

## **Multiphase Loop-Reactor with In-Situ Extraction: CFD Simulation and 2D Compartment Modeling with Population Balances**

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A multiphase loop-reactor, which combines a gas-phase for oxygen supply and an in-situ liquid-liquid extraction for a fermentation process, is developed via CFD simulation. In addition, a two-dimensional compartment model is developed and presented. It applies the results of the CFD simulation to calculate mass-transfer and the local drop distribution in the two-dimensional area of the liquid-liquid extraction part of the reactor. The drop population balances are solved via a n-Monte-Carlo approach. Results of the compartment model show good agreement of the drop motion in comparison with the CFD simulation. In addition, a chemical test-system is used to calculate the mass-transfer in the liquid-liquid extraction area.

### **1. Introduction**

Resulting from the foreseeable shortage of fossil resources and climate change, a feed stock change from fossil resources to renewable educts is inevitable. Therefore, white biotechnology has seen a substantial advancement in recent years. One promising opportunity offer optimized microorganisms from biotechnology combined with efficient processes. However, the separation and purification of products from fermentation media lead to new challenges for process engineers concerning product purity, bio-compatibility and economic as well as ecologic feasibility. Moreover, even if biotechnological processes are on the one hand more selective compared to chemo-catalytic processes they can on the other hand exhibit inhibition at low product concentrations leading to low space time yields.

To overcome product inhibition in aerobic fermentation processes an innovative multiphase loop-reactor was developed [1, 2]. The reactor set-up combines both oxygen supply as well as in-situ liquid-liquid extraction. Thus, the extraction offers the possibility to remove continuously a toxic or inhibitory product and to increase space-time yield of a fermentation. The advantages of the reactor set-up result from locally separated zones of oxygen supply and product separation in one apparatus. The oxygen rich gas-phase induces a loop flow whereas in the outer downcomer the extraction solvent rises counter-currently to the aqueous fermentation broth. The general set-up of the reactor is depicted in Figure 1.

In previous studies we already proposed a possible reactor set-up [2]. The reactor was operated with a test-system composed of water as continuous phase, air and a kerosene fraction (Shellsol T) as

dispersed phase. The experimental results of the fluid dynamics and the performed CFD simulations were in good agreement and proved the performance and functionality of the new reactor concept. For comparison the hold-up and an optical evaluation of the flow were used.

For the description and modeling of the multiphase-loop reactor containing a liquid-liquid extraction part and a bubble column part the drop/bubble size distribution and the mass transfer are of main interest. Depending to the energy input drops or bubbles can coalesce or break. Since simple thermodynamic models like the

equation of Kremser [5] do not account for interfacial area and mass transfer, several detailed models were developed. On the one hand, there are one-dimensional models that calculate the drop size distribution with population balances [6, 7]. Due to their one-dimensional set-up, the local flow pattern is calculated with models for axial dispersion along with additional terms including the effects of internals. On the other hand, detailed flow conditions are accessible via CFD simulation [8-10]. A coupling of population balances with CFD leads to a local resolution of drop size distribution and mass transfer according to the local exchange area. This method offers highly accurate results but needs high computational effort since the population balance equations have to be solved for each grid cell. Therefore, researchers have started to use compartment models that combine the advantages of both methods [11]. In the compartment grid, that uses only a small fraction of elements in comparison to the CFD grid, the continuous phase is modeled as ideally mixed and the population balances for the dispersed phase are solved. With a CFD simulation, the local flow characteristics are described neglecting mass transfer and population effects like coalescence and breakage of drops or bubbles. With the flow-data of the CFD simulation the exchange flow between the compartments, the mean convection flow in each compartment and the local energy dissipation are set. The advantages of the compartment model are the employment of detailed flow characteristics based on CFD simulations and less computational effort. Consequently, back-mixing and detailed drop motion is simulated on a physical basis (CFD) without empirical approximations as in one-dimensional population balances. In addition, population balances including drop diameter distribution and mass transfer can be applied with acceptable computational effort.

This methodology has been used with classical population balances for crystallization processes [11], batch reactors [12] and bubble columns [13]. To the best of our knowledge only for crystallization processes, a Monte-Carlo simulation was used to calculate the particle population [14].

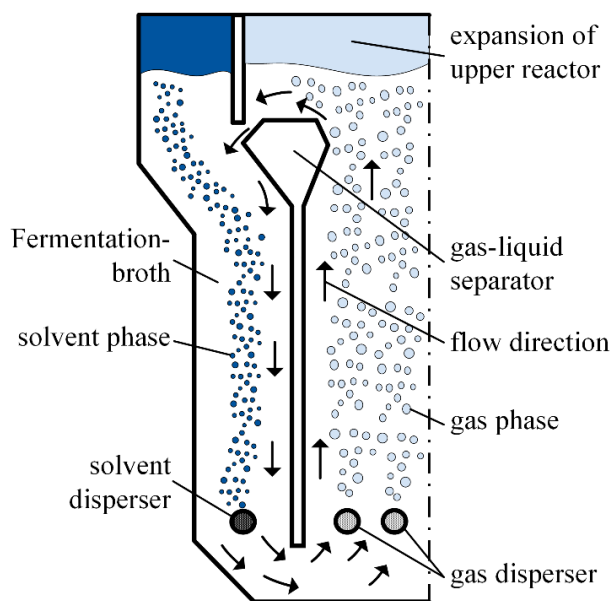


Figure 1. Concept of multiphase loop-reactor

In this paper, we present a new compartment model approach for the liquid-liquid extraction part of the recently developed multiphase-loop reactor [2], which solves the population phenomena with a n-Monte-Carlo simulation. Results of the simulation in the multiphase-loop reactor with a chemical test-system are introduced.

## 2. Experimental work and Simulation set-up

## 2.1 CFD Simulation

In this study we improved the reactor design via three-phase CFD simulation model as already used and validated in [2]. The properties of the used chemicals are listed in Table 1. The inlet volume flows of Shellsol T (drop diameter 2 mm) as solvent and air (bubble diameter 5 mm) for oxygenation are set constant to 0.034 m<sup>3</sup>/h and 0.8 Nm<sup>3</sup>/h. The gaseous phase is dispersed in the center of the reactor and the extraction solvent rises in the gap between inner cylinder and outer reactor wall (Figure 1). The top of the reactor is expanded to include a gas-liquid separator at the top of the internal cylinder. The CFD model includes a two dimensional axisymmetric Euler-Euler approach in combination with a standard k-ε turbulence model. The simulation considers gravitation, drag (model of Schiller and Naumann [3]) and turbulent dispersion (model of Simonin and Viollet [4]) as additional forces. Further information about the simulation set-up can be found in [2]. In addition, a new pilot-reactor has been designed and built in technical scale. The final dimensions of the improved reactor are given in Figure 2.

Table 1. Chemical properties

|   |                                 |                            |
|---|---------------------------------|----------------------------|
|   | Density<br>[kg/m <sup>3</sup> ] | Dynamic viscosity<br>[Pas] |
| Shellsol T  | 780,00                          | $2,40 \cdot 10^{-3}$       |
| Water   | 993,56                          | $1,03 \cdot 10^{-3}$       |
| Air   | 1,20                            | $17,10 \cdot 10^{-6}$      |
| Interfacial tension (water –<br>Shellsol T) [N/m] |                                 | 0,0263                     |

## 2.2 Two-dimensional compartment model

The liquid-liquid extraction part of the reactor (marked with a red rectangle in Figure 2) is modelled with a two-dimensional compartment model calculating drop size distributions and mass transfer. At the top of the domain, water loaded with  $0.025 \text{ kg/kg}_{\text{water}}$  of an exemplary transfer

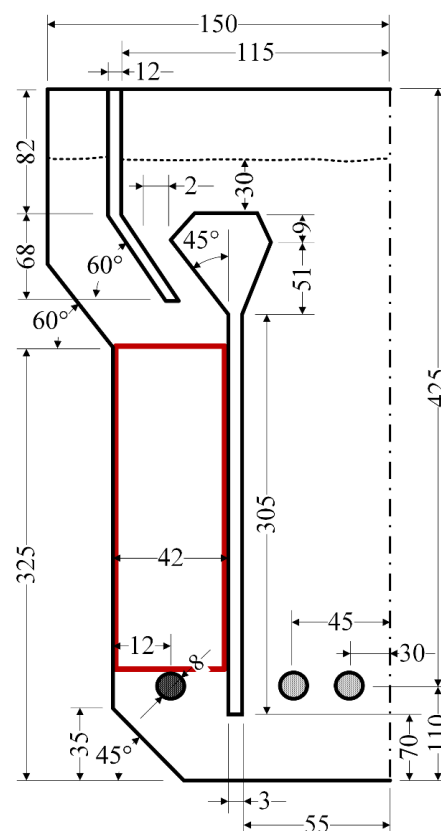


Figure 2. Dimensions of multiphase loop-reactor in mm

component flows into the extraction area. The unloaded dispersed phase enters at the lower domain at a radius of 0.08 m with a drop diameter of 2 mm. The compartment grid has a resolution of equally distributed compartments (three compartments in radial direction and eight in vertical direction). The performed CFD simulations supply the compartment model with flow characteristics, specifically velocity fields and exchange flows between the compartments as shown in Figure 3. Here the water velocity profile (result of CFD simulation) is shown. Overlapped is the grid in red and the vertical water velocity vectors are added at the boundaries of the compartments. In each compartment, the water velocity is averaged and saved in the model.

In the compartments, a representative population of the total amount of drops (50 drops per compartment) is calculated with a n-Monte-Carlo simulation approach. The advantage of a n-Monte-Carlo (fixed amount of drops per compartment) in comparison to a v-Monte-Carlo (fixed volume of apparatus) simulation approach is that in case of high droplet breakage the amount of calculated drops is kept constant. Consequently, this model can be used for example for strong dispersing systems.

In the here presented compartment model, sub-models are used for each representative droplet to account for motion, mass transfer and coalescence. The same chemicals as in the CFD simulation are used (Table 1). In the simulations with the compartment model an exemplary transfer component is added that is extracted into the dispersed phase. A sub-model for drop breakage is neglected since the drops can rise freely in the simulated domain. The sub-models of the one-dimensional population balance model ReDrop [6] are transferred into the compartment model and described in more detail in the following paragraphs. The model parameters and physico-chemical properties are given in Table 2. The motion of the drops is simulated with a superposition of the rise-velocity of a single drop and the averaged velocity vector of the continuous phase in one compartment. For the single drop rise-velocity the model of Henschke [6] is applied, which considers different droplet states (rigid, circulating, oscillating and deformed). The transition diameter  $d_{um}$  describes the transition of a rigid to a circulation drop. Both,  $a_{15}$  and  $a_{16}$  parametrize the drop-oscillation and the corresponding reduction of rise-velocity.

The mass-transfer into the drop is given according to the mass-transfer coefficients as

$$\Delta m = \Delta t \cdot \pi d^2 \cdot \frac{1}{\frac{1}{\rho_d \beta_d} + \frac{K}{\rho_c \beta_c}} \cdot (y - Kx) \quad (1)$$

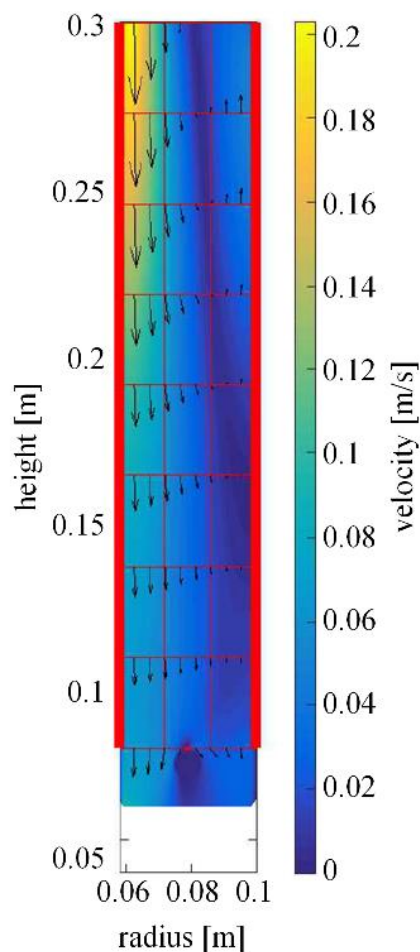


Figure 3. Compartments with velocity vectors at boundaries and velocity

where  $\Delta t$  is the time step,  $d$  the drop diameter,  $\rho_d$  and  $\rho_c$  the density of the disperse and continuous phase respectively,  $\beta_d$  and  $\beta_c$  the corresponding mass-transfer coefficients,  $K$  the distribution coefficient and  $y$  and  $x$  the concentrations of the disperse and continuous phase. The mass-transfer resistance in the continuous phase is neglected. The mass-transfer coefficient of the dispersed phase is defined by

$$\beta_d = \frac{D_{\text{eff}}}{d} \sqrt{\frac{4d^2}{\pi D_{\text{eff}} t} + \pi^4} \quad (2)$$

where  $D_{\text{eff}}$  represents an effective diffusion coefficient and  $t$  the drop lifespan. It describes an enhancement of the diffusion of the drop due to convection (inner circulation) and is calculated with

$$D_{\text{eff}} = D_d + \frac{v_{\infty} d}{C_{\text{IP}} \left(1 + \frac{\eta_d}{\eta_c}\right)} \quad (3)$$

where  $D_d$  is the diffusion coefficient of the dispersed phase,  $v_{\infty}$  the rise-velocity,  $C_{\text{IP}}$  the mass-transfer enhancement factor and  $\eta_d$  and  $\eta_c$  the viscosities of the disperse and continuous phase respectively.

The idea of the applied coalescence model is that every drop has a statistic lifespan  $\tau_c$  until it coalesces with another drop.  $\tau_c$  is determined by

$$\tau_c = \xi \frac{\eta_c}{\epsilon \sigma^{1/3} H_{\text{cd}}^{1/6} (\Delta \rho g)^{1/2}} \quad (4)$$

where  $\xi$  is the coalescence parameter,  $\epsilon$  the hold-up,  $H_{\text{cd}}$  the Hamaker coefficient (for organic-aqueous systems  $\sim 10^{-20}$  Nm),  $\Delta \rho$  the density difference and  $g$  the gravitation. The probability  $Z_c$  of coalescence of a drop is then calculated with

$$Z_c = \frac{\Delta t}{\tau_c} \quad (5)$$

The drop is marked for coalescence when an equally distributed random number  $s$  (between 0 and 1) is  $\leq Z_c$ . When two or more drops in one compartment are marked for coalescence randomly these two drops form a new bigger drop. Afterwards the coalescence marker is deleted.

Table 2. Model parameters and physico-chemical properties

| single drop sedimentation |        | mass-transfer   |  |
|---------------------------|--------|-----------------|--|
| $d_{\text{um}}$           | 7.1 mm | $C_{\text{IP}}$ | 9445                                       |
| $a_{15}$                  | 1.917  | $D_d$           | $2.199 \cdot 10^{-9} \text{ m}^2/\text{s}$ |
| $a_{16}$                  | 4.5    | $K$             | 0.9  |
| coalescence               |        | time step size  |  |
| $\xi$                     | 200    | $\Delta t$      | 0.1 s                                      |

### 3. Results and Discussion

#### 3.1 CFD Simulation

Figure 4 depicts the volume-fraction of air and solvent for a volume flow of 0.8 Nm<sup>3</sup>/h air and 0.034 m<sup>3</sup>/h solvent entering the reactor. The air rises in the center of the reactor and leaves the aqueous phase above the gas-liquid separator. The solvent rises counter currently to the aqueous phase and is not

entrained into the area of the gas phase. The operation in a pilot reactor (dimension in Figure 2) proved the simulated results in a similar manner as already presented in [2]. Therefore, with this setup the reactor can be operated in a preferred manner so that the dispersed phases leave the reactor in different areas.

### 3.2 Two-dimensional compartment model

The above explained setup of the two-dimensional compartment model including coalescence and mass-transfer is simulated using a n-Monte-Carlo algorithm for the solution of the drop dispersion. The result after reaching steady state (after 1500 s) is shown in Figure 5. In red the compartment grid is drawn. The drops are marked as circles. The size of the circle represents the drop diameter. The color of the circle represents the amount of transfer component in the drops. During the calculation in each compartment only up to 50 representative drops are considered. Size, concentration and position distribution of the drops are scaled up to the total amount of drops in the reactor. This scaling is also used in Figure 5 so that the drop number represent the hold-up in the reactor.

Considering the drop motion the general flow characteristic of the multiphase flow is captured well by the two-dimensional compartment model. It can be observed that during their rising they move to the outer part of the reactor. This is also shown in Figure 4 in the CFD simulation. The main deviation is found in the first height elements of the compartment grid. In the CFD simulation the drops first move nearly horizontally to the wall, whereas in the compartment model they move directly upwards with a slight motion towards the wall. This is caused by averaging the velocity field of the water phase in each compartment. Especially above the disperser high

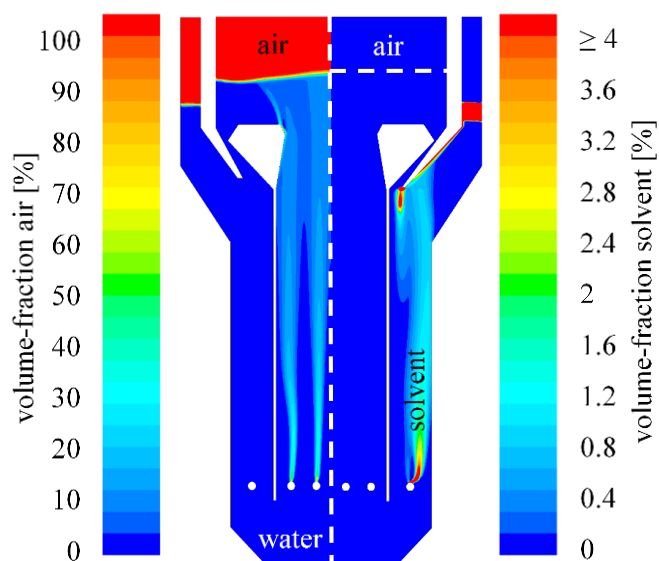


Figure 4. Results of CFD simulation:  
Volume-fractions in multiphase loop-reactor

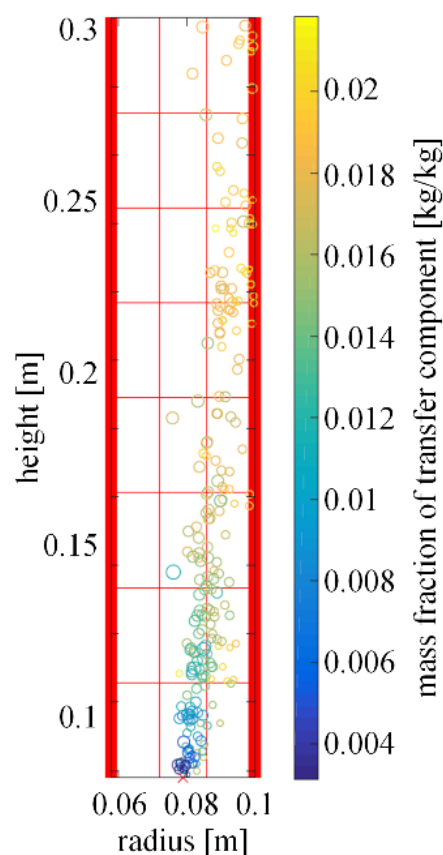


Figure 5. Representative drops in compartment model with position in reactor and concentration of transfer component



local horizontal water velocity vectors exist which are less due to the averaging over the compartment. At the top of the simulation domain in Figure 5, only next to the wall drops are present. However, in the CFD model drops are entrained (Figure 4) into the center of the extraction area. This is missing in the compartment model since the upper area with the backflow is not modeled. During the drop-motion, the transfer component moves into the drops (colored drops in Figure 4). Since only little dispersed phase flows into the reactor only few transfer component is extracted from the continuous phase. Therefore, the concentration of the transfer component in the drops nearly reaches equilibrium when leaving the reactor.

#### 4. Conclusion

The flow direction in the multiphase-loop reactor is modified and improved via CFD simulation in comparison to earlier investigations [2]. A two-dimensional compartment model was successfully developed describing the drop size distribution and mass-transfer into the drops. It uses a n-Monte-Carlo method to solve the population balance calculating only a fixed representative number of drops. It was used for the multiphase flow in the liquid-liquid extraction part of the multiphase-loop reactor. Results show good agreement with the CFD simulation. The advantages of the compartment model are the employment of detailed flow characteristics based on CFD simulations and reduced computational effort when calculating mass transfer, reactions and population balances in comparison to CFD simulations.

In further investigations, a detailed sensitivity analysis regarding number of representative drops and compartment resolution has to be performed. In addition, the whole reactor including an additional gas phase (air) and a bio catalytic reaction will be calculated with the compartment model to investigate the potential of the multiphase-loop reactor. When including models for drop/bubble breakage the two-dimensional compartment model can also be used for different multiphase reactors or extractors like liquid-liquid extraction columns.

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