Control System of Storage Containers Filling in the Uranyl Nitrate Crystallization Process in a Linear Crystallizer

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This article presents a model of a storage container of linear crystallizer. On the base of this model two independent control system of storage containers filling in a uranyl nitrate crystallization process were suggested. The use of the proposed control systems is necessary for the timely switching of the storage container to an empty one. For the proposed control systems, simulation results are presented in the SimSar software. The developed mathematical model and the control system could be used for optimization process during the design and adjustment of equipment.

1. Introduction

The development of modern technologies for the processing of spent nuclear fuel (SNF) involves the production of substances in pure form. The principal peculiarity of SNF processing technology is its high radioactivity, which requires the organization of biological protection for plants and a high level of automation and control over processes. Radiochemical technology is associated with great difficulties in maintenance and repair of equipment. Most technological schemes involve the use of water processes (liquid extraction, ion exchange sorption technology), in which the reagents are largely exposed to radiation, which reduces the possibility of their repeated use. At the moment PUREX-technology of SNF processing, i.e. regeneration of uranium and plutonium by extraction with 30% solution of tributyl phosphate (TBF) in kerosene, is the most widespread. This technology is the most well-studied and effective among all developed to date.

Separation by crystallization has several advantages compared to the PUREX-process. According to Chikazawa et al. (2008), the crystallization process does not require the use of flammable organic solvents, the use of this method is much safer. In addition, this fact removes some requirements for the SNF recycling station. Also, some solvent-related processes, such as washing, can be eliminated, which leads to simplification of SNF recycling technology, as well as reduction of liquid radioactive materials.

The results of application of crystallization in addition to PUREX process as method of uranium purification after its separation from plutonium are described in articles of Henrich et al., 1987 and Shepherd et al., 2015. At the moment works are being conducted in Russia on the launch of the refinery stand to develop the extraction-crystallization technologies of SNF recycling as part of the Proryv (Breakthrough) project. The current state of the development of fast neutron reactors and closed NFC technologies in Russia is considered and major problems are highlighted by Adamov et al. (2017).

The linear crystallizer (Figure 1) is a metal vertical column of constant cross-section, which is connected to two Piston batchers on top and two storage containers on the bottom. According to the Veselov et al. (2015) crystallizer can be divided into three main sections: crystallization section (I), crystal washing section (II), and storage section (III). Piston batchers provide a dosed supply to the upper part of the crystallization section of nitric acid solution containing the target product in dissolved form, heated to a certain temperature. In the

process of moving of the solution down the crystallization column, it is cooled by heat exchange with a cooling jacket. As a result, the saturation concentration of the target product in the solution decreases, the solution is oversaturated and the target product enters the crystalline phase. Due to the higher density of crystals, they move in a vertical volume faster.

In the washing section, partial dissolution of crystals of the target product occurs for the purpose of removing of fission products deposited on the surface of the crystal.

The settling crystalline product is accumulated in the storage container, when filled; it is replaced by switching the rotary valve.
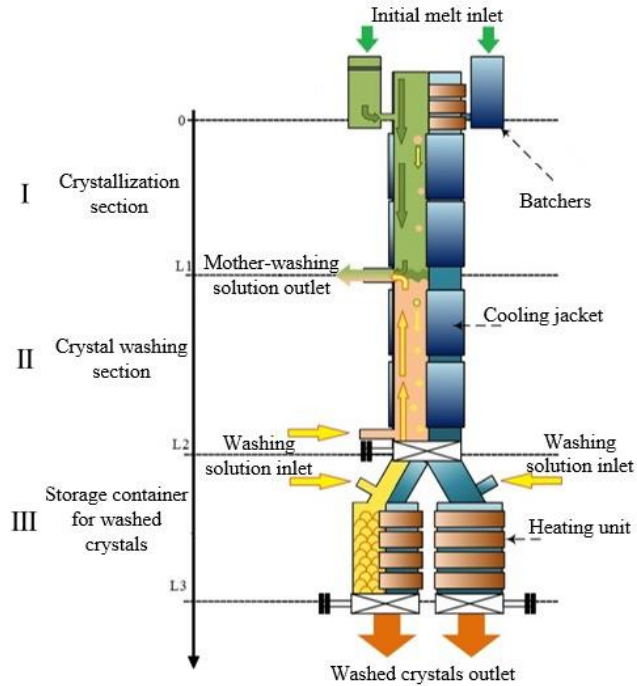

Figure 1: Structure of a linear-type crystallizer

Within the Proryv project a mathematical description of the crystallization process and washing processes was carried out. A cellular model was used to describe the crystallization process in these works. Detailed description and simulation results obtained using this model are described by Ochoa Bique et al. (2016). This work is devoted to the mathematical model of the storage containers and design of control system of storage containers filling in the uranyl nitrate crystallization process in a linear crystallizer.

2. Storage containers

Storage containers (Figure 2) – are cylindrical tanks with a volume of 8 liters, manufactured in accordance with nuclear safety standards. Containers are connected to the linear crystallizer by metal connecting tubes. The uranyl nitrate crystals enter the storage container from the wash zone through the outlet pipe. The product is withdrawn from the container through the opening at the bottom of the container. Before collecting the product, the storage container is heated by heating unit in order to melt the crystallized phase. Two branch pipes are provided for cleaning the storage container and filling it with washing solution.
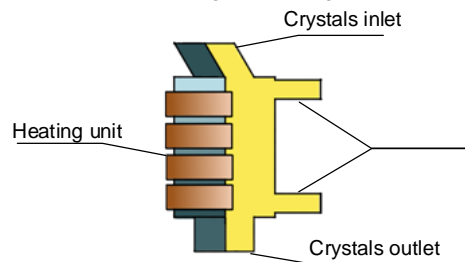

Figure 2: Structure of a storage container

3. Mathematical model of storage containers

The mathematical model of the process of uranyl nitrate crystallization in a linear crystallizer, described by Ochoa Bique et al. (2016), was limited to the description of crystallization and washing section only. As part of this work, this model has been supplemented by a mathematical description of storage containers. The main requirement for this model was its ability to reflect the proportion of products contained therein and the weight of the storage container. It was assumed that the ideal mixing takes place in the container and the crystalline phase is represented in the liquid phase in the form of a homogeneous suspension. Based on the objectives and assumptions made, the capacity was presented as a concentrated model. The informational structural diagram of the storage container is shown in figure 3. the Input variables of this model are the mass flow and density of uranyl nitrate crystals.

Figure 3: Storage container information structure

Table 1: Variable classification

Variable	Name	Note
Q_{cryst}	Mass flow of uranyl nitrate crystals	Input variable
ρ_{cryst}	Density of uranyl nitrate crystals	Input variable
W	Storage container weight	Controlled variable
V_{cryst}	Volume of uranyl nitrate crystals in storage container	Controlled variable
ρ_{cryst}	Density of uranyl nitrate crystals in storage container	Controlled variable
V_{liq}	Volume of liquid phase in storage container	Controlled variable
ρ_{liq}	Density of liquid phase in storage container	Controlled variable

The system of equations describing the storage container has the following form:

$$\begin{cases} W = W_{empty} + V_{liq} \cdot \rho_{liq} + V_{cryst} \cdot \rho_{cryst}; \\ \dfrac{dV_{cryst}}{dt} = \dfrac{Q_{cryst}}{\rho_{cryst}}; \\ \dfrac{dV_{liq}}{dt} = -\dfrac{dV_{cryst}}{dt}. \end{cases} \tag{1}$$

where W_{empty} – weigh of empty storage container;

4. Control systems of the degree of filling of storage containers

One of the important tasks that must be solved by the control system of the uranyl nitrate crystallization process in a linear crystallizer is to control the degree of filling the storage containers of the crystallizer. Nuclear safety design makes it impossible to use standard control methods. The lack of such control can lead to overfilling of the storage container and crystallization of the connecting tubes, the rotary valve and the washing section of the crystallizer, which in turn can significantly disrupt the efficiency of the device or even lead to an emergency. Two independent methods are proposed to be used to ensure that the filled storage container is replaced by an empty container in a timely manner.

The first method is to determine the degree of filling by measuring the weight of the storage container using strain gauge sensor. Application of this method requires a change in the design features of the device, namely the replacement of the connecting tubes of storage containers with a crystallizer for flexible ones. Also, one of

the possible drawbacks is the need for nuclear-safe placement. The second method involves the use of differential pressure sensor which is being developed within the Federal government-sponsored program «Science».

The given method is based on measuring of the pressure difference in the upper and lower part of the storage container. One of the advantages of this method is the absence of the need to change the design features of the device. Connection of the differential pressure gauge is made by means of capillary lines, which will be carried out through the fittings on containers branch pipes (figure 4). It is also worth noting that the capillary lines will allow to install a differential pressure sensor in another room, which is important from the point of view of nuclear safety.

Mathematical modeling of the methods described above, taking into account the error which will be given by real measuring sensors, was made in the software complex KT-NIMFA.

The strain gauge sensor readings were calculated using the following formula:

$$W_{sensor} = W \pm \Delta A \cdot A_{max};$$ (2)

where ΔA – strain gauge sensor measurement error; A_{max} – strain gauge sensor measurement range.
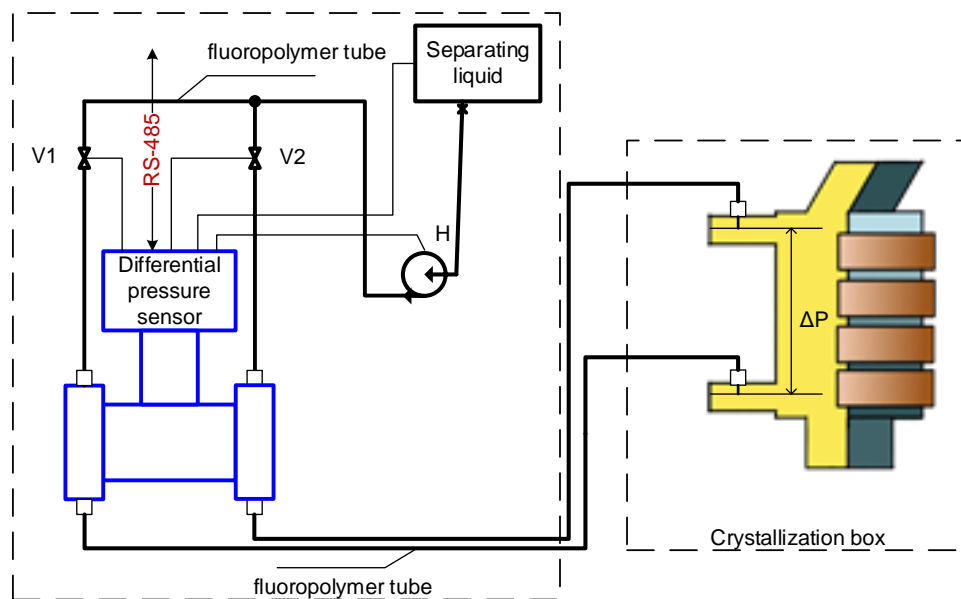

Figure 4: The connection scheme of the differential pressure sensor to the storage container

Based on the previously accepted assumption that the liquid and crystalline phase are in the storage container in the form of a homogeneous mixture, the differential pressure ΔP can be calculated by the Eq(3).

$$\Delta P = \bar{\rho} \cdot g \cdot h;$$ (3)

where g is the gravitational acceleration; h is the height between fittings (in the model is assumed equal to containers height); $\bar{\rho}$ – the average density of the substance in the column.

The average density of the substance was calculated by the Eq(4):

$$\bar{\rho} = \frac{V_{cryst}}{V_{sc}} \cdot \rho_{cryst} + \frac{V_{liq}}{V_{sc}} \cdot \rho_{liq};$$ (4)

where V_{sc} – internal volume of storage container.

Taking into account the error made by the measuring device, the measurement results of the differential pressure sensor can be calculated by the next formula:

$$\Delta P_{sensor} = \Delta P \pm \Delta A \cdot A_{max};$$ (5)

where ΔA – differential pressure sensor measurement error; A_{max} – differential pressure sensor measurement range;

5. Results and discussion

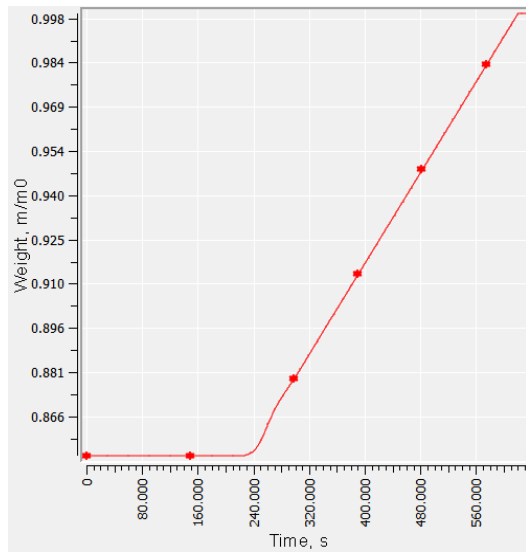The following graph shows the results of the modeling of storage container.

Figure 5: The storage container weight

The results of the measurement were given to the maximum weight of the filled capacity and are presented in conventional units This result allows to track the rate of weight change of the storage container and estimate the time of its filling, taking into account the actual mass flow obtained with the model of the linear crystallizer. On charts in the Figure 6a and Figure 6b presented the result of simulation of control systems based on the strain gauge measurement method and the differential pressure measurement method respectively. The both control system are based on the measurement of the derivative of the corresponding value.
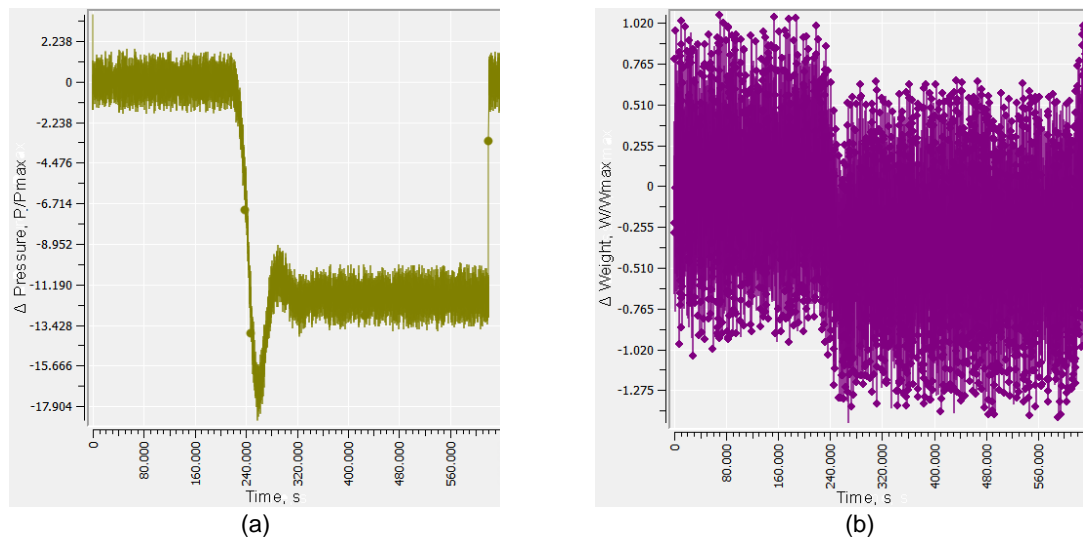

Figure 6: Simulation results of control system based on pressure (a) and weight (b) measurement

Based on the measurement simulation results, it can be said that the application of the control system, based on pressure measurement method allows us to accurately track the time of the full filling of the storage container. The accuracy of the differential pressure measurement by the proposed sensor is sufficient to determine the point at which the pressure ceases to change, which means that the capacitance is completely filled. On the other hand, the application of the filling control system based on weight measurement has not yielded positive results, since the rate of weight change is comparable with the measurement error. Based on the results obtained, it is proposed to supplement the already existing control system of crystallization process by a control

system of storage containers filling, based on a differential pressure sensor. The resulting system is shown in Figure 7. In this diagram, the dashed lines show control signals.
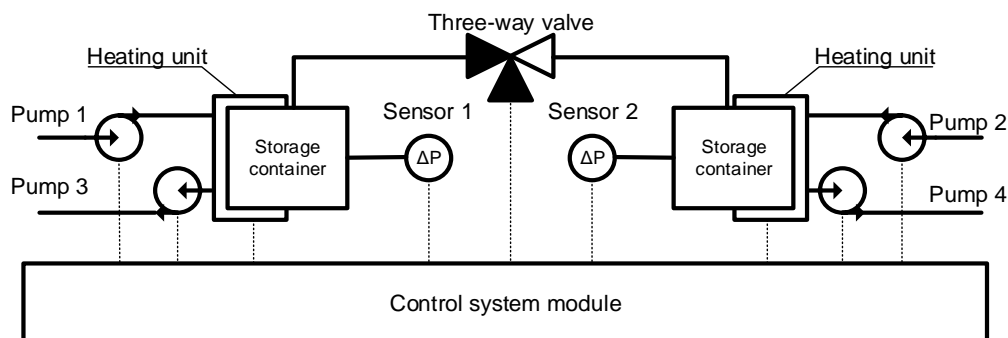

Figure 7: Schematic diagram of the storage container filling control system

The proposed control system operates according to the following algorithm:
- Waiting for the signal "active container is full" from the differential pressure sensor;
- Changing the active capacitance by switching the three-way valve;
- Heating of the filled container by heating unit;
- Emptying the filled container (pumps 3 or 4);
- Filling with washing solution (pumps 1 or 2);
- Waiting for valve switching.

6. Conclusions

A model of the storage container of the linear crystallizer was developed. On the basis of the received model, simulation of two independent from each other measurement method of storage container filling were provided. The results of the simulation showed that the application of the control system based on the differential pressure sensor allows to track the moment of full filling of the storage container. For this method, a control system is proposed and an algorithm for its operation is described.

The overfilling with crystals of storage container and washing section of the crystallizer can cause jamming of the linear crystallizer mixer or other emergencies. Switching storage tanks with partial filling will result in loss of efficiency. Timely switching by the control system of the filled container to empty will avoid these problems and improve the economic benefits of the crystallizer.

The results presented on the article will be used in the preparation of the experimental test program of the linear crystallizer in the Proryv project.

Acknowledgments

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