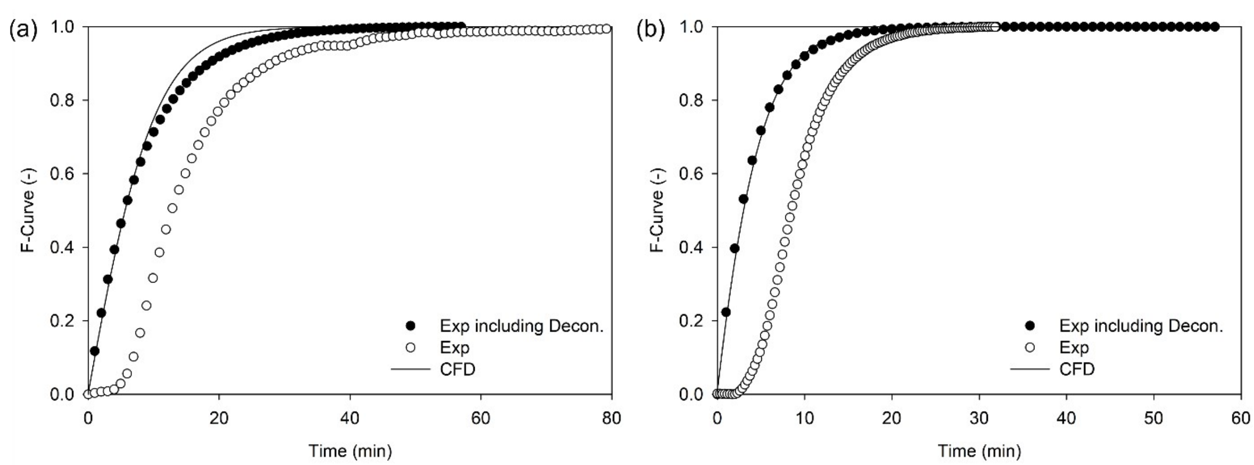


Figure S4: The F-curves from the 3D-CFD model and experimental data with and without convolution integral method under the temperature of 35 °C, atmospheric pressure, and impeller speed of 1896 rpm at the total flow rates of (a) 39.4 and (b) 84 cm^3 min^{-1}.

Figure S4 shows the F-curves from the 3D-CFD model and experimental data with and without convolution integral method under the temperature of 35 °C, atmospheric pressure, and impeller speed of 1896 rpm at the total flow rates of (a) 39.4 and (b) 84 cm^3 min^{-1}. When comparing the F-curves of the experimental data between with and without deconvolution method (black and white circles), it was found that the F-curve of the experimental data with the deconvolution method is significantly different from the case without the deconvolution method for both 39.4 and 84 cm^3 min^{-1}. In the case without the deconvolution method, the non-symmetrical S-shaped response curves were observed, while the F-curves of the case with the deconvolution method represented the mixed flow characteristics according to Levenspiel (1999).[2]

After the deconvolution, the flow characteristics of the system is more close to mixed flow behavior, and the RTD curve of the Berty reactor is revealed by extracting the signal distortion of the gas chromatography. When the F-curves from experimental data with the deconvolution method were compared with those from the 3D-CFD model for both flow rates (black circle and solid line), it was found that the F-curve of the 3D-CFD model showed good agreement with the F-curve obtained from the experimental data including deconvolution method. There is less than 5% of an error on both F-curves.

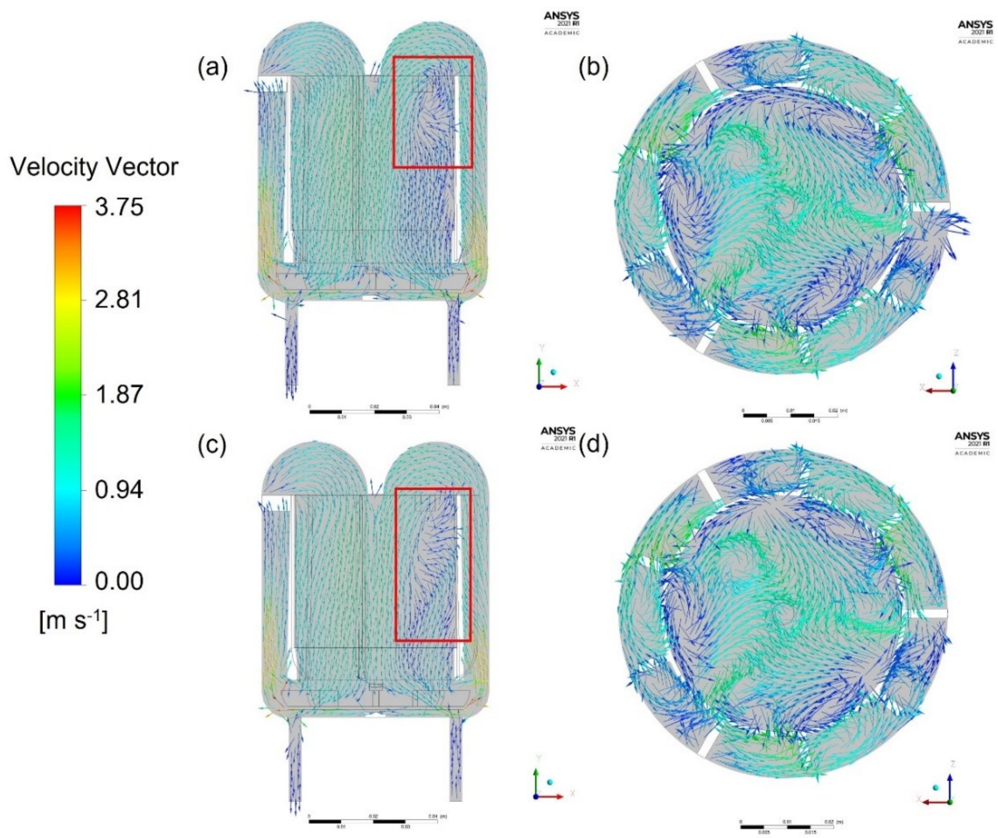


Figure S5: The velocity vector in vertical and horizontal planes (Y = 6.57 cm) of the Berty reactor under steady-state condition at the temperature of 35 °C, atmospheric pressure, the total flow rates of (a, b) 150 and (c, d) 39.4 cm^3 min^{-1}, and impeller speed of 1896 rpm.

Figures S5(a) and (b) show the velocity vectors in vertical and horizontal planes (Y = 6.57 cm) of the Berty reactor at a total flow rate of 150 cm^3 min^{-1}. Whereas Figures S5(c) and (d) represent those of the total flow rate at 39.4 cm^3 min^{-1}. The flow characteristics in Figures S5(a) and (b) were described as same as Figure 6. Nevertheless, the figures compare the two different CFD simulations of the Berty reactor at low and high flow rates. It was found that the velocity vectors of both cases are similar but the vortices area at the low flow rate (red box, Figure S5(c)) is bigger than that at the high flow rate (Figure S5 (a)). It refers to the higher dead volume for the low flow rate case. Moreover, it corresponds to the RTD results in Figures 9 and 10 that the dead volume can be reduced when the total flow rate increases.

References

1. J. A. Conesa, *Chemical Reactor Design: Mathematical Modeling and Applications*, 2020.
2. O. Levenspiel, *Chemical Reaction Engineering*, John Wiley & Sons, United States of America, 3th edn., 1999.