UDC 621.039.74

V. Kovalchuk, Ph.D., Assoc. Prof.,
I. Kozlov, DSc, Prof.
O. Dorozh, Ph.D., Assoc. Prof.,
T. Usova
Odessa National Polytechnic University, 1 Shevchenko Ave., Odesa, Ukraine, 65044; e-mail: odorog13@gmail.com

SPECIALFEATURES OF OPERATION OF THE HIGH-TEMPERATURE FILTER OF THE NWT-1

В.І. Ковальчук, І.Л. Козлов, О.А. Дорож, Т.Ю. Усова. **Особливості експлуатації високотемпературного фільтру СВО-1.** Розглянуто особливості експлуатації байпасної системи очищення теплоносія першого контуру (СВО 1), зокрема високотемпературного механічного фільтру з зернистим завантаженням титановою крихтою. Звернуто увагу на високу механічну, термічну і хімічну стійкість. Вказано, що більш ніж 30-річний досвід експлуатації високотемпературних сорбентів виявив їх позитивні і негативні властивості. Зокрема суттєвий вплив кількості виконаних циклів сорбції-десорбції на здатність фільтруючого шару титанової крихти затримувати домішки теплоносія. Досить висока міцність зчеплення конгломерату продуктів корозії конструкційних матеріалів першого контуру с губчастим сорбентом не дозволяє повноцінно використати гідності сорбенту. Наведені рівняння, які враховують одночасне протікання процесів осадження дисперсних частинок на сорбенті та їх відривання, що може бути причиною низької ефективності очищення теплоносія. Описано основні проблеми та недоліки накопичення залишкового забруднення шару сорбенту при його зворотному промиванні. Розглянуто причини зниження сорбційної здатності при накопиченні залишкового забруднення шару. Проаналізовано вплив залишкового осаду, який залишився після попереднього промивання, на характеристики фільтрації. Розглянуто інтенсивність втрати сорбційної здатності в залежності від кількості проведених циклів сорбції-десорбції. Отримано рівняння залежності сорбційної ємності шару від кількості проведених циклів сорбції-десорбції. Підтверджено, що критерієм вичерпання сорбційної ємності фільтруючого шару повинно бути не підвищення перепаду тиску на ньому, а зниження ефективності очищення по радіонуклідам корозійного походження.

Ключові слова: фільтрування, суспензія, осад, фільтро цикл, високотемпературний фільтр

V. Kovalchuk, I. Kozlov, O. Dorozh, T. Usova. **Special features of operating the high temperature filter of the NWT-1.** The features of operation of the bypass cleaning system of the first circuit coolant (NWT 1), in particular high-temperature mechanical filter with granular loading of titanium crumb are considered. Attention is paid to high mechanical, thermal and chemical resistance. It has been stated that today there is a greater 30 years increase in the use of high-temperature sorbents in terms of positive and negative power, and a constant supply of heat to the temperature of the heating and cooling cycle. It's not possible to give a viscous mixture to a conglomerate of products in a corroded construction material with a perpetual sorbent, and it is not permissible to add an additional sorbent. More than 30 years of experience in the operation of high temperature sorbents have revealed their positive and negative properties. In particular, the significant effect of the number of cycles of sorption-desorption on the ability of the filter layer of titanium crumb to retain impurities of the coolant. The sufficiently high adhesion strength of the conglomerate of corrosion products of structural materials of the first contour with the sponge sorbent does not allow to fully utilize the advantages of the sorbent. The equations that take in to account the simultaneous deposition processes of the dispersed particles on the sorbent and their separation, which can be caused by low efficiency of coolant cleaning.The main problems and disadvantages of accumulation of residual contamination of the sorbent layer during its backwashing are described. The reasons of decrease of sorption capacity at accumulation of residual pollution of a layer are considered. The effect of residual sediment remaining after pre-washing on the filtration characteristics is analyzed. The intensity of sorption capacity loss is considered depending on the number of performed sorption-desorption cycles. The equation of dependence of the sorption capacity of the layer on the number of cycles of sorption-desorption was obtained. It is confirmed that the criterion of exhaustion of the sorption capacity of the filtering layer should not be an increase in pressure drop on it, but a decrease in the efficiency of purification of radionuclides of corrosive origin.

Keywords: filtering, suspension, sediment, filter cycle, high temperature filter

Introduction

Purification of uncooled primary coolant from suspended activated corrosion products of structural materials of the primary circuit is carried out by a special water treatment plant (SWT-1), consisting of 4 bypass lines of the main circulation pump (MCP) [1]. Each of them includes a high-temperature mechanical filter (HTF) and a filter trap (FT), providing a capacity of 100 m^3/hour. Cleaning part of the coolant allows to reduce the specific activity of the primary coolant to levels $10^{-4}…10^{-5}$ curie/dm^3, to reduce the accumulation of corrosion products in the circuit, to reduce radiation fields from active equipment by 5…10 times.

Primary coolant contaminants are represented mainly by corrosion products of structural materials and fuel shells in an amount of, on average, about 50 $\mu g/dm^3$ with a particle radius of 0.2 to 2 microns (Fig.1) [2].

DOI: 10.15276/opu.2.58.2019.05

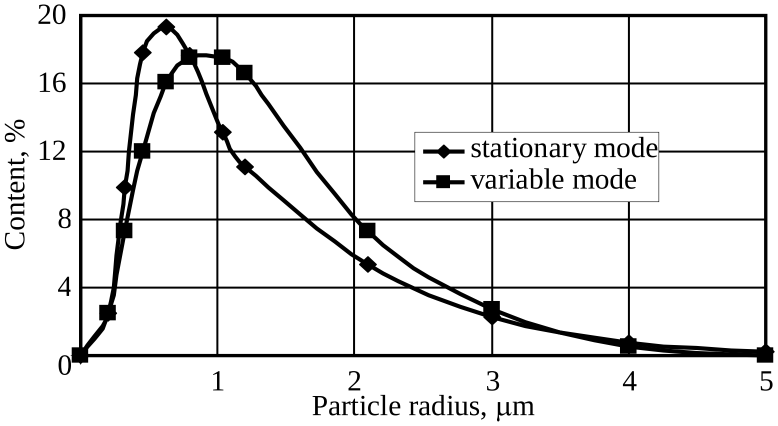

Fig. 1. Particle size distribution in the coolant of1 circuit

As a filtering material for HTF, titanium chips TP-VS-1 based on sponge titanium with fractions of 2+0.63 mm in size are used. It contains Ti and impurities as a base: iron 0.9 %, chlorides 0.02 %, nitrogen 0.05 %, hydrogen 0.1 % etc. It is obtained as a result of a reducing chemical reaction of $TiCl_4$ when interacting with magnesium in a neutral gas medium (argon, xenon) at a high temperature (950…1000°C). The finely divided granular substance obtained in this way has a porous structure (sponge) and a density of 1400 kg/m^3.

It possesses high mechanical strength and chemical resistance, retain their properties (specific surface, mechanical strength, exchange capacity, etc.) at a temperature of the treated water up to 330 °C during operation up to 10 thousand hours. The sorbent is used for water purification, including radioactive, at temperatures from 20 to 350 °C with a pH value of 5…9.5. Under the action of ionizing radiation to a fast neutron fluency of $5 \cdot 10^{25}$ neutrons/(m^2·sec), the properties of the sorbent practically do not change. Solubility is less than 1g/m^3. The presence of chlorides in the crumb composition requires pre-washing the freshly loaded sorbent.

The hydraulic resistance at a filter layer height of 1 meter and a filtration rate of 2.8 cm/s (100 m/h) is 0.007 MPa. With the accumulation of impurities, the sorbent resistance can increase to 0.04 MPa.

The high sorption ability of the sorbent ensures the extraction of substances from the solution in any state of aggregation: coarsely dispersed, colloidal and truly dissolved. The coefficient of purification from the most common pollution in the coolant, estimated by the ratio of the concentration of radioactive substances before and after decontamination, is for: iron – 2.0; copper – 1.5; hardness salts – 1.6; silicic acid – 1.4.

When the sorbent is saturated with contaminants, it is subjected to loosening washing with water or water with air by reverse current and chemical regeneration. Oxyethylene diphosphoric, oxalic and nitric acids are used as regenerating substances.

Formulation of the problem

More than 30 years of experience of operating high-temperature sorbents in the SWT-1 system showed a number of features [2, 3].

The main delay of finely dispersed corrosion products is observed in the upper (frontal) layer of the sorbent with a height of up to 400 mm. This layer is capable of retaining from 1.5 to 3 kg of active corrosion products. During the operation of the HTF, pores are clogged and fractions coarsened to 5…9 mm, which requires chemical decontamination and washing of the sorbent.

Sufficiently strong adhesion of the conglomerate of corrosion products of the structural materials of the first circuit with a sponge sorbent prevents their removal by washing and causes irreversible saturation of titanium chips with corrosion products – Fe_2O_3, NiO, Cr_2O_3, etc.

Studies of the impact of the SWT-1 installation on the radiation situation in the primary VVER-1000 circuit at the main power unit of the Zaporizhia NPP showed that the efficiency of the WTF during operation decreases and their impact on the radiation situation only affects the initial period of operation [4].

The purpose of the study

The experimental water washing of the sorbent, the effectiveness of which was estimated by the number of radionuclides removed from the filter layer, of a corrosive origin was not high. In particular, after the second period of operation of the filters, 1.6 out of almost 7.5 Ku were removed with wash water, i.e. the only about 20 % of the activity contained in the sorbent.

Chemical regeneration of the sorbent was never carried out on any of the blocks. At the same time, the need to regenerate or replace the sorbent after a certain period of its operation in connection with the loss of sorption properties and a decrease in the purification efficiency is evidenced by the entire filtration experience. The refusal from chemical regeneration of the sorbent at VVER-1000 NPPs was caused by two circumstances: a low-tech, from the point of view of providing sorbent hydraulic discharge, filter design and selection of the filter cycle termination criterion. The HTF provides for an upper hydraulic discharge using a special sorbent hydraulic discharge devices (SHDD), lowered into the filter and moved after the level of the sorbent layer during unloading (water for transporting the sorbent is supplied from below). Due to complexity, the process was not even tested at any nuclear power plant.

The purpose of the study is to quantify the effect of the duration of the filter load on the efficiency of cleaning the coolant. Therefore, the research objective is to determine the structure of the model of the sorption process and the compliance of the operational data of the selected model.

The choice of hydraulic and physico-chemical structure of the model

An indicator of the end of the working process in the filter cycle was initially traditionally accepted to achieve a pressure drop across the filter of 0.02 MPa. But high-temperature filtration, due to the turbulent flow through the bed, is characterized by the absence of a noticeable increase in the hydraulic resistance of the sorbent bed. Therefore, the criterion for the end of the filter cycle should be a decrease in the cleaning efficiency for radionuclides of corrosive origin, and a complete restoration of the properties of the sorbent should be carried out annually during the unit shutdown for repair [5].

The filtration process is characterized not only by mechanical retention of impurity particles in the intergranular space of the load. An important role in it is played by the processes of sorption of impurities by the surface of grains, especially colloidal and dissolved particles, capable of forming poorly soluble compounds with a sorbent [5].

The choice of the water-chemical regime of the coolant 1 circuit remains relevant. According to the data of the Beloyarsk NPP, it was found that the HTF operation is negatively affected by the hydrazine contained in the purified water, the concentration of which in the purified water during the experiment reached 2 mg/kg [4]. As a strong reducing agent, hydrazine promotes the dissolution of the sediment accumulated by the layer and it washing into the filtrate (as if chemical regeneration “on the go” occurs), as evidenced by the results of the analysis of the sorbent layer. Calculations show that the use of a water-chemical regime with a constant pH value of 7.1…7.2 can significantly increase the efficiency of HTF filters and reduce the return of activated corrosion products to the coolant.

To study the possibility of restoring the sorption ability of a titanium load, it is necessary to have information about the hydrodynamics of the bulk layer in the filtering and loosening washing regimes. For this, it is advisable to apply a mathematical model [6].

In normal mode, filtering is carried out at a constant speed. For this case, the clarification process is described by the equations [7]:

of balance –

$$m\frac{\partial C}{\partial t}+V\frac{\partial C}{\partial x}+\frac{\partial \rho}{\partial t}=D\frac{\partial^2 C}{\partial x^2}\,, \tag{1}$$

and

linear kinetics –

$$\frac{\partial C}{\partial x} = -bC + \frac{a}{V}\rho, \tag{2}$$

where $C=C(x, t)$ is the concentration of suspended solids in water;

$\rho=\rho(x, t)$ is the sediment concentration in the load;

x, t – spatial and temporal coordinates;

V is the filtration rate;

$m(x, t)$ is the loading porosity;

D is the diffusion coefficient;

a, b are kinetics coefficients, which respectively determine the intensity of separation and adhesion of suspended particles from the loading surface (or from previously adhering particles).

System (1, 2) is solved under boundary conditions:

$$\begin{cases} x = 0;\ C = C_0 \\ t = 0;\ \rho_0 = \rho_0(x) \\ t \to \infty;\ \partial C/\partial x = 0;\ C = C_0;\ \rho = \rho_{\max}, \end{cases}, \tag{3}$$

where C_0 is the concentration of suspension in the source water;

$\rho_0(x)$ is the concentration of sediment in the filter layer at the initial moment of time, which may be variable along its length;

$\rho_{\max}$ – maximum saturated pore space delayed by suspension at a long filtering duration.

To simplify the problem and the possibility of obtaining analytical solutions is usually assumed that the pollution layer at the beginning of filtering no ($\rho_0(x)=0$) that does not meet the conditions of cyclic operation because download never washed completely [8].

The calculations performed under the assumption of residual contamination layer about 0.06 % by mass and uniform distribution on height ($\rho(x)$=const) showed that when the concentration of the slurry of 0.5…1.0 g/dm^3 retention capacity of the layer is not more than 50…70 % of the initial value in one cycle. This suggests that remaining after washing, the precipitate is not uniformly distributed over the height of the layer and the effect of flush is the same for each layer. The concentration of the precipitate after washing the thus determined ratio:

$$\rho(x, 0) = \rho(x, T_f)(1 - E_{ew}), \tag{4}$$

where $\rho(x, T_f)$ is the distribution of sediment concentration over the layer height at the time of the end of filtration (T_f);

E_{ew} is the washing efficiency equal to the ratio of the mass of contaminants washed for washing to their initial amount.

Analysis and formalization of the results of pilot washes

Experimental flushing of operating filters suggests that the sediment remaining in the pore space after flushing has a higher density compared to the washed mass and a lower value of the kinetic coefficient a, which determines the intensity of separation of contaminants from the loading grains [9]. The dependence of washing efficiency on the number of worked cycles n, approximated by a polynomial of the form:

$$E_r = 0.1153 \cdot n^4 - 0.7885 \cdot n^3 + 1.9205 \cdot n^2 - 2.0568 \cdot n + 1,$$

indicates the advisability of replacing the sorbent after working out one cycle (Fig. 2).

This confirms the mechanism of the active influence of sediment on the deposition of suspended matter in the pore space of the load, described in [7]. On the one hand, it increases the number of vacancies for suspension particles and thereby intensifies clarification, especially at the initial stage. On the other hand, the formation of sediment gradually fills the vacancies that originally existed in the load, and adversely affects the separation of the suspension.

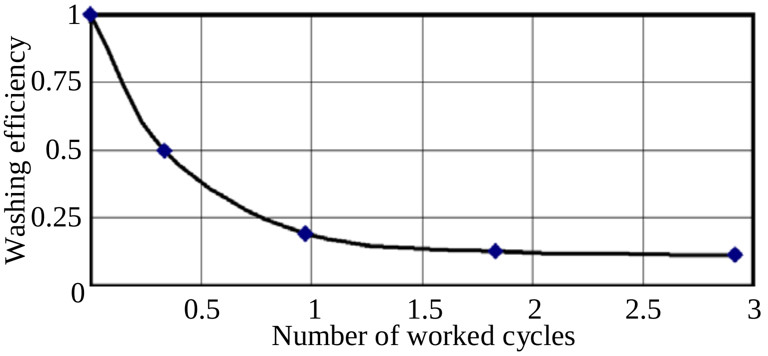

Fig. 2. The washing efficiency of titanium sorbent

The implementation of this mechanism is due to the combined influence of many factors: characterizing the linear dimensions and porosity of the layer, the mode of suspension flow through the layer, the shape of the suspension particles, their roughness, as well as the orientation of the particles, etc.

Conclusions

1. The operation of high-temperature filters in the conditions of the 1st circuit of the NPP revealed the active influence of the residual, after washing the pollution layer, on the sorption capacity of titanium chips used as a filter load.

2. Strong adhesion of the conglomerate of corrosion products of structural materials of the first circuit with a sponge sorbent prevents their removal by washing and causes irreversible saturation of titanium crumbs with corrosion products – Fe_2O_3, NiO, Cr_2O_3.

3. The selected mathematical model, taking into account the kinetic equation of sorption, including the simultaneous processes of deposition and separation of dispersed particles, can be taken as a base for developing washing regimes for a high-temperature filter.

Література

1. Ковальчук В.И., Козлов И.Л. Основы обращения с радиоактивными отходами на атомных электростанциях; Учебное пособие для студентов специальности 6.05060302 «технология теплоносителей и обращение с РАО на АЭС» Уч. пособие, Одесса : Бахва, 2013. 193 с.
2. Анализ эффективности работы высокотемпературного механического фильтра на действующих АЭС с ВВЭР-1000 / В.Г. Крицкий, В.В. Царев, Н.А. Прохоров и др. *Теплоэнергетика.* 1992. № 7. С. 15–19.
3. Гусев Б.А., Семёнов В.Г., Ефимов А.А., Панчук В.В. Поведение продуктов коррозии в первом контуре ЯЭУ с водным теплоносителем. *Вестник СПбГУ. Сер. 4.* 2012. Вып. 4. С. 110–118.
4. Ефимов А.А Влияние высокотемпературной фильтрации на состав примесей теплоносителя первых контуров энергоблоков с ВВЭР-1000 / А.А. Ефимов, Л.Н. Москвин, Б.А. Гусев и др. *Теплоэнергетика.* 1992. № 10. С. 49–52.
5. Гусев Б.А., Красноперов В.М. Влияние системы высокотемпературной очистки теплоносителя на АЭС с ВВЭР-1000 на формирование дозовых нагрузок. *Теплоэнергетика.* 2011. № 5. С. 34–36.
6. Повалишин Н.Б., Щелик С.В., Гузеева Г.И. Внутрикорпусная регенерация сорбента высокотемпературных фильтров СВО-1 блока 3 Калининской АЭС. *Материалы 7-го Международного научно-технического совещания.* (Тверь, 17–19 октября 2006 г.). Тверь, 2006. С. 13–15.
7. Грабовский П.А. Фильтрование воды через зернистый слой с убывающей скоростью. *Доп. НАН України.* 2016. № 8. С. 40–45. DOI: https://doi.org/10.15407/dopovidi2016.08.040.
8. Грабовский П.А., Гуринчик Н.А. Остаточные загрязнения при математическом описании фильтрования воды через зернистую загрузку. http://ogasa.org.ua. 2005, С. 49–53. URL: http://mx.ogasa.org.ua/bitstream/123456789/1475/1/Грабовский%20П.%20А.%2C%20Гуринчик%20Н.%20А.%20Остаточные%20загрязнения%20при%20математичес...pdf. (дата звернення 20.09.2019).

9. Поляков В.Л. Теоретический анализ длительности фильтроцикла. *Химия и технология воды*. 2009. 31, № 6. С. 605–618.

References

1. Kovalchuk, V.I., & Kozlov, I.L. (2013). *Fundamentals of radioactive waste management in nuclear power plants: textbook.* Odessa: Bahva.
2. Kritsky, V.G., Tsarev, V.V., & Prokhorov N.A et al. (1992). Analysis of the performance of a high-temperature mechanical filter at existing NPPs with WWER-1000. *Thermal Engineering, 7*, 15–19.
3. Gusev, B.A., Semenov, V.G., Efimov, A.A., & Panchuk, V.V. (2012). The behavior of corrosion products in the primary circuit of a nuclear power plant with a coolant. *Bulletin of St. Petersburg State University. Ser. 4, 4*, 110–118.
4. Efimov, A.A. (1992) The effect of high-temperature filtration on the composition of the coolant impurities of the first circuits of power units with WWER-1000. Efimov, A.A., Moskvin, L.N., Gusev, B.A. et al. *Thermal engineering, 10,* 49–52.
5. Gusev, B.A., Krasnoperov, V.M. (2011). The influence of the system of high-temperature cleaning of the coolant at nuclear power plants with VVER-1000 on the formation of dose loads. *Thermal Engineering. 5.* 34–36.
6. Povalishin, N.B., Shchelik, N.E., & Guzeeva G.I. (2006). Intra-shell regeneration of the sorbent of high-temperature filters sv-1 of unit 3 of Kalinin NPP. *7th International Scientific and Technical Meeting "Water-chemical regime of nuclear power plants"* (VNIIAES, October 17–19, 2006). Tver, pp. 13–15.
7. Grabovsky, P.A. (2016). Filtering water through a granular layer at a decreasing rate. *Nac. akad. nauk Ukr.*, 8, 40–45. DOI: https://doi.org/10.15407/dopovidi2016.08.040.
8. Grabovsky, P.A. & Gurinchik, N.A. (2005). *Gurinchik Residual pollution in the mathematical description of filtering water through a granular charge.* http://ogasa.org.ua. pp. 39–53. Retrieved from: http://mx.ogasa.org.ua/bitstream/123456789/1475/1/Грабовский%20П.%20А.%2С%20Гуринчик%20Н.%20А.%20Остаточные%20загрязнения%20при%20математичес...pdf. (Last accessed: 20.09.2019).
9. Polyakov, V.L. (2009). Theoretical analysis of the duration of the filter cycle. *Chemistry and Water Technology, 6*, 605–618.

Ковальчук В'ячеслав Іванович; Kovalchuk Vyacheslav, ORCID:https://orcid.org/0000-0001-8696-4414
Козлов Ігор Леонідович; Igor Kozlov, ORCID: http://orcid.org/0000-0003-0435-6373
Дорож Ольга Анатоліївна; Olga Dorozh, ORCID: https://orcid.org/0000-0001-8495-2911
Усова Тетяна Юріївна; Usova Tatiana, ORCID: https://orcid.org/0000-0002-0297-2925

Received September 05, 2019

Accepted October 03, 2019