UDC 539.21; 539.12.04

[1*]Mukashev K.M., [1]Muradov A.D., [2]Umarov F.F., [3]Michalenkov V.S.

[1]Faculty of Physics and Technology, al-Farabi Kazakh National University, Almaty, Kazakhstan
[2*]Kazakh-British Technical University, Almaty, Kazakhstan
[3*]Institute of Metal Physics Academy of Sciences, Kiev, Ukraine
[*]e-mail: kanat-kms@mail.ru

# Possibility of separation of elastic and inelastic channels radiation damage metals

**Abstract.** In this work we have performed a systematical investigation of energy dependence of the radiation defects distribution profile in three different materials – tantalum, molybdenum and stainless steel of type 10X18H10T–VD irradiated by high energy protons. It has been shown that in the stainless steel and tantalum, regardless of proton energy, the vacancy complexes similar by configuration appear which are described by the slightly expressed elastic channel. The defects recover in one annealing stage with different migration activation energy. At the same time the molybdenum radiation damageability consists of two components in each of which exists its own mechanism of defects formation. For high energy protons what's important is the inelastic channel of interaction and formation of sub cascades, which are created by primarily knocked-on atoms of considerable energies. However, for low energy protons, the processes of elastic interaction with lattice atoms and emergence of atomic hydrogen in the end of run important.
**Key words**: positron annihilation, tantalum, molybdenum, proton irradiation, point defects, vacancy complex.

## Introduction

It is known that the effects of nuclear radiation on a matter is accompanied by a number of new phenomena [1-4]. The most important among them are nuclear reactions and a change associated with them in the elemental composition, formation and the emergence of clusters of point defects, damage to the matrix caused by cascade atom – atom collisions, etc., and as a result, disturbance in the integrity of the crystal. In order to learn the phenomena related to the changes in the material's crystal structure in the active zone of the reactor, it often suffices to conduct simulation experiments at the accelerators of charged particles. At the same time a quite important task is to study the profile of the defect distribution along the depth of the damaged layer. In the past, a computer program was developed for this purpose on the basis of theoretical research in order to calculate the profile of the displaced atoms by the depth of passing of heavy ions in the material [5]. But any computer program, regardless of how universal it is, yet cannot take into account all aspects of the complex process of interaction of charged particles with the real crystal lattice, furthermore, it cannot be acceptable in situations when the object of study is multicomponent alloys.

Charged particles lose energy when moving in the matter. Energy losses of the incident particle can occur by various ways, including ionization and excitation of the electron shells, the atoms' polarization of the medium, the radiation losses and nuclear stopping, whose role in the formation of structural defects may be different. Consequently, the profile of the defect distribution in the depth may also be different, the location of which depends on the type and parameters of the bombarding particles of the target material, and the irradiation temperature. In this regard, experimentally obtained parameters of the defect structure may differ considerably from those calculated theoretically.

The average path traveled by a charged particle in matter to a full stop is called its range $R$. The latter depends on the particle energy and the properties of the target's material. The range of the particle is usually expressed through the length of the path, $d$ and density of the material, $\rho$:

$$R = d \cdot \rho \ [g \cdot cm^{-2}] \qquad (1)$$

 

To estimate the interaction of particles with the matter in the reference books we can find the following: particles energy – $E$ in the lab coordinates and expressed in MeV's; particle ranges $R(E)$, expressed in $g \times cm^{-2}$; stopping power $S(E)$in $MeV \times cm^{-2} \times g^{-2}$; derivatives of stopping power by energy $D(E)$, used as a correction factor [6,7]. In order to calculate the range of a particle whose energy lies between the tabular data, one has to use the formula:

$$R(E)\Delta E) = R(E) + \frac{\Delta E}{S(E)} - \frac{1}{2}\left|\frac{\Delta E}{S(E)}\right|^2 \cdot D(E), \quad (2)$$

where $E$ is the nearest table value of energy. On the contrary, for the calculation of energy corresponding to range, the value of which lies between the values in the table, you can use the formula:

$$E(R+\Delta R) = E(R) + S(E)\cdot\Delta R - \\ -\frac{1}{2}S^3(E)\cdot D(E)\cdot(\Delta R)^2 \quad (3)$$

To find intermediate values of $S(E)$, which are absent in the table, a linear interpolation relationship is used:

$$S(E+\Delta E) = S(E) + D(E)\cdot\Delta E, \quad (4)$$

where $S(E)$ is the closest value of the stopping power. Similarly, an intermediate value $D(E+\Delta E)$, which is not listed in the table, can be found by the relation:

$$D(E+\Delta E) = D(E) + \frac{\delta D}{\delta E}\cdot\Delta E, \quad (5)$$

Under these conditions, the accuracy of calculation of the tabulated data is ~ 1%.

**Experimental**

Because the passage of charged particles in the matter is accompanied by its successive energy losses, the study of the defect distribution profile by depth is, in principle, a task of studying the energy dependence of metal radiation damageability. One way to solve this problem is to study metal defect structure by sequential etching of the surface. From the other hand, you can solve it on the basis of variable thickness of the absorber method. Obviously the most acceptable way to solve it is the latter, non-destructive method of research, the essence of which is to irradiate by the charged particles of high energy and studying the stack of foils, the total thickness of which is greater than the mean free path of the particles in the material. By using such a technique, each foil is irradiated by particles of different energy and contains the respective structural damages typical for a given depth of the material. As the test objects we used polycrystalline Mo and Ta, and stainless steel-VD 10H18N10T as a foil with a thickness of 100 μm and a diameter of 17 mm each. The initial state of the metals achieved by annealing at T = 1200°C and for steel the temperature was at 1050°C for 1.5 hours in a vacuum of $10^{-5}$ Pa. The thickness of each foil $\Delta d$ defines a path element $\Delta x_i = \Delta d_i \rho$, on which there happens a loss of energy of protons $\Delta E_i = s_i(E)\Delta x_i$; the mean energy of the protons on the other side of each foil will be $E_{xi} = E_i - \Delta E_i$. Consequently, each successive foil is irradiated with protons of different energies. If one studies the degree of damage, its energy dependence can be established. Irradiation was carried out by protons flow $1.2 \times 10^{13}\ cm^{-2}c^{-1}$ up to $2 \times 10^{17} cm^{-2}$fluence.

The main research tool in this case is the electron-positron annihilation (EPA) method. Being one of the major nuclear-physical methods of studying the state of the condensed medium, the method of electron-positron annihilation is a very sensitive tool to various kinds of damages of crystal structure. Spectrum shape of the angular distribution of annihilation photons (ADAP), caused by annihilation of positrons with electrons of the material, changes significantly at localization of positrons near the defects in the crystal lattice, as well as from the atomic environment of defective regions. Slow positrons also react to the change of the structure's order. Therefore positron probe is an ideal tool for studying electronic states of the metallic materials local microregions [8-10].

The study of structural defects of materials was performed by the spectrometer with a linear-slit geometry with an angular resolution of 0.5 mrad. As a positrons source we used the isotope $^{22}$Na with 10 mCi activity. The measurement of ADAP spectrum allows to determine the relative contribution to the annihilation of positrons process with the conduction electrons and ion core electrons. To this effect, we experimentally measures the intensity of the annihilation gamma rays as the dependence of

counting rate of pulses that are coincident in time of 2 photons, which are detected by opposing detectors on the displacement angle of the movable detector $\theta$. ADAP spectra measured for different states of the material, are normalized to a single space. It is not difficult to establish that the spectrum for the defective material has a higher intensity at the maximum and the narrow width at half maximum (Fig. 1).

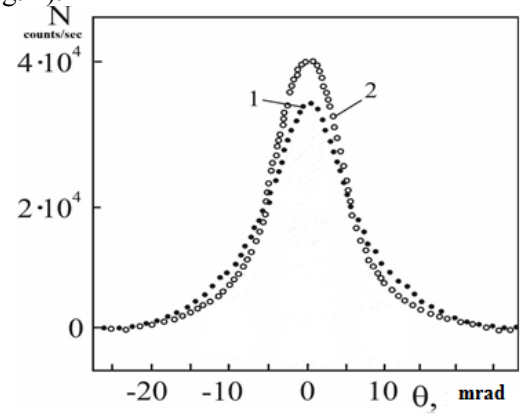


**Figure 1** − Experimental ADAP spectra: 1 - for annealed materials; 2- for irradiated materials

## Results and discussion

To interpret the results of studies used the following structure-sensitive annihilation parameters: $F$ – positron annihilation probability redistribution between the conduction electrons and bound electrons, as well as its corresponding increment $\Delta F$ relative to values for the initial state, ~~th~~at are recovered by processing the spectrum of the angular correlation of annihilation radiation [8]. The results of studying stainless steel according to these conditions by the annihilation parameters change data are summarized in Table 1.

High energy protons irradiation change 3 considerably the spectrum's shape of annihilation photons angular distribution. The spectrum becomes narrower in half-width and maximal intensity raises caused by the redistribution of positrons annihilation probability with conduction and ion core electrons. These factors are visible in annihilation parameters change. If for annealed sample the value $F = 0.15$, then after irradiation it increases almost as twice as much. The spectrum half-width (FWHM) for the initial state is equal to 6.1 mrad. The protons irradiation reduces it in average down to 5.6 mrad. In spite of the considerable changes in the parameters, the evident regularity between them and protons' energy in this case is not visible. Though, the certain tendency in the annihilation parameters behavior yet can be established. Thus, for example, the average value of the positrons annihilation relative probability $F$ with particles energy increase steadily decreases. This can testify that the main contribution in the steel radiation damageability is carried in the low energy protons, which experience the elastic collisions with steel components atoms.

**Table 1** − Parameters of steel annihilation by the depth of protons passage

$E_{initial} = 30\ MeV$ (Q= $2 \times 10^{17} cm^{-2}$ – protons fluence; Ep – current value of proton energy )

| Sample number | $E_P$, MeV | $X$, μm | $F=S_P/S_g$ | $f=N(0)/N(8)$ | FWHM, mrad. |
|---|---|---|---|---|---|
| annealed | - | - | 0.15 | 3.1 | 6.1 |
| 19 | 0 | - | 0.27 | 3.9 | 5.6 |
| 18 | 2.33 | 1750 | 0.24 | 3.9 | 5.6 |
| 17 | 6.40 | 1650 | 0.24 | 3.8 | 5.7 |
| 16 | 9.09 | 1550 | 0.28 | 3.4 | 6.0 |
| 15 | 11.29 | 1450 | 0.25 | 3.7 | 5.7 |
| 14 | 13.19 | 1350 | 0.31 | 3.6 | 5.8 |
| 13 | 14.91 | 1250 | 0.26 | 3.4 | 5.9 |
| 12 | 16.49 | 1150 | 0.23 | 3.2 | 5.9 |
| 11 | 17.97 | 1050 | 0.27 | 3.5 | 5.8 |
| 10 | 19.35 | 950 | 0.28 | 3.8 | 5.7 |
| 9 | 20.67 | 850 | 0.27 | 3.5 | 5.7 |
| 8 | 21.92 | 750 | 0.29 | 3.6 | 5.7 |
| 7 | 23.12 | 650 | 0.27 | 3.5 | 5.9 |
| 6 | 24.27 | 550 | 0.21 | 3.4 | 5.9 |
| 5 | 25.39 | 450 | 0.25 | 3.6 | 5.8 |
| 4 | 26.47 | 350 | 0.23 | 3.3 | 5.9 |
| 3 | 27.51 | 250 | 0.25 | 3.4 | 5.9 |
| 2 | 28.52 | 150 | 0.24 | 3.7 | 5.7 |
| 1 | 29.50 | 50 | 0.26 | 3.7 | 5.7 |
| error ± | 0.05 | 1.00 | 0.02 | 0.1 | 0.1 |

Therefore we can assume that the structural damages arising meanwhile in the steel samples are not much different between each other both by configuration and by positrons capture efficiency and correspond to the traps of one type. The latter is confirmed by the shape of isochronal annealing curves from the stack of the samples irradiated by different energy protons (Fig. 2). At EP = $29.5 \div 13.2\ MeV$ protons energy the materials basic properties recovery ends in the temperature range 350-600°C and at EP $\leq 6\ MeV$ – in the range of 250 – 550°C, that is existence of a certain relationship

between protons energy and defects annealing temperature is evident.

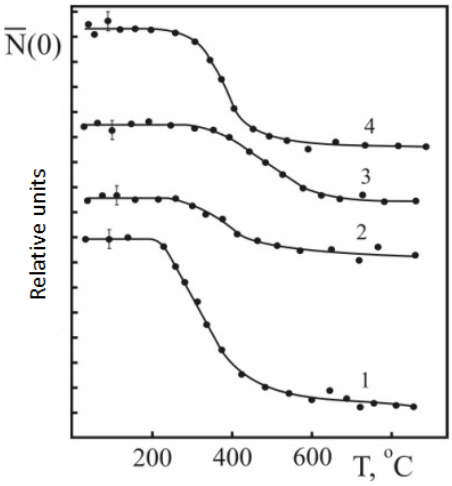


**Figure 2** − The energy dependence by protons irradiated 10X18H10T – ВД steel annealing kinetics
1. $E_P$ = 4.6 MeV; 2. $E_P$ = 31.2 MeV;
3. $E_P$ = 23.1 MeV; 4. $E_P$ = 29.5 MeV

Apparently of the defects migration activation energy value, irradiation by high energy protons causes the dislocation loops with $E_a = 2.1 - 2.2\ eV$. It is evident that in the low energy protons case the vacancy complexes as a small subcascades or connected vacancy-impurity complexes with $E_a = 1.7 - 1.8\ eV$ are created. The most likely is the formation of connected vacancy-Cr atom state whose decomposition at annealing is trickier than that of others [9].

Unlike stainless steel, the polycrystalline tantalum irradiation at the same conditions leads to some characteristic changes in the annihilation parameters. It is very difficult to define a spectrum parabolic component for this metal in consequence of insignificant portion of free charge carriers. Therefore, the basic equivalent (instead of F) parameter is the ratio between the counting rate in the maximum of spectrum $N(0)$ to its value at the angle $\theta = 8$ mrad, that is $f = N(0)/N(8)$. The dependence of this parameter on protons energy is given in Fig.3a. The maximal spectrum narrowing, as well as considerable increase of the parameter $f = N(0)/N(8)$ is observed at low energies $\sim 5 - 8\ MeV$. With particles energy increase these parameters possess a steadily increasing or decreasing pattern.

Such changes in parameters, which characterize the spectrum shape, are possible only at corresponding changes of capture efficiency of positrons by defects, created by protons. As far as the effectiveness maximum corresponds to low energy protons, then we can suppose that the largest damageability tantalum is suffered consequently of elastic interactions. The nuclear reactions influence activated by inelastic interactions in the high energy region in this case is negligible. The actual picture of the structure damages in tantalum ~~is~~ can be determined as a result of isochronal annealing of individual samples from the stack irradiated by different energy protons (Fig. 3b).

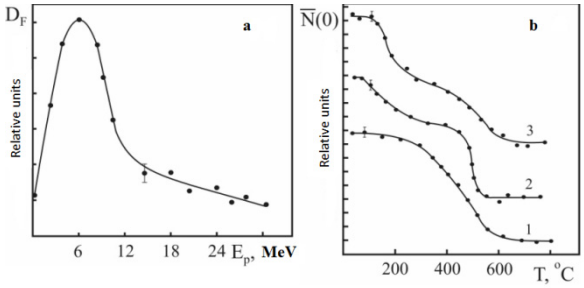


**Figure 3** − The energy dependence of damageability (a) and annealing kinetics (b) of Ta, irradiated by protons with 30 MeV initial energy:
1. $E_P$ = 6 MeV; 2. $E_P$ = 25 MeV; 3. $E_P$ = 30 MeV

One can see that the sample irradiated by low energy protons recovers in one stage in the temperature range of 250 – 600°C with curve amplitude ~15 % (curve 1), whereas at high energy irradiation the structure damages in Ta emerge with two recovery stages (curves 2,3). And with particles energy decreasing from 30MeV to 25MeV the beginning of the point defects migration is shifted sideways towards lower temperatures with simultaneous increase in portion of vacancy defects. This confirms the decisive contribution of elastic interactions in the process of defect formation. So, at protons irradiation with $E = 30\ MeV$ the vacancy stage portion makes up 41% from the damages common level. However, with energy decreasing to up to 25MeV it increases to up to 47%. Meanwhile the recovery second stage gets more relief appearance than in the first case. The migration activation energy's mean value for vacancy components is equal to $E_{a1} = 1.41 - 1.45\ eV$, and for dislocation and more complicated components, which are annealed during the second stage – $E_{a2} = 2.33 - 2.35\ eV$.

Still large interest in these experiments is presented by the study results of damageability distribution profiles of molybdenum, irradiated at the same conditions. For molybdenum in the

annealed state, the annihilate photons angular distribution is approximated by one Gaussian, as far as the probing positrons are annihilated basically with ion core electrons. As a result, it becomes difficult to identify a parabolic component in the spectrum. But as a result of protons irradiation, the emerging structure damages enable the parabolic component responsible for positrons annihilation with free charge carriers to appear in the spectrum. Meanwhile the energy dependence of the relative annihilation probability $F = S_P/S_g$have a more complicated and non monotonic nature (Fig.4). After certain energetic region 5 - 10 MeV in such the relative probability $F$ is stay constant, in the sequel it is sharply decreased and achieve the minimum at about 18 - 20 MeV is increasing again up to $E_P$= 30 MeV. Such annihilation parameter dependence is can be activated by the appropriate defects distribution profile which is determine the defects both concentration and configuration along the trajectory of charged particles in matter.

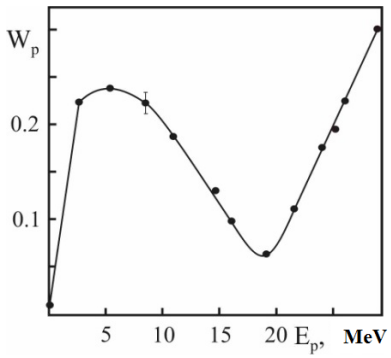


**Figure 4** − The energy dependence of the Mo damageability irradiated by protons, with initial energy 30 MeV

The probable reason of such dependence can be difference in contribution in the process of radiation defects generation of the elastic interactions at $E_P < 18\,MeV$ energies and subcascade regions formation at$E_P > 20\,MeV$ on the one hand, and also the nuclear reactions specified role on the other hand. But in both cases the defects structure must be of vacancy, which is revealed by isochronal annealing results. Regardless of energy, as a result of the proton irradiation there emerge the vacancy complexes in Mo and in the annealing temperature range below 800°C only one incomplete return stage has been observed. Meanwhile, as the proton energy increases the defects migration temperature threshold ~~is~~ gradually shifts towards higher temperatures ranges – from 300 – 350°C to up to 450 – 500°C, which testifies emergence of more stable in terms of temperature defects in the metal. In [12], it is stated that the full annealing of Mo irradiated by protons is completed at approximate temperatures of 300 – 320°C. But this temperature is considerably lower than Mo recrystallization temperature, and this gives rise to a doubt in the above mentioned conclusion in [13]. However, annealing temperature that was reached in this investigation (850°C) is also not sufficient for a completion of even the first stage of recovery (III-stage), whereas according to [13] the recrystallization temperature of Mo is higher than 1000°C.

## Conclusion

Thus, investigation of the defects distribution profiles by charged particles passing depth in three different materials has shown that in the stainless steel and Ta, regardless of protons energy, there appear configuration vacancy complexes similar by configuration, which are recoverable in one annealing stage with migration activation energy $E_a = 1.7 - 1.8\,eV$. The total damageability of stainless steel and Ta in the whole range of protons energy is formed basically by one low-grade elastic channel, whereas for molybdenum it is created from two components in each of which its defined mechanism of defects formation is in place. If for high energy protons, the inelastic channel of interactions and subcascades appearing are the basic, then for low energy protons the elastic interactions with lattice atoms and vacancy complexes and atomic hydrogen formation in the end of run are of high importance. The probable cause of such difference in radiation damageability of Mo and Ta polycrystals is the appreciable difference in atomic masses of these elements. Almost two timesexcess of Ta atomic mass over atomic mass of Mo is evidently the reason of implicitly expressed damageability at the expense of inelastic channel, situated below the response limit of the testing method. The basic mechanism of the radiation defects formation in stainless steel and Ta are evidently expressed elastic interactions. The role of nuclear reactions in the structure damages creation is weakly expressed in this case. The defects created by low energy protons are annealed in one stage, whereas high energy particles influence causes emergence of the radiation defects, which are of vacancy and dislocation nature and recover in two stages.

**Acknowledgements**

The authors would like to thank Dr. Umarov T.F. for his very useful help.

**References**

1. Agarvala R.P. Radiation Damage in Some Refractory Metals // Materials Science Foundations. − 2005. − Vol. 25. − P. 125-130.

2. Zinkle Steven L., Wiffen F.W. Radiation Effects in Refractory Alloys // Space Technology and ApplicationsInternational Conference Proc.− 2004. − Vol. 699. − P. 733-740.

3. Leonard K.J. 4.06-Radiation Effects in Refractory Metals and Alloys // Comprehensive Nuclear Materials. − 2012. − Vol. 4. − P. 181-213.

4. Grossbeck M.L. 1.04-Effect of Radiation on Strength and Ductility of Metals and Alloys // Comprehensive Nuclear Materials. − 2012. − Vol. 1. − P. 99-122.

5. Antonov A.G. The investigation of tritium entry in environment at liquid leady-lithium irradiation eutectic through channel wall from stainless steel // The liquid metal application in the national economy. – Obninsk. – 1993. − P. 181-183.

6. Myllyia R., Xostomaova J., A time-to amplitude converter with constant-fraction timing discriminators for short time interval measurements // Nucl. Jnstr. and Methods. – 1985. − P. 568-578.

7. Ibragimov Sh. Sh., Reutov V.F., Abdurashitov I. Yu. Radiation damageability of Mo at high temperature irradiation by protons with initial energy 30 MeV // VANT, ser. FRP and RM. − 1977. − Vol. 1. – No 21. − P. 76-79.

8. Mukashev K.M. Low positrons Physics and positron spectroscopy. – Almaty. – 2007. − P. 508.

9. Mukashev K.M., Tronin B.A. and Umarov F.F. Behavior of Structure Defects and Hydrogen in neutron-irradiated stainless steels studied by positron-annihilation method // Rad. Effects & Defects in Solids. − 2009. − Vol. 164. – No 10. − P. 611-618.

10. Mukashev K.M. and Umarov F.F. Positron annihilation in Titanium Alloy modified by proton irradiation // Rad. Effects & Defects in Solids. − Vol. 167. – No 1. − P. 1-11.

11. Berestetsky V.B., Livshits E.M., Litaevsky A.P. The relativistic quantum theory. − M. Nauka, 1968. − Vol. 1. − P. 480.

12. Dextyar I.Ya., Mukashev K.M., Rustamov Sh.A., at all. The positron annihilation in irradiated by chromium- ruthenium alloys // Ukr. Fis.Jurnal.− 1984. − Vol. 29. – No 11. − P. 1679-1681.

13. Belyaev V.N., Zemlerub P.A., Kovalev V.Yu. at all. The positrons lifetime spectrometer // Preprint ITEF – 95, − M., – 1980. − P. 26.