# A model of multicomponent extraction process in the pulsating column

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**Abstract**. The model of multicomponent process of extraction in pulsating column is presented. Model analysis on adequacy to real apparatus was made. The received results have shown, that the presented model adequacy to the real nuclear fuel-reprocessing apparatus.

## 1. Introduction
In nuclear reactors there is no complete combustion of fuel, which is determined by either physical destruction of fuel (fuel elements), or premature deterioration of reactivity. Spent fuel elements contain a significant amount of fissile material, the extraction of which is economically profitable.

Nowadays spent nuclear fuel is processed using an extraction technology, which can provide separation of substances. Hardware design of this technology is based on counter current columns connected in cascade.

In order to achieve high performance and high quality products with minimum production costs, the automated control systems (ACS) are used. The implementation of a good-quality automated control system of the extraction process will enhance its safety, improve technological and economic indicators of production as well as reduce the impact on the environment, due to decrease of toxic waste. In the development of automated control systems, it is advisable to use simulation models for the selection of the output variables, for the regulator settings and for the estimation of control quality.

The aim of this work was to develop a universal model of the extraction process in a common extraction column with its further testing by ACS synthesis of extraction process in a real apparatus.

## 2. A mathematical model of uranium extraction in pulsating columns
The following modeling methods of extraction in column reactors are widely used: the theory of similarity, hydraulic modeling and mathematical modeling [1].

The team of Department of Electronics and Automation of Nuclear Plants and Technology of Tomsk Polytechnic University applied mathematical models for the development of column models.

The equations of convective diffusion nonsteady in time at substance transfer from one phase to another are as follows [1, 2]:

$$\begin{cases} \dfrac{\partial x}{\partial t} = D_x \dfrac{\partial^2 x}{\partial h^2} - U_x \dfrac{\partial x}{\partial h} + W \\[2mm] \dfrac{\partial y}{\partial t} = D_y \dfrac{\partial^2 y}{\partial h^2} - U_y \dfrac{\partial y}{\partial h} + W \end{cases} \tag{1}$$

where x and y - uranium concentration in the dispersed and continuous phases; Ux, Uy - the rates of phase flow; Dx, Dy - diffusion coefficients of liquids.

$$W = \beta_1 a(x^* - x) = \beta_2 a(y - y^*) \qquad (2)$$

where β1 and β2 - mass transfer coefficients from one phase to another; a - the contact surface of two phases by a unit of volume; x*, y*- equilibrium values of concentration.

The diffusion coefficient of liquids D is so small that even at low rates of liquids the convective substance transport considerably dominates over molecular diffusion. In order to simplify the expression (1) the term of the equation containing D is usually neglected.

The resulting simplified equation of convective diffusion is a partial differential equation, which at a first approximation describes the processes in counter-current columns [2, 3, 4]. However, this equation assumes that the rates Ux, Uy are constant. Even in the first extraction model the flow rate changes depending on time, location and concentration of uranium. In this regard, the equation (1) becomes a nonlinear differential equation, analytical solution of which is difficult.

The analytical method for calculation by extraction stages was applied to solve this problem [1]. It takes into account the spatial distribution in the height of the column. In this method, an extraction column is divided into zones - stages. The extraction stage in packed columns is a part of the column in which equilibrium is established, i.e., the concentration of one of the considered components of the output counter flows at this stage, is characterized by a curve of the equilibrium distribution. A differential equation of material balance is used for each stage. The stages are connected in series. The coefficients are recalculated at each time point.

$$\begin{cases} \dfrac{d}{dt}\left(x^{out} \cdot V_x + y^{out} \cdot V_y\right) = x^{in} \cdot Q_x^{in} + y^{in} \cdot Q_y^{in} - x^{out} \cdot Q_x^{out} - y^{out} \cdot Q_y^{out} \\ y^{out} = K_p \cdot x^{out} \end{cases} \qquad (3)$$

where xin and и yin - input uranium concentration in the dispersed and continuous phase; xout and yout - output uranium concentration in the dispersed and continuous phases; Kp - coefficient of equilibrium distribution of uranium; Vx,Vy - volume of the dispersed and continuous phases; Qx Qy - input and output flow rates of the dispersed and continuous phases.

The developed model is adequate under certain conditions [5], and was used to develop several ACS of uranium concentration in pulsating columns of the "Utjos" unit at the Radiochemical plant of Siberian Chemical enterprise.

However, to enhance functionality and the universalization of ACS development it was necessary to eliminate the following restrictions on the model.

- The model is single-component. In this model it is possible to calculate only uranium concentration in the organic and aqueous phases apart from influence of other components.
- In the model it is impossible to change the number of stages into which the column height is divided. Therefore, the required accuracy of the components distribution by height of the column cannot be obtained.
- Due to the constancy of the uranium distribution coefficient Kp, the model is adequate for a very narrow range of input coordinates.

The developed model does not have these drawbacks. As an example a two-component model will be considered. However, the number of components can always be increased by adding equations to the system which doesn't significantly increase complexity of the model

This mathematical model of extraction column will take the following assumptions:

- The model of ideal displacement is used for the continuous phase [1].
- At each stage equilibrium between dissolved substance content in both phases is established [1, 6].
- Extraction rate is very high (equilibrium is established instantly) [6].

- Fluid temperature in the column is constant. Temperatures of all input and output flows are the same, the heat exchange between the phases and environment is not taken into account.
- It is considered that convective counter-current transfer does not affect the extraction process and, therefore, is not modeled.

The basic equations describing the process of mass transfer in a dynamic mode of the column operation are the equations of the system (1), as well as the equations describing the extraction of nitric acid.

$$
\begin{cases}
\dfrac{\partial x}{\partial t} = D_x \dfrac{\partial^2 x}{\partial t^2} - U_x \cdot \dfrac{\partial x}{\partial h} + \beta_1 \cdot \alpha \cdot \left( x^*(y) - x \right) \\[2mm]
\dfrac{\partial y}{\partial t} = D_y \dfrac{\partial^2 y}{\partial t^2} - U_y \cdot \dfrac{\partial y}{\partial h} + \beta_2 \cdot \alpha \cdot \left( y^*(x) - y \right) \\[2mm]
\dfrac{\partial N_x}{\partial t} = D_x \dfrac{\partial^2 N_x}{\partial t^2} - U_x \cdot \dfrac{\partial N_x}{\partial h} + \beta_{N1} \cdot \alpha \cdot \left( N_x^*(N_y) - N_x \right) \\[2mm]
\dfrac{\partial N_y}{\partial t} = D_x \dfrac{\partial^2 N_y}{\partial t^2} - U_y \cdot \dfrac{\partial N_y}{\partial h} + \beta_{N2} \cdot \alpha \cdot \left( N_y^*(N_x) - N_y \right)
\end{cases}
\tag{4}
$$

where  x and Ny - nitric acid concentration in the dispersed and continuous phases; $\beta_x$, $\beta_y$ , $\beta_{N1}$ and $\beta_{N2}$ individual mass transfer coefficients; $N_x^*$ and  $N_y^*$ - equilibrium concentrations of nitric acid.

To simplify the expression (4) the term of the equation containing D is usually neglected. The system of equations describing the extraction process with regard to the material balance is shown below (5).

The model can be composed from the upper settling section (USS), the lower settling section (LSS) and the extraction section which number of stages is determined by height.  Modeling the processes which are observed in the extraction column, the elements are calculated successively at each iteration: USS, the extraction section, LSS, calculation of sensor readings.

$$
\begin{cases}
\dfrac{\partial x}{\partial t} = -U_x \cdot \dfrac{\partial x}{\partial h} + \beta_1 \cdot \alpha \cdot \left( x^*(y) - x \right) + \dfrac{1}{V_x} \cdot \left( Q_x^{in} \cdot x_{in} - Q_x^{out} \cdot x \right) \\[2mm]
\dfrac{\partial y}{\partial t} = -U_y \cdot \dfrac{\partial y}{\partial h} + \beta_2 \cdot \alpha \cdot \left( y^*(x) - y \right) + \dfrac{1}{V_y} \cdot \left( Q_y^{in} \cdot y_{in} - Q_y^{out} \cdot y \right) \\[2mm]
\dfrac{\partial N_x}{\partial t} = -U_x \cdot \dfrac{\partial N_x}{\partial h} + \beta_{N1} \cdot \alpha \cdot \left( N_x^*(N_y) - N_x \right) + \dfrac{1}{V_x} \cdot \left( Q_x^{in} \cdot N_x^{in} - Q_x^{out} \cdot N_x \right) \\[2mm]
\dfrac{\partial N_y}{\partial t} = -U_y \cdot \dfrac{\partial N_y}{\partial h} + \beta_{N2} \cdot \alpha \cdot \left( N_y^*(N_x) - N_y \right) + \dfrac{1}{V_y} \cdot \left( Q_y^{in} \cdot N_y^{in} - Q_y^{out} \cdot N_y \right)
\end{cases}
\tag{5}
$$

where $x_{in}$, $y_{in}$ - input uranium concentration in the aqueous and organic phases, respectively; $N_x^{in}$ , $N_y^{in}$ - input concentration of acid in the aqueous and organic phases, respectively.

## 3. Synthesis of the automated control system of uranium concentration in the raffinate of EC1 column

The head column of Radiochemical plant of Siberian Chemical enterprise was chosen for checking the adequacy of the model to the real device and for ACS development.

The following parameters can be measured in the column:

- average density of  fluid in the column U (1);
- volume of the dispersed phase in the column $L_{здф}$ (2). Measuring vessels are cut in the column.  Buoy level meters are installed in the upper parts of the column. They measure weight of liquid columns and their average. In its turn, the average density in the column is proportional to the volume (fraction) of the dispersed phase, which is known as volume of the dispersed phase or dispersed phase delay (DPD) [1, 5].

- local concentration of the metal in the place of installation $c$U (3).

Model adequacy was checked using F-test and Pearson's chi-squared test. The modeling results correspond to the experimental data in a wider range of parameters than the results presented in [7].

An extraction column can be referred to multi-loop controlled objects. In such objects, each output variable depends on two or more input values. Multi-loop automated control systems with cross-connection are used for automated control of such objects [7] In this paper we study single-loop and cascaded ACS, so it is necessary to choose such an output variable, the control of which could give the best characteristics of the output product.

The analysis of the input variables in the EC1 column revealed the following:

- OP consumption cannot be a controlling variable, as it is set for the whole cascade. Its change will cause some change in an operation mode, which is unacceptable.
- AP consumption can be selected as a controlling variable. Moreover, currently it is the one.
- AP composition is prepared in advance; uranium concentration and acid in it cannot be changed promptly. Therefore, these variables are not satisfactory.
- It is fairly hard to stabilize pulsation amplitude, so this variable also cannot be controlling one.

Thus, the controlling value in the column is AF consumption.

In EC1 column it is possible to control four parameters: the level of the delay dispersion phase in the upper part of the column and the whole column ($L_{3в}$, $L_{3н}$), extract density (which determines the uranium concentration in the extract - $c$U$^{EC}$) and the uranium concentration in the middle of the lower extraction section (comparable with uranium concentration in the raffinate - $c$U$^{РФ}$). As a controlling variable $L_{3в}$ ($L_{3н}$) or $c$U$^{РФ}$ is used. $c$U$^{EC}$ is not considered due to the fact that this parameter was applied in similar columns, and it gave poor results.

For the development of a control system it is necessary to determine the transfer functions of the aqueous phase consumption, as this parameter is a control action. By all the output variables, TCO can be replaced with aperiodic links of the second order with different time response and delay.

The $L_{3н}$ output parameter will not be used for the ACS development. The character of its response to the changes in AP consumption is comparable to $L_{3в}$, but it is less sensitive and more inertial.

In order to improve the quality of control it is necessary to turn from the single-loop automated control systems to more complex systems using additional (correction) perturbations impulses or secondary output coordinates. In this case, we used a cascade ACS. Block diagram of the cascade ACS $c$U$^{РФ}$ is presented in Figure 1.
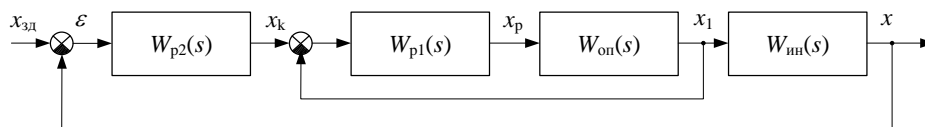


$c$U$^{РФ}$; $W_{р1}(s)$, $W_{р2}(s)$ - transfer functions of correcting and stabilizing regulators, respectively; $W_{on}(s)$ - transfer function of the advanced part of the object; $W_{on}(s)$ - transfer function of the inertial part of the object

**Figure 1.** Block diagram of the cascade ACS

From the approximate determination of the stabilizing regulator settings we turn to determination of correcting regulator, which circuit includes a regulator with known setting parameter [8].

Experimental study of EC-1 ACS was carried out using a computer model ACS. Perturbations were supplied to system input by all channels, and control quality was estimated.

The analysis of the experiments showed that regulator of the inner loop of cascade $c$U$^{РФ}$ ACS and the regulator of DPD ACS cannot be applied with the received settings. This system could damage the actuator, so the control coefficient was reduced in order to obtain a smoother signal at the regulator output.

The results of the experiments are presented in Figure 2 – 5. All perturbations were supplied at time moment $t_0 = 1000$ c.

In comparison to the single-loop ACS the cascade $c$U$^{РФ}$ ACS showed better quality indicators for the perturbations response of OP consumption, and the change of uranium concentration in the AP.
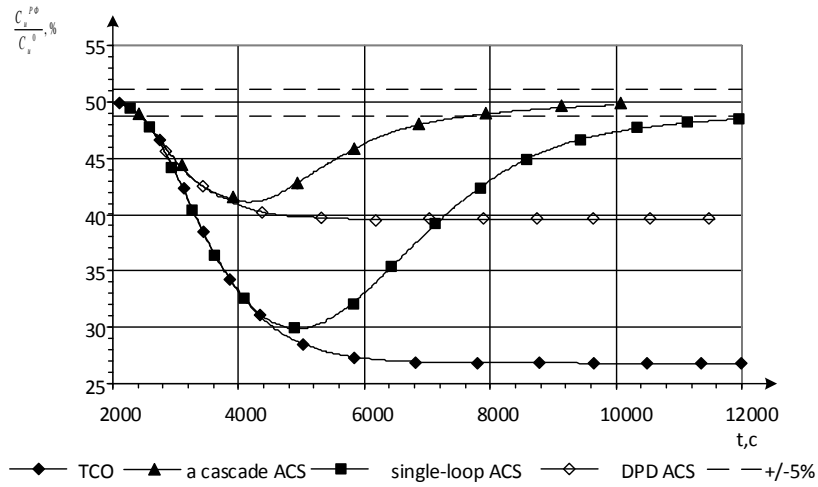
**Figure 2.** Response of TCO to 5 % perturbations and its influence on OP flow in the cascade, single- loop $c$U$^{\text{РФ}}$ ACS and DPD ACS
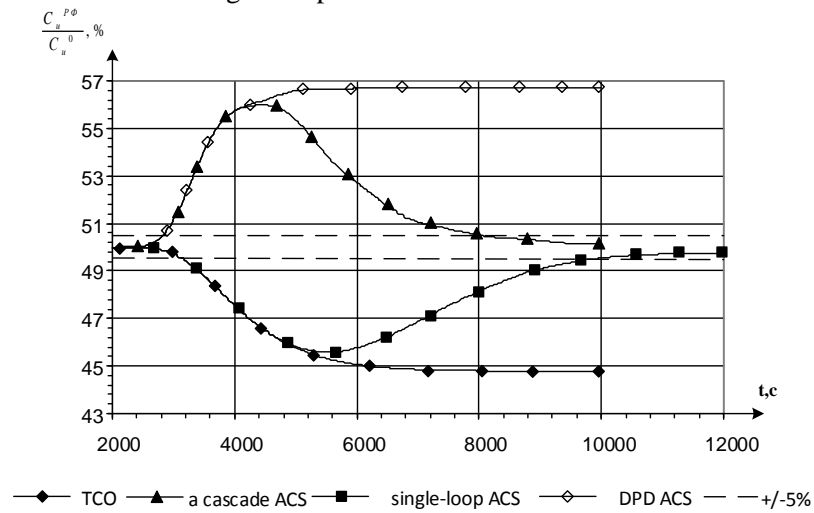


**Figure 3.** Response of TCO, cascade ACS, single-loop $c$U$^{\text{РФ}}$ ACS and DPD ACS to the 5 % perturbations and its influence on uranium concentration in AP
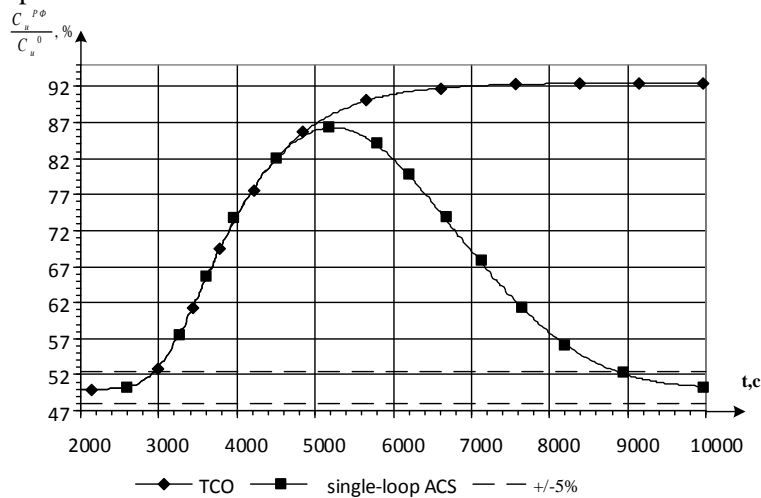


**Figure 4.** Response of TCO and single-loop $c$U$^{\text{РФ}}$ ACS to 7 % perturbations and its influence on AP6 flow rate
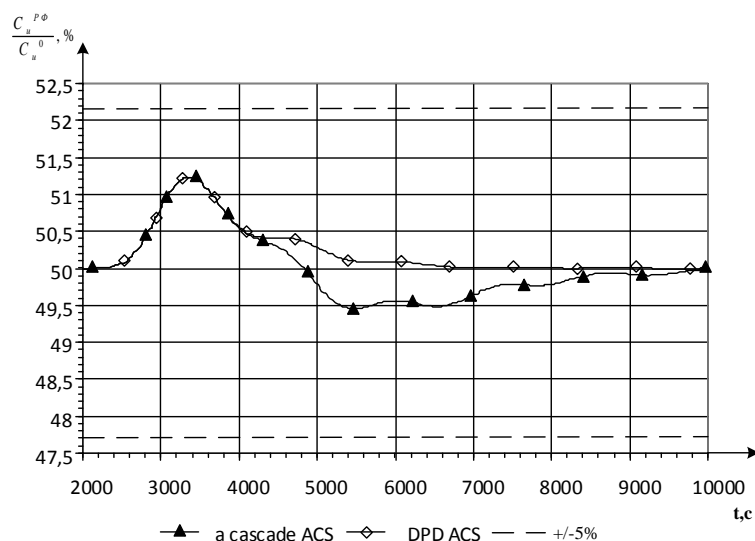
**Figure 5.** Response of cascade $c\mathrm{U}^{\mathrm{РФ}}$ ACS and DPD ACS to 7 % perturbations and its influence on AP6 flow rate

## 4. Conclusion

A dynamic multi-component model based on the solutions of the revised convective diffusion equations under the accepted assumptions, was developed. Hydrodynamic processes in the column were calculated using the results of work.

The comparison of the model to the real apparatus proved its adequacy in a wide range of coordinate variation. On this basis, uranium concentration ACS in EC-1 raffinate was synthesized.

The developed automated control system of the extraction process will significantly improve technical and economic indicators of production by increasing the purity degree of the produced uranium and reducing the amount of uranium in the raffinate. ACS is recommended for implementation.

The work resulted in the development of a universal multi-component model which can be used for the extraction columns operating in different modes and with different types of spent nuclear fuel.

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