Simulation of Uranyl Nitrate Crystallization Process in Linear Crystallizer Using Simsar Software

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Abstract. The paper deals with simulation of linear crystallizer work process for the research of technic operating modes and searching the most effective for material's nano-purity achievement. The model is realized by using SimSar software. Importance of device's geometry and process variables are marked. The model was included in the complex's composition of closed nuclear fuel cycle.

1. Introduction

Development of modern technology for nuclear waste handling is focused on getting of material in the pure state (nano-purity). In chemical industry, extraction [1] and crystallization [2] processes are used for this thing. The crystallization process is a complex process involving solid phase in the crystal form. Phase transformations and heat and mass transfer processes forming the conditions at the interface play a key-role in the formation of the material properties. To properly grow uranyl nitrate hexahydrate (UNH) crystals and operate uranium crystallization processes, it is important to carry out comprehensive studies of the phenomena involved in. For these reasons, crystallization of uranyl nitrate was studied in the papers of scientists from Russia and Japan [3-7]. A prominent role here is given to the theoretical studies using different mathematical model. The relevance of the task is related to the modeling of the crystallizer work process and detection by the method of the virtual experiment of unsteady regimes of its behavior. The virtual experiment implementation is due to the complexity of setting real experiments and a lack of knowledge about the process. At the present time a stand for refining of extraction and crystallization treatment process of spent nuclear fuel (SNF) is developing in Russia in the framework of "Proryv" project stream. In needs a computation-intensive for the model developing of all stand's parts and concurrent operation. It is expected of chemical process modeling and hardware modeling at the same time. The scope of the paper is the modeling of UNH crystals growth in linear crystallizer. SimSar software developing for modeling of closed nuclear fuel cycle (CNFC) production is used for end of purpose.

2. Mathematical Model of UNH Crystals Growth in Linear Crystallizer

A liquid nitric-acid uranyl nitrate solution (liquid solution) is ideally considered as a three-component liquid:

$$C^m_{UN} + C^m_{HNO_3} + C^m_{H_2O} = \sum_{i=1}^{NC} C^m_i = 1 \quad (1)$$

where C^m_i is a mass content of the i-th component in the liquid solution. Mass content values of uranyl nitrate (UN) and nitric acid (HNO_3) are evaluated by initial conditions of uranium (U) and HNO_3 concentrations.

The dependence between C^m_{UN} and $C^m_{HNO_3}$ can be described [7]:

$$C^m_{UN} = A - \psi \cdot C^m_{HNO_3},\ A = 1/(1+\alpha) = 0.785 \quad (2)$$

where the parameter α is the ratio of six molar mass of water to the molar mass of UN (it needs six molecules of water per the one molecule of UN to get the one crystal of uranyl nitrate hexahydrate). $\psi = (A - C^{m,int}_{UN}) \cdot C^{m,int\ -1}_{HNO_3}$ - the slope of the line, determined by the composition of the initial melt.

The growing rate of the crystal radius R is stated as follows:

$$\frac{dR}{dt} = k(C^m_{UN} - C^{m,sat}_{UN}) \cdot \frac{L}{U_s} - R \quad (3)$$

where k - the growth rate of the crystal phase inclusions (in the general case k is the function of temperature and liquid phase composition); U_s - a speed of solid phase movement; $C^{m,sat}_{UN} = C^{m,sat}_{UN}(T, \psi)$ – mass fraction of UN saturation in liquid solution (the function of temperature and composition).

Liquid and solid phases in the crystallizer active volume move with different velocities owing to their mass densities difference. The velocity of solid phase movement $U_s(t)$ can be written by the following expression considering Stokes law:

$$U_s(t) = U_{liq} + \frac{2}{9}\frac{g}{\upsilon} R^2 \left[\frac{\rho_s}{\rho_{liq}} - 1 \right] \quad (4)$$

where U_{liq} - liquid velocity; g – gravity; υ - liquid phase kinematic viscosity (function of the temperature and composition); ρ_s and ρ_{liq} – mass densities of solid and liquid phases, respectively.

The density of the liquid phase [5]: $\rho_{liq} = [1.023 + 2.936 \cdot 10^{-2} \cdot C_{HNO3} + 1.313 \cdot 10^{-3} \cdot C_U - (4.681 \cdot 10^{-4} + 3.475 \cdot 10^{-5} \cdot C_{HNO3}) \cdot T] \cdot 10^3$.

The conservation principle applied for UN and HNO_3 leads to the Eq. (5) and Eq. (6).

$$\frac{dC^m_{UN}}{dt} = \frac{Q_0 \cdot C^{m,int}_{UN} - U_{liq} \cdot S \cdot (1 - 4/3 \cdot \pi R^3(t) n) \cdot C^m_{UN} - U_s \cdot S \cdot 4/3 \cdot \pi R^3(t) n \cdot A}{U_{liq} \cdot S \cdot (1 - 4/3 \cdot \pi R^3(t) n)} \quad (5)$$

$$\frac{dC^m_{HNO3}}{dt} = \frac{Q_0 \cdot C^{m,int}_{HNO3} - U_{liq} \cdot S \cdot (1 - 4/3 \cdot \pi R^3(t) n) \cdot C^m_{HNO3}}{U_{liq} \cdot S \cdot (1 - 4/3 \cdot \pi R^3(t) n)} \quad (6)$$

where Q_0 is an initial volumetric flow rate; S – sectional area; n - amount of crystallization centers.

The energy balance for the active volume of the crystallizer is stated as follows and provide the temperature variation due to enthalpic bulk contributions, internal heat exchange and external (cooling jacket) heat exchange contributions:

$$\frac{dT}{dt} = T_{in} - T - \frac{\pi \cdot D \cdot \chi \cdot (T - T_{cj})}{\rho_{liq} \cdot C_{liq} \cdot Q_0 \cdot (1 - 4/3 \cdot \pi R^3(t) n) + 4/3 \cdot \pi R^3(t) n \cdot C_s \cdot \rho_s \cdot S \cdot U_s} \quad (7)$$

where D – diameter of the cross section of the crystallizer active volume; C_s and C_{liq}– specific heat of solid and liquid phases; χ – heat conductivity of the media (solid + liquid phases) in the considered crystallizer working volume section; T_{cj}- temperature of the cooling jacket.

A resulting model of crystallization process was adapted for modeling of a concrete device:

- achieved differential equations is represented in Cauchy's form;
- device's geometrics are taken into account;
- a device's work process is implemented for all source and target faucet conditions;

- a heat exchange model is implemented.

3. Virtual Experiments

A resulting model is implemented in SimSar software. Figure 1 and 2 show a model general form. Rate is 1 l/h, temperature of cooling jacket in the 1 and 2 zones is 2 and 1.5°C properly.

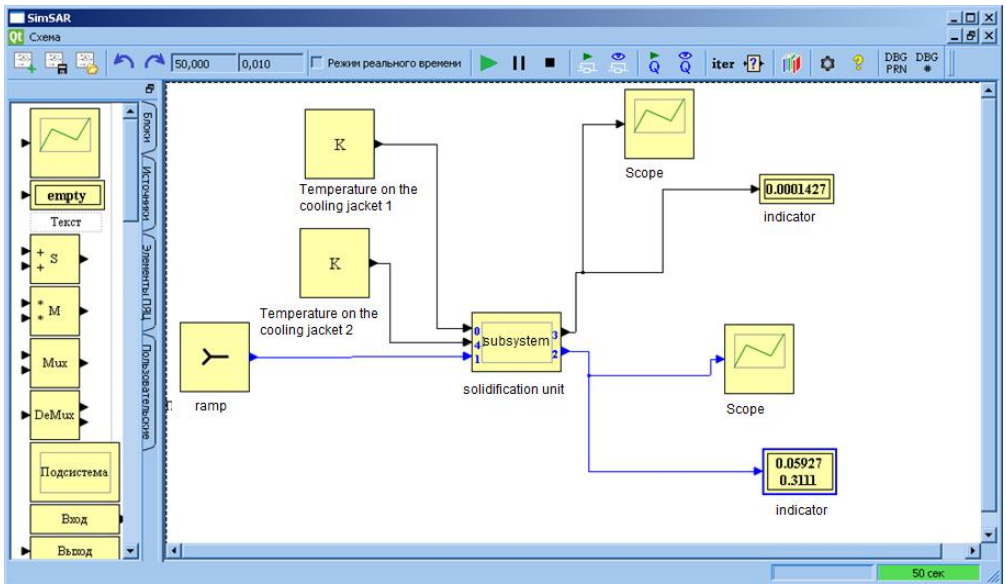

Figure1. A general form of crystallizer in SimSar software.

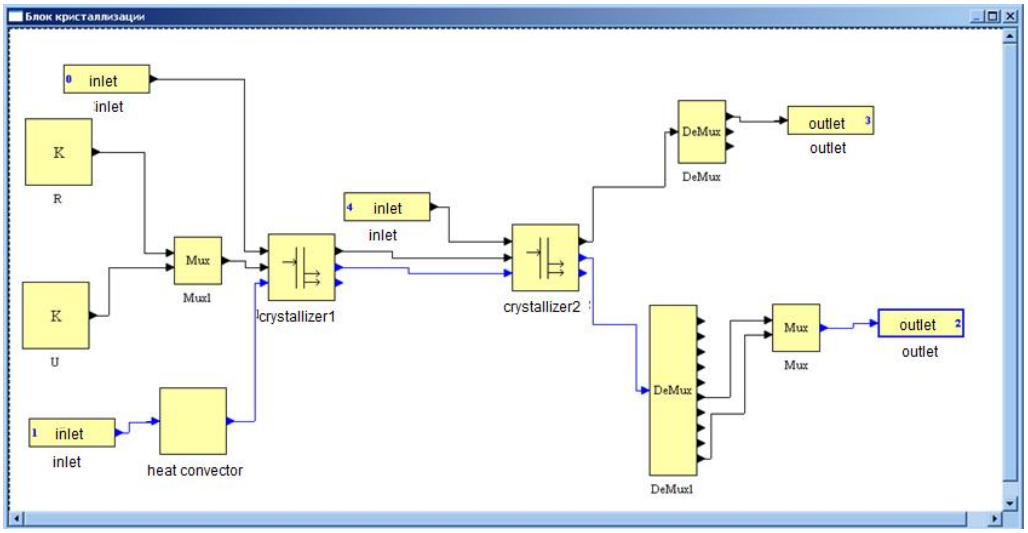

Figure 2. A decomposition of crystallizer's active volume on zones in SimSar software.

Results of simulation are showed in Figures 3 and 4. Figure 3 presents the trends of UN and HNO_3 mass fractions in the liquid phase % depending on time. A step-function response is not monotonous due to dependence between UN and HNO_3 concentrations. Figure 4 presents trend of UNH crystal growth.

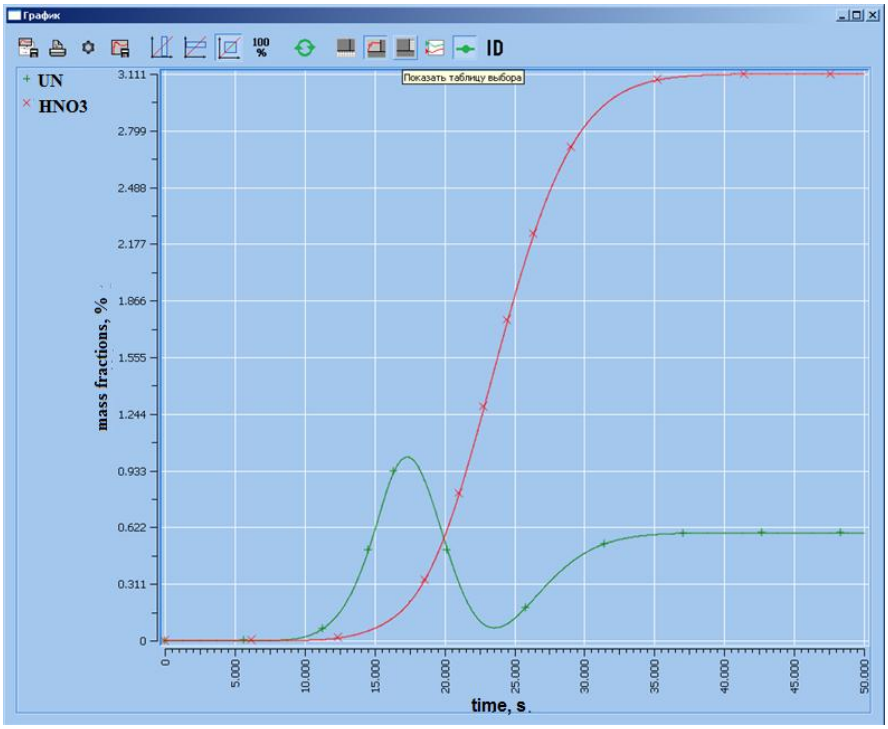

Figure 3. UN and HNO3 mass fractions changing over time.

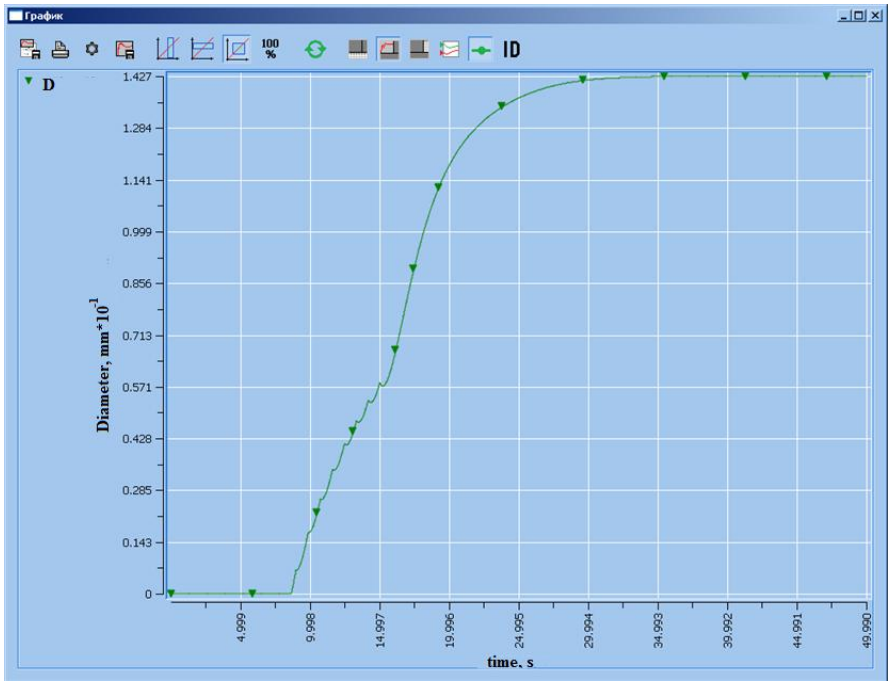

Figure 4. UNH crystal growth on time.

Model of linear crystallizer was implemented in Matlab Simulink for validity check of SimSar. (Figure 5)

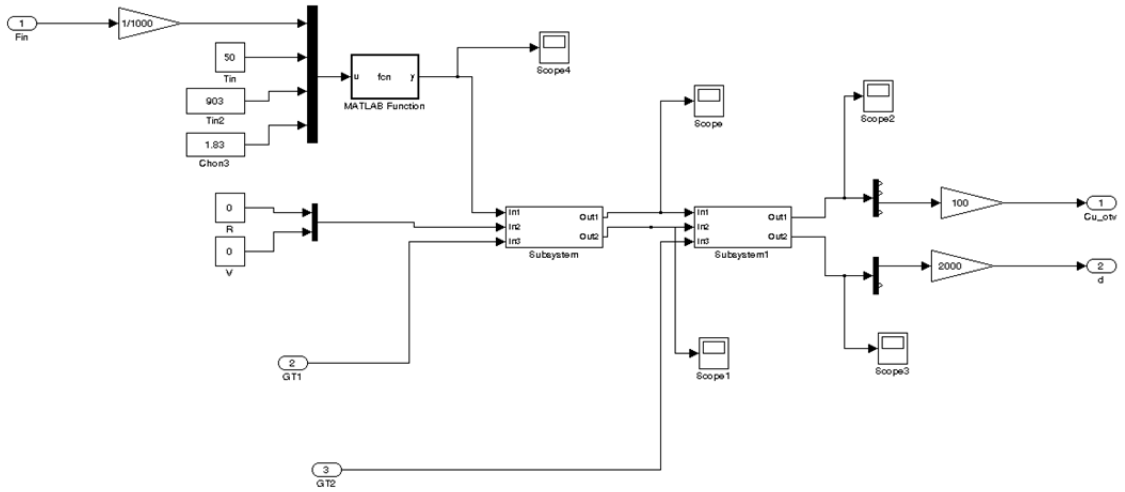

Figure 5. A model's implementation in Matlab Simulink.

Results of simulation in SimSar software and Matlab Simulink are equal (Figure 6). There are used the same ode1 equation solver and the one of simulation step. SimSar's rate of computing 26.2 times faster than Matlab (Table 1). The model of linear crystallizer in SimSar is developed on C++, but in Matlab Simulink it is developed by s-function. The model is failed verification due to deficiency of the real device, but results of simulation are equal with results getting in A. A. Bochvar All-Russian Scientific Research Institute for Inorganic Materials.

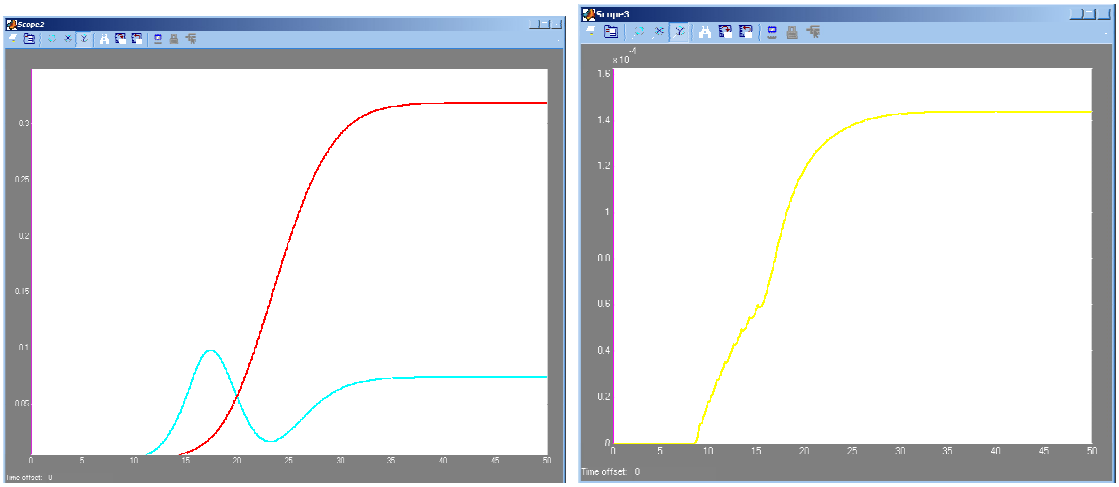

Figure 6. Results of simulation in Matlab Simulink.

Table 1. Inlet and outlet simulation characteristics in different software

	SimSar	Matlab Simulink
Simulation time (s)	50	50
Equation solver	ode1	ode1
UN mass fraction is coming out in outlet (%)	0.006	0.006
HNO_3 mass fraction is coming out in outlet (масс. %)	0.031	0.031
Diameter of UNH crystals (mm)	0.14	0.14
Rate of computing (s)	0.678	17.76

4. Conclusion

The model analyzing different temperature regimes of crystallization process allows choosing the most effective operating modes of linear crystallizer for material's nano-purity achievement. Results of simulation are equal with results getting in A. A. Bochvar All-Russian Scientific Research Institute for Inorganic Materials. Simulation of crystallizer work process in SimSar software faster than Matlab Simulink, thus this fact allows to integrate the model of linear crystallizer in calculation complex of CNFC production. Further, it is planned to test model against real data.

5. References

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