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MATHEMATICAL MODEL OF A RECORD TYPE DEVICE FOR VALUED COMPONENTS RECOVERY FROM END PROCESS GASES OF URANIUM HEXAFLUORIDE PRODUCTION

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Mathematical model of the device for valued components recovery from end gases of sublimate production including hydrodynamics, thermodynamics and kinetics of interaction process of solid and gaseous phases realized in the package MATLAB has been presented. Static and dynamic characteristics of the device as a control object necessary for control algorithm synthesis are obtained and analyzed.

One of the aspects of federal program on Russian energy industrial complex development in 2007–2010 and in perspective up to 2015 is the accelerated development of country nuclear power engineering (RF government regulation from October 6, 2006 № 605). In this connection there is a task of increasing productivity and efficiency of functioning enterprises of nuclear fuel production for NPP and improving their indices in safety and ecological compatibility.

At the present time uranium hexafluoride (UHF) is one of the main intermediate products in nuclear fuel cycle. The unique properties of UHF, the only volatile gaseous compound of this element, are widely applied in gas-diffusion and centrifugal processes of uranium isotopes separation for obtaining the product enriched with isotope U^{235} .

In UHF production three main stages may be determined:

- UHF synthesis in flame fluoridation reactor;
- solid UHF extraction in desublimation junction;
- recovery of fluorine, hydrogen fluoride and UHF from end process gases in recovery junction [1].

Automated control systems of technological process in development and modernization of which Tomsk Polytechnic University takes part are successfully used and continuously improved at domestic sublimate plants. One of the variants of recovery junction is the device of combined type recovery (DCR), Fig. 1. The method of recovery realized in DCR is called «combined» by the device developers as it combines the main features of «boiling» layer with a general restricted particle fall in ascending gas flow [2]. Modernization of automated control system of DCR resulted in necessity of improving control algorithm that is impossible without developing its mathematical model describing both static and dynamic modes.

DCR mathematical modeling was carried out on the basis of the analyzing recovery process kinetics, hydrodynamics and thermal conditions in the device using modern computation techniques in the following sequence:

1. Compiling process mathematical description in the device taking into account the peculiarities of chemical conversion and thermal processes, flow hydrodynamics and reaction space structure [3, 4].

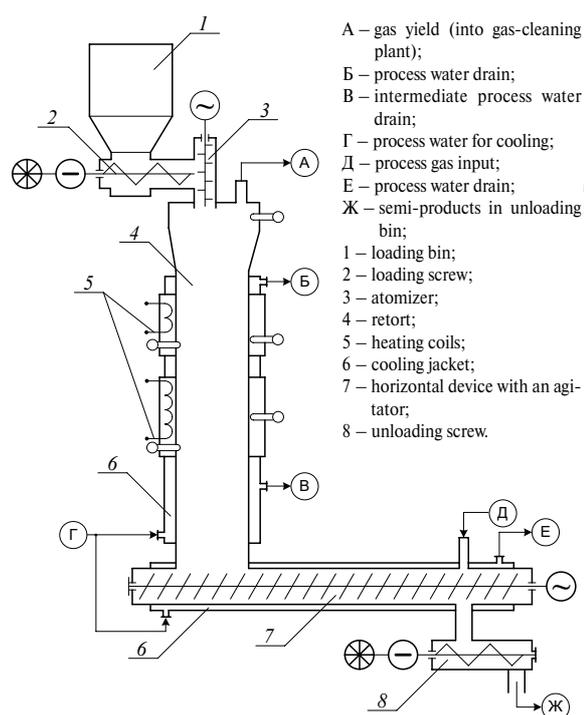


Fig. 1. Diagram of DCR device

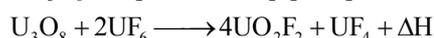
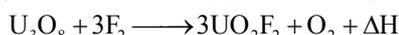
2. Partitioning physicochemical system into levels in each of which the regularities of process behavior are invariant to the scale; and studying the whole system in parts by means of consecutive transfer from lower level to the upper one [5].
3. Determining essential connections and parameters, ascertaining assumptions for simplifying the developed model.
4. Accounting the influence of space and time factors that requires in general case construction of dynamic model with distributed parameters.

DCR refers to the class of catalyst chambers with disperse flow and has a number of design and process features which should be taken into account at its modeling for studying static and dynamic properties as well as for solving the problems connected directly with automated control of recovery junction at sublimate production.

Hydrodynamic mode in DCR is complicated and characterized by the following peculiarities:

1. Low volume concentration of solid particles in reaction space ($\beta \leq 0,003$) that allows referring such disperse system to the class of gas suspension.
2. Gas variable velocity by retort length stipulated by component recovery from the original gas mixture and change of its temperature.
3. Counterflow of gas and solid phases which improves the conditions of heat- and mass exchange results in increasing contact time for solid particles, supports more uniform distribution of reaction velocities temperature along the reactor length in comparison with uniflow and increases the degree of reactor volume filling with solid phase.
4. Polydispersity of raw materials – particle size varies in wide range that results in different velocities of pseudo-motion for particles of different diameter and their multidirectional movement. This fact influences also thermal conditions in the device retort.

Recovery process in DCR is described by chemical reactions [1]



and characterized by intensive heat release; hydrodynamic regime combining properties of boiling and falling layer supports good conditions for interphase heat exchange and heat exchange with retort wall, therefore at small size of the particles of solid raw material heat equilibrium between gas and solid occurs practically simultaneously in comparison with the time of particle being in reactor and chemical reaction [6]. The whole system in this case may be presented by quasi-homogeneous media with efficient coefficients of diffusion and heat conductivity. Temperature at the reactor length has a distributed character and its dynamic in the range of maximal values – reactor «hot zone» stipulated by nonstationarity of fluorine concentration in gas (5...40 rev. %) should be especially taken into consideration. Temperature in this region may rapidly achieve critical values therefore, the task of its preliminary investigation at the stage of modeling and determination of maintenance method of DCR specified temperature conditions intended for industrial use is actual.

Block diagram of DCR as a technological control object is presented in Fig. 2. Degrees of valued component recovery and fluorine ion content in semi-product are output controlled variables (y). Consumption of solid raw material and coolant are control variables (u). Consumption of process gas, its temperature, concentration of gas components, entering to recovery as well as granulometric composition of solid raw material are disturbing variables (f). Regime variables: temperature of reaction volume and degree of its filling up with solid phase.

Studying ideal models [4]: ideal mixture and displacement revealed their disadvantages and resulted in necessity of accounting phase counterflow, heat transfer and temperature distribution, delays in «passing» perturbations and different parametric sensitivity by DCR

length. Therefore the quasi-homogeneous model of incomplete mixture was proposed for studying technological process in DCR, calculating possible steady-state and transient conditions. The model takes into account counterflow character of the system, reactor body cooling, retort height variable, gas velocity, probability of changing boundary and original conditions reflecting change of real variables of control object.

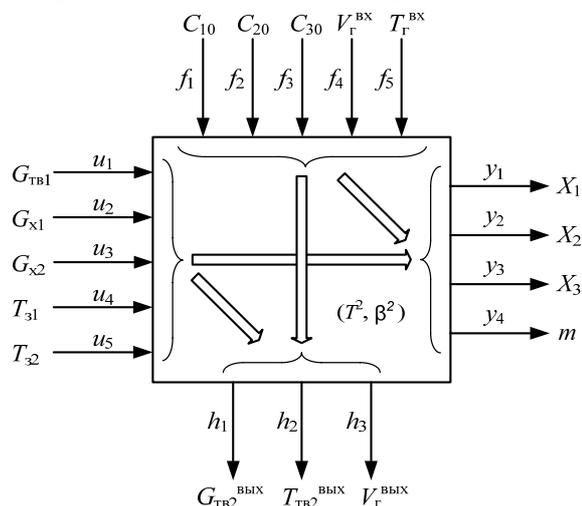


Fig. 2. Block diagram of DCR as a control object

The equation system has the form:

$$\left\{ \begin{array}{l} -u \frac{\partial X_i}{\partial l} + W_i(X_i, T) = \varepsilon \frac{\partial X_i}{\partial t}, \\ \lambda_{\text{эф}} \frac{\partial^2 T}{\partial l^2} - \frac{\partial [(\varepsilon c_r u - \beta c_r (u_b - u))]}{\partial l} + \\ + \sum_{i=1}^3 \Delta H_i C_{i0} W_i(X_i, T) - B(T - \Theta) + kP = c_p \frac{\partial T}{\partial t}, \\ -u_x \frac{\partial \Theta}{\partial l} + B_1(T - \Theta) = \frac{\partial \Theta}{\partial t}, \\ u = u_0 \left[\begin{array}{l} \frac{T}{T_{\text{bx}}} - 0,005 C_{10} X_1 - 0,00827 C_{20} X_2 - \\ -0,01265 C_{30} X_3 \end{array} \right], \\ B = \frac{K_T F}{V_p}, \\ B_1 = \frac{B V_p}{c_x V_x}. \end{array} \right.$$

Boundary conditions: $l=0: X_i=X_{i0}; \Theta=\Theta_0$,

$$\lambda_{\text{эф}} \frac{\partial T}{\partial l} = (\varepsilon c_r u - \beta c_r (u_b - u))(T - T_0),$$

$$l=L: \frac{\partial T}{\partial l} = 0.$$

Original conditions: $t=0: X_i=X_i^0; T=T^0; \Theta=\Theta^0$.

Description of variables included into equation system: u_0, u, u_x, u_e are the velocities of gas at retort input,

gas by retort height, coolant, solid particle soaring (m/s); X_i is the conversion degree of i component; W_i is the observed velocity of i reaction (s^{-1}); T is the temperature of disperse system (K); ε is the portion of free volume in retort; λ_{eff} is the effective heat conductivity coefficient ($W/(m \cdot K)$); c_p, c_r, c_s, c_x are the volumetric heat capacities of disperse system, gas, solid raw material and coolant ($J/(m^3 \cdot K)$); ΔH_i is the thermal effect of i reaction ($J/(m^3 \cdot rev. \%)$); C_0 are the input component concentrations (rev. %); Θ is the temperature of the coolant (K); L is the height of retort (m); V_p and V_s are the volumes of retort and cooling system (m^3); F and F_x are the areas of retort surface and cooling system (m^2); β is the degree of retort filling up with solid phase; K_r is the coefficient of heat transfer from disperse system to the coolant ($W/(m^2 \cdot K)$); X_0, Θ_0 are boundary values of recovery degree of i component and coolant temperature; X_i^0, T^0, Θ^0 are the initial values of recovery degree of i component, disperse system temperature and coolant temperature.

It is difficult to solve the equation system of the model by analytic methods therefore the finite-difference method was used for its solution [7]. To solve the finite difference equation system obtained by applying this method the distillation method was used [7]. Software implementation of the model was carried out in the package MATLAB 7.0.

Probable steady-state conditions of DCR were computed for different original data and parameters, Table 1, at stated peculiarities. The computed temperature distributions are given in Fig. 3.

Table 1. Original data for computing steady-state conditions in DCR

№	Perturbations					Control actions			
	C_{10}	C_{20}	C_{30}	u_0	T_{bx}	B	$B_i \cdot 10^4$	u_x	$\beta \cdot 10^4$
	об. %		м/с			Вт/(К·м³)		—	
1	20	20	2	0,11	540	2,500	57	0,014	28
2	20	20	2	0,21	520	0	0	0	28
3	20	20	2	0,21	505	0,250	5,7	0,030	28
4	20	20	2	0,21	505	0,150	3,4	0,014	28
5	10	20	1	0,14	480	0,150	3,4	0,014	20
6	5	10	1	0,18	310	0,085	1,8	0,004	18
7	17	32	1	0,12	430	0,125	2,7	0,010	18

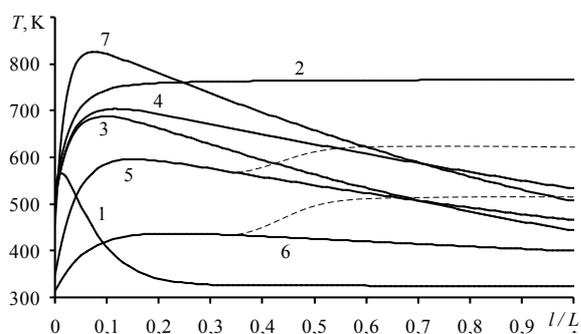


Fig. 3. Temperature distribution by retort height of DCR. Curves numbers correspond to Table 1

The analysis of the obtained results allows making the following conclusions:

1. There is a zone of maximum heat release and maximal intensity of recovery process, especially F_2 and UF_6 the length of which is approximately $0,25...0,3L$. Fluorine concentration C_{20} influences most of all the temperature value in this zone; heat release (values B and B_i) and gas input temperature influences weaker however their influence in the rest part of the reactor is significant.
2. In zone of maximal temperature $0,03...0,18L$ its dependence on concentration of the determinative component C_{20} close to linear one at constant parameter of heat removal B is observed. Changing concentration of C_{20} by 10 rev. % the temperature changes by 65...70 K. Range of maximal temperature change in this zone is from 453 to 753 K.
3. Influence of heat removal on temperature in DCR retort is illustrated.
4. Influence of probable reactor body heating on the area of its length $0,34...1L$ (dashed line in Fig. 3) is shown. Temperature of the heater Θ^* contacting with the wall amounted to 573 K.

Thus, applying cooling jacket of variable length and heating reactor body at $l \geq 0,3...0,37L$ and insignificant heating of solid raw materials in loading junction allows developing the required temperature profile in DCR.

To study dynamic characteristics of DCR as a control object the transient conditions in retort at different effects of stick-slip nature were computed. The values of component concentrations, gas consumption and temperature, the degree of filling up and parameters of heat removal corresponding to original steady-state conditions as well as stepwise changes of input variables or parameters are given in Table 2.

Table 2. Original data for computing transient conditions in DCR

№	Initial steady state								Change of variables	
	C_{10}	C_{20}	C_{30}	u_0	T_{bx}	B	$B_i \cdot 10^4$	$\beta \cdot 10^4$		
	rev. %		m/s		K		W/(K·m³)		—	
1	10	25	2	0,11	540	0,085	1,8	12	C_{20}	-15
2	10	10	2	0,11	540	0,085	1,8	28	C_{20}	15
3	20	20	2	0,14	540	0,100	2,1	28	C_{20} u_0	-10 -0,03
4	20	20	2	0,14	540	0,100	2,1	28	C_{20} u_0 B	-10 -0,03 0,02
5	5	10	1	0,18	340	0,125	2,7	18	C_{20} u_0	10 0,14
6	17	32	1	0,12	430	0,125	2,7	18	β	6
7	17	32	1	0,12	430	0,125	2,7	18	β	-6

The influence of stepwise changes of perturbation action was studied: concentration of fluoride C_{20} and gas velocity u_0 as well as control action: degrees of filling up DCR retort with solid phase β and parameter of heat removal B on temperature profile by retort height and degree of gas component recovery. Diagram of temperature reactions and degree of recovery of hydrogen fluoride

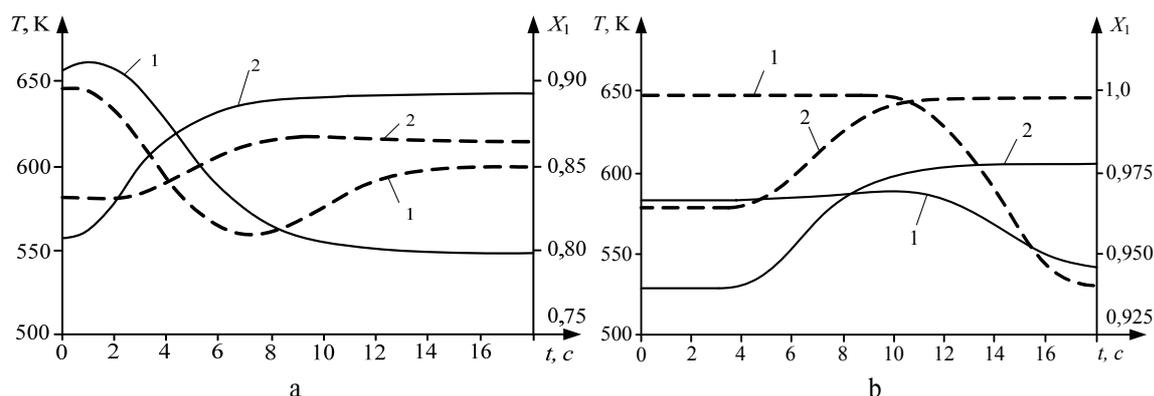


Fig. 4. Transients by temperature (—) and recovery degree X_1 (----): a) on the level 0,18L; b) at the output of the device

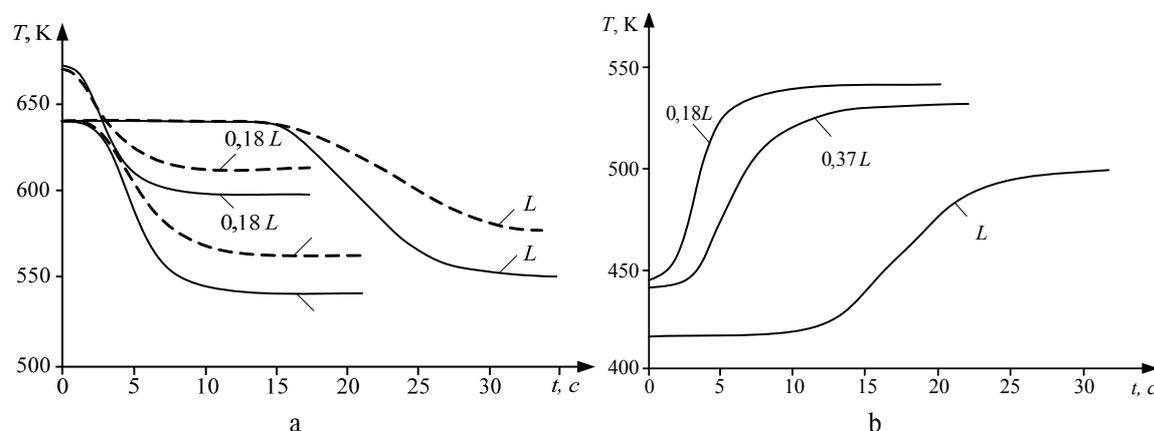


Fig. 5. Transients by temperature in various points of retort for variants of influences: a) 3 (—) and 4 (----); b) 5

as a component recovered rather worse than others on a change of stated effects (transients) for various points by retort height of DCR are given in Fig. 4–6.

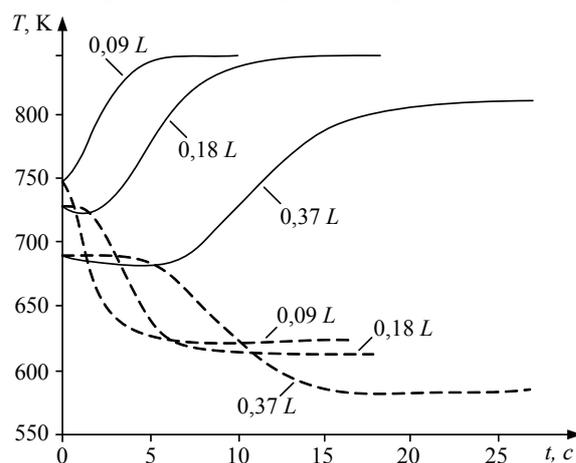


Fig. 6. Transients by temperature in three points of retort for variants of influences 6 (----) and 7 (—)

The highest sensitivity of temperature and recovery degree X_1 to the change of input actions is observed by retort length from 0,07 up to 0,3L, that is in the region of «hot point». Duration of transients on the level of 0,95 and delay on the level of 0,1 from the stated values of the variable in the channel « $C_{20} \rightarrow T$ » amount to:

- in the point 0,18L – $t_n = 7...15$ s; $\tau_n = 0,5...2$ s;
- at retort output – $t_n = 14...32$ s; $\tau_n = 7...20$ s.

For transients changing degree of filling up (Fig. 6) it was accepted that β changes step-wise along the whole reactor that is the passage time of solid phase from loading screw to retort concrete point was not taken into account. Value of this time was considered as a transportation lag τ_{tp} further summed with a value of pure time delay τ_n . Depending on gas velocity in retort the value τ_{tp} for point 0,18L amounts to 90...120 s; for retort output $\tau_{tp} = 40...50$ s. Value of pure time delay is rather lower: 1...2 s for point 0,18L and 8...15 s for retort output.

Investigation of transient conditions allows making two main conclusions:

1. Transients in the region of maximal temperatures differ in highest sensitivity to the input actions and the lowest inertia therefore it is necessary to control temperatures inside retort in this region by means of fast-response transducer.
2. Changing concentration by 15 rev. % corresponds to temperature change in this region by 90...110K, decrease of filling up degree by one third results in temperature increase by 110...120K.

Thus, temperature sensitivity in retort is comparable for the main input actions – perturbation – C_{20} and control G_M .

While modeling the influence of the main perturbation actions (fluoride concentration and gas consumption) and possible control actions (raw material loading, heat removal intensity and heating) on stationary and transient conditions in the device was estimated.

The developed mathematical model implemented in the package MATLAB 7.0 will be used for synthesizing modernized control algorithm of DCR. Improvement of algorithm is in automated stabilization of the required tempera-

ture distribution profile in the device retort by changing raw material loading in the device. Control algorithm will be implemented at modern software with software support at high-level object-oriented programming language. The model may be also used for studying other control objects in radiochemical technology of counterflow and uniflow devices with gas and disperse fluxes (for example, flame reactors for obtaining UHF, counterflow devices for processing product unreacted in fluoridation reactor etc.).

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MATHEMATICAL SIMULATION OF WATER AND METHANOL SEGREGATION PROCESSES AT FIELD PREPARATION OF GAS CONDENSATE

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Calculation modulus for water and methanol segregation processes from gas condensate has been developed. Applying the developed technological system the influence of process variables on segregation processes of water and methanol is studied. Modes of operation of liquid separators at which the most efficient segregation of water-methanol solution from unstable condensate occurs are recommended.

When producing and preparing gas condensate for transportation the supply of raised concentration methanol to the system is provided. When mixing hydrocarbon condensate with water methanol solution the mixture which should be divided is formed. Indistinctness of division of water-methanol and hydrocarbon phases results in considerable losses of methanol with unstable condensate and tank gas.

Applying methods of mathematical modeling and using computer analogues (A) allows solving these problems and many other ones rather efficiently.

On the basis of experience in developing analogues for various technologies we developed the mathematical models [1–4] of the processes of commercial treating of oil, gas and gas condensate on the basis of which the technological analogue (TA) for computing material balances and on-line analysis of operating practices of complex gas treatment device (CGTD) was developed.

The main blocks of the developed TA of complex gas and gas condensate treatment technology are modulus of computing processes of separation, tip leakage, liquid separation, throttling and heat exchange.

Constants of phase equilibrium were calculated by the technique proposed in papers [5, 6] on the basis of which there is the main Gofman-Kramp equation. For calculating constants of phase equilibrium of methanol and water the equation of Teck-Still was chosen [7].

The given correlation differs in high accuracy at calculating vapor pressures of polar substances and substances with hydrogen bonds at low temperatures. The equation of Teck-Still for calculating saturated vapor pressure of chemical substances has the following form [7]:

$$\ln P_{VP} = A \left(1,14893 - \frac{1}{T_r} - 0,11719 \cdot T_r - \right. \\ \left. - 0,03174 \cdot T_r^2 - 0,375 \cdot \ln T_r \right) + \\ + (1,042\alpha_c - 0,46284A) \times \\ \times \left[\frac{T_r^{5,2691+2,0753 \cdot A-3,1738 \cdot h} - 1}{5,2691+2,0753 \cdot A-3,1738 \cdot h} + \right. \\ \left. + 0,040 \cdot \left(\frac{1}{T_r} - 1 \right) \right], \quad (1)$$