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# INFLUENCE OF PULSED ION BEAM ON STRUCTURE AND PROPERTIES OF ZIRCONIUM ALLOY

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**Abstract**

*The impact of pulsed ion beam (PIB) with parameters the pulse duration 80 ns, energy of 200 keV and current density 120 A/cm$^2$ , pulse energy ~ 1,5 J/cm$^2$on the alloy Zr1%Nb has been studied. It was established that pulsed ion beam modification leads to zirconium alloy hardening for the depth of ~ 2 mc, decreases the probability of hydride phase forming and also decreases hydrogen penetration into alloy volume in up to 6 times.*

**Key words:** *zirconium alloy, pulsed ion beam, hydrogen*

## 1. INTRODUCTION

Beam methods of materials modification are being developed and implemented in industry in many countries [1]. These methods of materials surface layers modification increase durability, corrosion resistance, dynamic strength of products, realize surface polish and reduce the friction ratio, etc. The interest to these modifications is conditioned by the fact that their application in industry instead of traditional methods (heat treatment in ovens, acid and saline solutions hardening, galvanic coating) economizes electric power, increases productive efficiency, reduces or eliminates ecologically hazardous effects of production. In a number of cases plasma-beam technology allows to get such structure phase states of materials which cannot be realized with traditional methods application. With powerful pulsed ion beam impact on the material, the surface layer heats rapidly up to the temperature of phase transitions, for example, up to the melting temperature, then it cools rapidly due to heat extraction in material depth by the means of thermal conductivity. As a result of these processes the surface layer properties change, surface modification occurs. The following modifications in material microstructure are observed: grain size reduces from hundreds of microns to submicrons, transition to amorphous state is possible; phase composition changes, here metastable phases and compounds are possible to occur which cannot be formed with traditional material thermal treatment methods; phase state is homogenized, e.g., carbides are disintegrated and uniformly distributed in steels. Products made from zirconium are used in water-cooled power reactor cores. However during exploitation process under irradiation radiolysis of water occurs, atomic hydrogen which influences zirconium negatively is being released. Also there is a problem with hydrides formation. To exclude hydrogen penetration into materials doesn't seem possible due to large concentration of hydrogen in atmosphere or water environment, also due to technical conditions of materials exploitation. One of the ways to solve the problem of hydrogen penetration into metals is development of protective or barrier coatings on material surface [2,3]. Today there is vast experiential data array on research of metal and alloys structure and properties excited by pulsed ion beam impact on solid state [4]. However at present hydrogen influence on these materials is not sufficiently studied. Yet, pulsed ion modification of surface can provide significant influence on processes occurrence which are responsible for hydrogen

interaction with metals and alloys. In this work evaluation of possibility to use pulsed ion beams to form protective anticorrosion coatings has been performed. With this purpose series of research have been made to investigate the impact of ion irradiation on physical-mechanical properties of the alloy, hydrogen accumulation in Zr1%Nb has been studied.

## 2. EXPERIMENTAL TECHNIQUE

Formation of powerful ion beam was based on the ions acceleration on preformed blast emission plasma. In this case it is possible to get ions of practically all metals. The accelerator was designed on the Laboratory prototype of technological pulsed accelerator. Gas-filled diode type is magnetic isolated gas-filled diode of strip type with external magnetic field (Figure 1) [5].Nanosecond generator allows to form single voltage pulse and two heteropolar voltage pulses. Magnetic field in anode-cathode path of magnetic isolated diode is being formed due to current flow on cathode turn from complementing source. The design of cathode turn provides current flow close circuit throughout the whole cross-section of anode-cathode path.

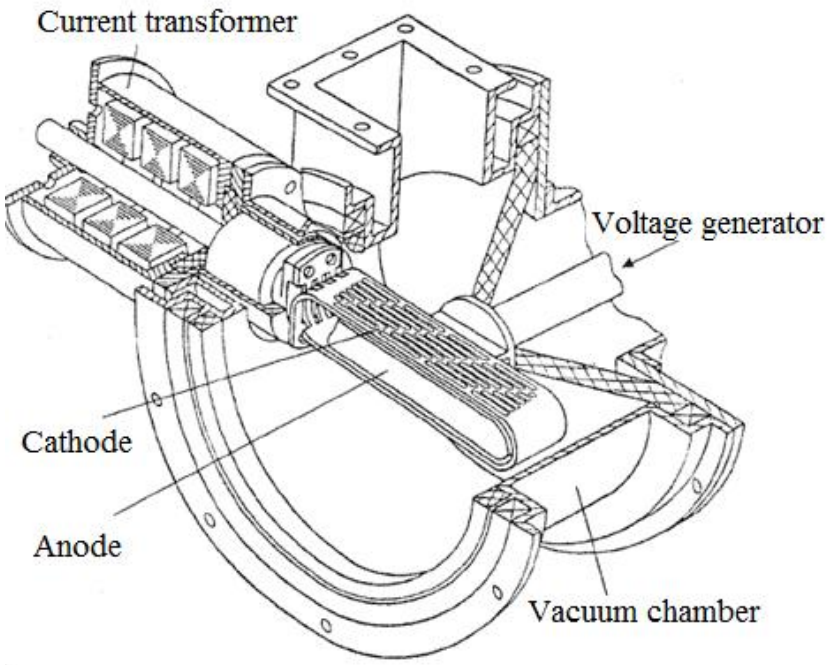



Figure 1 Magnetic isolated diode with closed accelerator drift

Figure 2 reveals the pulsed accelerator scheme. There NsG – nanosecond generator, GL – generating line, DL – delay line. TL – transmitting line, P1, P2 and P3 – gas dischargers, C1 – capacity of nanosecond generator primary storage, Tp1 – step-up pulse transformer, U1 – ignitron, SR – saturable reactor, C2 – primary storage of forming magnetic field system, Tp2 – step-up pulse transformer, MIVD – magnetic isolated vacuum diode, TU – timing unit

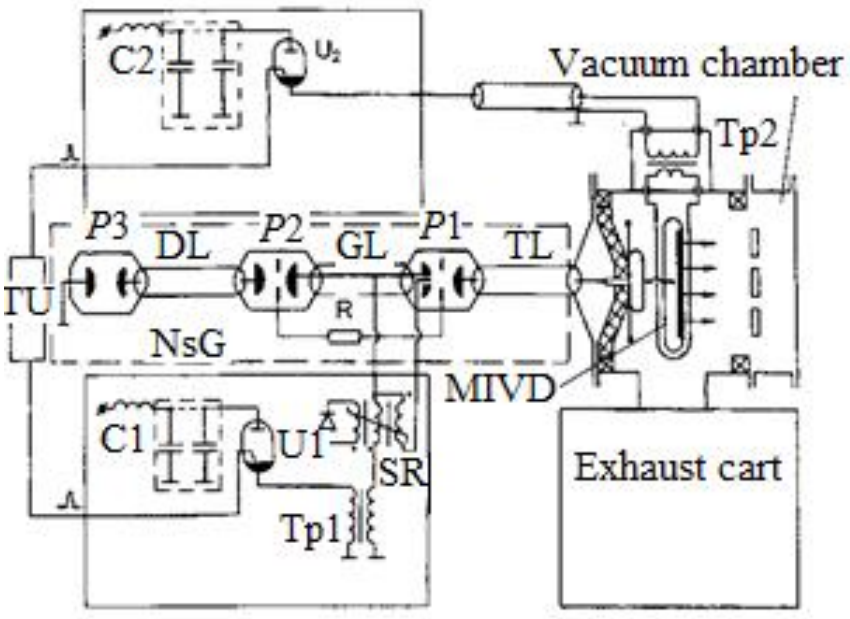



Figure 2 Scheme of pulsed accelerator and main nodes

Rectangular samples of Zr1%Nb alloy with the sizes of 5×10mm, 30×40mm and 40×60mm in the as-received condition were chosen for studies. Surface layer modification was performed with the help of pulse carbon ion beam accelerator with pulse duration 80 ns, energy 200 keV and current density 120 A/cm$^2$. Pulse energy constituted ~ 1,5 J/cm$^2$. Alloy modification was performed in two modes differentiated by number of pulses: initial mode - 1 pulse, second mode – 4 pulses. Surface structure was studied with the help of optical microscope «METAM LV» with magnitude 200, microhardness was studied on the microhardness meter «LOMO PMT-3M» with indenter capacity P = 0.1, 0.2 и 0.3 H, electron density was studied applying the thermoelectromotive method. Hydrogen saturation was performed from gaseous phase for one hour under the temperature 450°C and pressure 2 atm implementing Siverts method. Studies of hydrogen accumulation were performed on hydrogen analyzer produced by LECO (Figure 3).

## 3. THEORETICAL AND EXPERIENTIAL RESULTS AND THEIR DISCUSSION

Calculations of thermal fields were performed with pulse ion beam approximation with initial energy of particles in 200 keV. Main factors determining structure phase state and properties of near-surface layers of material under pulse heating are nonstationary thermal fields and thermomechanical stresses that occur as a result of transmitting energy of pulsed ion beam to the target. Thermal field is localized in the heat-affected zone, the thickness of which in the case of pulse ion beam does not exceed some mc. To solve thermal problem heat loss was taken into consideration for possible phase transitions (melting and evaporation). In this case it is quite proper to apply one-dimensional approximation to determine main patterns of thermal processes. According to the results of computational modeling with peak current of current density 120 A/cm$^2$ and specified pulse duration 80 ns distribution according to temperature depth is represented in Figure 4. The temperature on zirconium alloy surface reaches the value that increases its melting temperature (T = 2130 K). Herewith, the depth of molten layer does not exceed 3.5 μm. The surface layer of material possesses the highest heating speed.

Surface layer modification (change of its physical properties and also its geometry) under the pulsed ion beam influence with the named above parameters is conditioned by liquid phase formation and its consequent hardening after the pulse impact. Alloy cooling starts by the end of pulse impact and with the beginning of beam current density decrease.




Figure 3 Hydrogen analyzer produced by LECO

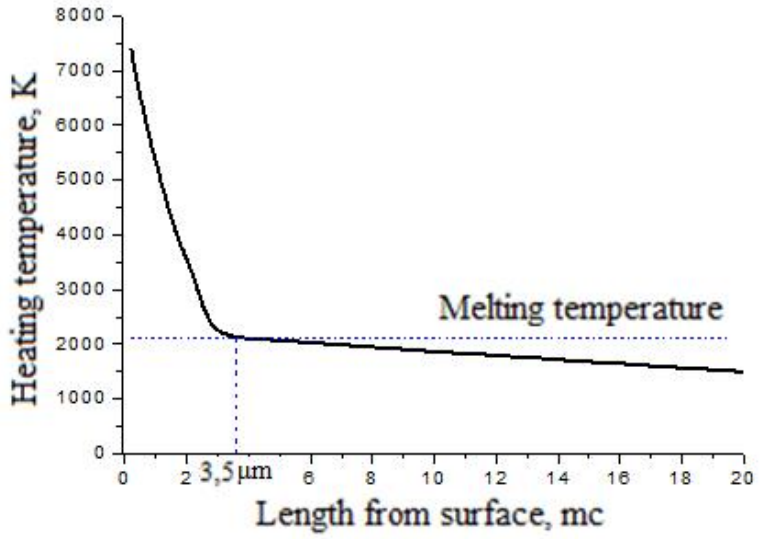



Figure 4 Dependence of melt alloy Zr1%Nb depth from heating temperature

Morphology research revealed that metals` treatment with surface flash forms sufficiently complex topological surface pattern (Figure 5). It is connected with microcraters formation on the target surface. Besides, it is necessary to point out scalloping surface structure presence after irradiation. The

geometry of this kind occurs as a result of metal fine surface layer melting. Microcraters are being formed as a result of gases yield from near-surface field of metal.

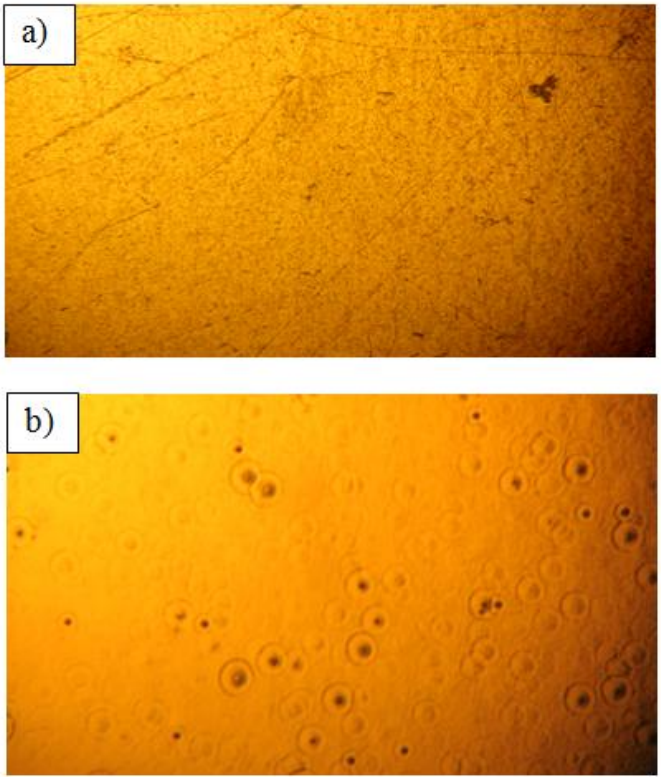



Figure 5 Photographs of Zr1%Nb alloy before (a) and after (b) irradiation by pulsed ion beam

Studies of physical-mechanical properties were performed applying the method of microhardness measurement and the thermoelectromotive method. Experiential results of microhardness measurements are given in Table 1. As it appeared from the presented results microhardness under stress of 0.2 and 0.3 H after pulsed ion beam modification practically didn`t change. Under stress 0.1 H microhardness in the modified sample is higher than in the virgin sample in almost one and a half time. Stress 0,1 H corresponds to the thickness of layer 2 μm, 0,2 H – 2,2 μm, 0,3 H – 2,4 μm. That is why surface layer hardening occurs up to the depth not less than 2 μm. This relates quite perfectly with the theoretical calculations of thermal fields.

Table 1

| | Stress 0,1 H | Stress 0,2 H | Stress 0,3 H |
|---|---|---|---|
| | Depth ~ 2 μm | Depth ~ 2,2 μm | Depth ~2,4 μm |
| Modified pulsed ion beam (mode 1) | 1030 MPa | 1250 MPa | 1570 MPa |
| Modified pulsed ion beam (mode 2) | 970 MPa | 1280 MPa | 1500 MPa |

| | | | |
|---|---|---|---|
| Initial | 770 MPa | 1260 MPa | 1600 MPa |

Material hardening under pulsed beam impact, apparently, is conditioned by structural changes of the surface layer. They are characterized by recrystallization layer formation which prevents advancement of existing microcracks and motion of dislocations. Due to dissolved and adsorbed carbon and mixing of surface layer components carbides are being formed. All these factors enable increase of microhardness.

Results of thermoelectromotive value studies in dependence with coordinate of sample are represented in Figure 6.

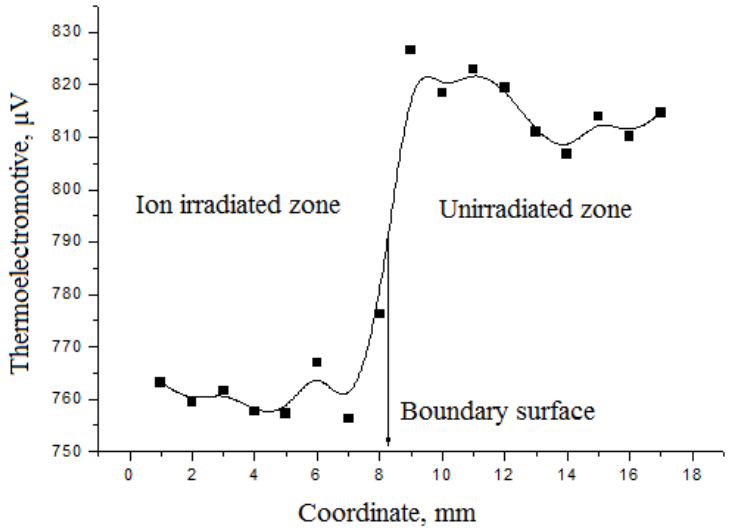



Figure 6 Dependence of coordinate of Zr Э110 sample before and after irradiation by the thermoelectromotive method

From the dependence presented it is possible to observe that electron density decreased in irradiated zone which is connected with the forming of other internal elastic stresses, the latter are generated by high cooling speed (hardening) of fine surface layer. There is possibility of coherent-scattering regions` forming in big amounts.

The X-ray structural analysis was performed in order to identify hydride phase formation in hydrogen saturation process of bearing and modified alloy. The obtained results signify that after one pulsed ion beam impact hydride formation is reduced in two times in comparison with the initial state and after four pulsed ion beam impacts hydride phase practically is not formed. Results of hydrogen accumulation studies are represented in Table 2.

Table 2

| | Hydrogen concentration mass.% |
|---|---|
| Zr1%Nb + PIB + $H_2$ Mode 1 | 0,0159 |
| Zr1%Nb + PIB + $H_2$ Mode 2 | 0,0102 |
| Zr1%Nb Bearing + $H_2$ | 0,0648 |

After comparing the obtained results it follows that pulsed ion beam impact in Mode 1 (1 pulse) reduces hydrogen penetration into the volume in more than 4 times in comparison with the bearing alloy and modification by pulsed ion beam in Mode 2 (4 pulses) reduces concentration in 6 times. This signifies that formed layer by modification of Zr1%Nb alloy pulsed ion beam can serve as effective protective coating against hydrogen penetration into alloy volume.

## 4. CONCLUSIONS

The impact of pulsed ion beam (PIB) leads to surface layer modification of Zr1%Nb alloy. The depth of modified layer with pulse duration 80 nc, energy 200 kiloelectronvolt and current density 120 A/cm$^2$, pulse energy of ~ 1,5 J/cm$^2$ constitutes about 2 μm, which corresponds to theoretical calculations of thermal fields. Modified by PIB layer possesses increased mircohardness, lower electron density and can serve as effective protective coating against hydrogen penetration into alloy volume. Impact of PIB modification in one pulse reduces hydrides formation in two times as well as it reduces hydrogen penetration into the volume in four times in comparison with the initial and with the impact of four pulses hydride phase practically is not forming and hydrogen concentration is reduced in six times.

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