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Changes in the temperature of polymorphic transformations under the action of radiation

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ABSTRACT

Studies on transport processes of ^{46}Sc, ^{57}Co and ^{188}Re impurity atoms in irradiated and non-irradiated samples of metallic titanium, iron and tungsten have shown that the temperature of polymorphic transition depends on the degree of radiation damage of the metal and is "moved" to the region of higher temperatures due to the presence of large number of structural radiation defects that make the restructuring of the crystal lattice difficult.

1. Introduction

In the course of numerous studies it has been shown that radiation exposure, which may cause deep and sometimes irreversible structural changes, will determine some physical-and-chemical properties of the irradiated solid, such as electroconductivity, thermal conduction, mechanical strength and reactivity; it will also substantially affect phase transformations and diffusion processes in radioactive materials (Freeman and Dienes, 1955; Spitsyn and Gromov, 1973; Gorbunov and Seleznev, 2001). In the work presented, these facts have been confirmed even further in the studies on transport processes of ^{46}Sc, ^{57}Co, and ^{188}Re impurity-atoms in the region of polymorphic transformation in irradiated and non-irradiated samples of metallic titanium, iron and tungsten.

2. Experimental methods

2.1. Preparation of targets

The initial samples were (a) thin metal foils (20–90 μm) of titanium and iron (natural isotope composition of a high chemical purity, 99.99%), which were obtained by rolling; prior to irradiation, the samples were subjected to recrystallization annealing (1330 ± 10 K, $\sim 10^{-7}$ Torr, 4 h) to remove structural deformation stresses; and (b) finely-dispersed powders (the grain size of from 1 to 3 μm) of metal isotopic tungsten (^{186}W, 99.78%).

2.2. Irradiation techniques

Irradiation of titanium and iron foils was carried out in an injected beam of an U-120 accelerator of the St. Petersburg State University (SPbSU) with the initial energy of deutrons, 13.5 MeV.

The energy, intensity of deuteron flux and activation time for the samples varied as follows:

(a) for titanium targets: $E_d = 13.3/11.5$ MeV, deuteron currents = 2 and 5.3 μA, fluence = 1.5×10^{20} m^{-2} and 3.1×10^{21} m^{-2}, heat dissipated within the target – 3.6 and 9.54 Wt, respectively; the production of ^{46}Sc by reactions ^{46}Ti(d, 2p), $E_{threshold}$ = 3.98 MeV; ^{48}Ti(d, ^{4}He), $E_{threshold}$ = 0 MeV; ^{49}Ti(d, ^{4}He+n), $E_{threshold}$ = 4.33 MeV; and
(b) for iron targets, $E_d = 7.3/5.3$ MeV; fluence = 1.0×10^{22} m^{-2}, beam current = 10 μA; heat dissipated within the target – 20 Wt; the production of ^{57}Co via the ^{56}Fe(d, n) channel, $E_{threshold}$ = 0 MeV.

To reduce the energy of deuterons down to optimal level, one needed to recover the structurally sensitive Mössbauer ^{57}Co nuclei (National Nuclear Data Center), a tantalum ~100 μm thick filter was used (such irradiation technique enabled the yield of ^{56}Co to be substantially reduced ($T_{1/2}$, 78.76 days, nuclear reaction, ^{56}Fe(d, 2n), $E_{threshold}$, 7.85 MeV) – see Fig. 1).

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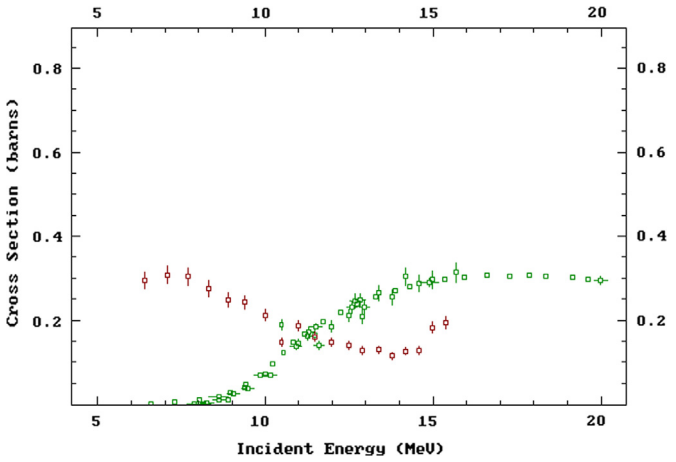

Fig. 1. Cross section of nuclear reactions (National Nuclear Data Center,): ^{56}Fe(d, n)^{57}Co – brown points, and ^{56}Fe(d, 2n)^{56}Co – green points. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

For dissipation of heat generated during irradiation, the samples under study were mounted by a tight mechanical hold-down, on a massive water-cooled supporting aluminum substrate that could withstand a pressure gradient of several atmospheres (the maximum pressure of cooling water that acts on the substrate was 4 atm).

In order to determine the integral current and total beam flux of charged particles through the surface of targets, Al or Cu beam position monitors were used, being placed along the path of the charged-particle beam in front of the targets.

Irradiation of tungsten targets (the production of ^{188}Re: ^{186}W(n, γ)^{187}W(n, γ)^{188}W→β$^-$-decay) was carried out in the WWR-M reactor of the St. Petersburg Institute of Nuclear Physics named after B.P. Konstantinov (the Russian Academy of Science), in quartz ampoules that were evacuated down to 10^{-3} Torr; the flux of thermal neutrons being 3.1×10^{14} neutron cm^{-2} s^{-1}; fluence= 8×10^{23} m^{-2}; target temperature during irradiation – 333–343 K; and the specific radioactivity of ^{188}W as at the end of irradiation being equal to 0.04 Ci/g.

2.3. Experimental methods used to study products of nuclear reactions and irradiated targets

In order to study products of nuclear reactions, the radionuclide composition of irradiated targets and to determine the concentration of "hot" impurity-atoms and their diffusion coefficients, gamma-ray spectrometry has been used with semiconductor detectors (an HPGe detectors GX1018 and GUL0035 manufactured by Canberra Industries Inc., U.S.A.). Analysis of spectra and data processing were done using the Genie-2000 peak-analysis software (developed by Canberra Industries Inc., U.S.A.).

To measure the diffusion coefficients of impurity ^{57}Co atoms in Fe metal we used the absorption method. The method consists in the following.

Irradiated foils (specifically those used in parallel for recording the Mossbauer spectra) were subjected to a series of isothermal annealings at 823–1423 K in quartz ampules evacuated to 10^{-4} mm Hg. After each annealing, from the X-ray peak at 6.4 keV (National Nuclear Data Center) (with the experimental geometry and recording efficiency of the detector, GUL0035, preserved), we determined the activity on both sides of the samples.

In the course of measurements, samples were contained in a special cell which was placed over the detector in a strictly fixed position. The ^{57}Co coefficients were calculated from the activity ratio (Boltaks, 1961)

$$\frac{I(t)}{I(0)}=1-\frac{2}{\sqrt{\pi}}\sqrt{Dt/h},$$

where $I(0)$ and $I(t)$ are the γ-radiation intensities on one of the sample sides before and after thermal annealing; h is the sample thickness; t is the annealing time; and D is the diffusion coefficient of the impurity.

To obtain the transport characteristics of ^{188}Re impurity atoms a special technique has been used to study transport processes, which is based upon a study of time dependence of gamma-radiation intensity of daughter atoms under conditions of a disturbed equilibrium in genetically related radioactive chains (Alekseev and Antropov, 2002).

The essence of this method is as follows. Samples under investigation, uniformly doped by an impurity owing to the decay of a parent nuclide (formed by a nuclear reaction, e.g., ^{188}Re: ^{186}W (n, γ)^{187}W(n, γ)^{188}W→β$^-$-decay), are subjected to diffusion annealing in the examined temperature interval. The annealing stimulating diffusion of an impurity to the phase boundary is followed by precision (with the experimental geometry and recording efficiency of the detector, GX1018, preserved) measurements of the time dependence of the radioactivity of the samples treated (in some cases) with a specially chosen reagent to remove impurity atoms that diffused to the surface. The diffusion coefficients D of impurities in the metal under consideration are calculated from the experimentally measured effect of the equilibrium perturbation.

The quantity D is determined from the equation for the diffusion in a finite-size body with uniform initial distribution of the diffusing substance (Boltaks, 1961)

$$Q=\frac{8}{\pi^2}N_0 l e^{-\frac{\pi^2 Dt}{l^2}},$$

where N_0=constant is the initial (at t=0) concentration of the diffusing substance; t is the diffusion time; l is the sample thickness; D is the diffusion coefficient of an impurity; and Q is the amount of the diffusing substance at given time.

It should be emphasized that such distribution is valid for a body that has been uniformly saturated with a diffusing substance which leaves the body through either evaporation or binding on a phase boundary.

In view of the formation of a number of radioactive products in the deuteron beam (most of the reactions under deuteron bombardment are either exothermic or have a low energy threshold), which makes it impossible to measure absorption spectra, Emission Mössbauer Spectroscopy (EMS) was used to observe the after-effects of Fe-target irradiation and post-irradiation thermal annealing.

Mössbauer spectra (characteristics of the ^{57}Fe Mössbauer transition (Barb, 1980) are given in Table 1) were measured with an electrodynamic apparatus that operated in a constant acceleration mode – Commercial Mossbauer Spectrometer CM 2201 (made in the Institute for Analytical Instrumentation of the Russian Academy of Sciences).

Table 1
Characteristics of the ^{57}Fe Mössbauer transition (Barb, 1980).

Parent isotope	$T_{1/2}$ (days)	Gamma-transition energy (keV)	Radiation yield (%)	Level lifetime (ns)	Natural line width (2Γ) (mm s^{-1})	ICC[a]
^{57}Co	271.74	14.4	9.16	98.3	0.194	8.18

[a] The internal conversion coefficient.