The role of ion and thermal sputtering processes of radioactive deposits in applying the ion-plasma deactivation technology for nuclear power plants

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One of the actual problem of nuclear engineering is the decommissioning of reactors and decontamination of reactor equipment, as well as the decontamination of the surfaces of the primary coolant system during scheduled shutdowns of nuclear power plants. To solve this problem, we develop the ion-plasma „dry" deactivation technology. The calculation of ion and thermal sputtering rates of reactor graphite and radioactive deposits from deactivated of the internal circuit equipment surfaces of nuclear power plants are presented.

Keywords: decommissioning, surface radioactive deposits, nuclear power plants, ion-thermal deactivation technology, metal structure surface decontamination, reactor graphite.

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The problem of decommissioning of nuclear power plants (NPPs) sets an objective of finding efficient processes for deactivation of metal structures of NPPs and reactor graphite. A total of 15 VVER reactors and 11 RBMK reactors are planned to be decommissioned by the year 2049 [1]. It is also known that ~ 260 thousand tons of irradiated reactor graphite have been accumulated worldwide ($\sim 50-60$ thousand tons are located in Russia). Various techniques for graphite deactivation proposed in recent years, which include acid etching, abrasive treatment, grinding into fractions, thermal evaporation, etc., entail a manyfold increase in the volume of secondary radioactive waste (RAW) [2,3]. However, the data [4] on spatial distribution of dose-forming radionuclide ^{14}C (with a half-life of 5700 years), which constitutes 95% of activity in a graphite assembly, indicate that it is localized primarily in the near-surface layer (at a depth up to 5 mm). This opens the way for development of new approaches to deactivation of reactor graphite.

Radioactive contamination of the inner surfaces of NPP pipelines is caused by activated products of corrosion of metal structures of the reactor and, in certain cases, fission products (due to their transfer by the coolant). For example, corrosion of stainless steel structures produces deposits in the form of spinels $Ni_nCr_mFe_{3-n-m}O_4$ ($n = 0-2$, $m = 2-n$), $Ni_nFe_{3-n}O_4$ ($n = 0-2$) or mixed cobalt and nickel ferrites $(CoO)_x(NiO)_y(FeO)_{1-x-y}Fe_2O_3$, where $1 > y \gg x$ [5]. However, traditional techniques for deactivation of internal circuit equipment of NPPs (chemical, ultrasonic, electrolytic, etc.) increase the volume of secondary RAW and induce corrosion of fittings and pipelines; in addition, the choice of a specific solution, its concentration, and deactivation mode depends in a complex and ambiguous way on the nature of the radioactive deposit and the metal being deactivated [6,7].

In the present study, we propose a „dry" ion-plasma deactivation technique that allows one to remove the ^{14}C-enriched surface layer of reactor graphite and the radioactive layer from NPP metal structure surfaces.

This ion-plasma technique relies on ion and thermal sputtering of radioactive deposits from the deactivated surface by igniting a shortened discharge (with a discharge gap length on the order of $1-5$ mm) between the surface to be deactivated (cathode) and a renewable collector electrode (anode) in an inert gas (argon) at a pressure of $0.1-1$ atm. The operating voltage across the discharge gap is $100-1000$ V, and the current density is $0.1-1$ A/cm^2. A mobile sputtering device [8], which is shifted by a manipulator along the deactivated surface, is used to implement this technique. A sputtering device design with a renewable collector electrode (a plate similar in shape to the inner pipeline surface) [8,9], which is also moved inside the pipes by a manipulator, is used to deactivate the inner surface of NPP pipelines. Radioactive contaminants are removed by means of ion and thermal sputtering of surface atoms in a shortened discharge; ion sputtering is provided in this case by argon ions that acquire an energy in excess of 100 eV in the cathode drop region. The ion sputtering rate and the thickness of the sputtered layer depend on the power input into the discharge. Atoms sputtered from the deactivated surface are deposited onto the collector electrode (anode) in the form of a solid precipitate in the mass transfer mode. It should be emphasized that the process in question does not produce secondary RAW, since sputtered radionuclides do not react with inert gas (argon). A detailed description of this technique is presented in our

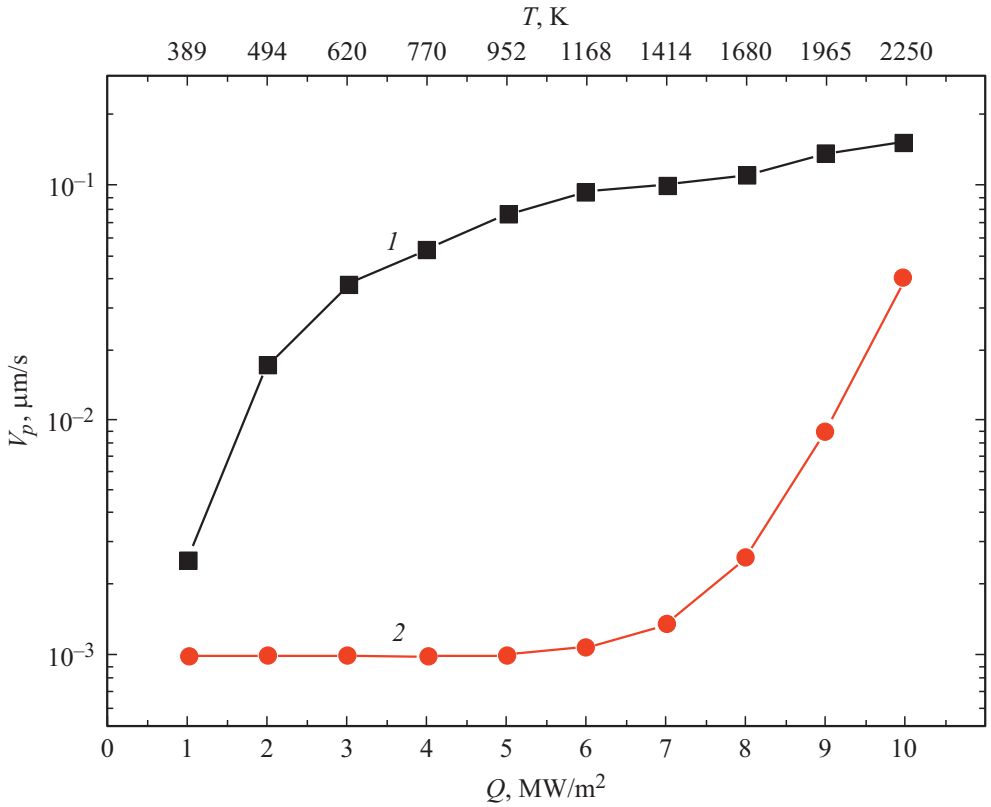

Figure 1. Dependences of rate V_p of ion (*1*) and thermal (*2*) sputtering of reactor graphite on power input Q into the discharge and the corresponding temperature T of the processed graphite surface.

patent prepared in collaboration with State Atomic Energy Corporation Rosatom [10].

The heat conduction problem was solved to estimate the technological rate of thermal sputtering of reactor graphite, and the maximum temperatures on the surface of an RBMK graphite (grade GR-280) assembly in the process of its deactivation by the sputtering device were determined as functions of discharge power (10^6–10^7 W/m^2). The obtained values of temperature of the treated graphite surface fall within the 400–2250 K range. Using the data from [11] on rates of graphite evaporation in an argon environment at a pressure of 1 atm within the temperature range of 2200–2600 K, we determined the rate of thermal sputtering of reactor graphite within the temperature range of 400–2250 K by extrapolation. The rate of ion sputtering of reactor graphite was calculated as

$$V_p = KjM_C/eN_a\rho, \qquad (1)$$

where K is the graphite sputtering ratio, e is the electron charge, ρ is the reactor graphite density [g/cm^3], j is the ion current density, M_C is the mass of carbon atoms, and N_a is the Avogadro number. In a shortened discharge used in deactivation, 90% of the 100–1000 V voltage in the discharge gap actually drop at the cathode; therefore, argon ions near the cathode acquire an energy in excess of 100 eV. The sputtering ratio of reactor graphite by argon ions with an energy of 100–1000 eV was calculated using the SRIM (Stopping and Range of Ions in Matter) program [12], which relies on the Monte Carlo method and the approximation of binary collisions between ions and target atoms. The dependences of rates of ion and thermal sputtering of reactor graphite on the discharge power and the corresponding temperatures of the treated graphite surface are shown in Fig. 1. The obtained data make it clear that thermal sputtering produces a contribution when the temperature of the deactivated surface exceeds 2000 K; under such conditions, the process of thermal diffusion of other radionuclides from the bulk of a graphite assembly to the surface is also accelerated, facilitating decontamination of the graphite matrix.

The mechanism of ion sputtering of radioactive deposits also contributes to deactivation of metal structures of NPPs. The rates of sputtering by argon ions with energies of 200–600 eV were calculated for metal structures with radioactive $NiCrFeO_4$, $NiFe_2O_4$, Fe_2O_3, TiO_2, and U_3O_8 deposits; the sputtering ratios were obtained using SRIM. Figure 2 presents the dependences of rates of ion sputtering of the above-indicated example deposits on the power input into the discharge. As was demonstrated in [10], it is crucial to avoid overheating of the treated surface in the process of deactivation of metal structures of NPPs. In view of this, the optimum deactivation mode is pulsed ignition of a discharge with a duration of 1–2 s and a power no higher

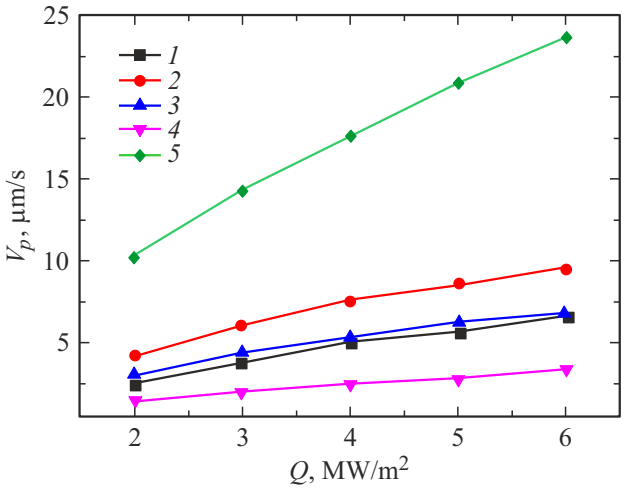

Figure 2. Dependences of rate V_p of ion sputtering of radioactive deposits (*1* — $NiCrFeO_4$, *2* — $NiFe_2O_4$, *3* — Fe_2O_3, *4* — TiO_2, *5* — U_3O_8) on the internal circuit equipment of NPPs on power input Q into the discharge.

than $6 \cdot 10^6$ W/m^2 input into it. Under such conditions, the deactivated surface temperature does not rise above 1000 K. Therefore, the ion mode of sputtering of radioactive deposits on the inner surfaces of the internal circuit equipment is preferable.

Thus, two pressing issues in decommissioning of NPPs (deactivation of the internal circuit equipment and handling of irradiated reactor graphite) were considered. A mobile ion-plasma technology allowing for the removal and localized collection of radionuclides directly at the NPP site with the volume of secondary and liquid radioactive waste kept at a minimum is proposed as a solution to these problems. The process of deactivation of GR-280 reactor graphite was used as an example to calculate the rates of ion and thermal sputtering of the surface layer; the total sputtering rate at a power input into the discharge of $\sim 1 \cdot 10^7$ W/m^2 was found to be on the order of 0.1 μm/s. It was demonstrated that thermal sputtering produces a contribution to reactor graphite deactivation at temperatures above 2000 K. The rates of ion sputtering of radioactive $NiCrFeO_4$, $NiFe_2O_4$, Fe_2O_3, TiO_2, and U_3O_8 deposits on the inner surfaces of the internal circuit equipment of NPPs were also calculated. These rates varied within the 5–25 μm/s range depending on the deposit type. The obtained rates of sputtering of radioactive deposits will be used to estimate the time of deactivation of NPP facilities during scheduled shutdowns and decommissioning.

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Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] B.K. Bylkin, G.B. Davydova, E.A. Zhurbenko, Atom. Energy, **110** (3), 203 (2011). DOI: 10.1007/s10512-011-9411-7.
[2] V.F. Myshkin, E.V. Bespala, A.O. Pavlyuk, Yu.R. Bespala, I.Yu. Novoselov, Izv. Vyssh. Uchebn. Zaved. Yad. Energ., No. 3, 154 (2019) (in Russian). DOI: 10.26583/npe.2019.3.14
[3] D. Vulpius, K. Baginski, B. Kraus, B. Thomauske, Nucl. Eng. Des., **265**, 294 (2013). DOI: 10.1016/j.nucengdes.2013.09.007
[4] D. LaBrier, M.L. Dunzik-Gougar, J. Nucl. Mater., **460**, 174 (2015). DOI: 10.1016/j.jnucmat.2015.01.063
[5] S.N. Orlov, A.A. Zmitrodan, V.V. Krivobokov, Therm. Eng., **68** (5), 361 (2021). DOI: 10.1134/S0040601521040042.
[6] S. Liu, Y. He, H. Xie, Y. Ge, Y. Lin, Z. Yao, M. Jin, J. Liu, X. Chen, Y. Sun, B. Wang, Sustainability, **14** (7), 4021 (2022). DOI: 10.3390/su14074021
[7] R.N. Yastrebinsky, V.I. Pavlenko, A.A. Karnauhov, IOP Conf. Ser.: Mater. Sci. Eng., **687** (6), 066053 (2019). DOI: 10.1088/1757-899X/687/6/066053
[8] A.S. Petrovskaya, A.B. Tsyganov, RF Patent 2771172 (2022) (in Russian).
[9] A.S. Petrovskaya, A.B. Tsyganov, Phys. Solid State, **65** (12), 2095 (2023). DOI: 10.61011/PSS.2023.12.57674.5167k.
[10] A.S. Petrovskaya, A.B. Tsyganov, M.R. Stakhiv, RF Patent № 2711292, EP 19888171.6, US 20210272715, CA3105179A1, CN112655056A (2020).
[11] A.E. Chalykh, *Tekhnologiya i oborudovanie elektrodnykh i elektrougol'nykh predpriyatii* (Metallurgiya, M., 1972) (in Russian).
[12] J.F. Ziegler, M.D. Ziegler, J.P. Biersack, Nucl. Instrum. Meth. Phys. Res. B, **268** (11), 1818 (2010). DOI: 10.1016/j.nimb.2010.02.091

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