Model of mass transfer processes in the cascade of centrifugal extractors

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Abstract. The paper describes a mathematical model of mass transfer processes in a cascade of reverse-flow centrifugal extractors. Model of operation of each extractor is given as tightly coupled system of mixing and separating chambers. All model units are represented by systems of differential equations. The article presents the results of testing of the developed model, which confirmed the validity of the assumptions made in the model. The authors assessed the impact of the overflow of dense phase level on the hydrostatic position of phase interface level in the extractor. The research showed that a change in the volume of dense and light phases occurs in each apparatus of a cascade even in the steady mode. Operation of the cascade consisting of 12 series-connected centrifugal extractors was simulated in order to verify the model. Computer simulation results confirm the adequacy of the developed model.

1. Introduction

Currently, there are various methods of chemical treatment, but extraction technologies are the most widespread because of their efficiency. Pulse column extractors connected by a cascade are used in production as the hardware implementation of the extraction technologies. In recent years, centrifugal extractors have been introduced into the technological process instead of column extractors. This is due to the factor that centrifugal extractors provide low contact time of phases, high coefficient of substances interface, increased nuclear safety. They also allow increasing production volume without expansion of workspaces. Such production aspects are important especially when dealing with radioactive materials. Although the extraction processes occurring in the pulse columns and centrifugal extractors are comparable, there are significant differences in the process flow diagram. These technological differences lead to the necessity for the development of extraction technologies. A number of technological schemes of extraction refining are being worked out within the framework of the project “PRORYV”. These schemes include a large number of processing units with centrifugal extractor as the main hardware unit.

The search for optimal technological schemes of refining from the standpoint of safety and minimization of production costs is performed basically according to experimental research data. It is difficult to conduct experimental research at production site because of the reagents with high reactivity. In this regard, there is a need for mathematical and later computer models describing the operation of centrifugal cascade of extractors taking into account occurring basic physical and chemical processes. Moreover, computer simulation will evaluate the performance of centrifugal extractors, as separate devices or as a cascade of elementary units. This aspect will allow the introduction of safe technologies into chemically reactive production.

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2. Mathematical model

The technological scheme of the extraction refining process contains blocks of cascades of centrifugal extractors with centrifugal extractor as elementary unit. Depending on industrial purposes, the number of extractors in the blocks varies. Since all the devices included in the blocks are identical structurally and by the principle of operation, we will describe the operation of one extractor.

2.1. Description of the simulation object

Operation of the centrifugal extractor is based on two processes. The first is the mixing of initial fluids in the mixing chamber of the apparatus, which provides mass transfer of the component. The other is the process of interface of the formed emulsion under the influence of centrifugal field on the light and dense phases in the separating chamber of the device. Therefore, the centrifugal extractor can be represented in the form of tightly coupled system of two parts, namely the mixing and separating during the simulation. The principle of operation of the extractor is as follows. After starting the drive initial mutually immiscible liquids are fed into the lower part of the mixing chamber where they are mixed with stirrer. After the liquids are mixed biphasic emulsion is formed in which the process of mass transfer occurs. Formed emulsion is fed from the mixing chamber into the separating chamber by bladed transfer unit. Emulsion is divided into dense and light phases in the mixing chamber under the influence of centrifugal field. At the same time the dense phase enters the manifold for collecting the dense phase through the water seal. Light phase is fed into the manifold for collecting light phase through the tubes of light phase output. Then, light and dense phases are removed from the machine through respective vertical channels.

Analyzing occurring technological processes in the centrifugal extractor, we concluded that the main input variables are the concentrations of the target components entering the extractor flow, volumetric flow and temperature of input streams, the number of rotations of stirrer and bladed transfer unit of extractor. Volumetric flow and temperature of the streams leaving the extractor, as well as the concentration of the target components in each of the streams are the output variables. Volumetric flow of resulting emulsion entering the separating chamber, the component wise concentration composition of emulsion refer to disturbing parameters. In addition, disturbing influences on the system should include the volumetric flow rate, temperature and concentration of the components of the poured solution, if it exists in the technological flow diagram.

Because the main processes taking place in the centrifugal extractor are mixing and interface processes, the mathematical model is based on the processes describing diffusion mass transfer and hydrodynamics. In addition, we will present the developed model as a cell model, where all the workspace is divided into *n* cells. The mechanism of the construction of this type of model is well represented in the [4].

2.2 The mathematical model of heat transfer processes in a centrifugal extractor as the basic unit of the cascade

According to [1-3] the extraction rate of the target components in the conditions of intensive mixing in a centrifugal extractor is determined by the rate of diffusion mass transfer. But the rate of the mass transfer process is influenced not only by physical and chemical processes occurring in the apparatus, but also by the rotation speed of internal moving parts of the machine, such as a stirrer or bladed transfer device. Therefore, it is possible to estimate the character of flowing of working media in the process of mixing using centrifugal similarity criteria such as Reynolds, Prandtl, Nusselt criteria presented in detail in [2, 5]. In practice, it is quite difficult to define the criteria, which include speed, because according to [1], the mode of developed free turbulence occurs in devices having internal rotating components. Velocity distribution in such devices is complex, therefore at the stage of design conventional velocity equal to the peripheral velocity of the stirrer blade end, excluding factor π=3.14 is used in centrifugal criteria [2].

Initial streams entering the extractor form a two phase emulsion during the mixing process in the mixing chamber. One liquid is dispersed as droplets and forms the dispersed phase in such a two-phase system. The other liquid forms a continuous phase in which the droplets of dispersed phase liquid move. According to [1, 3, 5] form of droplets depends on the rotational velocity of stirrer in apparatus. The greater the velocity, the greater the deformation of a drop. In this regard, the droplet

diameter can only be determined experimentally. In the simulation, we will assume that the drops have a spherical shape and can only change its diameter. Dispersed phase will be denoted as X phase and continuous phase will be denoted as Y phase when developing the model. This denotation will be valid both for the mixing chamber unit and for the separating chamber unit. In the mixing chamber, due to the presence of the internal rotating mechanism, convective diffusion processes dominate over the processes of molecular diffusion. Therefore, the basic mass transfer equations for the block representing the mixing chamber can be represented as follows:

$$\begin{cases} V_i^x(t)\dfrac{\mathrm{d}\left[C_i^x(t)\right]}{\mathrm{d}t}+C_i^x(t)\dfrac{\mathrm{d}\left[V_i^x(t)\right]}{\mathrm{d}t}=Q_{\mathrm{in}}^x(\mathrm{t})C_{\mathrm{in},i}^x(t)-Q_{\mathrm{f}}^x(\mathrm{t})C_{\mathrm{f},i}^x(t)-\Delta m_i^x \\ V_i^y(t)\dfrac{\mathrm{d}\left[C_i^y(t)\right]}{\mathrm{d}t}+C_i^y(t)\dfrac{\mathrm{d}\left[V_i^y(t)\right]}{\mathrm{d}t}=Q_{\mathrm{in}}^y(\mathrm{t})C_{\mathrm{in},i}^y(t)-Q_{\mathrm{f}}^y(\mathrm{t})C_{\mathrm{f},i}^y(t)+\Delta m_i^x \end{cases} \quad (1)$$

where Q_{in}^x and Q_{in}^y – volumetric flow rates of phase X and phase Y respectively at the input of the mixing chamber; Q_{f}^x and Q_{f}^y – volumetric flow rates of phase X and phase Y respectively at the output of the mixing chamber; V_i^x and V_i^y – volumes of phase X and phase Y respectively; $C_{\mathrm{in},i}^x$ and $C_{\mathrm{in},i}^y$ – initial concentrations of i-th component in phase X and phase Y respectively; $C_{\mathrm{f},i}^x$ and $C_{\mathrm{f},i}^y$ – current value of concentration of i-th component in phase X and phase Y respectively; Δm_i^x and Δm_i^y – amount of a substance transferred from one phase to the other.

Volume of phase X and phase Y included in the system of equations (1) we will define as follows depending on the total volume of the chamber:

$$V_{\mathrm{m}}{}^x=\frac{Q_{\mathrm{in}}^x V_{\mathrm{m}}}{Q_{\mathrm{in}}^x+Q_{\mathrm{in}}^y} \qquad V_{\mathrm{m}}{}^y=V_{\mathrm{m}}-V_{\mathrm{m}}^x \quad (2)$$

where V_{m} – the working volume of the mixing chamber.

In the area of intensive mixing mass transfer occurs primarily by convective diffusion. Therefore, the amount of matter carried at each moment of time from one phase to the other presented in the system (1) can be defined as follows:

$$\begin{cases} \Delta m_i^x=6\cdot\beta_i^x\cdot\dfrac{V_{\mathrm{m}}^{\mathrm{d}}}{d_x}\left(C_i^x-{}^*C_i^x\right) \\ \Delta m_i^y=6\cdot\beta_i^y\cdot\dfrac{V_{\mathrm{m}}^{\mathrm{d}}}{d_x}\left({}^*C_i^y-C_i^y\right) \end{cases} \quad (3)$$

where $V_{\mathrm{m}}^{\mathrm{d}}$ – volume of dispersed phase; $\mathrm{d_x}$ – diameter of the disperse phase droplet; C_i^x и C_i^y – concentrations of i-th component in phase X and phase Y respectively; ${}^*C_i^x$ – concentration of i-th component in phase X equilibrium with the concentration C_i^y in the basic mass of phase Y; ${}^*C_i^y$ – concentration of i-th component in phase Y equilibrium with the concentration C_i^x in the basic mass of phase X; β_i^x and β_i^y – coefficient of mass transfer of i-th component in phase X and in phase Y respectively. Mass transfer coefficients in the system of equations (3) depend on the physical, geometrical and hydrodynamic factors and can be found mainly experimentally. But, according to [1], mass transfer coefficient is proportional to the product of stirrer rotation frequency to the diameter of blades of the stirrer and inversely proportional to the linear geometric size of the chamber in a turbulent flow mode of working environments, which is created in the mixing chamber. Based on this, and the fact that values of the equilibrium concentrations of target components in the set state are equal ${}^*C_i^x={}^*C_i^y$.

The values of the equilibrium concentrations in the system (3) are mainly determined by experimental data, curves of distribution are presented in literature. The disadvantage of such determination is that the reference data is presented according to the specific modes of operation of the device and composition of working environments, which often does not fully correspond to the investigated data. Therefore, based on the studies mentioned in [6], we will use a polynomial dependence for determination of the values of equilibrium concentrations of the target components in each phase.

The emulsion formed in the mixing chamber by means of a bladed transfer device is fed into the separating chamber. The process of interface of the emulsion is a complex hydrodynamic process, which is influenced by various factors, such as angular velocity of the rotor, emulsion supply rate, threshold level of phase interface, internal hydraulic resistance due to the geometry of the machine, change in the density of formed phases and distribution of the temperature gradient across the device in whole.

Based on the data in [1], we can assume that there are two liquid layers separated by phase interface level in separating chamber. The unblended layer is dispersed phase liquid layer and the second layer is the layer of continuous phase. Depending on the type and composition of emulsion entering the separating chamber, each of the layers can be either light or dense. Another assumption is that is a complete phase interface without mutual entrainment of components in the separating chamber. In this case, conical chamber can be replaced with a cylindrical one with equivalent volume. Phase volume ratio of X and Y will influence both the phase threshold position and value of the delay of matter concentrations in the chamber.

Volumes of light phase V_s^{l} and dense V_s^{h} phase in the separating chamber according to [1], are a function from the density of the light and dense phases, composition and concentration of the temperature gradient of the emulsion coming for interface, as well as from the structural features of the camera. However, our research showed that we could determine the amount of light and dense phases, without loss of accuracy in the following way:

$$V_{\mathrm{s}}^{\mathrm{l}} = \frac{V_{\mathrm{s}} \cdot V_{\mathrm{m}}^{x}}{V_{\mathrm{m}}^{y} + V_{\mathrm{m}}^{x}} \qquad V_{\mathrm{s}}^{\mathrm{h}} = V_{\mathrm{s}} - V_{\mathrm{s}}^{\mathrm{l}} \tag{4}$$

where V_{s} – working volume of the separating chamber, V_{m}^{x} – the volume of the dispersed phase of the mixing chamber, V_{m}^{y} – the volume of the continuous phase of the mixing chamber.

Since phases X and Y are continuous and incompressible, then we will put the model of ideal displacement as the basis of the separating chamber model. In general terms, taking into account all the chamber settings and the processes occurring in it, we receive a system of higher order equations. High order equations make the process of modeling and verification processes more complicated. Since concentrations of target components are significant parameters of the separating chamber, it is possible to reduce the order of the resulting model, for example up to the first. Accordingly, we will describe the concentration distribution within the separating chamber by the following system of differential equations:

$$\begin{cases} \dfrac{\mathrm{d}\left[C_i^{x}(t)\right]}{\mathrm{d}t} = \dfrac{C_{\mathrm{in},i}^{x}(t) - C_{\mathrm{f},i}^{x}(t)}{T_{\mathrm{s}}^{x}} \\ \dfrac{\mathrm{d}\left[C_i^{y}(t)\right]}{\mathrm{d}t} = \dfrac{C_{\mathrm{in},i}^{\mathrm{y}}(t) - C_{\mathrm{f},i}^{\mathrm{y}}(t)}{T_{\mathrm{s}}^{\mathrm{y}}} \end{cases} \tag{5}$$

where $C_{\mathrm{in},i}^{x}$ and $C_{\mathrm{in},i}^{y}$ – concentrations of i-th component in phase X and phase Y respectively in the input to separating chamber; $C_{\mathrm{f},i}^{x}$ and $C_{\mathrm{f},i}^{y}$ – concentrations of i-th component in phase X and in phase Y respectively at the output of separating chamber; T_{s}^{x} and T_{s}^{y} – concentration delay in phases X and Y respectively.

If the phase X at the output of mixing chamber is dispersed, then the concentration delay in phases X and Y we will estimate as follows:

$$T_s^x = k \cdot \frac{V_s^h}{Q^h};\ T_s^y = k \cdot \frac{V_s^l}{Q^l};\ \ Q^h = Q^x;\ \ Q^l = Q^y$$

where V_s^h and V_s^l – volumes of dense and light phases respectively, k – correction factor.

We will determine the change of the flow rate of phases in the output from separating chamber at the following way:

$$Q^l = \frac{V_s^l \cdot \left(Q_{s_f}^x + Q_{s_f}^y\right)}{k \cdot V_s}$$
$$Q^h = \frac{V_s^h \cdot \left(Q_{s_f}^x + Q_{s_f}^y\right)}{k \cdot V_s} \qquad (6)$$

where k – correction factor, $Q_{s_f}^x$ и $Q_{s_f}^y$ – volumetric flow rates of phases X and Y at the output of the mixing chamber.

3. Simulation results

We have tested the developed model with the assumptions made using Matlab/Simulink software, since the software provides extensive instrumental basis for modeling physical systems. We will take the statement of the coincidence (with a possible deviation within 15%) of the experimental data with the simulation results for the target components, such as U and Pu as a criterion for the adequacy of the developed model.

When testing the developed model we have taken technological scheme of the extraction assembly of 12 consequently connected centrifugal extractors as a basic scheme. We have verified the model by the values of concentrations of target components both in the extract and in the raffinate at the cascade output.

The result of verification of the developed model is shown in Figure 1 and Figure 2 for the raffinate and the extract, respectively. For visual display of the results of simulation verification data are presented in relative units.

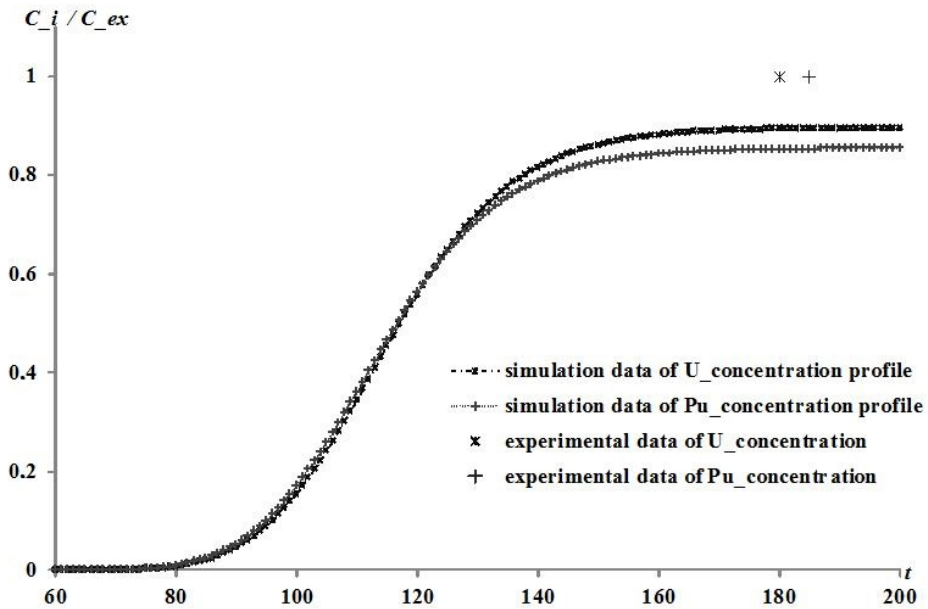

Figure 1. Distribution of the concentrations of U and Pu in the raffinate.

As we can see from the figure, the deviation of the distribution of the concentrations by target components, such as U and Pu in the raffinate does not exceed 15% from the experimental data, which meets production requirements.

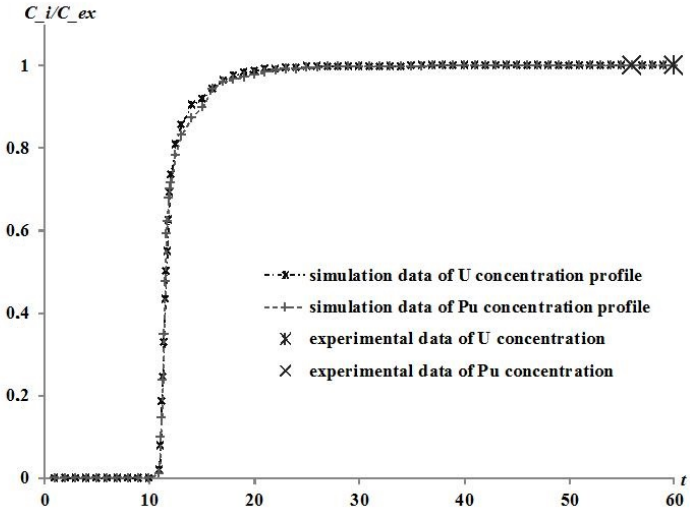

Figure 2. The distribution of concentrations of U and Pu in the extract

Figure 2 shows that the values of the concentrations of U and Pu in the extract obtained in the simulation are the same as the experimental data.

4. Conclusion

In the development of the mathematical model we presented the centrifugal extractor, the elementary unit of the cascade, in the form of tightly coupled system of two objects. We conducted analysis of technological process, on the basis of which we identified the main parameters influencing the process of mass transfer both in the extractor, and in the cascade. Preliminary testing of the model showed that an increase in radius of overflow of dense phase level in the water seal leads to reduce of the hydrostatic level position of phase interface in the separating chamber. If we change the density of the light phase, the volume of light phase also changes. This fact confirms the importance of considering changes in the volume of each phase in both chambers. The obtained verification data show that the concentration distribution of the target components at the cascade output both in the raffinate and in the extract, does not exceed the production limits. This fact confirms the admissibility of the assumptions made in the mathematical model.

References

[1] Kuznetsov G I, Pushkov A A, Kosogorov A.V 2000 *Centrifugal extractors CENTREX* (Moscow: RCHTU D.I. Mendeleev)

[2] Strenk F 1975 *Stirring and Stirred Apparatuses* [in Russian] (Leningrad: Khimiya)

[3] Goryunov A G 2011 Dynamic model of a multi-component nonequilibrium extraction process in the mixing chamber of centrifugal extractor *Scientific Technical Bulletin of Volga region* **4** pp 129-132

[4] Goryunov A G and Mikhaylov V S 2012 The automatic control system of a multi-component nonequilibrium extraction process in the pulse column *J. of Process Control* vol **22** pp. 1034-1043

[5] Vedantam S Wardle K E Tamhane T V Ranade V V and Joshi J B 2012 CFD Simulation of Annular Centrifugal Extractors *Int. J. of Chemical Engineering* vol **2012** 31 (doi:10.1155/2012/759397)

[6] Zilberman B Ya Fedorov Yu S Puzikov E A Goletskii N D Ryabkov D V Kudinov A S Blazheva I V and Kukharev D N 2014 Preliminary Studies of Dynamic Regimes in the Extraction–Scrubbing Groups of Units of the Experimental and Demonstration Center (EDC) Using Mathematical Modeling *Radiochemistry* vol **56** iss **6** pp 583-593