The energy dependence of refractory metals and alloys radiation damageability

K M Mukashev¹, F F Umarov²

¹Almaty,050100, Kazakhstan, National Pedagogical University after Abai, 13, Dostyk Avenue,
²Almaty,050000, Kazakhstan Kazakh-British Technical University, 59, Tole bi str.
e-mail: mukash_kanat@mail.ru

Abstract.

In this work the systematical investigation of the radiation defects distribution profile energy dependence in three different materials - tantalum, molybdenum and 10X18H10T–VD stainless steel irradiated by high energy protons has been performed. It has been shown that in stainless steel and tantalum, independently of proton energy, the vacancy complexes related by configuration appear which are described by the slightly expressed elastic channel. The defects are recovered in one annealing stage with different migration activation energy. At the same time the molybdenum radiation damageability is composed of two components in each of which its specified defects formation mechanism takes action. For high energy protons the inelastic channel of interaction is the basic and subcascades appearance is created by primary knocked-on atoms of considerable energies. For low energy protons the processes of elastic interaction with lattice atoms and atomic hydrogen in the end of run creation are major.

Introduction

As is well known, the effects of nuclear radiation on matter is accompanied by a number of new phenomena. The most important among them are nuclear reactions and change associated with these reactions 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, violation of the integrity of the crystal. For the study of phenomena related to changes in the crystal structure of the material in the active zone of reactor, it is often sufficient to conduct simulation experiments at accelerators of charged particles. It is very important task of studying the profile of the defect distribution along the depth of the damaged layer. At one time, for this purpose on the basis of theoretical research program has been developed for calculations on computer of the displaced atoms profile in depth passage of heavy ions in the material [1]. But any program, as it was neither universal, yet cannot take into account all aspects of the complex process of interaction of charged particles with the real crystal lattice, the more it cannot be acceptable when the object of study is the multicomponent alloys.

Charged particles during moving in matter lose energy. The energy loss of the incident particle can occur in 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 the theoretically calculated.
The average path traversed by a charged particle in matter to a full stop, called its range \( R \). The latter depends on the particle energy and material properties of the target. The range of the particle is usually expressed through the length of the path \( d \) and density \( \rho \) material:

\[
R = d \cdot \rho \left[ \text{g \cdot cm}^{-2} \right] \tag{1}
\]

To assess the interaction of particles with the matter in the directories are: energy particles - \( E \) in the lab coordinates, expressed in MeV; particle ranges \( R(E) \), expressed in \( \text{g \cdot cm}^{-2} \); stopping power \( S(E) \) in \( \text{MeV \cdot cm}^{-2 \cdot g}^{-1} \); derivatives of stopping power by energy \( D(E) \), used as a correction factor [2,3]. To calculate the range of a particle whose energy lies between the tabular data, use the formula:

\[
R(E)\Delta E = R(E) + \frac{\Delta E}{S(E)} \left( 1 + \frac{\Delta E}{2S(E)} \right) \cdot D(E), \tag{2}
\]

where \( E \) - 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:

\[
E(R + \Delta R) = E(R) + S(E) \cdot \Delta R - \frac{1}{2} S'(E) \cdot D(E) \cdot (\Delta R)^2 \tag{3}
\]

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

\[
S(E + \Delta E) = S(E) + D(E) \cdot \Delta E, \tag{4}
\]

where \( S(E) \) - the closest value of the stopping power. Similarly, absent in the table intermediate value \( D(E + \Delta E) \) can be found by the relation:

\[
D(E + \Delta E) = D(E) + \frac{\delta D}{\delta E} \cdot \Delta E, \tag{5}
\]

Under these conditions, the accuracy of calculation of the tabulated data is \( \sim 1\% \).

**Experimental**

Because as the passage of charged particles in matter, there is a consistent discharge its energy, the study of the profile of the defect distribution in depth, in principle, is the task of the study of the energy dependence of metal radiation damageability. To solve this problem, you can go the way of studying of metal defect structure by sequentially etching the surface, or on the basis of variable thickness of the absorber method. Naturally, the most acceptable is the second, 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 characteristic for given depth of the material. The test objects were used polycrystalline Mo and Ta, and stainless steel-VD 10H18N10T as a foil thickness of 100 µm and a diameter of 17 mm each. The initial state of metals achieved by annealing at \( T = 1200^\circ \text{C} \) and steel - at \( 1050^\circ \text{C} \) for 1.5 hours in a vacuum of \( 10^{-5} \text{ Pa} \). The thickness of each foil \( \Delta d \) defines a path element \( \Delta x_i = \Delta d \cdot \rho \), on which there is a loss of energy of protons \( \Delta E_i = s_i(E) \Delta x_i \); the
The mean energy of the protons on the other side of each foil will be \( E_{\text{f2}} = E_{\text{f1}} - \Delta E_{\text{f}} \). Consequently, each successive foil is irradiated with protons of different energies, studying the degree of damage that can be installed by its energy dependence. Irradiation was carried out by protons with a flow of \( 1.2 \cdot 10^{13} \text{ cm}^{-2} \text{ s}^{-1} \) up to \( 2 \cdot 10^{17} \text{ cm}^{-2} \) 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), resulting from the 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 react also to change the order of the structure. Therefore positron probe is an ideal tool for studying the electronic states of the metallic materials local microregions. The study of structural defects of materials produced by the spectrometer with a linear-slit geometry with an angular resolution of 0.5 mrad. As a positrons source the isotope \( ^{22}\text{Na} \) of with 10 mCi activity was used. 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. For this experimentally measures the intensity of the annihilation gamma rays as the dependence of counting rate of pulses coincident in time 2 photons 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. 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 (Figure 1).

![Figure 1. Experimental ADAP spectra: 1 - for the source; 2- irradiated materials](image_url)
Results and discussion

To interpret the results of studies were 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 ΔF of relative values for the initial state is recovered by processing the spectrum of the angular correlation of annihilation radiation [4]. Results of studies of stainless steel according to these conditions by the annihilation parameters change data are summarized in Table 1.

Table 1. Parameters annihilation became the depth of passage protons $E_{\text{initial}} = 30 \text{ MeV} \ (F = 2 \times 10^{17} \text{ cm}^{-2})$

| 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        |

High energy protons irradiation is change considerably the spectrum shape of annihilation photons angular distribution. The spectrum is narrow in half-width and maximal intensity raise consequently the redistribution of positrons annihilation probability with conduction and ion core electrons. These factors are visible in annihilation parameters change. If for annealed sample $F = 0.15$, whereas after irradiation it increases almost twice as much. The spectrum half-width (FWHM) for initial state is equal 6.1 mrad. The protons irradiation is reduce to it decreases at the average up to 5.6 mrad. In spite of existing parameters considerable change the evident regularity between them and protons energy does not visible in this case. Though, the certain tendency in the annihilation parameters behavior yet it can be established. Thus, for example the positrons annihilation relative probability $F$ with particles energy...
increase is steadily decrease at the average. It can testify that the main contribution in the steel radiation damageability carried in the low energy protons are suffered the elastic collisions with steel components atoms.

Therefore it can suppose that the structural damages arising meanwhile in the steel samples are not differ practically between itself both by configuration and by positrons capture efficiency and correspond to the traps of one type. The last is confirmed by shape of isochronal annealing curves from the stack of the samples irradiated by different energy protons (Figure 2). At \( E_p = 29.5 \div 13.2 \text{MeV} \) protons energy the materials basic properties recovery is ended in the 350-600°C temperature interval and at \( E_p \leq 6 \text{MeV} \) - in the field of 250 - 550°C, that is the specified relationship between protons energy and defects annealing temperature existence is present.

![Figure 2. The energy dependence by protons irradiated 10X18H10T – ВД steel annealing kinetics](image)

1. \( E_p = 4.6 \text{MeV} \); 2. \( E_p = 31.2 \text{MeV} \);
3. \( E_p = 23.1 \text{MeV} \); 4. \( E_p = 29.5 \text{MeV} \)

Apparently of the defects migration activation energy value, consequently irradiation by high energy protons the dislocation loops with \( E_a = 2.1 - 2.2 \text{eV} \) are appeared. In the low energy protons case the vacancy complexes as a small subcascades or connected vacancy-impurity complexes with \( E_a = 1.7 - 1.8 \text{eV} \) are created evidently. The connected vacancy-Cr atom state which difficultly than other decomposed at annealing is most likely[5].

Unlike of stainless steel the polycrystallinetantalum irradiation at the same conditions is lead to same characteristic changes of the annihilation parameters. It is very difficult to separate out for this metal the spectrum parabolic component in consequence of insignificant part of the freecharge carriers. Therefore the basicequivalent (instead of F) parameter is the counting rate in the maximum of spectrum relation to its value at the angle \( \theta = 8 \text{ mrad} \), that is \( f = N(0)/N(8) \). The dependence of this parameter on protons energy is given in Figure 3a. The maximal spectrum narrowing and parameter \( f = N(0)/N(8) \) considerable increase correspondingly is observed at low energies ~5 - 8 MeV. With particles energy increase these parameters are possessed a steadily increase or decrease character.
Such changes of parameters characterized of 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 is fitted on field of low energy protons, that which 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, here is negligible. The actual picture of the structure damages in tantalum is can be determined consequently 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 tantalum, 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 is recover in one stage in 250 - 600°C temperature interval with curve amplitude ~15 % (curve 1), whereas at high energy irradiation the structure damages in Ta are appearing with two recovery stages (curves 2, 3). And with particles energy decreasing from 30MeV to 25MeV the point defects migration beginning is shifted sideways to lower temperatures with simultaneous increasing of vacancy defects part. This confirm the decisive contribution of elastic interactions in the process of defect formation. So, at protons irradiation with $E = 30$ MeV the vacancy stage part is compose 41% from the damages common level and with energy decreasing up to 25MeV it increasing up to 47%. Meanwhile the recovery second stage is receive the more relief appearance than in the first case. The migration activation energy for vacancy components is mean $E_{a1} = 1.41–1.45$ eV, and for dislocation and more complicated components are annealed pending second stage - $E_{a2} = 2.33–2.35$ eV.

Still large interest in these experiments are can present the damageability distribution profiles of molybdenum irradiated at the same conditions study results. For molybdenum in 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. The parabolic component in spectrum is does not succeeded. But consequently protons irradiation the nascent structure damages are promoted to the parabolic component responsible for positrons annihilation with free charge carriers appearing in the spectrum. Meanwhile the energy dependence of the relative annihilation probability $F = S_p / S_g$ have a more complicated and nonmonotonic character.
(Figure 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](image)

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

The probablereason of such dependence can be the different contribution in the radiation defects generation process 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 specifiedroleon the other hand. But in both cases the defects structure must be vacancy that is confirmed by isochronal annealing results. Regardless of energy the proton irradiation consequently the vacancy complexes are appeared in Mo and in annealing temperature region up to 800°C the only one incomplete return stage have been observed. Meanwhile as far as proton energy increasing the defects migration temperature threshold is gradually shift to more high temperatures sideways - from 300 - 350°C up to 450 - 500°C that testify about appearance in the metal the more stable temperature concerning defects. But in work [6] is stated that the full annealing of irradiated by protons Mo completed at temperature about 300 - 320°C. But this temperature is considerable lower than Mo recrystallization temperature, that give rise to doubt in above mentioned conclusion [6]. But annealing temperature reached in this investigation (850°C) is also does not sufficient for completioneven the first stage of recovery (III-stage), whereas the recrystallization temperature of Mo is higher than 1000°C[7].

**Conclusion**

Thus the defects distribution profiles investigation by charged particles passing depth in three different materials is shown that in stainless steel and Mo regardless of protons energy similar to configuration vacancy complexes are appeared which recoverable in one annealing stage with migration activation energy $E_a = 1.7 – 1.8$ eV. The steel total damageability in the whole of protons energy interval is formed basically by one low-
grade elastic channel, whereas for molybdenum it is added from two components in each of which its defined mechanism of defects formation is work. If for high energy protons the inelastic channel of interactions and subcascades appearing are the basic, that which the major processes for low energy protons are the elastic interactions with lattice atoms and atomic hydrogen formation in the end of run. The basic mechanism of the radiation defects formation in Ta is the evidently expressed elastic interactions. The nuclear reactions role in the structure damages creation are low-grade in this case. The defects created by low energy protons are annealing in one stage, whereas at high energy particles influence the radiation defects appearing which are recovery in two stages possessed the vacancy and dislocation nature.

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