Computational and experimental studies into the hydrodynamic operation conditions of container filters for ion-selective treatment*

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Abstract

Formation of radioactive waste (RW) is specific to the NPP operation. Liquid radioactive waste (LRW) forms in the process of the reactor plant operation, and in decontamination of equipment, rooms and overalls. The radionuclides found mostly in vat residues are ¹³⁴,¹³⁷ Cs in the form of ions and ⁶⁰Co and ⁵⁴Mn isotopes in the form of chelates including substances used for equipment decontamination. Among the well-known conditioning techniques, selective sorption provides for the greatest reduction of LRW amounts. The efficiency of using the amount of the filter material can be increased by supplying the treated medium simultaneously to several sorbent layers.

The paper presents computer simulation results for three proposed options of improved container filter designs for ion-selective treatment differing in the ways used both to separate the treated water flows and to deliver these to the sorbent layers. The improved efficiency of the sorption processes in the proposed designs was estimated using computer simulation in SolidWorks Flow Simulation.

Three sorbent grades from NPP Eksorb were used for the study. A series of experimental studies of the flow through the sorbent layer was undertaken to determine the hydraulic resistance of the studied samples. The obtained experimental data was added to the Solidworks Flow Simulation engineering database for simulation of the earlier presented designs. Representative parameters of the flow inside of container filters were obtained as a result of the simulation.

Keywords

Decommissioning, liquid radioactive waste, ion-selective treatment, sorbents, container filter, optimization of radiation protection, hydraulic resistance, porosity, computer simulation

* Russian text published: Izvestiya vuzov. Yadernaya Energetika (ISSN 0204-3327), 2022, n. 2, pp. 62–72.
Introduction

Generation of radioactive waste (RW) is specific to the operation of any NPP. The key condition for the nuclear power acceptability as a reliable low-carbon source capable to support the sustainable evolution of humankind is therefore addressing the problem of environmentally safe handling of RW, along with ensuring the safety of the NPP operation (Novikov et al. 2017).

Since 2013, a strategy has been implemented at Russian NPPs to reduce the amounts of the RW formed and reprocessed, and to ensure that RW meets the acceptability criteria (Tashlykov et al. 2021) as defined in the regulatory document NP-093-14 “Criteria of the Radioactive Waste Acceptability for Disposal”. As required by NP-093-14, the regulatory framework has been largely updated and requirements have been modified with respect to establishing solid (SRW) and liquid (LRW) radioactive waste reprocessing facilities. Earlier, the RW reprocessing rate was not large, and the reprocessed waste amount depended on if there were onsite reprocessing facilities. At the present time, RW reprocessing facilities are established taking into account the RW amounts formed both in the process of operation and as a result of the nuclear plant life extension and decommissioning (Adamovich et al. 2018).

Liquid radioactive waste (LRW) form in the process of the reactor plant operation and decommissioning (maintenance of water chemistry, equipment decontamination, etc.) (Tashlykov et al. 1995, Nosov et al. 2019). The radionuclides found mostly in vat residues are $^{134,137}$Cs, $^{60}$Co, and $^{54}$Mn. Ionic form is typical of cesium isotopes. Cobalt and manganese radioisotopes occur in vat residues in the form of chelates with compounds used for equipment decontamination (oxalic, formic, citric and ethylene diamine tetraacetic (EDTA) acids).

Cobalt is the most radioactively hazardous component of the NPP structural materials. The source for $^{60}$Co with a high gamma quanta energy (~1.2 MeV) and a long half-life ($T_{1/2} = 5.272$ years) is $^{59}$Co. Cobalt-60 and cesium-137 define the radiation background in the process of dismantling NPP units and following the shutdown and long-term cooling (Kropachev et al. 2019).

The key purpose in handling of LRW is final isolation of conditioned RW. Among the conditioning techniques, selective sorption provides for the greatest reduction of LRW amounts (Table 1, Fig. 1). This makes it possible to concentrate radionuclides in a small amount of sorbent (Arustamov et al. 2005).

| Table 1. Sorbent characteristics |
|-------------------------------|
| Parameter | Sample | SMET | RATsIR | MODIX |
|-------------|--------|------|--------|-------|
| Sorbent grade | | | | |
| Bulk density, kg/m³ | 1320 | 1092 | 959 |
| Porosity | 0.214 | 0.187 | 0.234 |
| Real density, kg/m³ | 1681 | 1343 | 1252 |
| Average particle size, mm | 0.9 | 0.8 | 1.4 |

Materials and techniques

The process of LRW treatment in an ion-selective treatment (IST) facility includes prefiltering and preparation of the initial solution, ozone treatment, filtration, and selective sorption in container filters (CF) using ferrocyanide sorbents. The occurrence of cobalt and manganese in a complex and, therefore, non-sorbing form defines the need for complexes to be broken to recover the above radionuclides from solutions. The ozone treatment stage is used to this end. Besides, the organic compounds contained in vat residues reduce the useful life of cesium ferrocyanide sorbents (Remez et al. 2016).

The LRW handling concept based on a technology for the LRW ion-selective treatment to remove radionuclides is used at the Kola NPP as part of the integrated LRW reprocessing facility put into operation in a phased manner in 2006-2009 (Avezniyazov and Stakhiv 2018).

The RW handling systems in operation or in the process of construction at Russian NPPs use state-of-the-art technologies to ensure that RW is handled safely at all stages, in a range from collection processes to reprocessing and preparation of the final package for delivery to the National Operator. One of the projects currently under way is to build an LRW reprocessing facility (LRW RF) at the Beloyarsk NPP for producing solidified RW in accordance with the acceptability criteria. One of the three LRW RF process lines will be a modular ion-selective treatment unit (ISTU) for reprocessing of vat residues from the tanks of the LRW storages (Bulatov et al. 2020).

A treatment technology using dedicated sorbents, with no ozone treatment stage, has been developed by NPP EKSORB for hard-to-ozone LRW containing stable cobalt chelates with ethylene diamine tetraacetic acid. The

[Figure 1. Microphotograph of sorbent particles: a. SMET; b. RATsIR; c. MODIX. Grid size: 0.1 mm.]
experiments conducted at the Kola NPP in 2016 were successful (Remez et al. 2016).

An important condition for implementing this technology is to optimize radiation protection in the process of placing conditioned LRW in casks (Tashlykov et al. 2015, Litovchenko et al. 2020, Mikhailova and Tashlykov 2020).

An important characteristic of the ion-selective treatment technique efficiency is that the maximum possible portion of the total sorbent amount is used for the LRW treatment. The existing filter designs sorb radionuclides non-uniformly as the treated medium moves through the sorbent, and the efficiency of filtering is much lower.

When single-section filters are used, the efficiency of treatment is at all times below 100%, that is, less than the thermodynamic capacity value. This is explained by kinematic factors mostly. A variety of methods are offered in literature to improve the efficiency. For example, it is proposed in (Venitsianov 2013) that the sorbent layers are broken down into sections. After the maximum saturation is reached in section 1 (this happens faster than in the event of the others), it is removed and section 2 becomes section 1, as the section filled with pure sorbent becomes the final one in the direction of the treated medium movement. Thus, using a two-section filter instead of a single-section filter leads to the protection time increased by 64%.

It is possible to increase the efficiency of using the filter material amount by feeding the medium being treated into several sorbent layers at a time increasing so the medium-sorbent contact area. One example of a similar design solution is improvement of cesium traps in the form of fuel assemblies, in a range from a design with one graphite layer for the BOR-60 reactor to the MAVR trap design with sodium delivery simultaneously to four layers (for the BN-350 and BN-600 reactors) (Beltyukov et al. 2013).

Three filter trap designs were developed for the study. In design 1, the medium being treated is delivered in the same manner as in the MAVR cesium trap (Fig. 2a). The treated medium is fed from below and distributed by the sorbent layers using the supply tubes installed inside the layers. After being filtered, the medium leaves the sorbent layer through the discharge tubes.

The medium flow in this design leads to a substantial local resistance at the distributing tube inlets. A design has been proposed to minimize these resistances using perforated rings (Fig. 2b). In this design, the medium enters the filter from above and, as it approaches the first layer, is divided using perforated rings. The same thing also happens with the further layers. After it passes through the sorbent layers, the medium enters the common drain channel. The advantage of this design is that it is possible to change the number of the sorbent layers.

A design was considered as option 3 with the distributing tubes outside the container filter body (Fig. 2c). The advantage of this model, as compared with design 1, is a smaller hydraulic resistance in the discharge channels thanks to the channel’s larger flow area. In this design, the medium is fed from above and distributed by four directions. The medium is delivered to the layers via the rear distribution channels.

Three sorbent grades (SMET, RATsIR and MODIX) from NPP Eksorb (see Table 1), one of the leading makers of sorbents for ion-selective treatment in Russia and in the world (Khomyakov et al. 2021), were selected for the study.

As can be seen, the three sorbent grades differ in the particle shape and the porosity value (the ratio of the pore volume to the entire layer volume). Each sorbent layer consists of many irregularly shaped components and has a complex and static geometrical structure. Where the flow scales are large as compared with the layer particle sizes, the flow is simulated as a quasi-homogeneous medium with one generalized characteristic (permeability).

Figure 2. Trap designs: a. Tube technique; b. Stinger ring technique; c. Off-body flow split technique; 1 – waste header; 2 – body; 3 – discharge tube; 4 – sorbent layers; 5 – supply tube; 6 – feeding header; 7 – perforated rings; 8 – drain channel; 9 – perforated flow split walls; 10 – partitions; 11 – distribution channels.
A series of experimental studies was undertaken for the flow to determine the hydraulic resistance of the investigated samples (Bessonov et al. 2021a, b, c, Mordanov et al. 2021).

The purpose of the study is to obtain the dependence of the sorbent layer hydraulic resistance on the water flow velocity. The action of the experimental set-up (Fig. 3) is as follows: the flow with initial pressure $P_0$ enters the column through the inlet chamber and passes through the sorbent layer. The sorbent inside the column is retained by the filters installed below and above the sorbent layer. As the flow passes through the sorbent layer, its pressure starts to decrease, because of the medium resistance, to value $P_f$. The differential pressure value at the sorbent layer inlet and outlet is recorded using a pressure difference gage (PD). This series of experiments used an NT-1890 digital pressure gage with an instrumental error of 0.3%. The flow velocity is controlled using valve 5. The liquid flow rate, $F$, through the column is measured using a variable flow area meter of the LZS-15 series.

Experimental studies were undertaken five times for each sorbent grade with an equal liquid flow rate to determine the measurement error. After the required number of measurements, the liquid flow rate was increased by 10 l/h.

Simulation

Flow Simulation is a computational fluid dynamics (CFD) module built in the SolidWorks software package. Flow Simulation offers general parametric flow simulation based on the finite element method. Simulation makes it possible to calculate hydrodynamic performance and heat-exchange conditions in isothermal and non-isothermal turbulent and laminar flows in the absence of phase transitions and chemical transformations. Among other things, Flow Simulation allows simulation of flows in porous media.

Flow Simulation is a problem solver for a variety of applications. This module allows one to simulate liquid and gas flows, use standard physical models of liquids and gases, and calculate thermal performance, and hydrodynamic and thermal models of devices. Flow Simulation is used extensively to find solutions to problems to be addressed (Aladyev et al. 2020).

For simulation of a liquid or gas flow in a porous medium, Flow Simulation treats the porous filler as a solid body with two characteristics: porosity that is equal to the ratio of the volumes of interconnected pores to the whole body volume, and permeability that is defined as hydraulic resistance $k = -\nabla(P)/\rho v$, where $P$ is the pressure, $\rho$ is the density of the flowing medium, and $v$ is its velocity.

The dependence of the permeability coefficient on velocity or flow rate is defined by the user as a table. Characteristics are entered in the system for a rectangular or cylindrical porous body, and are further automatically rearranged to match the model geometry in the process of the calculation. If the solid body characteristics were obtained for a liquid other than the design flowing medium, calibration viscosity (where coefficient $k$ depends on the liquid viscosity with the constant flow rate and differential pressure values), or calibration density (where resistance coefficient $k$ is proportional to the flowing medium density) is introduced additionally.

Flow Simulation’s initial engineering database presents just a number of porous bodies (wire mesh filters, foam filters, etc.). To analyze a flow in a particular porous medium, one needs to determine the characteristics of the medium in question. Two approaches can be used here. Where pores form a regular structure, a solid-body model can be built that reflects the porous body geometry and the required characteristics can be determined using CFD techniques. Where no such simulation is possible, the porous body characteristics are determined in a full-scale experiment.

The data obtained in the experiment for all sorbent grades was added to the SolidWorks Flow Simulation engineering database to be used in hydrodynamic simulations of the proposed filter trap designs.

Results and discussion

The purpose of the experimental study was to obtain the dependence of the sorbent layer hydraulic resistance on the water flow velocity. The experiment results are presented in Fig. 4.

It can be seen from the experiment results that hydraulic resistance of the layer increases in accordance with the quadratic dependence on the flow velocity, this being in agreement with the Ergun equation.

It can be observed in the diagram that the interval of velocities for the RATsIR sorbent differs from that for the other sorbents. This is explained by the low porosity of the sorbent, this leading to a high hydraulic resistance of the sorbent layer. The measured pressure difference exceeded the pressure gage measurement limit, so the velocity range was required to be reduced.
Fig. 5a presents the results of simulating the flow current process for design option 1 in the form of a field of pressures and velocity vectors. The medium with a velocity of 0.2 m/s enters the given container filter from below and is distributed among the supply tubes to different sorbent layers. It can be seen from the figure that the sorbent layer is the biggest contributor to the flow resistance. The local resistances that occur due to the flow through the distributing tubes are small as compared with the layer resistance. The total pressure difference at the structure inlet and at the container filter outlet was about 390 kPa.

The simulation results for option 2 are presented in Fig. 5b. Here, the medium is fed from above, a part of it entering the first sorbent layer, and the rest delivered to the further sections through the holes in the perforated rings. The drawback of this design is that an additional local resistance occurs when the flow passes through the perforated rings. This manifests itself in an eddy current forming at the inlet area of each sorbent layer. The negative effect from the eddy current, apart from the resistance growth, consists in stagnant zones formed potentially in the structure corners and the wall deposition intensified. On the whole, the design with perforated rings has the highest total hydraulic resistance among the options considered (492 kPa).

Fig. 5c presents the simulation results for design option 3. The flowing medium direction is downward: the medium from the side distributing header (on the right in the figure) is uniformly distributed by the sorbent layers, and then, after being treated, is fed into the side intake header (on the left). The pressure difference between the structure’s inlet and outlet cross-sections was 115 kPa. The sorbent layers are also the biggest contributors to the resistance. Thanks to the fact that the distributing tubes are outside the body, the hydraulic resistance value could be reduced.

All options considered had the same sorbent layer cross-section area and height. In terms of the efficiency of the flowing medium distribution among the sorbent layers, the designs do not have any major differences since, as shown by the simulation, the medium flow rate in all options was distributed by the layers approximately equally.

**Conclusions**

The dependence of the hydraulic resistance of different sorbents on the flow velocity through the sorbent layer was obtained using an experimental study. The data obtained was used as the basis for a computer model. Three trap designs have been proposed with an increased efficiency of sorption processes. The simulation results have shown that design option 3 has the lowest hydraulic resistance. The undertaken calculations are hydrodynamic. They do not consider chemical interactions of the filtered medium with the sorbent.
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