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Techniques for study of irradiated materials using the methods of positron spectroscopy

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Introduction

Nowadays, materials are subject to higher requirements in terms of reliability and durability. Whether it is reactor vessel steel or spacecraft solar cell plates, they all operate under extreme conditions of radiation and elevated temperatures [1]. When creating these materials, we must be sure that they will last their service life without failing. Because of this, it is necessary to conduct laboratory tests to investigate changes in structure and properties under irradiation and elevated temperatures [1]. Positron Annihilation Spectroscopy (PAS) can be a reliable and non-destructive method of material control [1]. PAS can also be used as a versatile tool to study the microscopic texture of materials. Using PAS, detailed experimental information about the electronic and atomic structure is obtained from volume regions [1].

Types of radiation

In physics, radiation is the transfer of energy in the form of waves or particles through space or through a material medium. Ionizing radiation is radiation, the interaction of which with the medium leads to the formation of ions of different signs. Such properties are possessed by radioactive radiation, high energy radiation, X-rays, etc.
There are several types of radioactive radiation:
- electromagnetic radiation - X-rays and gamma rays (γ);
- particle radiation - alpha-radiation (α), beta-radiation (β), neutron radiation;

Alpha radiation

Alpha radiation is a flux of alpha particles. An alpha particle, structurally equivalent to the nucleus of a helium atom, consists of two protons and two neutrons. Alpha radiation occurs when the nucleus of an atom becomes unstable (the ratio of neutrons to protons is too low) and alpha particles are emitted to restore the balance. This decay occurs in elements with high atomic numbers, such as uranium, radium and thorium. During nuclear decay, the energy released (decay energy) is divided between the daughter nucleus and the alpha particle [2]. The two neutrons of the alpha particle give it additional mass, which further contributes to ionization through coulomb interaction or even direct collisions of the alpha particle with the electrons of the atom [2]. During collisions mainly through ionization and electronic excitation, alpha particles, like other types of charged particles, dissipate their energy. Alpha particles are very energetic, but they are so heavy that they expend their energy over short distances and cannot travel very far from the atom [1]. The greater mass and charge of the alpha particle compared to other forms of nuclear radiation gives it a greater ionizing ability, but less ability to penetrate matter [2]. Therefore, the methods of protection from alpha particles are quite numerous and do not require large resources. In many cases, even rubber gloves are sufficient.

Beta radiation

Beta radiation arises as a result of β^- decay, which has two main variants:
In the case of β^- decay, the neutron n turns into a proton p with the emission of β^--particles (e^-) and antineutrinos $\bar{\nu}$ [3]. This is described by the reaction equation,

$$n \rightarrow p + e^- + \bar{\nu}.$$

In β^+ decay, the proton decays into a neutron releasing a positron e^+ (the antimatter equivalent of an electron) and a neutrino ν,

$$p \rightarrow n + e^+ + \nu$$

that can only occur within the nucleus with the energy available from the decay transition [3].
β-decay is due to the weak interaction and this is its feature. This process is not intranuclear, it is intranucleon. A single nucleon decays in the nucleus [4].

For radiation protection purposes, beta particles are considered more ionizing than gamma rays, but less ionizing than alpha particles. The higher the ionizing effect, the greater the damage to living tissues, but also the lower the penetrating power of radiation. Typical beta emitters are strontium-90, technetium-99, cesium-137, carbon-14, sulfur-35 and tritium. As an example of β^+ sources, we could point to 22Na, as well as the rapidly decaying isotopes used in PET scanners 11C, 13N, 150, 18F. Beta radiation is more penetrating than alpha radiation, but it still has a hard time penetrating our clothes. It has enough energy to enter our skin, but not enough to pass through it. Thus, unlike alpha particles, beta particles can penetrate a sheet of paper, but are easily stopped by a thin sheet of plexiglass or aluminum.

Gamma radiation

Gamma radiation is electromagnetic radiation that is emitted by an unstable nucleus of an atom during radioactive decay. A nucleus in an unstable state may fall to a more stable state by the emission of energy as gamma radiation [5].

This phenomenon consists in the fact that the nucleus emits a γ-quantum without changing the mass number A and the charge of the nucleus Z. Gamma radiation occurs during the decay of the excited state of the nucleus [6].

The decay processes of radionuclides often leave the product nuclide in an excited energy state. The product nuclide in such an excited state either falls directly into the ground state or descends stepwise to lower energy states due to energy dissipation in the form of gamma radiation [5]. A nuclear isomer is a nuclide in an excited energy state, and the transition (or decay) from a higher energy state to a lower energy state is called an isomeric transition. Gamma rays are emitted at discrete energies corresponding to the energy state transitions that a nuclide in an excited state may undergo [5]. An isomeric transition, as described earlier, is a decay process in which gamma radiation is the only energy release process from an excited nucleus [5]. This mode of decay is called an isomeric transition because neither the mass number A nor the atomic number Z of the nuclide ($^A_Z X$) change during the decay process, and the nuclides are considered to be in isomeric energy states [5].

Different energies and types of emitted elementary particles have different penetrating power and, accordingly, have different effects on living organisms [7]. Gamma rays are very penetrating and can penetrate anything less dense than a thick sheet of steel plate [7].

There are many varieties of interaction of photon radiation with the material environment. It can be talked about three main types of such interaction [8]:

- Photo effect - the energy of a gamma-quantum is absorbed by an electron in the shell of an atom, and the electron, doing the work function, leaves the atom (becomes positively ionized).
- Compton effect - a gamma-quantum is scattered when interacting with an electron, while a new gamma-quantum of lower energy is formed, which is also accompanied by the release of an electron and ionization of an atom.
- The effect of pair formation - gamma-quantum in the electric field of the nucleus turns into an electron and a positron.

X-rays

X-ray radiation is electromagnetic radiation with a wavelength of 10^{-12}–10^{-8} m and, accordingly, a photon energy of 10^{2}–10^{6} eV, resulting from the accelerated movement of electrons [9]. The ionization of atoms of matter is the main result of the interaction of X-ray radiation with matter. X-ray radiation can ionize not only valence electrons, but also electrons in the inner shells of atoms [10]. The high-energy excitations created in this way decay in times less than 10-13 s into low-energy ones, which are excitations of valence electrons [10]. The absorption coefficient of X-ray radiation is low, depending on the energy of the quanta and the chemical composition of the material. The depth of passage can vary from fractions of millimeters to tens of centimeters through a solid body with increasing photon energy. The penetrating power of x-rays depends on the wavelength and power of the radiation. The greater the energy and the shorter the wavelength, the deeper the rays penetrate into the substance [11].

X-rays are used to elucidate the structure of substances at the atomic level by means of X-ray diffraction scattering (X-ray diffraction analysis). Moreover , with the help of X-rays, the chemical composition can be determined. A non-destructive research method - X-ray spectral analysis allows you to determine the chemical composition of a substance, since all chemical elements have a strictly defined spectrum of emission lines of characteristic radiation. This technology helps to detect cracks, defects, foreign inclusions in cast products.

Neutron radiation

Nuclear radiation, consisting of streams of particles with a neutral charge (neutrons) is called neutron radiation. Occurs during nuclear reactions (in nuclear reactors, industrial and laboratory facilities, during nuclear explosions). A free neutron is an unstable, electrically neutral particle with a lifetime of about 15 minutes. The penetrating power of neutrons is very high due to the absence of charge and, as a result, weak interaction with matter. The penetrating power of neutrons depends on the energy and composition of the atoms of the substance with which they interact. The layer of half attenuation of light materials for neutron radiation is several times less than for heavy ones. Heavy materials weaken neutron radiation worse than gamma radiation. Neutrons, depending on the kinetic energy, are divided into fast (up to 10 MeV), ultrafast, intermediate, slow and thermal.
The main properties of neutrons used in neutron scattering:

- The energy of slow neutrons is comparable to the energy of atomic and molecular motions, and is in the range from meV to eV.
- The wavelength of slow neutrons is comparable to interatomic distances, which makes it possible to study the structure of matter in the range of 10-5 – 105 Å.
- Since neutrons are neutral particles, they interact with the nuclei of atoms, and not with diffuse electron shells. The neutron scattering cross section for nuclei of similar masses can differ significantly, which makes it possible to "see" light nuclei against the background of heavy ones, effectively apply the isotopic substitution method, and easily distinguish neighboring elements. This feature is a great advantage over the X-ray scattering method, in which radiation is scattered on the electron shell of atoms.
- The presence of a magnetic moment in neutrons makes it possible to study the microscopic magnetic structure and magnetic fluctuations, which determine the macroscopic parameters of matter.
- Neutron radiation is deeply penetrating deep into the substance, which makes it possible to study microscopic properties, such as microcracks, industrial objects. Such studies cannot be performed using optical methods, X-ray scattering, or electron microscopy.

The main difference between neutron radiation and X-ray radiation is that neutron scattering occurs on the nuclei of atoms. Therefore, there is no need to take into account the atomic form factor to describe the shape of the electron cloud of an atom; in addition, the scattering power of an atom does not decrease with an increase in the scattering angle, which is observed for X-ray scattering. X-ray scattering is practically insensitive to the presence of hydrogen atoms in the structure. But the nuclei of hydrogen and deuterium are strong scatterers for neutron radiation. Therefore, with the help of neutrons, it is possible to more accurately determine the position of hydrogen and its thermal vibrations in the crystal structure.

Radiation Resistant Materials

Under the action of irradiation, materials undergo structural transformations that lead to changes in properties in operation. Neutron irradiation has a strong effect. The influence of irradiation with α-particles, protons, β-particles and γ-radiation is weaker. In view of this, materials operated under irradiation conditions must be radiation resistant.

Radiation resistance is the stability of the structure and properties under irradiation [12]. Structural changes from irradiation have the greatest influence on mechanical properties and corrosion resistance [12].

Effect of irradiation on the structure

Irradiation leads to the formation of point and line defects, micropores and other structural damage to the material [12]. During irradiation, the atoms of the irradiated material are displaced into interstitials and vacancies are formed [12]. The density of point defects increases. One neutron particle, which has less energy than an α-particle and a proton, creates incomparably more structural damage [12].

When irradiated with high fluxes, neutrons not only displace material atoms into interstitials, but also excite them, transferring part of their energy. Excitation intensifies vibrations of the atom and its

neighbors at the lattice sites, which is accompanied by a local increase in temperature in a small volume of the crystal [12]. Vacancies and interstitial atoms interact - annihilate, which reduces the concentration of defects [12]. Under irradiation, the long-term strength always decreases, especially in aging alloys.

Metals such as tungsten are highly resistant to ion path formation, so material damage caused by heavy ion irradiation will be low [13]. Neutron damage can lead to the deterioration of mechanical properties and the appearance of structural defects, in some cases to a decrease in thermal conductivity [13]. Moreover, high energy neutrons can produce transmutation elements and cause bias damage during fusion reactor operation. Used for protective lining, the materials determine the durability of the remaining parts of the object that are directly exposed to radiation [13]. As a result, it is essential to study changes in the material structure after exposure to high-energy ions and neutrons. Tungsten is a promising material for use as a plasma lining material in nuclear fusion reactors due to its low sputtering, high melting point, and high thermal conductivity [13]. But, the brittleness and other well-documented disadvantages of pure tungsten have reduced its use in many applications, prompting researchers to explore tungsten-based alloys with better ductility. One of the promising materials is tungsten carbide (WC), which, in addition to well-studied advantages in terms of thermophysical properties, also has a combination of suitable nuclear properties (large thermal absorption cross section, high atomic packing, etc.) [13]. To find information about defects and voids in solids, there is a method of positron annihilation spectroscopy.

Positron annihilation spectroscopy

Positron annihilation spectroscopy is a powerful non-destructive characterization technique that can provide information on the defect structures of crystalline solids [14]. The positron interacts with the solid under study, losing energy due to in elastic scattering, ionization, and excitation of electrons. Deceleration to thermal energy takes a few picoseconds. A thermalized positron diffuses in a delocalized state for several hundred picoseconds until it eventually annihilates with an electron [14]. In places of vacancy defects, the electron density and electrostatic repulsion of the missing positive ions are lower than in defect-free regions of the lattice [14]. This means a higher probability of positron capture at the defect site and a longer lifetime compared to the lifetime of a positron in a defect-free lattice. Then, the measurement of the positron lifetime gives an idea of the concentration and type of vacancy defects. This measurement underlies the method of positron annihilation lifetime spectroscopy (PALS) [14].Three methods are commonly used in PAS measurements to characterize materials: positron lifetime spectroscopy, Doppler broadening analysis, and angular correlation measurements. Positron lifetime measurements provide information on the electron concentration at the annihilation site. On the other hand, measurements of Doppler broadening and angular correlation provide information about the electron momentum distribution [15]. Thus, the electronic configuration of the studied material is reflected in the parameters of positron annihilation [15].

PAS methods are complemented by similar methods used for material structure analysis (Raman, XRD, ND). In the same comprehensive study of WC + 6% Co irradiated with 167 MeV Xe, it was noted that the concentration of point defects in tungsten carbide grains decreased with dose, while the number of carbon grain boundaries on the surface increased [13]. PAS leads to the fact that the intensities of various components of the positron lifetime change nonmonotonically with the

irradiation dose, which may also indicate a rearrangement of the radiation type of the defect. The same result is confirmed by XRD and ND. We can say that the reorganization of defects occurs in planes formed by atoms of the same type, mainly in the carbon sublattice.

Experimental part

The two most frequently used techniques to detect and to investigate the properties of open volume defects and their interaction with the medium are the lifetime (PLT) and the Doppler broadening techniques (DPS). If fast positrons from a radioactive nuclide are used, the mean properties of the material under investigation are measured in a layer several tens of micron thick. Otherwise, if a slow positron beam is utilized, a depth scanning from the surface to a few microns is possible. The momentum of a thermalized positrons is a lot smaller than the momentum of electrons in a crystalline lattice. Therefore, the momentum seen in the Doppler broadening of the annihilation photons is that of an electron at the annihilation location.

PALS

Positron lifetimes were measured for 2 tungsten samples (99.95) using PLT. Following the logic that under natural operating conditions the materials would have many mechanical and structural defects from the very beginning of their operation, we deliberately left the samples without annealing. We used an initial sample and a sample irradiated with gamma (average value for the energy 1.25 MeV, intensity 171 Rad/s) for 30 days.

A positron lifetime of 195±2 ps was obtained for the starting sample. The sample after irradiation shows a positron lifetime of 190±1 ps. These results can be interpreted as the ability of the material to anneal with the help of radiation in local areas, where there is the presence of kinetic energy released after the capture of gamma quanta by the atoms of the material and the subsequent secondary ionization.

DPS

When a positron annihilates with an electron, two γ photons are created with the energies of $E = 511KeV \pm \Delta E$, where ΔE corresponds to the Doppler shift caused by the momentum of the annihilating electron-positron pair. These shifts are recorded experimentally with a high-purity germanium detector of a high energy resolution, and the shifts cause the characteristic broadening of the 511 keV annihilation peak.

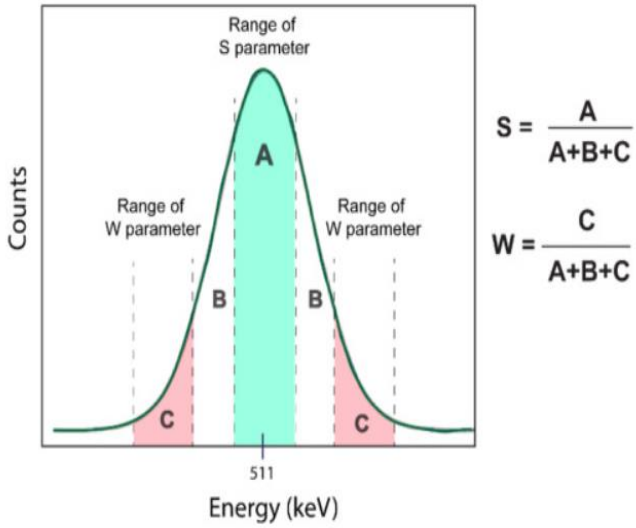

Fig. 1. *Definition of defect parameters S and W extracted from the 511 keV annihilation peak. A typical value for the windows are: from 510.2 to 511.8 keV for region A. For the wings C, from*

507.8 to 509.3 keV and from 512.7 to 514.8 keV.[16].

The shape of the Doppler-broadened 511 keV annihilation line is generally characterized by dividing it into two types of regions, as in Fig 1. The central region describes the count rate of the low momentum electrons, and the wing regions, taken from the peak edges, describe the count rate of the high momentum electrons. The peak shape is conveniently described by using the S and W parameters. The S parameter is defined as the counts in the central region divided by the total number of counts in the annihilation peak and the W parameter is defined accordingly as counts in the wing regions divided by the total number of counts in the annihilation peak. The S and W parameters are very sensitive to changes in the electron momentum distribution at the annihilation site. So, increasing the S parameter is evidence of increasing the number of defects.

Using DPS, a measurement was made at different values of the applied voltage for 4 different samples (W, WC, WC-6Co, WC-10Co), each of which was irradiated with 2 gamma doses (Fig. 2).

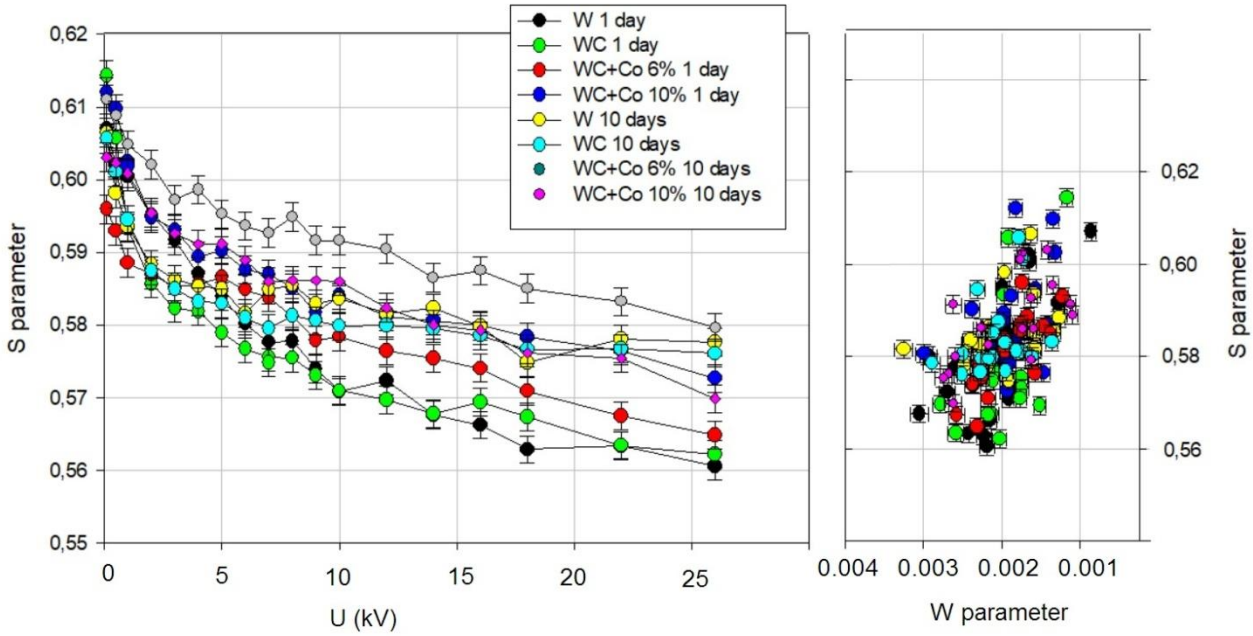

Fig. 2 *Shows the results of 4 different samples, each irradiated, at 2 different gamma doses - for 1 day and for 10 days.*

By increasing the voltage, one can analyze how the annihilation of the positron changes depending on the depth at which it takes place. For pure tungsten, with increasing gamma dose, an increase in annihilation processes in defects with depth is observed. If we compare tungsten with tungsten carbide in a dose of 1 day, we can conclude that the presence of surface defects in tungsten is significantly more than in tungsten carbide. This can ensure better strength properties for tungsten carbide. The one-day gamma dose picture for WC and WC-6Co is very close. From which it should be argued that with respect to gamma irradiation the two materials have a similar behavior. The surface defect situation at 1 day dose for pure tungsten and WC-10Co is also close. And this again determines close properties of the two materials. From the course of the graphs, comparing them

between the doses for 1 day and for 10 days, it can be concluded that there is actually a process resembling local annealing during gamma irradiation. Looking at the general course of the values in the S/W analysis, we can determine that it is precisely because of the restructuring of the defects from the surface to the volume.

Conclusions

- Defects in depth increase with increasing gamma dose.
- The radiation shielding properties of WC and WC-6Co are better than those of W and WC-10Co.
- By means of PLT, the possibility of radiation materials receiving natural relaxation under the action of intense gamma irradiation was examined. The same result it was confirmed by DPS.

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