

Post-Irradiation Effects of Tantalum by Charged Particles

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Abstract—Post-irradiation effects of metallic tantalum bombarded with proton beams and nuclear reaction products, as well as Ta self-ions knocked out from lattice sites, were investigated using probe emission Mössbauer spectroscopy and X-ray diffraction. A methodological approach is proposed for studying post-effect patterns of critical radiation exposure in structural materials.

Keywords: metal irradiation, radiation defect, probe emission Mössbauer spectroscopy, X-ray diffraction

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INTRODUCTION

Emission Mössbauer spectroscopy (EMS) allows one to obtain unique information about the charge and local environment of impurity atoms, to unambiguously identify the crystallographic positions they occupy, to study the processes of interaction of impurity probes with other atoms and defects of the crystal lattice, and to conduct research in the range from ultralow temperatures to 1000 K, which are practically inaccessible to traditional physicochemical methods [1–6].

In the last few decades, EMS has found wide application in studying the structural features of metals that have undergone extreme influences: irradiation with charged ion beams, severe plastic deformation, etc. [1, 3, 5, 6]. The use of EMS in experiments with metal matrices allows one to obtain undistorted information about the structural states of atoms before and after critical impact, since in metals there are a priori no post-effects of own radioactive transformation for radionuclides undergoing electron capture or converted isomeric transition (in particular, $^{57}\text{Co}/^{57}\text{Fe}$), used as Mössbauer probes during measurements.

This paper presents new experimental data demonstrating the fundamental possibilities of using impurity Mössbauer atoms as structure-sensitive physicochemical probes in the study of the post-effects of critical radiation exposure—the consequences of irradiation of metallic tantalum by proton beams, products of nuclear reactions, knocked out of the lattice sites by Ta self-atoms.

EXPERIMENTAL

Tantalum foils with a thickness of (40 ± 4) μm , prepared by cold rolling, were used for irradiation.

Annealing of deformation stresses after rolling was carried out for 60 min at (1123 ± 5) K in a high vacuum (pumping up to 10^{-7} Torr, a protective tantalum container inside an evacuated quartz ampoule, in which foils were placed, was used as an additional getter of oxygen). The rate of cooling of samples to room temperature did not exceed 5 K per min.

A hydrochloric acid solution of $^{57}\text{CoCl}_2$ was used to produce labeled tantalum foils without support.

Diffusion implantation of radioactive atoms of ^{57}Co evaporated onto the surface of tantalum substrates was carried out for 140 min at 1350–1370 K in a high vacuum (conditions are given above); the rate of cooling of the samples to room temperature also did not exceed 5 K per min.

Diffusion coefficients of ^{57}Co atoms at such annealing temperatures are $(4\text{--}5) \times 10^{-14}$ $\text{cm}^2 \text{s}^{-1}$ [7, 8], and their range in tantalum during annealing did not exceed 0.5 μm .

The maximum activity of ^{57}Co in the prepared sources was (37 ± 1) MBq with an active spot diameter of 2 cm. Thus, the maximum concentration of ^{57}Co in the surface layer of tantalum (0.5 μm thick) did not exceed 370 ng (less than 1 ppm).

Irradiation of the samples with proton beams was carried out on an MGC-20 accelerator using a target unit [9–11], the design features of which made it possible to control the temperature of the targets during irradiation. The conditions of target irradiation are given in Table 1.

Table 1. Irradiation conditions for ^{57}Co -labeled tantalum targets

Irradiation conditions	Foil 1	Foil 2
Fluence, particles m^{-2}	2.6×10^{21}	2.6×10^{21}
Integral beam current*, μA	11:45	11:45
Proton energy loss, MeV	6.95/5.24	5.08/2.86
Heat release**, W	19.6	25.4
Maximum target temperature***, K	(310 ± 3)	(310 ± 3)

* Iron monitors placed along the charged-particle beam before or after the irradiated samples were used for the estimate.

** Only due to passage of the proton beam through the entire target volume.

*** Measured with thermocouples placed inside the holder of the irradiated targets; the possibility of using thermocouples under intense radiation fields was confirmed in separate experiments with simultaneous measurement of the target temperature by an optical pyrometer.

To study the products of nuclear reactions and the radioisotopic composition of irradiated targets and determine the concentration of probe Mössbauer atoms, a secondary standard of the Russian Federation of units of radionuclide activity was used based on the GX1018 and GUL0035 detection units and the LYNX-MCA analyzer (all instruments from Canberra Industries, United States):

(1) HPGe detector GX1018: energy resolution for the 88 keV photopeak is no worse than 620 eV and for the 1332 keV photopeak, no worse than 1600 eV;

(2) HPGe detector GUL0035: energy resolution for the 5.9 keV photopeak is no worse than 130 eV.

Calibration of the secondary standard of the Russian Federation for the detection efficiency of X-ray and gamma radiation with a specific energy was carried out using sources whose activity was measured by absolute methods:

(1) $2\pi\alpha$ -accounts: ^{226}Ra , ^{227}Ac , and ^{228}Th (under conditions of radioactive equilibrium of the parent and daughter products);

(2) $4\pi\gamma$ -accounts: point sources certified as secondary standards of the Russian Federation ^{54}Mn , ^{60}Co , ^{88}Y , ^{137}Cs , ^{133}Ba , ^{152}Eu , ^{228}Th , and ^{241}Am [12];

(3) coincidences of X-ray, beta, and gamma radiation in the decay patterns of radionuclides ([13]) included in the set of spectrometric reference gamma radiation sources.

The measurement results were processed using the Genie-2000 software package (developer, Canberra Industries, United States).

The Mössbauer spectra were recorded using an MS-1104EM setup (Research Institute of Physics, Southern Federal University).

To calibrate the velocity scale and record the spectra, a source of $^{57}\text{Co}(\text{Cr})$ with an activity of 370 MBq, an iron 57-enriched $\text{K}_4[^{57}\text{Fe}(\text{CN})_6]$ scintillation resonance detector, and a set of iron absorbers ($\alpha\text{-Fe}$) with a thickness of 5–30 μm were used.

The obtained data were processed using the MossFit 3.2 and MossFit 3.6 programs developed in the NGR spectroscopy laboratory, Department of Analytical Chemistry, Faculty of Chemistry, St. Petersburg State University [14].

X-ray diffraction (XRD) analysis of tantalum samples before and after irradiation was performed on a BRUKER D2 PHASER diffractometer. X-ray tube voltage was 30 kV, and current was 10 mA. CuK_α -radiation was used (note that the total absorption layer of CuK_α -radiation with an energy of 8.05 keV in metallic tantalum does not exceed 8 μm). Spectra were recorded in the range of 2θ angles from 39° to 103° in scanning mode with a step of 0.02° and a speed of 0.5° per min.

The DIFFRAC.EVA.V5.0 program and the PDF-2 ICDD card files [15] were used to process the results.

EXPERIMENTAL RESULTS

The Mössbauer spectra before and after irradiation represent a superposition of two single lines (Table 2, Fig. 1), corresponding to different positions of ^{57}Co atoms in the bcc lattice of tantalum: singlet 1, ^{57}Co atoms in the crystal lattice nodes (isomeric shift characteristic of interstitial atoms) and singlet 2, ^{57}Co atoms at the grain boundaries of crystallites.

The crystallographic position of the impurity atom in this case is determined by the size effect:

Table 2. Parameters of emission Mössbauer spectra of proton-irradiated tantalum targets

Spectral lines	Before irradiation		Foil 1 (6.95/5.24 MeV)		Foil 2 (5.08/2.86 MeV)	
	I.S.*, mm/s	Γ^{**} , mm/s	I.S., mm/s	Γ , mm/s	I.S., mm/s	Γ , mm/s
Singlet 1	−0.06	0.40	−0.06	0.53	−0.05	0.54
Singlet 2	−0.64	0.34	−0.64	0.36	−0.65	0.35

* Isomer shifts, I.S. (± 0.02 mm/s), are given relative to $\alpha\text{-Fe}$.

** Experimental line widths Γ (± 0.02 mm/s).

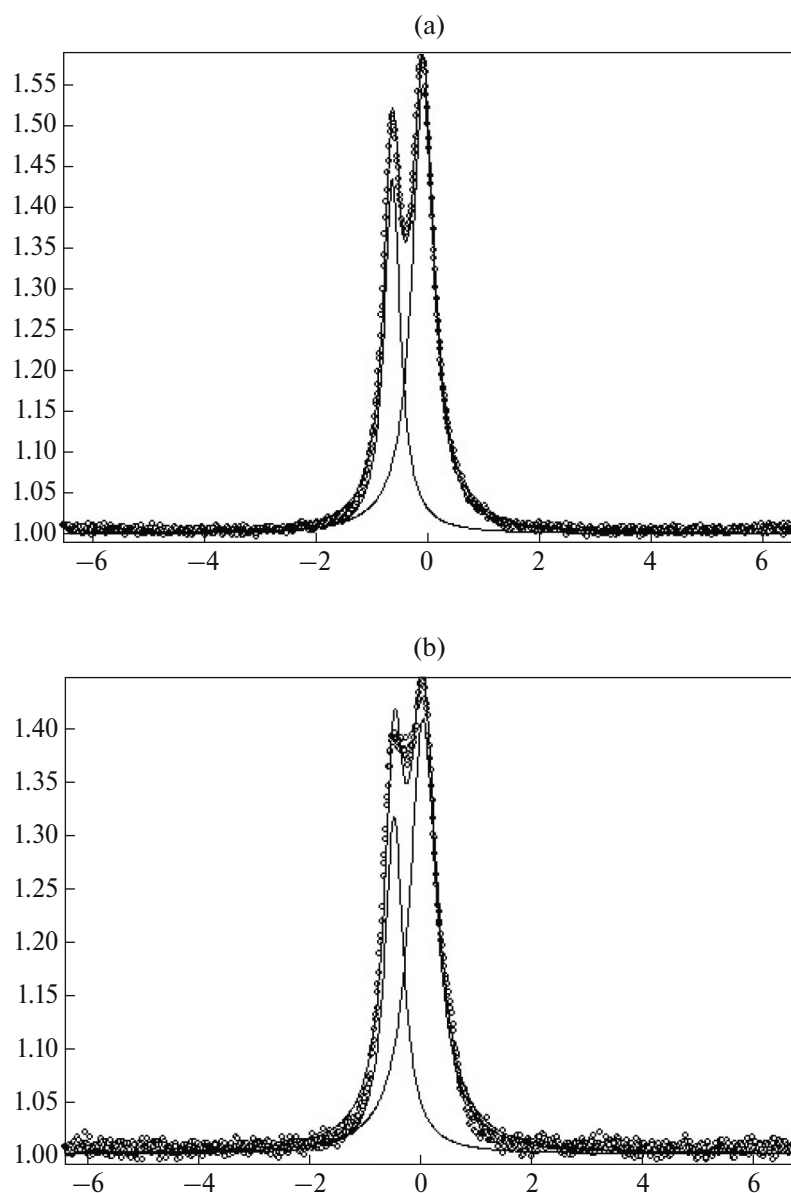


Fig. 1. Emission Mössbauer spectrum of ^{57}Co -labeled tantalum foils before (a) and after (b) irradiation.

(1) The atomic radius of cobalt (0.125 nm) is smaller than the atomic radius of tantalum (0.146 nm [16]).

(2) The radii of the octahedral and tetrahedral pores in the close-packed body-centered cubic lattice of metallic tantalum (0.060 nm and 0.033 nm, respectively [17]) are much smaller than both the atomic and ionic radii of Co/Fe: $\text{Co}^{2+}/\text{Fe}^{2+}$ 0.078/0.080 nm and $\text{Co}^{3+}/\text{Fe}^{3+}$ 0.064/0.067 nm.

(3) Therefore, impurity Co/Fe atoms can be found either at the nodes or at the grain boundaries of crystallites.

As mentioned above, the thickness of the surface layer of tantalum doped with Co^{57} atoms does not exceed 0.5 μm . This layer is so thin that protons practi-

cally do not create any direct radiation damage in it (the number of displaced atoms is 0.1 vac/ion, Fig. 2 [18]).

However, the products of nuclear reactions (^{181}W , ^{177}Hf , ^{178}Hf , ^{180}Ta) and Ta self-atoms knocked out of the lattice sites by protons (Tables 3, 4) create new cascades of defects and the number of displaced lattice atoms can reach 3900 vac/ion (Fig. 3 [18]).

The presence of a significant number of various point and bulk radiation defects leads to a broadening of the experimental lines for interstitial atoms, which are most sensitive to the local environment, by 25% (Table 2).

Low integral temperature of the targets during irradiation (310 K, Table 1) and a fairly high thermal conductivity of tantalum (57.6 $\text{W m}^{-1} \text{K}^{-1}$ at 293–1800 K

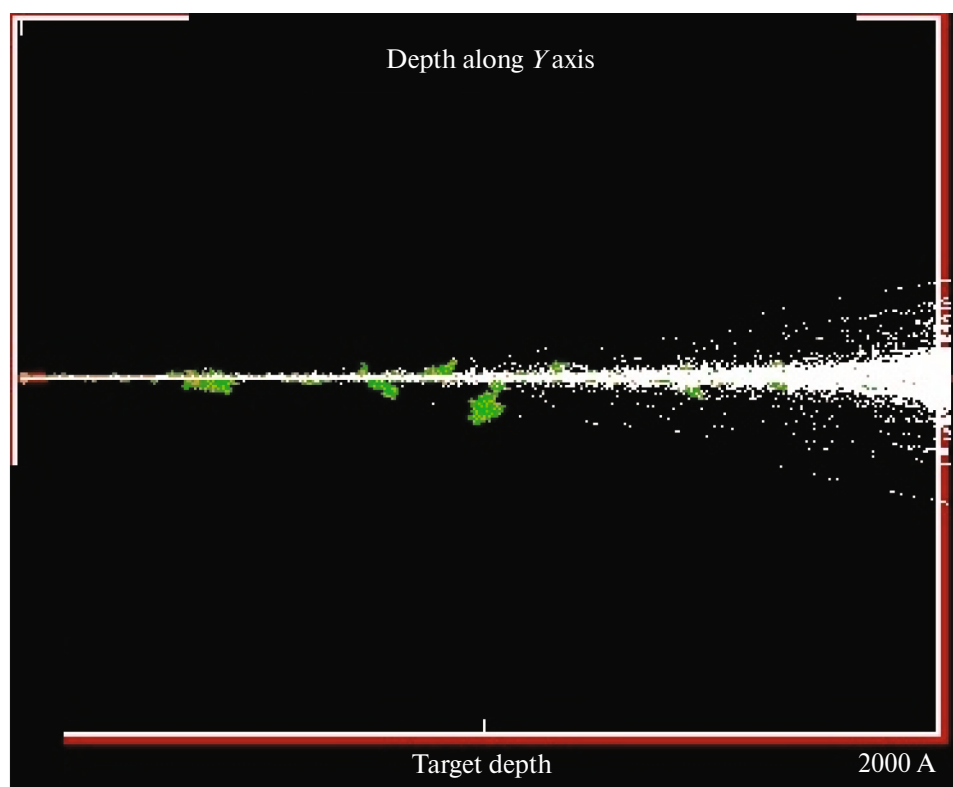


Fig. 2. Radiation damage zone of the metal surface layer doped with ^{57}Co atoms formed upon target bombardment with protons with an initial energy of 6.95 MeV.

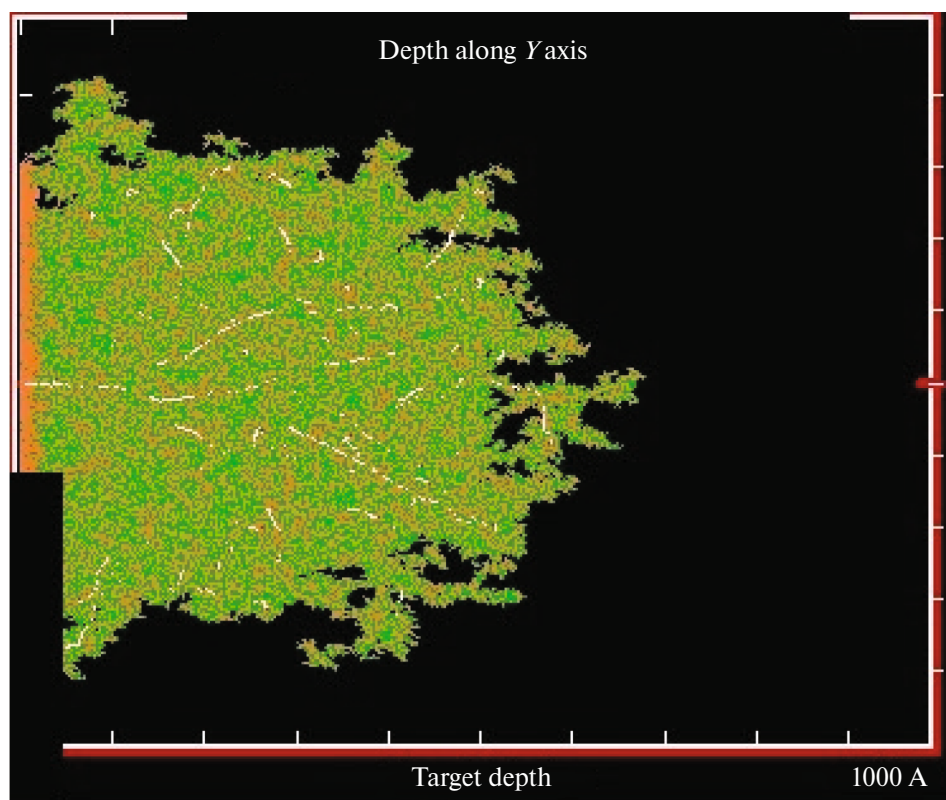


Fig. 3. Radiation damage zone formed upon target bombardment by tantalum atoms with an energy of 150 keV knocked out from crystal lattice sites by protons.

Table 3. Nuclear reactions occurring during irradiation of tantalum targets with 6.95 MeV protons

Nuclear reaction (^{181}Ta , 99.99%)	Reaction threshold, MeV	Reaction product (half- life, gamma-ray energy)
$^{181}\text{Ta}(p,n)$	0.993	^{181}W (121.2 days, 136.3 keV)
$^{181}\text{Ta}(p,2n)$	7.699	^{180}W (stab.)
$^{181}\text{Ta}(p,d)$	5.382	^{180}Ta (8.154 h, 93.3 keV)
$^{181}\text{Ta}(p,^4\text{He})$	0	^{178}Hf (stab.)
$^{181}\text{Ta}(p,^4\text{He} + n)$	0	^{177}Hf (stab.)

[19]) exclude the possibility of migration of impurity atoms in the volume of samples; therefore the ratio of lines in the spectrum is maintained: the proportions of singlet 1 and singlet 2 are (65 ± 2) and $(35 \pm 2)\%$, respectively.

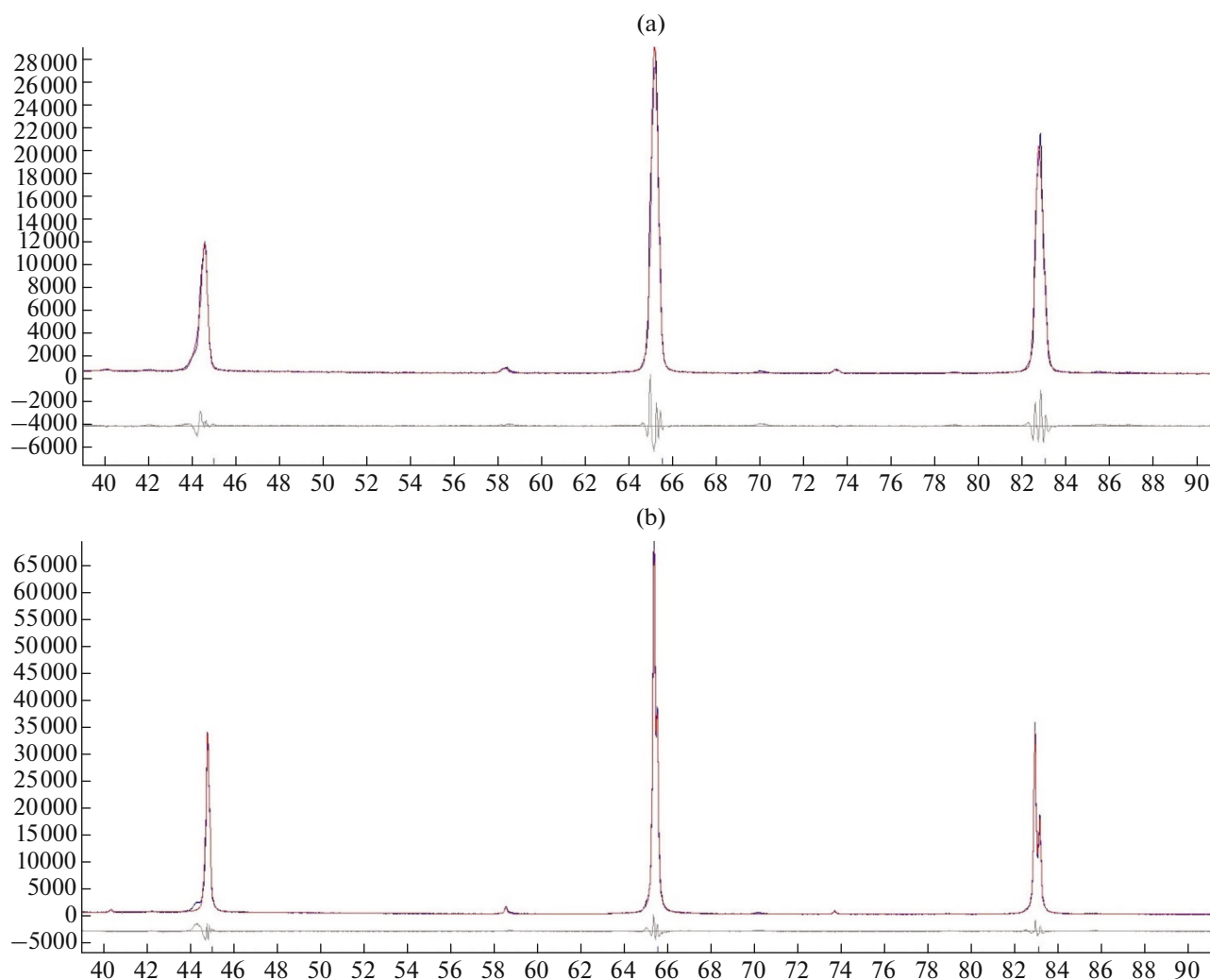
In the diffraction patterns of the samples after irradiation (Fig. 4), splitting of the main diffraction lines of the X-ray spectrum is clearly visible, which can only

Table 4. Maximum energies of nuclear reaction products and Ta self-atoms knocked out lattice sites by protons

Initial energy of protons, MeV	6.95	5.08
Maximum energy of nuclear reaction products, keV	38.2	27.9
Maximum energy of Ta self-atoms knocked out lattice sites by protons, keV	151.9	111.0

be explained by the presence of interfacial stresses [20–22] arising as a result of the cooling of displacement spikes [23] within zones disordered as a result of irradiation by products of nuclear reactions and tantalum atoms knocked out of the lattice sites.

With an increase in the scanning angle, the contribution of the damaged area to the total diffraction pattern increases, which is accompanied by a change in the intensities of the X-ray spectrum lines and an enhancement of the fine spectral structure.

**Fig. 4.** X-ray diffraction patterns of ^{57}Co -labeled tantalum foils before (a) and after (b) irradiation.

CONCLUSIONS

The post-effects of irradiation of metallic tantalum with proton beams with an initial energy of 7 MeV, products of nuclear reactions (^{181}W , ^{177}Hf , ^{178}Hf , ^{180}Ta), and Ta self-atoms knocked out of the lattice sites during bombardment of the targets were studied.

Minor layer of total absorption of $\text{CuK}\alpha$ -radiation (8.05 keV) in metallic tantalum (about 8 μm) and even smaller thickness (less than 0.5 μm) surface layers of the samples doped with ^{57}Co atoms made it possible to exclude from consideration direct radiation damage to the crystal lattice by proton beams and to study the consequences of irradiation of the metal by products of nuclear reactions and Ta self-atoms knocked out of the lattice sites by protons.

Irradiation of metal with heavy ions is accompanied by disordering of the crystal lattice and the formation of displacement wedges, the cooling of which leads to the splitting of the main lines of the X-ray diffraction spectra.

The presence of a significant number of various point and bulk radiation defects leads to a broadening of the experimental lines of the Mössbauer spectra for interstitial atoms, which are most sensitive to the local environment, by 25%.

The use of Mössbauer resonance atoms as structure-sensitive physicochemical probes may prove promising in the study of radiation effects arising in the surface layers of structural materials when irradiated with fission products, recoil nuclei, and heavy charged particles.

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AUTHOR CONTRIBUTION

I.E. Alekseev: preparation of ^{57}Co -labeled samples for irradiation, Mössbauer studies of the samples, processing of the measurement results, and writing the manuscript.
V.I. Zherebchevsky: irradiation of ^{57}Co -labeled samples.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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