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Optimization of Uranium Crystallization Process by Using MPC Approach

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This paper proposes optimization of uranium crystallization process to provide insight into the trade-offs between performance and robustness explicitly. Based on developed mathematical model, which describes the heat and mass transfer processes, behavioral analysis of using PID and MPC controllers was presented. Increasing of crystals outlet and simultaneous decreasing of uranyl nitrate concentration in the liquid outlet showed MPC advantages.

1. Introduction

Development of modern technology for nuclear waste handling is aimed at obtaining a final product in the pure state, and the crystallization process proposed in this paper is known as one of the effective methods for separating uranium (Chikazawa et al., 2008). Application of crystallization was attempted to uranium purification of PUREX process in Germany (Henrich et al., 1987). In addition, two concepts were proposed in Japan: U-Pu co-crystallization processes (Homma et al., 2006) and the New Extraction System for TRU Recovery (NEXT) (Takata et al., 2004) based on PUREX process.
The large project “Proryv” is ongoing in Russian Federation with the aim of the creation of the pilot energy complex. Rationalization a process layout and design the unit operations and the current phase of works is focusing on the development of computer-aided process engineering systems supporting the operations are as part of the project. Automation has implemented at the every processing step of the material production. It has explained of requirements to ensure high controllability and operability for safety purposes (Sayyaadi and Sabzaligol, 2009). In order to improve the automatic control system (ACS) efficiency of crystallizer, there was implemented controllers based on a predictive model. It allowed to predicted of controlled variable changes for the time being ahead and provided the best trajectory of the process (Manenti et al., 2015). The present paper is proposing optimization of crystallization process. The process is sketched in Section 2. Plant and ACS descriptions are reported in Section 3. Section 4 addresses results of experimental work. The sensitivity analysis of the systems is reported in Section 5. Section 6 includes results and general comments.

2. Process description

The crystallizer, whose structure is reported in Figure 1, represents as a vertical metal chimney, whose working volume can be divided in three sections as reported by Veselov et al. (2015): a crystallization section, a washing section, and a crystal collection section (storage container). Uranyl nitrate (UN) solution (initial melt) containing a dissolved product is heated up to the required temperature and it is delivered to the top of the unit operation. The external surface of the unit is cooled by a cooling jacket. Along the unit, the uranium melt is cooled and it becomes supersaturated in the content of the product. Due to the difference in mass densities, crystals acquire an additional speed as compared to the mother liquor move co-directionally with the solution. The nitric acid (HNO_3) washing solution is delivered to the bottom part of crystal washing section and it is

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pushed up along the unit by precipitated crystals. It is used for crystal surface purification from captured impurities. On the boundary of first and second sections, the washing solution and the mother liquor are removed from the unit. The design of the initial melt loading and the crystals unloading allows operating the crystallizer continuous work. The design of the cooling jacket provides independent cooling of several sections of the unit.

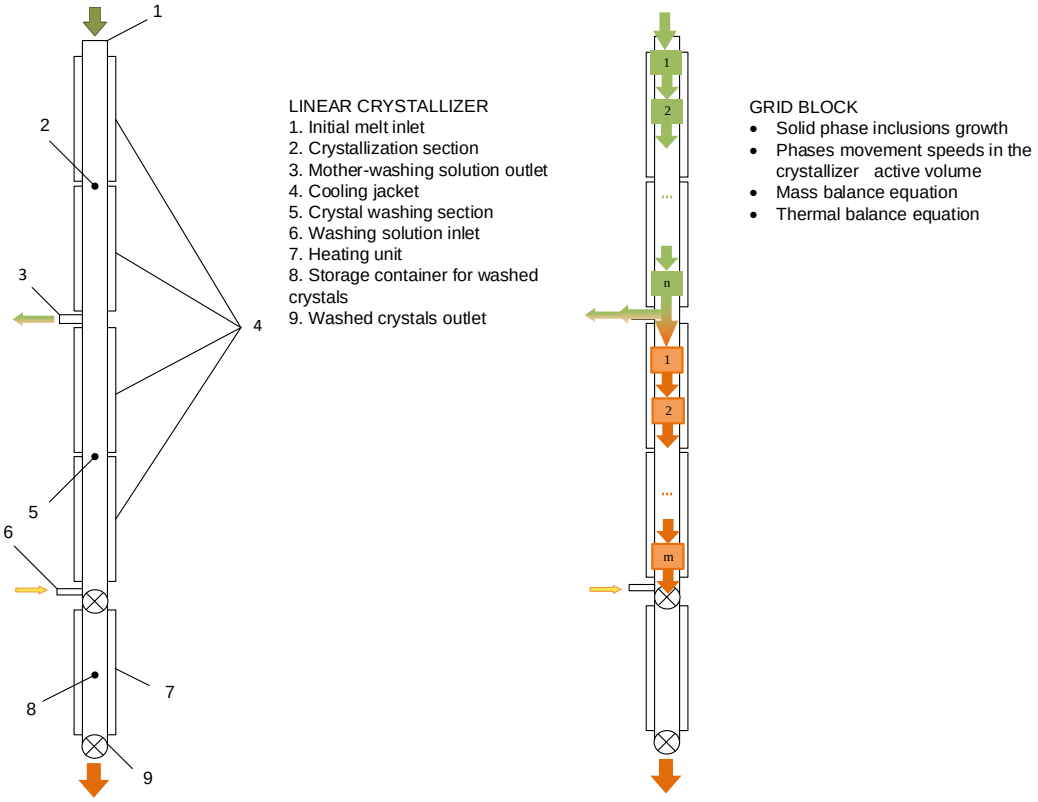

Figure 1: Diagram of a linear-type crystallizer

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 (Goryunov et al., 2011). To properly refine uranium and operate uranyl nitrate crystallization process (UNCP), it is important to carry out comprehensive studies of the phenomena involved in.

3. Control methodology

There is important to develop a mathematical model of the control plant. Using this model, regulator is predicted of controlled variable changes for the time being ahead and calculated the optimal control to provide for the best trajectory of the controlled variable.

At the initial instant, crystallizer is described according to Ochoa Bique et al. (2015), including the solid phase inclusions growth, mass and energy balance equations. The distributed nature of the system imposes to discretize the UNCP into a series of N interconnected blocks as schematically reported in Figure 1 by obtaining a diagonal-block numerical structure for the Jacobian matrix that can be efficiently solved by dedicate algorithms. Each block includes the equations for crystal growth, volume flow rates in the active volume of the crystallizer, mass balances and energy balances. It is assumed that the concentrations in the solution are ideal and the flowrate is constant along with the length of the crystallizer. The growing rate of the crystal radius is stated as follows:

$$\frac{dR}{dt} = k(C_{UN}^{m} - C_{UN}^{m,sat}) - R\,/\,L \cdot U_s \qquad (1)$$

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; $C_{UN}^{m,sat} = C_{UN}^{m,sat}(T,\psi)$ – concentration of UN saturation in liquid solution (the function of temperature and composition).

The conservation principle applied for UN and HNO_3 leads to the Eq.(2) and Eq.(3).

$$\frac{dC^{m}_{UN}}{dt} = \frac{Q_0 \cdot ((1-Win)\cdot C^{m,int}_{UN} - (1-W)\cdot C^{m}_{UN}) - U_s \cdot S \cdot W \cdot A}{V\cdot(1-W)} \quad (2)$$

$$\frac{dC^{m}_{HNO3}}{dt} = \frac{Q_0 \cdot (C^{m,int}_{HNO3} - C^{m}_{HNO3})}{V} \quad (3)$$

where Q_0 is an initial volumetric flow rate; S – sectional area; n - amount of crystallization centres; W - solid phase volume percent.

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

$$\frac{dT}{dt} = \frac{(\rho_{liq}\cdot C_{liq}\cdot Q_0\cdot(1-W) + W\cdot C_s\cdot\rho_s\cdot S\cdot U_s)\cdot(T_{in}-T) - \pi\cdot D\cdot\chi\cdot(T-T_{cj})}{(\rho_{liq}\cdot C_{liq}\cdot(1-W) + W\cdot C_s\cdot\rho_s)\cdot V} \quad (4)$$

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.

3.1 Controlled plant

Based on system analysis there were chosen 2 manipulated and 2 controlled variables. Figure 2 schematically represents information structure of crystallizer and Table 1 gives variables classification.

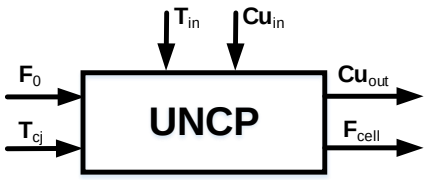

Figure 2: Crystallizer information structure

Table 1: Variables classification

Variable	Name	Note
Fo	Initial flowrate	manipulated variable
Tcj	temperature of the cooling jacket	manipulated variable
Cu_{out}	UN concentration in the liquid outlet	controlled variable
Fcell	Solid phase outlet	controlled variable
Tin	Temperature of solution	measurable disturbance
Cu_{in}	UN concentration in the liquid inlet	measurable disturbance

Linearization of the system was made by using simulation experiment approach. At the initial instant controlled plant is described by the transfer function of the first order. The dynamics of the controlled plant is characterized as a linear system with the transfer functions W1(s), W2(s), W3(s), W4(s) in Figure 3, where s – laplacian.

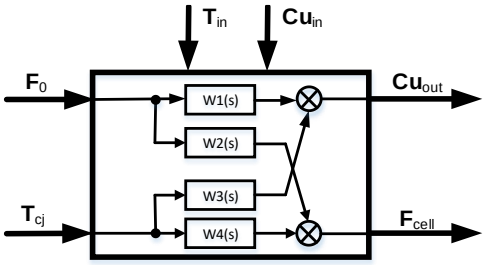

Figure 3: Information structure of linear system of control object

The linear model has high cross couplings, and using PID controllers to provide robust control is impractical for this model. Widely spread of MPC methodology with respect to the other rival control alternatives has recently used for optimization of unit operations and processes. MPC controller provides the optimal control to achieve minimum UN concentration in the liquid outlet from this linear model. It is intrinsically able to manage this system despite of high cross couplings.

4. Sensitivity analysis

ACSs with different controllers, describing in previous paragraph, have been simulated by MATLAB/Simulink. Parameters of control plant have been defined to configure PID and MPC regulators in ACS. There had been assumptions on manipulated variables. A desired value for the controlled variable (setup value) has been set at a given instant. Initial flowrate was manipulated variable. Resulting plots of transient processes are shown in Figure 4, where Cu0 and Fcell0 are setup values of UN concentration in the liquid outlet and solid phase outlet.

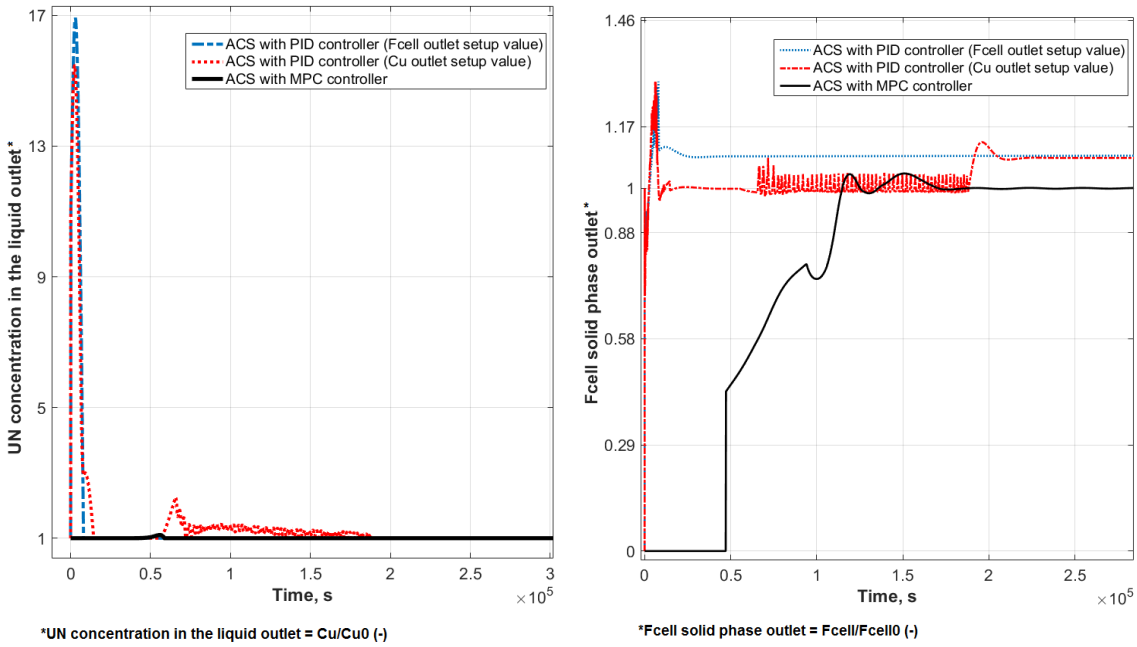

Figure 4: The transient processes for setup value

All of ACSs align of UN concentration in the liquid to a setup value. Settling time was $0.6*10^5$ s for the ACS with MPC controller to achieve 100 % of its final value, for ACS with PID controller (Cu setup value) - $1.7 \cdot 10^5$ s, for ACS with PID controller (Fcell setup value) – $0.1 \cdot 10^5$ s. Overshoot for both ACSs with PID were more than 20 %, but the control system with MPC-controller came out to a setup value without overshoot.
Settling time was $1.7 \cdot 10^5$ s for the ACS with MPC controller to achieve 100 % of its final value of solid phase outlet, ACS with PID controller (Cu setup value) – $2 \cdot 10^5$ s (bias: +0.082), ACS with PID controller (Fcell setup value) – $0.3 \cdot 10^5$ s (bias: +0.082). Overshoot for both ACSs with PID were 21 %, but the control system with MPC-controller came out to a setup value with 5 % overshoot. Table 2 gives comparison between controllers.

Table 2: Comparison of two PID and MPC controllers

Type of Controller	Tin solution	Cu output	Settling time, s	Overshoot, %	Fcell output	Settling time, s	Overshoot, %	Bias
MPC	1	1	$0.6 \cdot 10^5$	0	1	$1.8 \cdot 10^5$	5	0
PID (Cu)		1	$1.7 \cdot 10^5$	> 20	1.082	$2 \cdot 10^5$	21	+0.082
PID (Fcell)		1	$0.1 \cdot 10^5$	> 20	1.082	$0.3 \cdot 10^5$	21	+0.082

Then, the 20 % stepwise disturbance (T_{in}) has been introduced at the system at time 500,000 s. Resulting plots are presented in Figure 5.

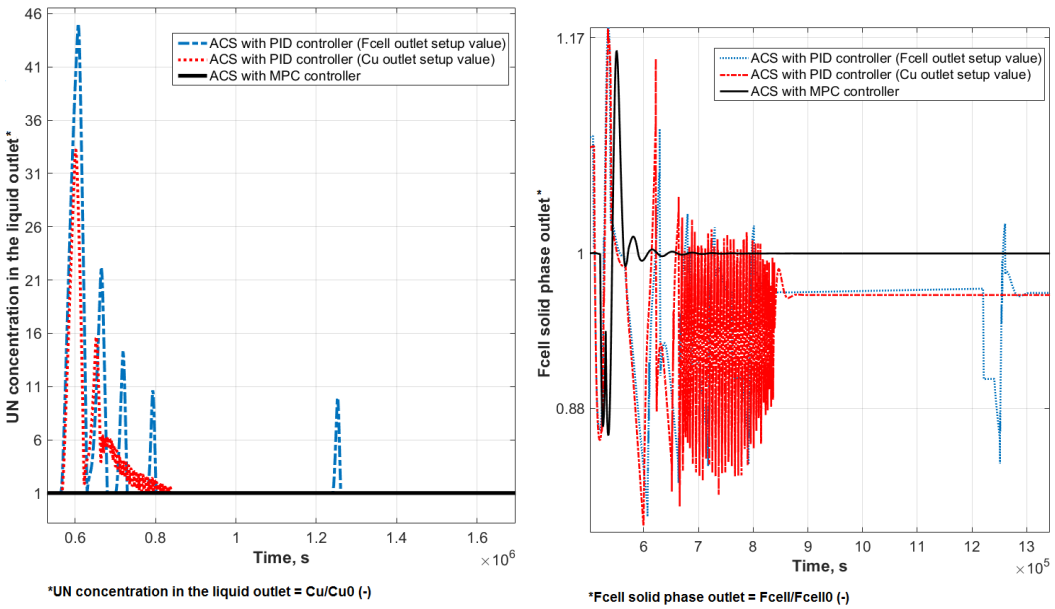

Figure 5: The transient processes for 20 % of a step disturbance

Settling time was $2.6 \cdot 10^5$ s for the ACS with PID controller (Cu setup value), the ACS with PID controller (Fcell setup value) was unstable, but ACS with MPC controller is stayed stable despite stepwise disturbance. Overshoot for the ACS with PID (Cu setup value) was more than 20 % and ACS with MPC-controller came out to a setup value without overshoot. Settling time was $1 \cdot 10^5$ s for the ACS with MPC controller to achieve 100 % of its final value of solid phase outlet, but ACS with PID controller (Cu setup value) – $3.5 \cdot 10^5$ s (bias: -0.032), the ACS with PID controller (Fcell setup value) was unstable. Overshoot for the ACS with PID (Cu setup value) was 21 %, but the control system with MPC-controller came out to a setup value with 16 % overshoot.
Table 3 gives comparison between controllers after the 20 % stepwise disturbance.

Table 3: Comparison of two PID and MPC controllers after 20 % step disturbance

Type of Controller	Tin solution	Cu output	Settling time, s	Overshoot, %	Fcell output	Settling time, s	Overshoot, %	Bias
MPC		1	0	0	1	$1 \cdot 10^5$	16	0
PID (Cu)	1.2	1	$2.6 \cdot 10^5$	> 20	0.968	$3.5 \cdot 10^5$	21	-0.032
PID (Fcell)		-	-	-	-	-	-	-

Analysis of step-response performance specifications shows advantage of the ACS with MPC controller.

5. Results

Changing setup values of controlled variable, ACS with MPC controller allows achieving their optimum. Figure 6 represents the increasing of solid phase outlet. All of ACSs align of UN concentration in the liquid and Fcell solid phase outlet to setup values. The best indicators quality.is achieved by the ACS with MPC controller. Settling times were near $2.6 \cdot 10^5$ s for both ACS with MPC controller to achieve 100 % of its final value of solid phase outlet before 20 % stepwise disturbance. Overshoot for both ACSs were less than 20 %. Numerical results are reported is reported in Table 4.

Table 4: Numerical results of solid phase outlet achievement

Fcell Setup value	Settling time, s	Overshoot, %	Settling time after stepwise disturbance, s	Overshoot, %	Cu output
1	$2.5 \cdot 10^5$	5	$6 \cdot 10^5$	15	1
1.17	$2.7 \cdot 10^5$	6	$8 \cdot 10^5$	16	1

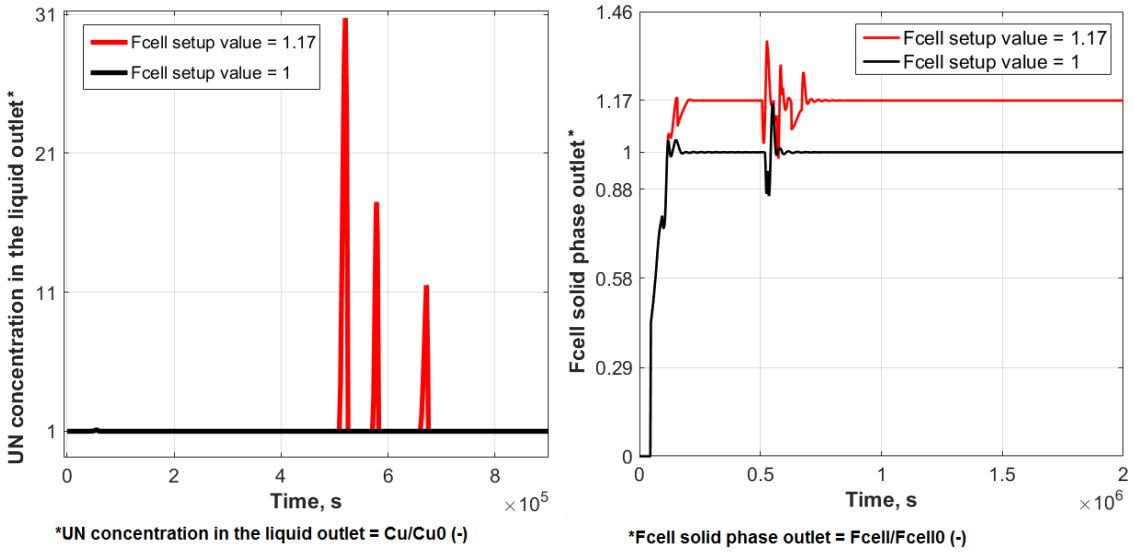

Figure 6: The transient processes with different setup values

6. Conclusion

The mathematical model analysis provided useful information for appropriately choosing the operating conditions of the crystallizer. It was a cause of optimal control system development to predict of system's behavior. Sensitive analysis ACS with different controllers showed MPC advantages. Using MPC controller allowed to predict of controlled variable changes for the time being ahead and provided the best trajectory of the process. Maximum solid phase outlet was achieved after process optimization. Further, it is planned to use Nonlinear Model Predictive Control (NMPC) techniques to manage nonlinearities in process dynamics and in profits. It could allowed solving effectively the real-time dynamic optimization by accounting for constraints on manipulated variables.

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