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Modelling of the Monitoring and Control System of Extraction Units with Nuclear-safe Tanks via Use of Capillary Impulse Lines with Differential Low-pressure Gages

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This paper focuses on the computer-aided simulation of extraction process in the cascade of reverse-flow centrifugal extractors with nuclear-safe tanks. The major objective of this work is to design an advanced control system for enhancement of extracting technology efficiency. The basis of the monitoring and control system is that the systems should be based on non-destructive measuring techniques. Two models of monitoring systems are being considered: the measuring system in nuclear-safe tanks where capillary impulse lines are used for measurement of differential pressure at various points in the process media, and model of spectrometric system. The density measurement in both presented systems is indirect. The experimental studying of monitoring spectrometric system have shown that the accuracy of experimental data less than 1 % in steady-state condition and more than 3 % in a dynamic one. The accuracy degradation is due to air bubbles presents in the solvent stream.

1. Introduction

In the last 10 years, waste refinery and nuclear fuel conversion are growing in importance. The radiation resistance of solvent to enhanced radiation doses is one of the important factors affecting the efficiency of available refinery technologies for spent nuclear fuel processing.
The processing equipment of conventional radiochemical technologies places in canyons where refining is a long-time process. This technology is not applicable for processing of spent nuclear fuel from fast neutron reactors since the solvent radiation loads in that equipment is increased hundreds times for one multistage cycle. Moreover, the prolonged exposure of radioactive emission to extraction systems leads to losses of separation efficiency and solvent destruction. There are two ways to avoid the above mentioned problems. The first one is to develop a new radiation-stable extraction system. The second is set new extractors with short phase contact time.
Innovative radiochemical plants that designed for nuclear fuel cycle closure have a significant technological feature. This feature is that most of the processing equipment uses nuclear-safe technological units that are placed in hot shops. Furthermore, up-to-day, the extraction technology is design on centrifugal extractors. The internal capacity of most these units less than 10 L, and fluid flow rates through it less than 30 mL/min. In this case, preferably to use a high-precision control and measuring devices, which components should be free of maintenance and can be placed on equipment. But, the implementation of new or additional equipment brought about significant changes into an existing technology and would lead to the necessity for the additional experimental studies. Due to the use of components of high reactivity in uranium and plutonium extraction and re-extraction processes, it is impossible to carry out an experimental research. Therefore, it is necessary to use computer-aided models at the development and research stages. In addition, there are many challenges in manufacturing devices for monitoring the level and density of the media, measuring the

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micro-flows. Thus the computer-aided modelling for design of control system and nuclear-safe technological units is relevant and has scientific and technical significance.
The computer-aided simulation of extraction process in the cascade of reverse-flow centrifugal extractors based on mathematical model fully presented in Zelenetskaya et al. (2016), since only a brief introduction of the model description is presented in this work.
The basic technological processes of the centrifugal extractor include mixing the initial solutions in the mixing chamber to ensure mass transfer and separating of the resulting emulsion in the rotor for obtaining the pure chemical compounds. Hence, each centrifugal extractor in the scheme represented as tightly coupled system of two parts: the mixing and separating. The numbers of technological schemes of extraction refining are being worked out within the framework of the major advanced global projects PRORYV in the nuclear power sector. The large numbers of processing units with centrifugal extractor as a main hardware unit are included in these schemes. All model of processing units are presented by systems of differential equations.
The cascade of centrifugal extractors is a complex plant hence the dynamical computer aided model of the cascade system had been made with the following assumptions:

- The mass transfer processes in both phases have variable rate, hence, the concentrations of chemical compounds circulating in the system are continuously calculated at different points in each centrifugal extractors.
- Values of concentrations are continuously calculated in two different conditions, one is the start-up and the other steady state.
- • The values of the flow rates of solutions are automatically calculate at each modelling time interval, since volumes of organic and water phases are variable during whole cascade.
- The amounts of phase capture less than 3 %.

The determination of the equilibrium concentration values of the target components in each phase in the extractor is based on Puzikov et al. (2014) and Zavalina et al. (2017) but with respect of plutonium distribution coefficient in the following:

$$lgE_{wf}^{0} = 1.2 + 3.04251lgC_{NO_3} - 0.36pH \quad (1)$$

In Eq(1), C_{NO3} is the total nitrate concentration in the water phase, mol/L.
The completed mathematical model proposed in Zelenetskaya et al. (2016) is applicable not only for extraction but also for re-extraction processes simulation. Moreover, these two processes could be simulated simultaneously. The computer-aided model has been made by taking into account the following considerations:

- It is possible to change structure of the cascade hence the number of centrifugal extractors in the model is variable.
- The dynamical model is based on some interlocks, such as opposite connection of apparatus, overflow.
- There are additional modules such as valves, pumps and tanks that could be added to the process or removed from it.

The technological scheme of the extraction refining process contains blocks of cascades of reverse-flow centrifugal extractors and tanks. All units of the scheme have nuclear-safe design.
By computer-aided simulation of different cascade configuration and conditions, it is possible to solve technological and optimization problems which occur in plants, explore effect of a disturbances that affect systems, carry out analysis, synthesis and design of automatic control systems. This study provides for creation of the new generation nuclear power technologies on the basis of closed nuclear fuel cycle using fast neutron reactors. Operation of the cascade consisting of 9 series-connected centrifugal extractors was simulated in order to verify the proposed models of monitoring systems.

2. Determination of metal density in the cascade

The conversion of nuclear fuel and waste treatment include processes for refining solutions containing high-level radioactive materials, thus determining the content of the target components throughout the whole technological scheme is an important and extremely complex process. In the literature, a numerous methods have been developed to determine the quantitative and qualitative composition of solutions and mixtures. But only a few numbers of them can be useful when working with systems containing fissile elements. The radioactive emission of solutions adversely affects immersion-type gage sensor, thus all measuring technology should base on non-destructive material control methods.

2.1 The model of density measuring of solution by spectrometric system

One of the most informative methods of non-destructive analysis of radioactive materials is gamma-spectrometric analysis. This method allows simultaneous determination of the isotopic composition of fissile elements, activity and masses of radioisotopes in the solution under study.
Based on Reilly et al. (1991) the change in the amount of target components in the solution can be determined by density changing of the solution, which is measured by the degree of attenuation of electromagnetic radiation of a given energy.

$$\rho = -\frac{ln\mathrm{T}}{\mu x} - \frac{\mu_0 \rho_0}{\mu} \tag{2}$$

In Eq.(2), μ, and μ_0 are mass attenuation coefficient and mass attenuation coefficient at a given gamma-ray energy respectively, ρ_0 is density of solvent, T is photon transmission coefficient at given gamma-ray energy, x is material thickness.
When measuring a two-component mixture the mass attenuation coefficient of i-th component measured at j-th energy described as following:

$$\mu_i^j = \mu_i(\mathrm{E}_j) \tag{3}$$

The transmission coefficient at j-th energy described as following:

$$T_j = \exp\left[-\left(\mu_1^j \rho_1 + \mu_2^j \rho_2\right) x\right] \tag{4}$$

The measurement of two transmission coefficients gives two equations for two unknown densities:

$$\begin{cases} -\frac{ln\mathrm{T}_1}{x} = \mu_1^1 \rho_1 + \mu_2^1 \rho_2 \\ -\frac{ln\mathrm{T}_2}{x} = \mu_1^2 \rho_1 + \mu_2^2 \rho_2 \end{cases} \tag{5}$$

In the proposed model the components detection is carried out on resonance peaks. The experimental data analysis gives a correct solution if, and only if components differ from each other in absorbing properties. When absorbing properties of components are similar, the values of analysed peaks would coincide with each other that lead to inaccurate measurement.
The accuracy of the measurement is estimated from the relative error of the density of each component, since the source of the stochastic measurement uncertainty is the statistical dispersion of the measurement results.

$$\begin{cases} \frac{\sigma(\rho_1)}{\rho_1} = \frac{1}{\mu_2^1 \ln T_2 - \mu_2^2 \ln T_1} \left[\left(\mu_2^2 \frac{\sigma(T_1)}{T_1}\right)^2 + \left(\mu_2^1 \frac{\sigma(T_2)}{T_2}\right)^2 \right]^{1/2} \\ \frac{\sigma(\rho_2)}{\rho_2} = \frac{1}{\mu_1^2 \ln T_2 - \mu_1^1 \ln T_2} \left[\left(\mu_1^2 \frac{\sigma(T_1)}{T_1}\right)^2 + \left(\mu_1^1 \frac{\sigma(T_2)}{T_2}\right)^2 \right]^{1/2} \end{cases} \tag{6}$$

Note that the thickness of the sample x should be constant. Moreover, the range of optimum value of transmittance coefficients can be attributed to the characteristic concentration.
The scheme of measuring density of uranium and plutonium by gamma radiation is shown in Figure 1.

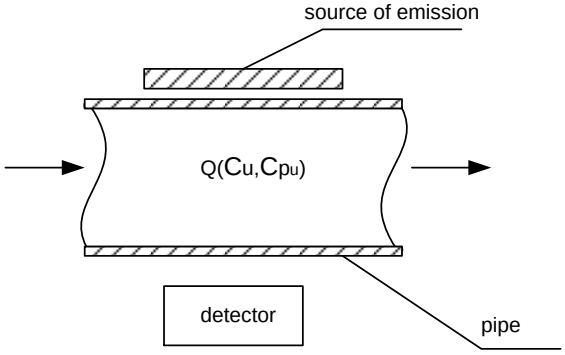

Figure 1: Scheme of measuring density of target components by gamma radiation

The emission source contains a collimating element that provides the straight gamma rays beam, and detector unit has a filter unit. The filter absorbs an emission which energy spectrum is less than photon energies emitted during the decay uranium and plutonium. The filter unit makes possible to unload the photon measurement channels and improve the accuracy of the experimental data.
The spectrometric system is portable and can be set in different points of the technological scheme. In this work the spectrometric system is set on the connected pipeline in the cascade.
Since the detector is discrete, in case of multi-component mixture, the counting rate of the pulses detected by the detector can be determined as follows:

$$n_p^i = I_\gamma^i \left(\frac{N^i ln2}{T_{1/2}^i} \right) \varepsilon_\gamma^i \tag{7}$$

In Eq(6) I_γ^i is quantum yield of radiation detected in the analysed peak of *i*-th component, ε_γ^i is radiation detection efficiency in the analysed peak of *i*-th component, $T_{1/2}^i$ is half-life of *i*-th isotope.
The experimental studies were carried out in order to verify the proposed model and obtained results is shown in Table 1. The intensity of source emission was 1.2 MeV.

Table 1: Mass attenuation coefficients for different samples

	water	air-liquid layer	uranyl nitrate solution
experiment	0.0301	0.0270	0.0619
model	0.0301	0.0280	0.0681

Experimental studies have shown that air bubbles are present in the solvent stream. In this model, the correction for absorption of the spectrum by air was not performed, which led to a peak shift and, as a result, distortion of the measurement results. Currently, the model has been adjusted to take into account the absorption of the spectrum by impurities. The influences of the air bubbles formation on the measurement accuracy and the reliability of the obtained data were shown by Manenti et al., 2013.

2.2 The model of density measuring of solution in tanks via use of capillary pulse lines with differential low-pressure gages

The current work uses the mathematical modelling of monitoring system of nuclear-safe technological units for manufacturing risk reduction. A pressure drop measurement technique for monitoring the level and density of solutions in tanks and vessels of extraction cascades is proposed. The capillary impulse lines are used for measurement of differential pressure at various points in the process media. The technique flexibility involves stable level and density measuring of a complex multiphase media.
The scheme of measuring the density of solution containing uranium and plutonium in tanks via the use of capillary pulse lines with differential low-pressure gages is shown in Figure 2.

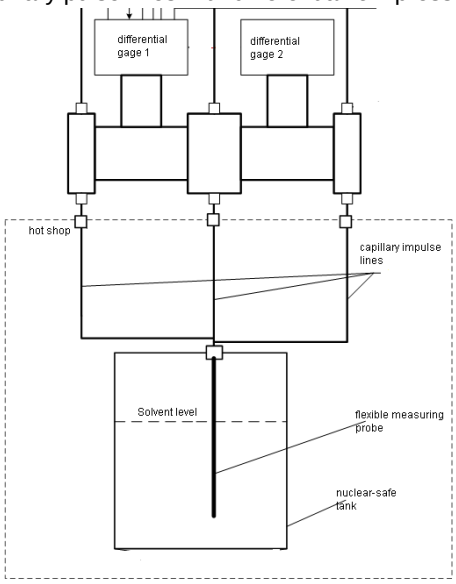

Figure 2: Scheme of measuring density of uranium and plutonium by gamma radiation

The capillary pulse lines are installed in the nuclear-safe tank through one hole from above and combined into a flexible measuring probe. The flexible probe consists of 2 parts. Inside the flexible probe, three capillaries are installed. Two out from three capillary lines are set at 30 cm and 45 cm respectively above the bottom surface of the tank. The third capillary line is set at 40 cm below upper level of the tank. To measure pressure differences, experimental digital differential pressure gauges are used. The solution density and liquid level in the tank can be determined as follows:

$$\rho = \rho_s + \frac{\Delta P_1}{g\Delta H_1} \tag{8}$$

Where ρ_s is density of solvent, ΔP_1 is indication of the first pressure gage (differential gage 1 in Figure 2), g is gravitational constant and ΔH_1 is the level difference between first two capillary lines in the flexible probe.

$$l = \frac{\rho_s g\Delta H_2 - \Delta P_2}{\rho g} \tag{9}$$

In Eq.(9), ρ and ρ_s are density of solution and solvent respectively, ΔP_2 is indication of the second pressure gage (differential gage 2 in Figure 2), and ΔH_2 is the level difference between second and third capillary lines in the flexible probe (see Figure 2). The solution of uranyl nitrate had been chosen in order to verify the proposed model. The testing results are presented in Figure 3 and Figure 4.

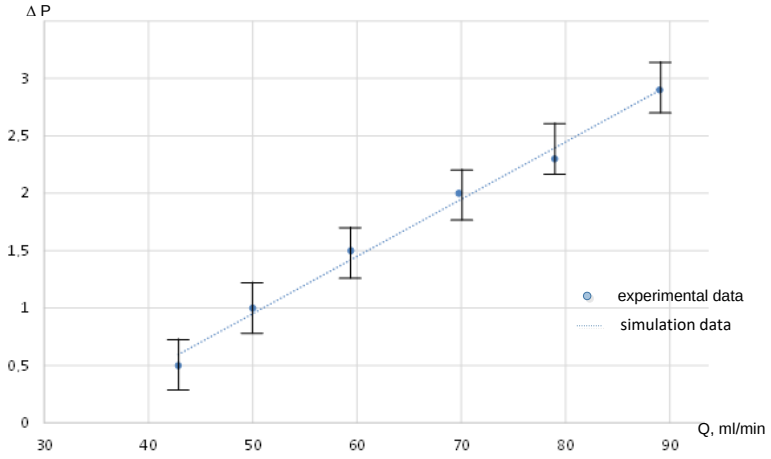

Figure 3: Dependence of pressure in the tank on ingoing flow rate

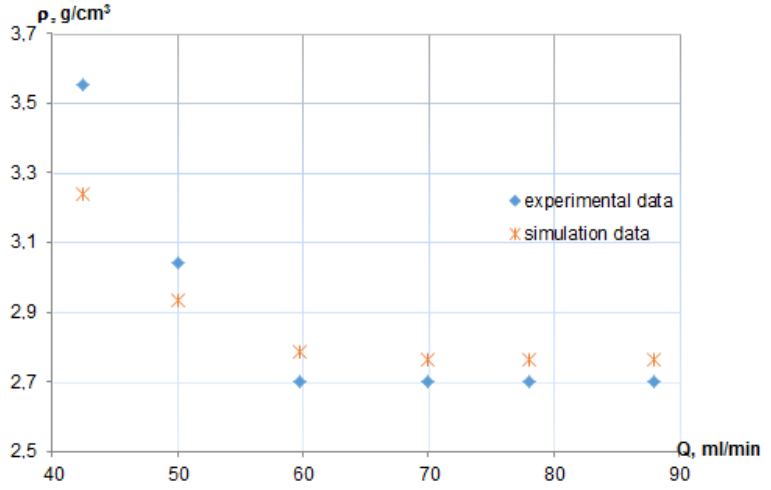

Figure 4: Dependence of solution density in the tank on ingoing flow rate

According to the testing summation result (Figure 3 and Figure 4) it is possible to say that the basic assumptions are correct.

3. Conclusions

This paper has outlined two models of monitoring system which allows calculate the solution density. First system is based on the non-destructive analysis. The second one is used the capillary impulse lines for measurement of differential pressure at various points in the process media. The current work shows the capability of technological processes in nuclear-safe extraction units. Solutions of the installation of the capillary impulse lines with low-level delays in sensing ports were explored. The dependence of multi-component mixture density in the tank on variation of ingoing flow rate would be study in order to develop an advanced automatic control system of refinery processes. The proposed model of spectroscopic system would be corrected and improved model would respect the spectrum absorption by air bubbles. Moreover, the adjusted model could differentiate the components with similar absorption properties. The components would be detected not only by resonance peaks but also by absorption on the wings of the L- or K-lines. This monitoring system would be applied on different steps of technological schemes of extraction refining in order to determine the density distribution in cascades. The developed systems are intended to be further used as basis for designing advanced automatic control system of the cascaded centrifugal extractors.

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References

Belova E.V., Dzhivanova Z.V., Tkhorzhnitskii G.P., Stefanovsky S. V., Myasoedov B. F., 2016, Kinetics of oxidation of 30 % TBP solutions in C13 diluent with nitric acid: Effect of temperature and γ-irradiation, Radiochemistry 58, 426–430.

Blake C.A., Davis J.V., Scmitt J.M. 1963, Properties of Degraded TBP-Amsco Solutions and Alternative Extractant–Diluent Sistems, Nuclear Science & Engineering, 17, 626-637.

Goryunov A. G., Mikhaylov V. S., 2012, The automatic control system of a multi-component nonequilibrium extraction process in the pulse column, Journal of Process Control, 22, 1034–1043.

Manenti F., Rossi F., Croce G., Grottoli M.G., Altavilla M., 2013, Intensifying air separation units, Chemical Engineering Transactions, 35, 1249–1254. DOI:10.3303/CET1335208.

Puzikov E. A., Zilberman B. Ya., Fedorov Yu. S., Blazheva I. V., Kudinov A. S., Goletskii N. D., Ryabkov D. V., 2014, A New Approach to Simulation of Extraction Equilibria in the PUREX Process, Solvent Extraction and Ion Exchange, 33, 1–23

Puzikov E. A., Kudinov A. S., Blazheva I. V., Sytnik L. V., Shishkin D. N., Goletskii N. D., Zilberman B. Ya., 2017, Mathematical modeling of extraction of nitric acid, actinides, and fission products into 30% TBP in halogenated diluents using A.M. Rozen’s model, Radiochemistry, 59, 143–151

Reilly D., Ensslin N., Smith Jr H., Kreiner S. (Ed), 1991, Passive Nondestructive Assay of Nuclear Materials, Nuclear Regulatory Commission, U.S.

Skvortsov I.V., Belova E. V., Nikitina Y. V., Rodin A. V., Zilberman B. Ya., Goletskii N. D., 2016, The dynamics of gas release at oxidation of TBP solution in diluent c13 in two phase systems, Nuclear and Radiation Safety, Vol. 2 (84), 8.

Shmidt O.V., Makeeva I.R., Liventsov S.N., 2016, Simulation of closed nuclear fuel cycle processes as a tool for the development and optimization of radiochemical technologies, Radiochemistry, 58, 366–373.

Zavalina O. A., Dvoeglazov K. N., Pavlukevich E. Yu., Stepanov S. I., 2017, Kinetics of Pu(VI) reduction with carbohydrazide in nitric acid, Radiochemistry, 59(4), 331-336.

Zelenetskaya E. P., Goryunov A. G., Daneikina N. V., 2016, Model of mass transfer processes in the cascade of centrifugal extractors, IOP Conf. Series: Materials Science and Engineering, 135, 012053, doi:10.1088/1757-899X/135/1/012053