Coaxial Source of Ar^+ Ions with a Wide Energy Spectrum for Modification of Long Cylindrical Surfaces

B.A. Kalin, N.V. Volkov, V.V. Osipov, S.Yu. Nakvasin, D.N. Altukhov,
S.N. Timoshin, I.V. Oleynikov, O.V. Shcherbakov

Moscow Engineering Physics Institute (State University),
31 Kashirsko shosse, Moscow 115409 Russia, (095)323-9210, (095)324-3165,
E-mail: nvvolkov@mephi.ru

Abstract – A construction of the type ILU-03 installation to treat long-length cylindrical details is presented. The construction of an ion coaxial source, in which the conditions of high-voltage discharge are realized in the axial-cylindrical magnetic field (the pressure of residual gases is lower than $1 \cdot 10^{-2}$ Pa), is considered as the main element of the installation. A possibility to form a radial flow of Ar^+ ions with a wide energy spectrum at the mean ion energy of 5–10 keV is illustrated. The feature of the ion source is application of mutually perpendicular magnetic and heterogeneous electric fields that raise density of a gas-discharge, which allows raising the ion current density up to 1 $\mu A/cm^2$ and achieving a uniformity of the surface treatment on sections up to 100–200 mm in the axial direction. The developed ion source is supposed to use for a multi-component alloying of the external surface of zirconium alloy fuel claddings to improve their corrosion resistance with reference to exploitation conditions of water-moderated reactors.

1. Introduction

One of the main methods making it possible to increase the corrosion resistance of fuel claddings is to use modern radiation-beam technologies, among which is the method of ion mixing that permits to carry out a simultaneous penetration of practically any alloying atoms in various combinations [1]. At present time, ion-beam installations are used, as a rule, with a narrow energy spectrum (Fig. 1) of ions in the beam – the monoenergetic beam (the ratio between the width of an energy distribution ΔE measured at a half-height and its mean value $<E>$ is equal to a value of the order $\sigma=\Delta E/<E>=0.1$–5 %). However, in a number of cases it is appropriate to irradiate by an ion beam with a wide energy spectrum – the polyenergetic beam (σ=10–50 %).

For example, differences are observed in the processes taking place under irradiation by a polyenergetic ion beam in comparison with those occurring under irradiation by a monoenergetic beam of the same ions at experiments on the study of the radiation erosion of materials (sputtering, formation of a surface relief) [2, 3] and radiation-enhanced migration of implanted atoms (Fig. 2) [4].

These differences depend above all on features of the energy release *Fq*, the distribution of radiation defects, and the gradient of internal mechanical stresses in the surface layers of irradiated materials.

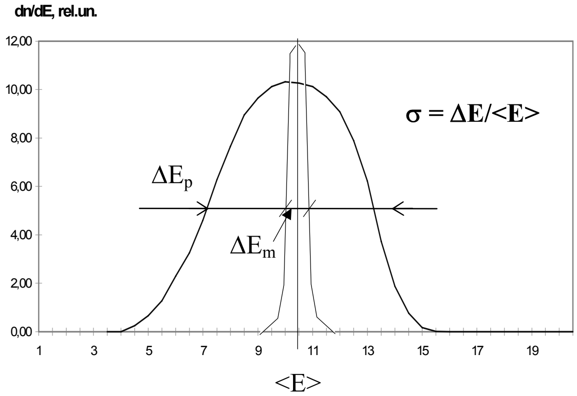

Fig. 1. The typical energy spectrum of ions in the beam: 1, a monoenergetic ion beam σ=1–5 %; 2, a polyenergetic ion beam σ=30–45 %

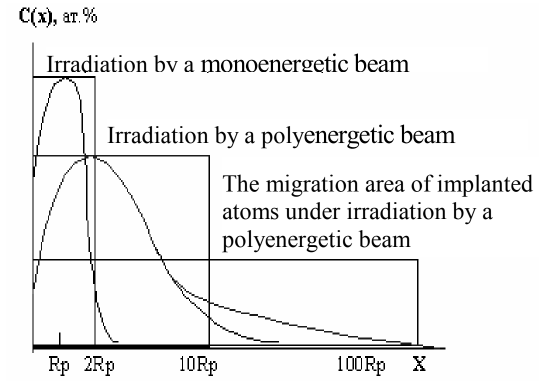

Fig. 2. The distribution of implanted atoms in depth of a material under irradiation by mono- and polyenergetic ion beams (*Rp* is the projective range of ions with a mean energy $<E>$)

The qualitative change of the energy release *Fq* in a surface layer under irradiation by monoenergetic (E=10 keV) and polyenergetic (with a mean energy $<E>$=10 keV) Ar^+ ion beams is presented in Fig. 3. It is seen from the figure that the energy release is higher under irradiation by a monoenergetic beam at depths less than Rp_m. At depths more than Rp_m, it appears to be higher under irradiation by a polyenergetic beam.

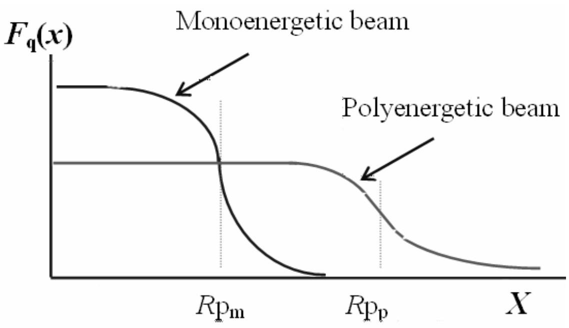

Fig. 3. The energy release *Fq* at the target depth: Rp_m, the projective range of Ar^+ ions of a monoenergetic beam (with the energy *E*); Rp_p, the projective range of ions of a polyenergetic beam (a mean energy $<E>$)

One of the methods to estimate the efficiency of ion-mixing B under penetration of atoms from the films (including multi-layer films) deposited on the matrix surface can be the condition suggested at the isotropic-mixing model [5]: $B=<X_m>^2/D^*F_q$, where $<X_m>$ is the maximum depth of atom penetration; *D*, the dose of irradiation; F_q, the energy release in the material of a substrate. Table 1 shows values *B* under irradiation of "film-substrate" systems (film – Al, Fe, Mo; substrate – Be, Zr) by monoenergetic and polyenergetic Ar^+ ion beams which illustrate that the value B is several times higher under irradiation by a polyenergetic beam under equal conditions of irradiation (the energy load per a unit surface, the dose of irradiation) [6, 7, 8].

Table 1. The efficiency of ion-mixing *B* under irradiation by monoenergetic (B_m) and polyenergetic (B_p) ion beams, (Ar^+, $<E>=5$ keV, $D=4\cdot10^{17}$ cm^{-2})

Film's atom	Substrate material	B_m, 10^{-3}, nm^5/eV	B_p, 10^{-3}, nm^5/eV
Al	Be	0.46	1.9
Ti		0.28	3.6
Mo		0.09	1.2
Al	Zr	0.02	0.7
Fe		0.01	5.2
Mo		0.01	8.4

Thus, the application of ion beams for alloying of surface layers with activation of radiation-enhanced migration is more preferable in comparison with merely radiation penetration. The application of polyenergetic beams is also expedient to reduce the radiation-induced formation of defects in the modified layer as the density of defect decreases, while the accumulation of penetrated atoms of a working gas in the surface layer appears to be inessential (including its outlet along drains).

The above-mentioned advantages to apply ion beams with a wide energy spectrum were found to be important at the development of a complex Al-Fe-Mo-alloying of zirconium alloys by ion mixing [9].

2. Construction of the installation

Because of a need to simultaneously alloy by Al, Fe, and Mo atoms of the external surface of long-length cylindrical details of Zr (Al, Ti, and others) with a diameter of the external surface up to 14 mm as applied to fuel claddings, a coaxial source of ions generating a radial ion beam with a wide energy spectrum was developed. The feature of this source is the construction of its discharge chamber (Fig. 4) composed of a cylindrical anode (1), a cathode (2) which is the most work piece, and polar tips of an electromagnet (3).

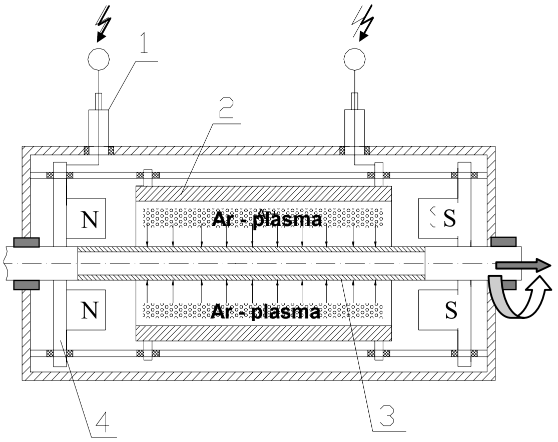

Fig. 4. The construction of a discharge chamber of the Ar^+ ion source with a wide energy spectrum: 1, the anode; 2, the cathode (a tube to be processed); 4, the area of the working gas ionization, 5, the high-voltage bushing

When the potential between the anode and the cathode is higher than U_{AC}=3–5 kV and the intensity of a magnetic field is about 0.3 T, free electrons move along circular orbits in the 30–50 mm radius range and ionize the working Ar-gas (4). To increase the efficiency of ionization, the polar tips (3) are at a negative potential about – (50–100 V) in relation to that of the cathode (2). It appears to be sufficient to block electrons at the anode-cathode discharge space (4). As shown by experiments (Fig. 5), the discharge current is stable at the 10^{-2}–10^{-4} Pa pressure range. At that, its maximum value is achieved at the working gas pressure of $3...5\cdot10^{-3}$ Pa. Oscillations of the ion current, which seem to be caused by the features of a cyclotron resonance of the electron constituent of a gas discharge, occur at the pressure less than $2\cdot10^{-4}$ Pa [10, 11].

To satisfy the necessary vacuum conditions, the discharge chamber is placed into the vacuum chamber with a diameter *Dy*=160 mm. The magnet windings are at atmosphere pressure, and the magnetic flow closes through the chamber wall.

Because of such a composition, the load on the vacuum system reduces and the presence of admixtures significantly decreases. It positively influences the processing of such a reactive material as Be, Al, Ti, Mo, and Zr. The optimal parameters of alloying by ion mixing [12, 13, 14, 15, 16], obtained as a result of carrying out the experiments on alloying of polycrystalline matrixes by a polyenergetic Ar^+ ion beam on the installation "VOKAL" [17], were taken into account during development of the installation.

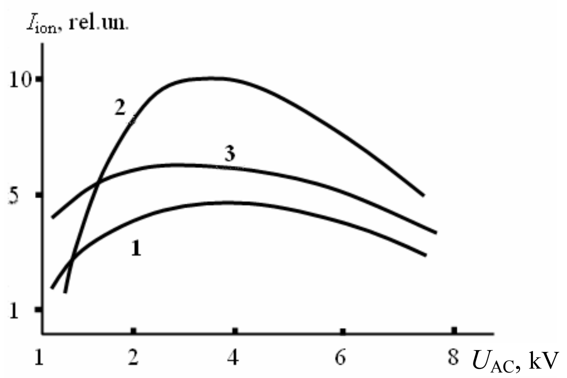

Fig. 5. A change of the ion current on the cathode surface for other U_{AC} at the pressure of a working gas: 1, $10\cdot10^{-4}$ Pa; 2, $5\cdot10^{-4}$ Pa; 3, $2\cdot10^{-4}$ Pa

The fundamental difference of the developed source construction, which is supposed to apply on the installation ILU-03, consists in the possibility to carry out the processing of long-length cylindrical details (fuel claddings).

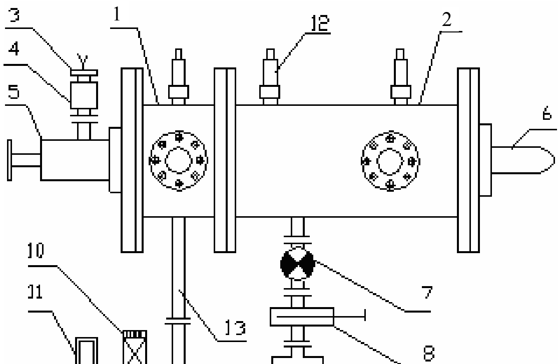

Fig. 6. The construction of a vacuum chamber of the installation ILU-03: 1, the chamber 1; 2, the chamber 2; 3, the air inlet valve; 4, the filter; 5, the mechanical lead in; 6, the auxiliary chamber; 7, 8, mechanical closing valves; 9, the reservoir for filling gas into the system; 10, a sensor of low vacuum; 11, a sensor of high vacuum; 12, the electric lead-in; 13, the vacuum lead

The construction of the vacuum chamber of the installation ILU-03, which consists of two discharge chambers, is presented in Fig. 6. The first chamber is necessary for purification of the cylindrical surface of tubes in a glow-discharge plasma (Ar^+). The second chamber is necessary to deposit films on a cylindrical surface and its subsequent processing at a high-voltage discharge that generates a radial ion flow with a wide energy spectrum. The filling of these chambers by a working gas occurs through the central working chamber 2 by mechanical closing valves (7, 8). The pumping-out of vacuum volumes is realized by a vacuum hose (13) connected with an H-0.5 type vacuum pump (supplied by a liquid nitrogen trap).

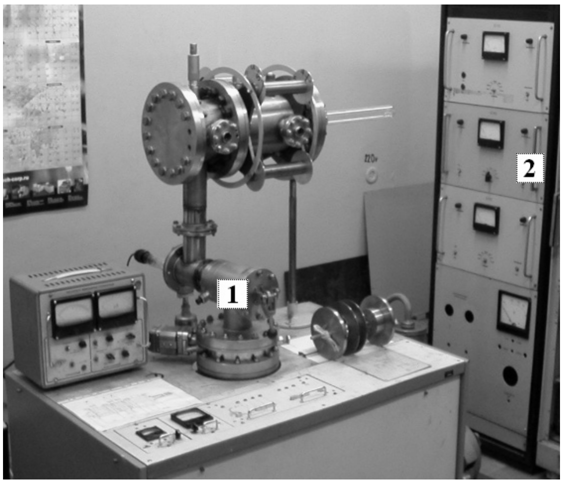

Fig. 7. The appearance of the installation ILU-03: 1, the vacuum block; 2, the high-voltage block of power supply; 3, the low-voltage power block (is not presented)

A photo of the installation ILU-03 is presented in Fig. 7. Its has the following main components: a vacuum block (1), a high-voltage block of power supply (2) for generation of a discharge in both the chambers, and a low-voltage power block (3) that provides the power supply from a power system and control of the installation's units.

The main parameters of the installation for complex alloying of cylindrical surfaces of long-length tubes are presented in Table 2.

Table 2. Optimal parameters for alloying the external surface of fuel claddings

Parameter	Value
The pressure of residual gases in the working chamber, torr, 10^{-3}	1
The discharge voltage, V	3.0–10
The discharge current, A	0.1–5
The temperature of a cylindrical sample, °C	50–150
The ion energy in beam, keV	0.3–8
The Ar^+ ion current, μA	10–500
The irradiation dose, cm^{-2}, 10^{18}	0.5–2
Maximum penetration depth, μm	≤1

Conclusion

Thus, the construction of a coaxial ion source has been designed on the basis of earlier realized experiments on complex alloying of cylindrical surfaces of zirconium alloys and obtained dependences on the discharge parameters at axial magnetic and radial heterogeneous electric fields. The developed source generates a radial flow of Ar^+ ions with a wide energy spectrum at the 0.3–8 keV energy range at the pressure of residual gases of 10^{-2}–10^{-4} Pa. The developed ion source is supposed to be used for multi-component alloying of the external surface of zirconium alloy fuel claddings to increase their corrosion resistance as applied to exploitation conditions at water-moderated power reactors.

References

[1] F.F. Komarov, *The ion implantation in metals*, Moscow, Metallurgy, 1990, p. 76.

[2] B.A. Kalin, N.V. Volkov, *in Proc. 15th Int.Conf. on Ion-surface interaction ISI-2001, 2001*, V. 1, pp. 124–127.

[3] B.A. Kalin, N.V. Volkov, Questions of Atomic Science and Technics, Ser. Nuclear Fusion, 2, pp. 72–76 (2001).

[4] A.M. Shalaev, *Radiation-enhanced diffusion in metals*, Moscow, Atomizdat, 1972, p. 148.

[5] B.M. Paine, R.S. Averback, Nucl. Instr. Meth., V. 7/8 B. pp. 667–675 (1981).

[6] B.A. Kalin, N.V. Volkov, V.K. Egorov, Physics and chemistry processing of materials, 1, pp. 13–13 (2000).

[7] B.A. Kalin, N.V. Volkov, I.V. Oleynikov, Physics and chemistry processing of materials, 3, pp. 25–29 (2004).

[8] B.A. Kalin, N.V. Volkov, Questions of Atomic Science and Technics, Ser.Nuclear Fusion, 1, pp. 61–65 (2000).

[9] A.S. Zaymovskiy, T.V. Nikulina, N.G. Reshetniko, *Zirconium alloys in nuclear energy*, Moscow, Energoatomizdat, 1994. p. 256.

[10] I.D. Abroyan, A.N. Andronov, A.I. Titov, *The physical fundamentals of electron and ion technology*, Moscow, High School, 1984, p. 320.

[11] M.D. Gabovich, N.V. Pleshivtsev, N.N. *Semashko, The beams of ions and atoms for controlled thermonuclear fusion and technological objects*, Moscow, Energoatomizdat, 1986, p. 248.

[12] A. Gras-Marti, P. Sigmund, Phys. Stat. Sol (a), V. 30, p. 621–632 (1983)

[13] B.A. Kalin, N.V. Volkov, V.V. Osipov, I.V. Oleynikov, j. Physics and chemistry processing of materials, № 1, pp. 13–21 (2004).

[14] B.A. Kalin, N.V. Volkov, I.H. Atalikova, V.F. Ribalko, G.D. Tolstoluckaya, Proceedings of the Academy of Sciences, Ser. Physical, v. 62, 7, pp. 1477–1482 (1998).

[15] P. Sigmund, A. Gras-Marti, j. Nucl. Instr. Meth., v. 168, p. 389–394 (1980).

[16] B.A. Kalin, N.V. Volkov, I.V. Oleynikov, Surface, 8, pp. 48–52 (2004).

[17] N.V. Volkov, B.A. Kalin, *Science of materials questions of nuclear engineering*, Moscow, Energoatomizdat, 1991, pp. 64–72.