

Experimental and Theoretical Studies of Distribution of Vacancy Clusters in Depth of Material Irradiated by High-Energy Particles

A.A. Kupchishin¹, A.I. Kupchishin^{1,2,*}, N.A. Voronova^{1,2}, V.I. Kirdiashkin^{1,2}, AM Bogdan³

¹Abay Kazakh National Pedagogical University, Almaty, Kazakhstan

²al-Farabi Kazakh National University, Almaty, Kazakhstan

³Tomsk Polytechnic University, Tomsk, Russia

E-mail: ankupchishin@mail.ru

Abstract. Non-destructive method of positron depth sensing of the radiation defects in solids was proposed and implemented. The depth distribution of vacancy clusters in molybdenum irradiated by protons with energy of 30 MeV was found based on the experimental data. Theoretical calculations of this value were made and sufficient agreement between calculations and experiment was obtained.

Keywords: vacancy cluster, material, radiation, particle, proton, primary knock-on atom, energy spectrum, concentration, positron, annihilation, method, energy.

1. Introduction

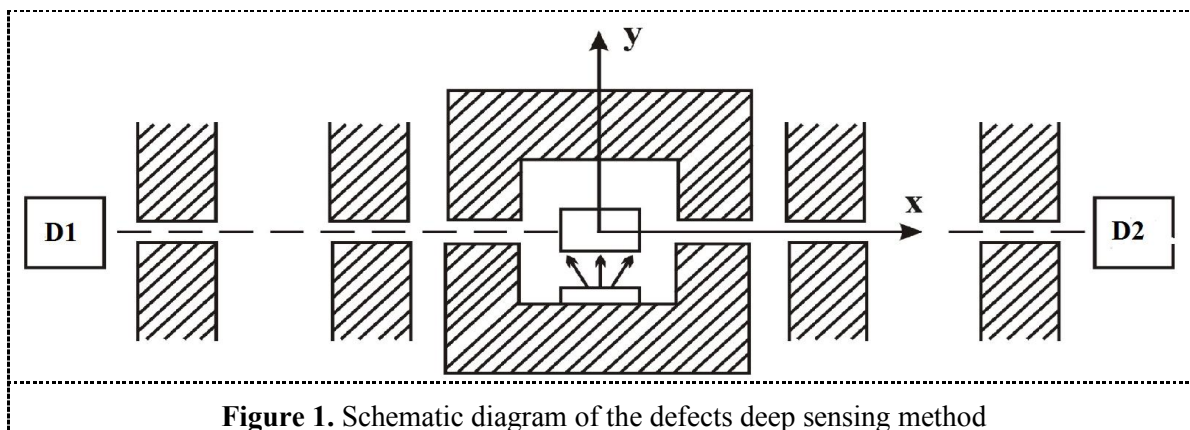
Due to the intensive development of nuclear power, various studies of the physical processes taking place in near-Earth space, including in radiation belts around Earth, study of the influence of electron, gamma, and ion irradiation on the properties of materials is currently one of the most important trends in radiation physics and solid state chemistry. As a result of bombardment of solids by charged particles, such as ions, not only light particles such as secondary electrons are generated (when power is lost, mainly through ionization of the atoms and molecules), but also heavy atoms and ions of the medium, which are the progenitor of the atomic nuclear cascades, can be generated along the path of their movement. Neutrons interacting with matter also form primary knock-on atoms (PKA). However, due to their great penetration capability they create radiation damage over much greater distances unlike charged particles. The energy spectrum of PKA at different depths in the material depends on the energy, the mass of the incident particles and the type of target (atomic mass, density), integral and differential cross sections of particle-atom system and atom-atom collisions, and the energy losses due to ionization and excitation. Propagating through the medium, PKAs generate secondary knock-on atoms, and then tertiary ones, etc. Ultimately, after the first stage of the process in the solid, there is a formation of radiation-induced defects such as vacancy-interstitial atom, divacancy, two knock-on atoms and so on. As a result of the passage of particles and development of diffusion processes defects interacting with each other transform into vacancy complex, clusters of interstitials. Types of defects and their distribution by size and depth of the irradiated sample depend not only on the type and energy of particle, material type, but also on the impurity concentrations,



dose, temperature, uniformity and time of the irradiation, and are responsible for a change of almost all properties of the irradiated material [1]. The complexity of the processes occurring in solids under irradiation by different particles leads to considerable difficulties in the interpretation of the experimental results, especially because at the moment there is not enough complete theory of radiation damage of materials, particularly metals that form the basis of nuclear power plants.

2. The method of positron depth sensing of radiation defects

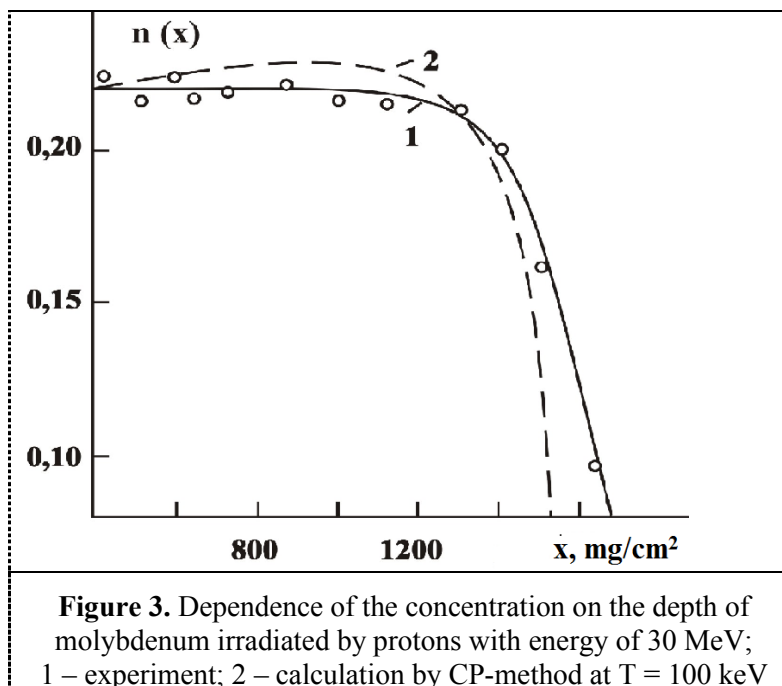
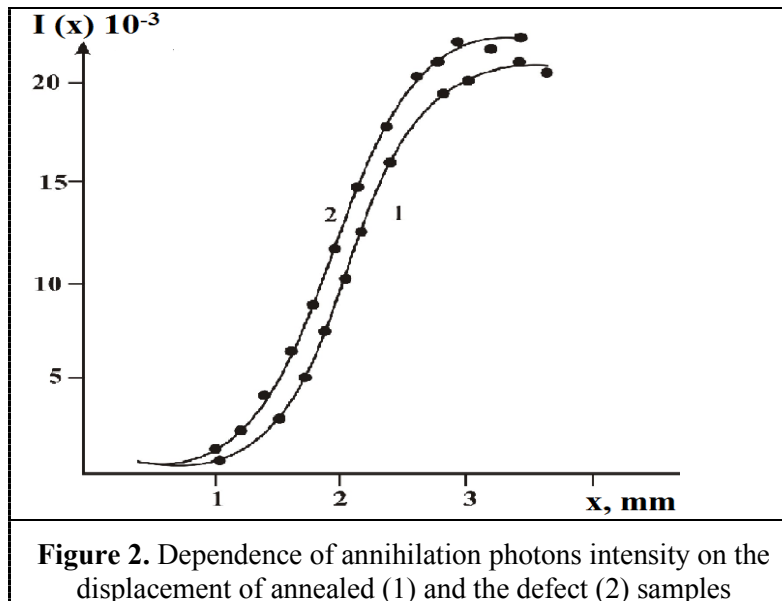
In [2], it was shown that in many experiments on the annihilation of positrons, the distribution of the concentration of radiation-induced defects in depth can be determined in defective materials by varying thickness of the absorber involving selection of the irradiated material in the form of a stack of foils. The number of foils is desirable to choose as equal to $N \geq N_0$ ($N_0 \sim 10-15$), and their thickness – from the condition of $d = x_{\max}/N$, where x_{\max} is a maximum depth of material where there are defects, annihilating in which positrons are still able to contribute to the spectra of photons. Since actual experiments often have to deal with whole materials (rather than with a stack of foils), there is a need for a method to determine the profiles of radiation-induced defects in materials having a greater thickness. To this end, we propose a method of defects depth sensing by high-energy positrons. The maximum depth of x_{\max} , where the defects can be explored, is determined by the energy of positrons, which is desirable to be changed. The method of deep sensing of radiation defects can be carried out using both units of angular correlation (2γ , 3γ , etc. matches) and the unit of the lifetime or the Doppler effect with small changes in the patterns of these experiments. Schematic diagram of the experimental depth sensing is shown in Fig.1. It is based on the principle of gradual displacement of the annealed sample the first and then the defective sample perpendicular to the line D1 - D2 (connecting the detectors) to a relatively small distance Δx . The principle of moving the samples had previously been used by researchers to study the process of passage of positrons through materials [3]. However, our experiment has entirely different purpose - to establish the spatial distribution of defects in the depth of the irradiated sample. However, ultimately distribution (2γ -matches) of $J(x)$ for both annealed and for defective samples is measured.



We investigated samples representing monolithic tablets of polycrystalline molybdenum with diameter of 20 mm, annealed in a vacuum of $1.3 \cdot 10^{-3}$ Pa at 1100 °C for 1 hour. The thickness of the sample corresponds to the full path of the protons with an initial energy of 30 MeV in molybdenum. Irradiation was carried out on a cyclic accelerator by the flow of $9 \cdot 10^{12}$ h/cm² to the integral dose of $6 \cdot 10^{17}$ particles/cm². The sample temperature during irradiation was maintained at 80 °C. The measurement of the angular distributions of annihilation photons was carried out using UKAF automated instrumentation with program software [3]. During this experiment, the angular resolution of the instrumentation was 1 mrad, and the pitch of the movable detector was 0.5 mrad.

3. Experimentals

Fig. 2 shows the dependences of $J_1(x)$, $J_2(x)$ on x annealed Mo samples and samples irradiated by protons with an energy of 30 MeV to the fluence of $6 \cdot 10^{17}$ r/cm². It can be seen that the irradiation leads to a shift of the curve to the left, and to a substantial increase in the intensity of annihilation photons at the same values of displacement x . Analysing the change in the intensity of $J(x)$ for annealed and irradiated states in different layers of Δx , a change in the concentration of these defects can be also studied. In addition to moving the sample, the method can be based on moving the slits with width Δx . There are other options, but the essence will not change.



In the case of defects in the material of only one type (or by the separation of the photon flow to i -component, for example during the annealing) the following formula can be obtained [3]:

$$n(h) = \frac{(J_{ir} - J_{an})}{\Delta x} n_0, \quad (1)$$

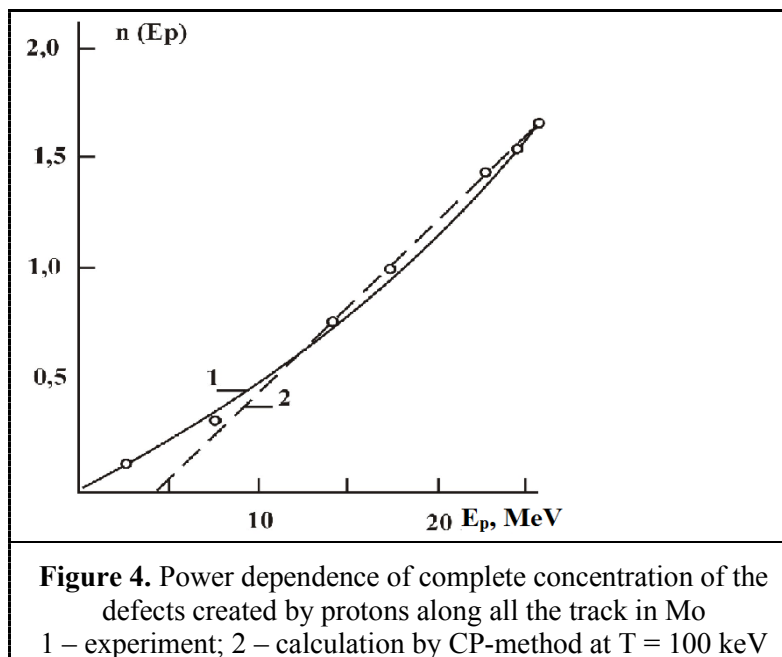
where $n(h)$ is the concentration of defects at a depth h ; J_{ir} and J_{an} are normalized values of the intensity of photons in the maximum of the curve for irradiated and annealed states; n_0 is distribution constant.

Dependence of the defect concentration on the depth has been calculated according to the formula (1) (Fig. 3). Fig. 3 shows that $n(h)$ is constant up to a certain depth $h = h_{cr}$ and then sharply decreases to zero. These results are also confirmed by electron-microscopic studies. Then, using the data on positron annihilation (Fig. 4), the energy dependence of the total concentration of defects produced by proton along the track in Mo has been calculated.

Further, CP-method was used for calculation of the spectra of PKA in molybdenum irradiated by protons with energy of 30 MeV. It was assumed that between the elastic collisions, leading to the displacement of PKA, the proton loses its energy continuously (described by the formula such as the Bethe-Bloch as in the case of electrons) and after each collision it retains its direction of motion. Introducing the concept of minimum energy of the formation of cluster T_k , in the first approximation the concentration of clusters $C_k(h)$ at the depth of h can be found from the expression [1]:

$$C_k(E_1, h) \approx \frac{1}{\bar{\lambda}(E_1)} \frac{E_d}{T_k} \left(1 - \frac{T_k}{E_d(E_1)} \right), \quad (2)$$

where $\bar{\lambda}(E_1)$ is the average value of the displacement path; p is the average value of atomic collisions along the track of the proton up to the depth of h ; $E_d(E_1)$ is damage energy.



According to this formula at $T = 100$ keV, spatial distribution of vacancy clusters and energy dependence of the total concentration of defects produced by protons along the track in Mo (Fig.4, dashed line 2) were calculated. It is clear that $C_k(h, E_1)$ is essentially constant and then changes

abruptly at the end of the track. Fig. 4 shows the energy dependence of $C_K(E_1)$ for Mo calculated by the formula (2). Comparison of calculations with experimental data on positron annihilation shows that there is a satisfactory agreement for both the spatial distribution of $C_K(h, E_1)$ (Fig.3) and the energy dependence of $C_K(E_1)$ (Figure 4, dashed curve 2).

Thus, the expression for $C_K(h, E_1)$ found by CP-method in the first approximation explains the experimental data on positron annihilation and can be used by researcher in their work.

4. Conclusion

1. Non-destructive method of positron depth sensing of radiation defects in solids was developed. Distribution of intensities of gamma rays directly in the annihilation zone was experimentally measured for irradiated (by the protons with energy of 30 MeV) and non-irradiated samples of molybdenum. These data and the proposed method were used for studying the distribution of vacancy clusters in the depth of the material.

2. Cascade-probabilistic method was used for calculation of $C_K(h)$, and a satisfactory agreement with experiment was obtained. The value of the concentration of vacancy clusters initially hardly changes with depth, and then abruptly falls to zero at the end of the path.

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