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METHODS OF MATERIALS IRRADIATION WITH HIGH ENERGY ($\geq$1 MeV/nucleon) CHARGED PARTICLES

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Introduction.

The interest to the development of methods of irradiation with high energy charge particles (HECP) manifested itself in full measure in early 1970 and was caused first of all both by the problem of radiation material swelling and development of a new trend of researches of thermonuclear materials science.

One of the deterrent of scientifically justified and effective use of material irradiation with HECP in solving both fundamental problems of radiation physics and applied ones of thermonuclear materials science was the absence of solving the complicated tasks of irradiation procedure with accounting the specific peculiarities of forming the energy profiles of radiation damage and ion alloying.

In this connection the determining conditions providing the possibility of using the controlled HECP irradiation for solving the wide complex of scientific applied problems of radiation physics and thermonuclear materials science are the elaboration of the following methods: radiation dosimetry, solid samples of ion alloying uniform in volume, space formation in samples of energy damage profiles and ion alloying for their further studying both with the help of transmission electron microscopy and wide complex of physico-mechanical methods of investigation.

At present new systematic methods of sample irradiation with HECP performed at the FLNR accelerators (JINR) are described.

Radiation dosimetry.

Radiation dosimetry refers to the main parameters determining the reliability and reproducibility of the results obtained under the influence on the objects of charged particles accelerated at cyclotron. In this case due to specific properties of charged particle beam connected with its space inhomogeneity one needs the measurement of density distribution of particle flux over the surface of the irradiated sample. Because of instability of a beam with time and space it is very difficult to perform the precision dosimetry "in situ". In practice the methods of post-irradiation dosimetry are often used, in particular the method of photometry of autoradiography from the irradiated sample and also the method of induced radioactivity of the monitor-material. For the last method there was proposed the way of using the multi-isotope monitor-material in order to unify it in the wide range of the HECP energies [1]. For α-particles with energies in the range from

20 to 50 MeV which are widely applied in solving the problems of radiation resistance of structural materials for TNR we have justified and propose to use the multiisotope molybdenum for monitor. It has been found experimentally that isotope ^{97}Ru has the most activity in molybdenum slightly depending on α-particle energy in the range of 20–50 MeV. In Fig.1 there is illustrated the dependence of production cross section of ^{97}Ru of α-particle energy in natural molybdenum. One should note that a multiisotope natural molybdenum has a combination of properties which are necessary for creating the unified monitor for α-particles: it is convenient in performing the measurements with radioactive isotope ^{97}Ru (half-life is 2,89 days), constant in the wide energy range of α-particles with high sensitivity, high melting, cheap and can be used many times.

Uniform ion alloying of solid samples.

One of the main advantages of HECP is the capability to penetrate into the objects at great depths (tens or hundreds μm). However, the peculiarities of physics of damage and alloying of HECP do not provide the conditions of the uniform distribution of radiation defects and ion accumulation along the way of particle motion in the sample without resorting to specific modification of the irradiation procedure.

It is known that ions falling on the sample stick at the end of their motion in so-called straggling zone the width of which is some percents of the length of the projective particle range in material. To obtain the uniform sample alloying with thickness less than or equal to the length of the projective range one should provide the conditions for straggling zone to move in the irradiated sample changing the length range of the projectiles in sample material. More often a set of thin absorbing foils [2] or absorbing filters with varying thickness, e.g. in the form of a wedge [3], is applied. These methods have a disadvantage – preparation of absorbing filters is a time-consuming task.

In the present paper three methods of HECP irradiation simple in practice are described. They provide conditions for forming the uniform volume concentration of implanted ions in solid samples.

The basis for the first method is the fulfilment of the condition for bilateral irradiation of a plane sample when it constantly rotates in the flux of projectiles [4]. In this case the necessary conditions for forming the uniform volume concentration of implanted ions are:

- the thickness of the irradiated sample should be less than or equal to the projective ion range of the given energy in material;
- the uniformity of particle flux in cross section of the beam falling on a sample;
- the size of ion beam should be more than or equal to the square of the irradiated sample.

The basis for the second method of the uniform ion alloying is the use of free air as a filter with the varying absorbing capability due to reciprocating motion of the irradiated sample along the ion beam extracted into the atmosphere [5]. The depth of the ion penetration into a sample depends on the energy of incoming ions which in its turn is determined by their path length through the air. In this case, except straggling zone, the linear connection takes place between the particle range through the air and in the sample material under the condition of its uniform motion in the particle flux extracted e.g. into the atmosphere.

The third method of forming the uniform concentration of implanted ions in solid sample excludes the time-consuming process of preparing the absorbing filters of the varying thickness in the form of a wedge. It involves the object irradiation through the absorbing filter, as a foil curved by the parabolic law, moving along its bombarding surface. Parabolic foil curvature meets the requirements for obtaining the linear dependence of changing the thickness of absorbing filter. Varying the foil thickness one can change the alloying depth.

The first method is the most effective under irradiation of a large quantity of samples, e.g. for mechanical tests, the second one – for irradiation in different gaseous media, and the third one – for reaching high concentrations of implanted ions under controlled thermal and deformation conditions.

Method of changing space disposition of ion alloying profiles.

It is known that under irradiation of a plane sample with the accelerated ion beam falling perpendicular its bombarding surface, at a depth equal to the projective particle range (R_p) the region of the ion alloying is formed. The size of this region ($2\Delta_{||}$) is determined by so-called longitudinal particle straggling, and the profile of concentration of implanted ions is approximated in it by the Gaussian distribution function.

High gradients both damage rates and concentration of alloying ions influence upon the of radiation-induced defect structure formation behavior at the end of particle range providing the mobility of radiation defects and implanted ions.

In connection with the above it is of scientific and practical interest to study of the formation processes of the profiles of radiation damage and ion alloying under such nonequilibrium conditions of the defect formation.

For these purposes both the indirect (e.g. Rutherford scattering) and direct (transmission electron microscopy (TEM)) methods are widely used. However their use, especially under object irradiation with high energy ions, has experimental limitations connected both with a great attitude depth of the doped region in a sample and with impossibility for most materials to prepare the TEM-objects following by so-called "cross section" method [6].

In the present work a new method of irradiation revealing limitless possibilities of its application in simultaneous studying of structural changes in TEM in the region of high energy ion alloying of any materials is discussed.

The basis for the proposed method is the plane sample irradiation with high energy particles through absorbing filter of cylindrical shape [7] which provides the conditions for forming cross section profile of ion alloying in a sample volume (Fig.2).

Upon irradiation, using the usual method of preparing thin foil with thickness δ parallel to the bombarding surface one can obtain the TEM-objects from any depth of a sample, involving sections with the unirradiated region (A), zone of the ion alloying (B) and region with the known energy profile of radiation damage (C).

Experimental test for performing the given method in practice was realized at macroscopical and microscopical levels. In the first case the copper plate was irradiated with α-particles with the energy of 50 MeV through the absorbing filters in the form of copper wires 230 μm in diameter. The last case was chosen less than the projective range of α-particles (360 μm) with the aim of forming the whole alloying profile in the sample volume. Upon irradiation the sample was annealed at temperature 750^{0}C. In Fig.3 the macrostructure of the copper sample on the surface parallel to the flux of bombarding α-particles is presented. The analysis of alloying profiles manifesting in the form of sample regions filled with helium pores allows to distinguish two cases. First, the size of the alloying region formed with the help of longitudinal straggling of α-particles is 2,5–3 times less than the alloying region formed by cross straggling. In this case the width of the latter practically doesn't change towards particle migration in a sample. Secondly, the size of pores and their concentration in the alloying region formed by longitudinal straggling decreases with increasing the depth in a sample.

The exact determination of the function of volume distribution of the ion concentration in the alloying region in terms of the longitudinal and cross straggling is of a difficult problem. To a first approximation, using the TRIM-90 program one can show the function of distribution in two-dimensional case as:

$$P[x,z/x_0,t(x_0)] = \frac{\Phi}{2\pi\Delta_{||}\Delta_{\perp}}\cdot\frac{1}{2}\left\{\exp\left[-\frac{(x_0-R_{\perp}-x)^2}{2\Delta_{\perp}^2}\right]+\right.$$
$$\left.+\exp\left[-\frac{(x_0+R_{\perp}-x)^2}{2\Delta_{\perp}^2}\right]\right\}\cdot\exp\left[-\frac{(t(x_0)+R_{||}-z)^2}{2\Delta_{||}^2}\right],$$

where Φ – a particle flux, x, z – a coordinates of the point in a sample, x_0 – a coordinate of particle entrance in a sample, $t(x_0)$ – a function describing the change of thickness of the absorbing filter, $\Delta_{||}$ and $\Delta_{\perp}$ – longitudinal and cross straggling, respectively, $R_{||}$ and $R_{\perp}$ - longitudinal and cross range particles, respectively. In the case of using the filter of cylindrical shape with diameter D its thickness towards x changes according to the law $t(x) = 2\cdot\sqrt{2Dx - x_0^2}$. For α-particles with energy of 50 MeV the longitudinal projective range in the copper is 360 μm ($\Delta_{||}$=9,6 μm and $\Delta_{\perp}$=22,3 μm) this is saticfactory agrees with experimental data. Let's note that in the last case one can take into account both helium pore diffusion and gas sweeling of the alloying region.

In the Fig.4 there is presented an example of experimental realization of the proposed method of irradiation for simultaneous observation of structural changes in TEM in the energy damage profile and ion molybdenum alloying under neon ion irradiation with the initial energy equal to 113 MeV. In Fig.4a the defect area in which the damage in the form of dislocation loops changes in accordance with changing the neon ion energy from 80 to 0 MeV is illustrated.The general picture of structural changes along the concentration profile of the alloying molybdenum (region B, see Fig.2) upon annealing at 1100°C is given in Fig.4b. One can see from the unirradiated sample section (region A, see Fig.2) that the distribution of dislocation loops on size and concentration has a asymmetric character: the average size and quantity of dislocation loops is more and less, respectively, to our opinion this is due to the relative shifts of the damage profiles and ion alloying in it.

Thus, in order to obtain the information on radiation-induced or stimulated structural changes along the profiles of radiation damage and ion alloying in any material under high energy ion irradiation ($\geq$1 MeV/nucleon) without invoking the very complicated so-called "cross section" methods there has been proposed a simple practical method of forming the longitudinal damage profiles by ion irradiating the plane samples through the absorbing filters of cylindrical share.

Changing the size (diameter, length) and geometrical forms of cylindrical filter in the object plane one can obtain ion alloying zones, arbitrary in size and shapes, in its volume, this method can be used with another purposes in technology of different regions employing the ion material modification.

Formation of the energy damage profiles and ion alloying on the bombarded surface.

Because of very small sizes of the ion alloying zone, even for ions with the energy of some tens MeV studing the radiation effects at the end of their range in so-called straggling zone with the use of the majority of classic methods of measuring physico-mechanical properties is practically impossible. This is due to the fact that straggling zone size is many times smaller than that of sounding section.

In this connection there exists an actual problem associated with the development of such irradiation method which would satisfy the conditions of forming the radiation damage profiles and alloying along the bombarded surface to the scale essentially exceeding such a lever in depth.

The main point of the proposed method lies in the fact that the studied object (foil or plate) is irradiated with HECP through the absorbing filter in the form of foil curved according to the given law [8]. The thickness of absorbing foil and the profile of its curvature are

chosen from the conditions of the initial energy of projectiles and from the required law of their energy distribution on the irradiated sample surface.

The thickness of foil from which the absorbing filter is prepared is chosen from the condition of forming the damage profile on the sample surface formed within the given range of projectile energies. For example in order to form the damage profile changing on the sample surface because of the falling ion energy in the range from 0 to E_{max} (where E_{max} should correspond to the initial ion energy as close as possible to E_0, i.e. $E_{max} \simeq E_0$), one has to use the foil with thickness by far less than the ion range in filter material R_p, e.g. about $(0{,}05\text{–}0{,}2)R_p$. If it is necessary to widen the damage zone in straggling region the thickness is taken somewhat less than R_p, e.g. $(0{,}8\text{–}0{,}9)R_p$. Reasonable limitation in applying the given method is associated with a necessary use of ions with the energy providing their range in a material at least of 1 μm.

As an illustration of the proposed method of the object irradiation through the curved absorbing filter (CAF) with high energy particles in Fig.5, there are presented the calculated profiles of the damage level and helium concentration on the molybdenum surface, irradiated in vacuum through CAF (thickness 110 μm, curvature radius - 42 mm) with α-particles with the initial energy of 29 MeV up to fluence of $3.8 \cdot 10^{20}$ m^{-2} and the results of measurement of relative change of the lattice parameter and the microhardness value.

We do not dwell on physical peculiarities of changing the lattice parameter and microhardness in space increased straggling zone of α-particles, just note, that the number of points of measuring the lattice parameter and microhardness value are of some tens whereas under the usual method of irradiation one can make just one point.

Thus, the proposed method at the given initial energy of bombarding ions allows: first, to form the energy profile of ion alloying along the bombarded sample surface, second, to increase space interval of changing their energy in the studied sample many times (hundreds times). This enables one to study in detail not only the regularities of changing one or another property of the material versus the energy of bombarding ions with the methods just used but to apply other nuclear-physical methods which are not available for these goals earlier also as well.

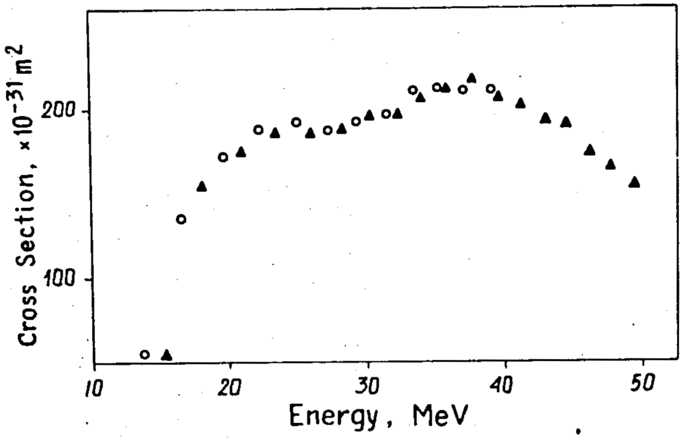

Fig.1. Change of cross-section of registered radioactive isotope (^{97}Ru) formation in suggested material of monitor (Mo) as a function of α-particle energy.

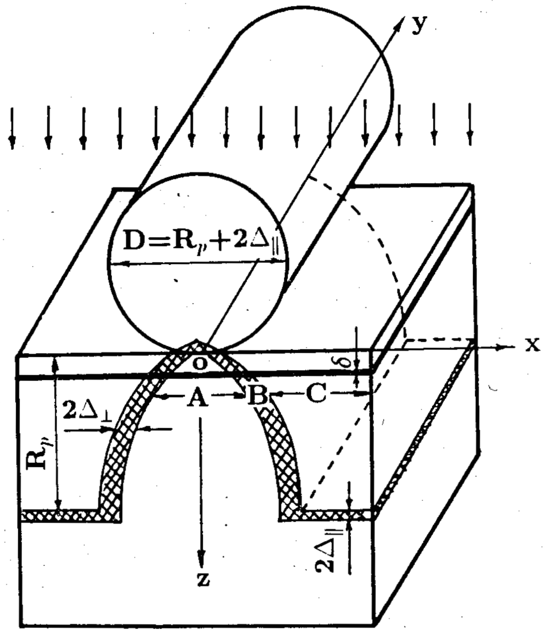

Fig.2. Schematic view of a sample irradiated with high energy ($\geq$1 MeV/nucleon) ions through the absorbing filter of cylindrical shape. D- filter diameter, R_p - projective range, $\Delta_{\perp}$ - cross straggling, $\Delta_{\parallel}$ - longitudinal straggling. Region A - unirradiated sample part, B - zone of ion alloying, C - damage region from the ions with a varying energy, δ - TEM-object thickness.

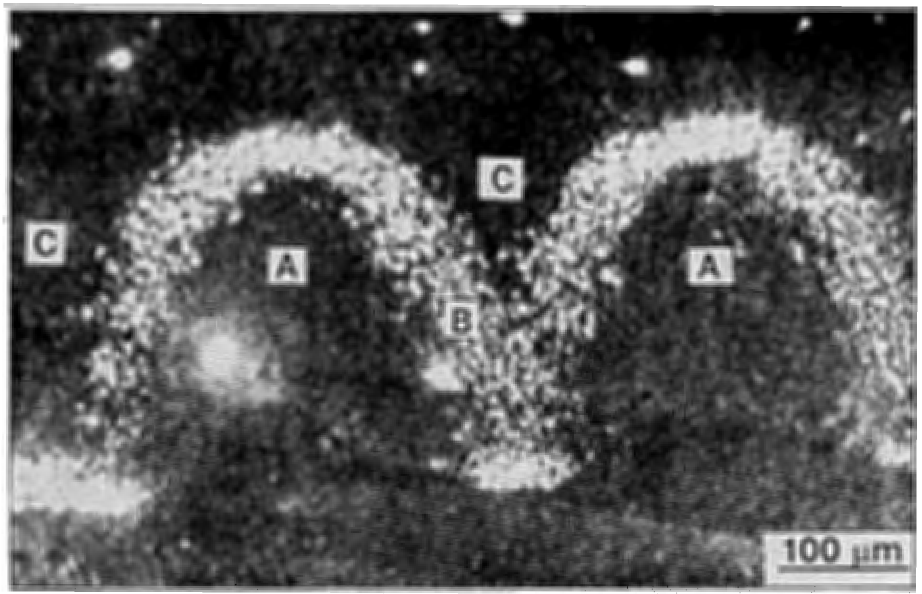

Fig.3. Profiles of zones in the copper sample doped with helium upon α-particle irradiation with the energy of 50 MeV through absorbing filter of cylindrical form 0,23 mm in diameter and upon post-irradiation annealing at 750^{0}C. (A, B and C - see Fig.2).

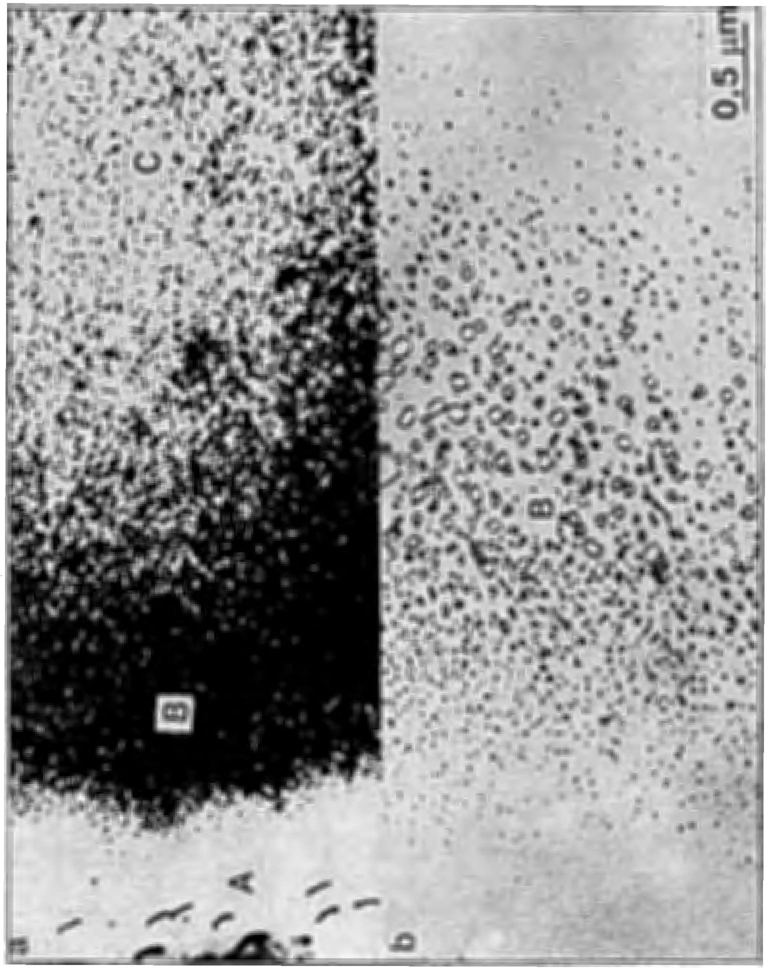

Fig.4. Energetic profile of radiation damage upon $^{+4}Ne^{22}$ ion irradiation with the energy of 113 MeV (A, B and C - see Fig.2) -(a); damage profile of Ne ions in straggling zone (region B) upon 1100^{0}C post-irradiation annealing - (b).

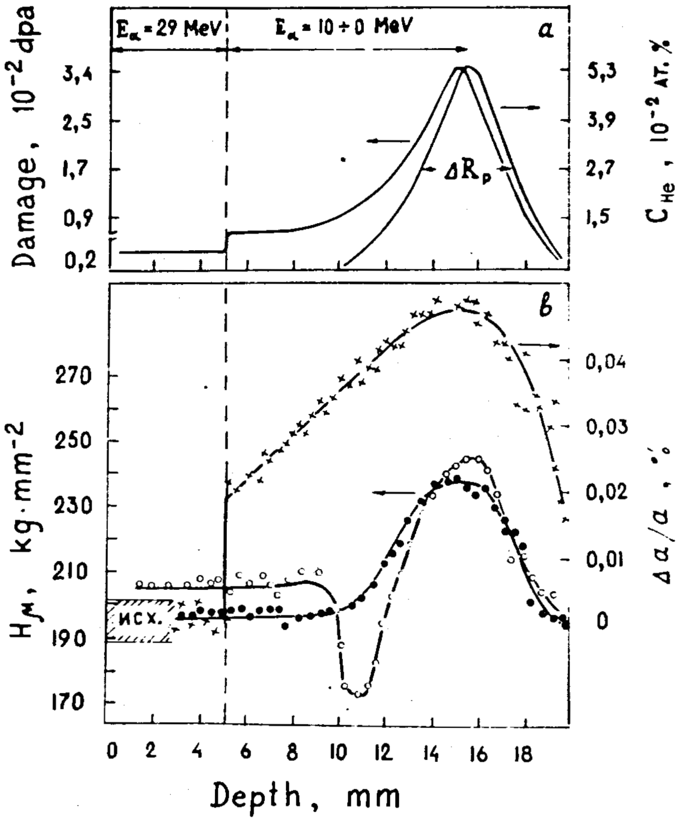

Fig.5. Change of molybdenum lattice parameter (x) and microhardness (o) in straggling zone of α-particles with 29 MeV energy – (b). Real size of straggling zone is 10 μm, the suggested method gives ΔR_p=$8 \cdot 10^3$ μm – (a).

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Методы облучения материалов высокоэнергетичными (≥1 МэВ/нуклон)
заряженными частицами

Высокоэнергетичные заряженные частицы (ВЭЗЧ) с энергиями более 1 МэВ широко используются для решения многих фундаментальных проблем радиационной физики твердого тела и практических задач ядерного, термоядерного и космического материаловедения. Выполнены и реализованы следующие методические разработки вплоть до использования на циклотроне У-200 (ОИЯИ, ЛЯР): 1) три метода облучения заряженными частицами, создающие условия ионного легирования тонких плоских объектов (с толщиной пластинки меньше или равной длине пробега ВЭЗЧ) без использования поглощающих фильтров в форме клина или набора фольг; 2) метод изменения пространственного положения энергетического профиля ионного легирования в объеме образца, создающий условия для последующего изучения в ПЭМ радиационных эффектов вдоль энергетического профиля повреждения и ионного легирования во всех материалах; 3) метод многократного (в сотни раз) пространственного увеличения энергетического профиля заряженных частиц на облученной поверхности образца, создающий условия для экспериментального изучения радиационных эффектов вдоль зоны страгглинга ВЭЗЧ практически всеми ядерно-физическими методами.

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Methods of Materials Irradiation with High Energy (≥1 MeV/nucleon)
Charged Particles

High energy charged particles (HECP) with energies more than 1 MeV/nucleon are widely applied for solving a lot of fundamental problems in radiation physics of solid state and practical tasks of nuclear, thermonuclear and cosmic materials study as well. The following complex of methodical developments has been founded and realized up to practical use at cyclotrons U-200 (JINR, FLNR): 1) three methods for volume uniform ion alloying of thin plate solid samples (plate thickness is less or equal to projective HECP range) without using absorption filters in the shape of wedge or a set of folis; 2) a method for changing spatial position of ion alloying to TEM-examination of radiation effects along energetic profile of damage and ion doping in any material; 3) a method of multiple (by hundreds of times) spatial increase of energetic profile of charged particles on bombarded sample surface, providing conditions for experimental study of radiation effects along straggling zone of HECP by practically all nuclear-physical methods.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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