About inert gas selection for the application of ion-plasma technology deactivation of reactor equipment

© A.S. Petrovskaya, A.B. Tsyganov

InnoPlasmaTech LLC, Saint-Petersburg, Russia
E-mail: anita3425@yandex.ru

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The deactivation of reactor equipment during scheduled shutdowns and decommissioning is one of the actual problems of nuclear power engineering. The „dry" ion-plasma deactivation technology that we are developing to solve this problem and allows to avoid the formation of secondary liquid radioactive waste. In this paper, we calculate the sputtering yields and rates of ion sputtering of deposits in the oxides and spinels form for different ions of inert gases in order to select the most effective inert gas for deactivation. It was found that argon is the most effective for implementing our technology.

Keywords: decommissioning, surface radioactive deposits, nuclear power plants, ion-thermal deactivation technology, reduction of secondary radioactive waste, reactor equipment.

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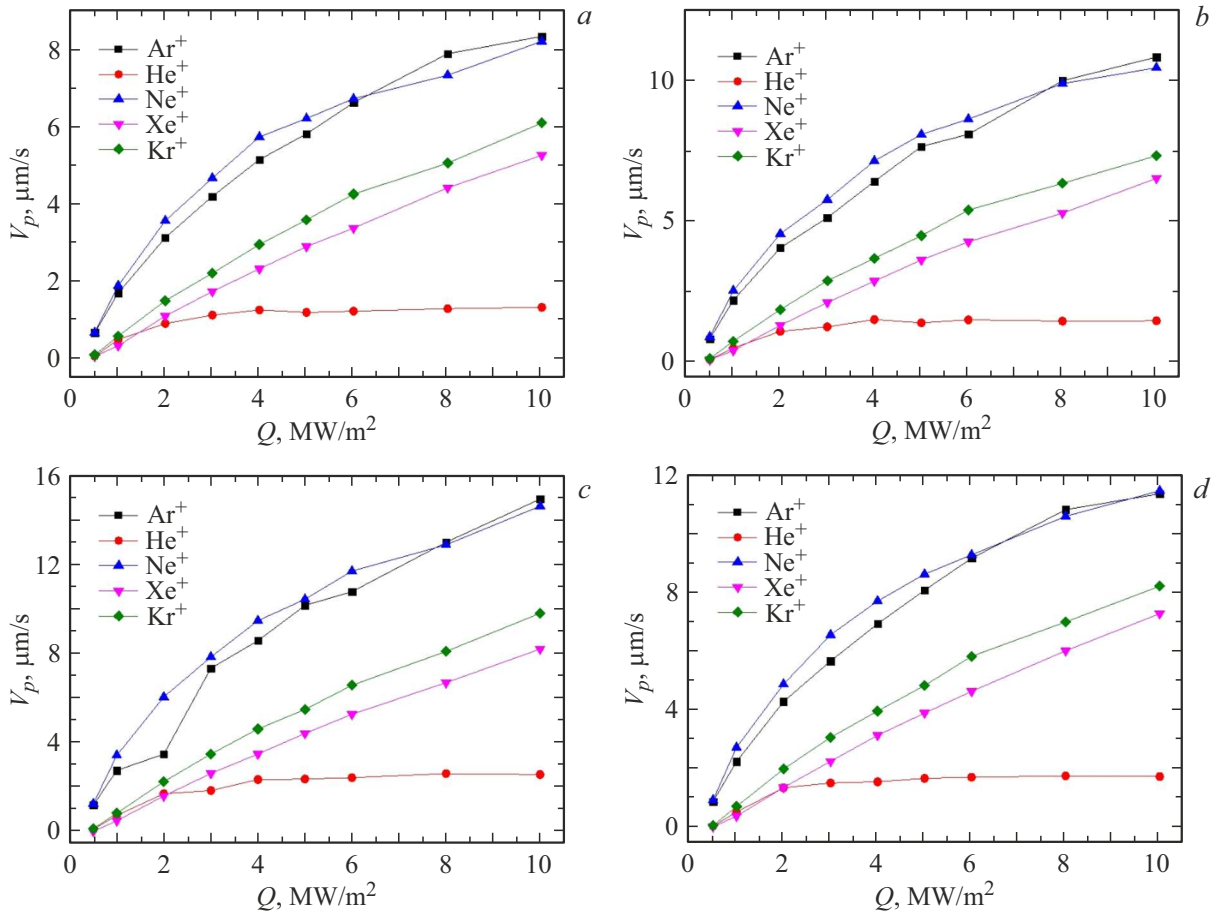
In order to resolve the pressing nuclear engineering problem of deactivation of internal equipment of reactor plants (RPs), we are developing a fundamentally new „dry" ion-plasma technology based on the use of a shortened discharge in an inert gas for removal of surface radioactive deposits. Radiation contamination of the inner surfaces of RP pipelines takes the form of activated corrosion products: compounds of metal oxides and spinels with general formula $Ni_nCr_mFe_{3-n-m}O_4$ ($n = 0-2$, $m = 2-n$), $Ni_nFe_{3-n}O_4$ ($n = 0-2$) or mixed cobalt–nickel ferrites $(CoO)_x(NiO)_y(FeO)_{1-x-y}Fe_2O_3$, where $1 > y \gg x$ [1]. For example, deposits in the active zone of a water-moderated power reactor for steam generator tubes made of nickel alloys consist of an inner $Ni_xFe_{1-x}Cr_2O_4$ ($0 < x < 1$) and an outer $Ni_xFe_{3-x}O_4$ ($x = 0.6$) layers (or magnetite Fe_3O_4 in the case of steam generator tubes made of stainless steel). In the VK-50 reactor (a 50 MW multipurpose research boiling-water reactor), the composition of deposits is dominated by the γ-Fe_2O_3 oxide; in transport nuclear power plants, deposits contain Fe_3O_4, α-Fe_2O_3, γ-Fe_2O_3, and γ-FeOOH compounds with admixed alloying elements (Cr, Ni, Mn) in various ratios [2]. The chemical processes of surface radioactive deposits are rather complex, and their chemical composition is determined by the RP type and the conditions of its operation. For example, the formation of ferrous oxide hydrate Fe_3O_4 is dominant at low temperatures in a deaerated water coolant. In oxygen-saturated water, iron is oxidized to γ-FeOOH, which is transformed into loose hematite α-Fe_2O_3 powder at $T > 520$ K. This powder is reduced by the metal to magnetite Fe_3O_4, which is oxidized by oxygen to α-Fe_2O_3. Wustite (FeO), maghemite (γ-Fe_2O_3), goethite (α-FeOOH), akaganeite (β-FeOOH), and spinels of the $NiO(Cr,Fe)_2O_3$ type [3] also form upon contact with a water coolant. Radioactive deposits often form duplex scales with spinel dispersed in the simple oxide matrix of the main element. When Fe–Cr alloys are oxidized, FeO and $FeCr_2O_4$ first form around the metal. Following that, FeO transforms into Fe_3O_4, which reacts with $FeCr_2O_4$ to produce spinel $Fe(FeCr)_2O_4$. Thus, Fe_2O_3 is produced on the outer oxide surface, and spinel is transformed into $(FeCr)_2O_3$ [4]. The most frequently and widely used methods for deactivation of internal RP equipment are radiochemical ones. These techniques lead to an increase the secondary liquid radioactive waste (LRW) volumes and corrosion of fittings and pipelines; the type of solution, its concentration, and deactivation process parameters depend on the radioactive deposit type and the deactivated metal [5,6].

We propose a fundamentally new „dry" ion-plasma technology, which does not entail the production of secondary LRW and allows one to remove a radioactive deposit layer of arbitrary composition by ion sputtering, as an effective solution to the problem of the internal equipment deactivation.

Physically, the ion-plasma deactivation is based on the ion sputtering of radioactive deposits from the deactivated surface with a shortened discharge (discharge gap on the order of 1–5 mm) ignited between the deactivated surface (cathode) and the replaceable collector electrode (anode) in an inert gas (argon) under the pressure of 0.1–1 atm, the discharge gap of 100–1000 V, and the current density of 0.1–1 A/cm^2 [7,8]. Deposits are sputtered by inert gas ions that have acquired kinetic energy in the cathode drop region of the shortened discharge; sputtered atoms are deposited on the collector electrode (anode) in the form of a solid precipitate. The detailed description of ion-plasma technology is presented in our patent prepared in collaboration with State Atomic Energy Corporation Rosatom [9]. The deactivation time and the rate of ion sputtering depend on the shortened discharge parameters,



Dependences of the ion sputtering rate V_p of surface deposits on the power input into the discharge. *a* — Fe_2O_3, *b* — Fe_3O_4, *c* — $NiCrFeO_4$, *d* — $FeCr_2O_4$.

the type of inert gas used, and the formula of the compound being removed. To determine the most effective gas medium for deactivation, we used the SRIM (Stopping and Range of Ions in Matter) program [10] to perform the numerical calculation of ion sputtering yields for radioactive deposits on internal RP equipment in the form of oxides and spinels FeO, Fe_3O_4, Fe_2O_3, $NiFe_2O_4$, $NiCrFeO_4$, $FeCr_2O_4$, and $NiCr_2Fe_2O_4$ in various inert gases (He, Ar, Ne, Xe, Kr). Calculations were performed for He^+, Ar^+, Ne^+, Xe^+, and Kr^+ ions with energies within the range of 50–1000 eV and were aimed at finding the optimum inert gas supporting the most efficient ion sputtering of the indicated deposits. The obtained sputtering yields for He^+, Ar^+, Ne^+, Xe^+, and Kr^+ ions were used to calculate the sputtering rate:

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

where K is the sputtering yield, e is the electron charge, ρ is the density of the sputtered deposit (oxide and/or spinel) [g/cm^3], j is the ion current density, M_C the oxide and/or spinel molar mass [g/mol], and N_a is the Avogadro number.

The figure presents the dependences of the ion sputterinf rates of Fe_2O_3, Fe_3O_4, $NiCrFeO_4$, and $FeCr_2O_4$ surface deposits by inert gas ions He^+, Ar^+, Ne^+, Xe^+, and Kr^+ on power input Q into the discharge.

The presented data make it clear that the highest values of sputtering yields and, consequently, ion sputtering rates are achieved when the shortened discharge is ignited in argon and neon and surface deposits are sputtered with Ar^+ and Ne^+ ions, respectively. Similar results are observed when other oxides and spinels (e. g., FeO, $NiFe_2O_4$, $NiCr_2Fe_2O_4$, etc.) are sputtered. Specifically, the sputtering yield for spinel $NiFe_2O_4$ and Ar^+ and Ne^+ ions is on the order of 2 at the energy of 500 eV, and the layer sputtering rates are 8 and 9 μm/s at the ion current density of 1 A/cm^2. Thus, since the physical parameters of sputtering with argon and neon are virtually identical, cheaper argon is preferable from the economic point of view. The efficiency of sputtering of various model alloys (stainless steel, zirconium, non-radioactive surface deposits on stainless steel in the form of oxides and spinels) and reactor graphite with argon ions was demonstrated experimentally using the laboratory installation prototype. The following process parameters ensuring efficient sputtering in experiments with argon used for igniting the shortened discharge and an Ar^+ ion source were determined: argon pressure, 0.65 atm; discharge gap,

no wider than 2 mm; the discharge gap, 500 V; current density, 1 A/cm^2. To avoid overheating of the deactivated surface, one should use the pulsed discharge ignition mode (see our study [11]). The technology was tested in laboratory conditions in experiments on deactivation of reactor graphite. Specifically, the sputtering of the graphite surface layer and the sputtered graphite deposition onto nickel and tantalum collector electrodes (anodes) were demonstrated. It is known that dose-forming isotope ^{14}C is localized predominantly on the graphite blocks surface in spent graphite stacks of high-power channel-type reactors (RBMK). Responding to the IAEA Secretariat invitation, we have presented earlier results of research on ion-plasma technology as applied to the deactivation of reactor graphite the a technical meeting [12].

The present study is focused on the internal reactor equipment deactivation during scheduled shutdowns and their decommissioning, which is the pressing problem in nuclear power engineering, and the search for effective techniques for removing radioactive deposits from the surfaces of RP equipment. To solve this problem, we have proposed a fundamentally new approach: „dry“ ion-plasma deactivation technology, which does not involve radiochemistry and the use of solutions, does not produce secondary LRW, and allows one to remove surface deposits by ion sputtering in an inert gas medium and deposit them in solid form onto collector electrodes. The sputtering yields for radioactive deposits in the form of oxides and spinels FeO, Fe_3O_4, Fe_2O_3, $NiFe_2O_4$, $NiCrFeO_4$, $FeCr_2O_4$, and $NiCr_2Fe_2O_4$ and inert gas ions He^+, Ar^+, Ne^+, Xe^+, and Kr^+ with energies falling within the range of 50−1000 eV were calculated in order to choose the most effective inert gas medium for deactivation. It was found that the highest values of sputtering yields and, consequently, ion sputtering rates are observed when a shortened discharge is ignited in argon and neon. Thus, since the physical parameters of sputtering in argon and neon are virtually identical, cheaper argon is preferable from the economic point of view. The obtained ion sputtering rates of radioactive deposits ($0.5-20\,\mu$m/s) may be used at the next stage of research work to set up the experiment on measurement of the sputtering rates of spinels, compare theoretical and experimental sputtering rate values, and estimate the RP deactivation times during scheduled shutdowns and decommissioning.

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Conflict of interest

The authors declare that they have no conflict of interest.

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