ABSTRACT

of the dissertation paper under the title:

"SIMULATION OF HIGH-DOSE RADIATION DAMAGES OF STRUCTURAL REACTOR MATERIALS BY PROBE MESSBAUER ATOMS",

submitted for the degree of Doctor of Philosophy (PhD) in the specialty 6D071000 - "Materials Science and Technology of New Materials"

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Assessment of the current state of the fulfilled scientific or scientific-technological tasks

Currently, the modernization is taking place in the nuclear power and the transition from the slow neutron reactors to the fast neutron reactors, with the damaging doses of more than 400 displacements per atom (dpa). In this regard, it is required to develop the perspective reactor materials with high reliability, and to replace the traditional ferritic-martensitic steels by the steels of austenitic grade. Such steels are characterized by low induced radioactivity, weak vacancy gas swelling, high resistance to creeping and high temperature embrittlement.

The gradual transition to nuclear and thermonuclear power engineering leads to the fact that development of the advanced reactor materials, resistant to radiation destruction, control of their structure and properties is the important scientific and technical task of the Radiation Materials science. The complexity of such works is associated with the induced radioactivity of the materials, spent in the reactor and the long-term accumulation of the damaging radiation dose (several years). Its successful solution requires a fundamentally new approach to the methodological support of the experiments with irradiated materials, characterized by the lack of such drawbacks.

The priority position in the development of advanced reactor materials is occupied by the method of imitation irradiation with the ions of heavy metals, which reduces the duration of service life tests and provides the radiation safety of the experiments. The non-destructive method of irradiation with Mössbauer iron atoms is characterized by optimal capabilities. It simultaneously provides registration of the materials radiation damages, and their simulation (creation) at the level of the crystal lattice.

Basis and initial data for development of the topic

The practice of the advanced countries development shows that only nuclear and thermonuclear energy can provide the growing energy consumption. The issue of the NPP construction in the Republic of Kazakhstan is a matter of time and there are all prerequisites available for this. According to the IAEA data, the country's uranium reserves are estimated as 900 thousand tons. In addition, there is experience available in operating of the nuclear power plant with the fast neutron reactor, which has been operated in Aktau city for more than twenty years.

Justification of the demand for this research work

Increasing the power of the nuclear reactors with high-dose irradiation requires the development of reactor materials characterized by high operability and operational reliability. There are International Programs implemented which give the priority to chromium-nickel steels of the austenitic grade, capable of operating at the damage levels above 200 dpa in a wide range of operating temperatures. The need to intensify the accumulation of the damaging doses, reduce the duration of the service life tests and provide the safety of the scientific experiments is the main content of this research work. Taking into account the global trends in the nuclear energy development, it is reasonable for Kazakhstan to expand its own participation in the development of the perspective and improvement of the existing reactor materials.

Information about the planned scientific-technical level of development

The search and development of the effective and safe methods of irradiation, which create damage of the same type as in the decay of nuclear fuel, determines the planned level of scientific-technical development. In this respect, the simulation tests by irradiating materials in the accelerator with the high-energy flows of metal ions or inert gases are promising. The use of high-dose Mössbauer iron atoms in the work makes it possible to simultaneously fulfill two tasks - to simulate (create) the radiation damages and to analyze their type.

Information about metrological support of the dissertation thesis

The experimental studies during the works were carried out on the equipment and devices that comply with the "Law on Provision of Measurements Uniformity" and have passed state calibration. The measurement units, that correspond to the metrological rules and standards of the International Units System of the Measurement Instruments, were used in all cases.

The following instruments were used for research work: the Mössbauer spectrometer MS-110Em; the diffractometer D 8 ADVANCE of the BRUKER Company (Germany), the nanohardness tester "Nanoscan-compact" (Russia); the scanning electron microscope LYRA3 TESCAN (Czech Republic).

The used equipment and instruments are certified and have the documentary evidence issued by the relevant control organizations (Almaty branch of JSC "National Center of Expertise and Certification", the accreditation certificate No.KZ.P.02.0687 dated 04.05.15).

Relevance of the topic

The relevance of the topic is defined by the change in the energy policy of the world powers and a gradual transition to the nuclear and thermonuclear energy with higher damaging doses and more severe operating conditions for structural materials. The need of the conversion to the reactor materials of new generation requires a search for effective and safe irradiation methods that simulate the radiation damage of the same type as in nuclear decay. The simulation tests with the high-dose fluxes of heavy metal ions, produced in the accelerators of various types, can be a definite alternative.

Novelty of the topic

The development of the simulation technique for irradiation using ^{57}Fe Mössbauer atoms makes it possible to simulate the radiation damage in the reactor materials in the accelerator of heavy elements. This technique is capable to create the radiation damage of the same type as in the decay of nuclear fuel, to identify its type, as well as to exclude induced radioactivity, to reduce the time of the damaging doses accumulation, to determine the effect of radiation on the structural-phase state and properties of the irradiated materials. Selection and development of the simulation modes for obtaining of the objective information about radiation damages characterizes the novelty and scientific-practical significance of the research topic.

Relationship of this work with other research works

The dissertation paper was prepared at the RSE Institute of Nuclear Physics of the Republic of Kazakhstan in the Laboratory of Nuclear Gamma-Resonance Spectroscopy in the framework of the Scientific-Technical Program "Development of nuclear power in the Republic of Kazakhstan" (Grant No.0381/GF4) jointly with the M.V. Lomonosov Moscow State University (Department of Atomic Nuclear Physics, RINP).

Goal of research: to develop the simulation method for modeling of the radiation damages during irradiation of reactor materials in the charged particles accelerators with high-dose probe ^{57}Fe atoms.

Objects of research: the foils of molybdenum and tantalum, structural stainless steels of austenitic grades – 12Cr18Ni10Ti, AISI 304 and AISI 316.

Subject of research: the effect of high-dose irradiation with heavy ^{57}Fe ions on the type of radiation damages, the structural-phase state and properties of molybdenum, tantalum and real structural steels 12Cr18Ni10Ti, AISI 304 and AISI 316, used in the core of the nuclear reactor.

Tasks of research, their place in implementation of the research work in general

1) To prepare the pilot samples of pure metals and steels of three grades 12Cr18Ni10Ti, AISI 304 and AISI 316 in the form of thin foils and perform irradiation with ^{57}Fe iron ions at the energy of 1 MeV and the dose of $5x10^{16}$ ions/cm^2;
2) To obtain and calculate the main characteristics of the interaction of ^{57}Fe iron ions irradiation according to the SRIM-2008 software, to determine their structural-orientational arrangement in the crystal lattices of molybdenum and tantalum, to calculate the number of the formed defects;
3) To obtain and process the Mössbauer spectra of all steel grades, to perform the X-ray and electron diffraction studies, to process, analyze the obtained data and to make the justified conclusions;
4) To measure nanohardness in the initial and annealed state, to evaluate the effect of the state on the amount of radiation hardening of different types of steels, depending on the penetration depth of the indenter.

Fulfillment of the specified tasks will make it possible to work out the method of simulated irradiation with the high-dose flow of ^{57}Fe iron ions to simulate the radiation damages similar to those formed during the decay of nuclear fuel.

Methodological framework of research

For fulfillment of the task and achieving the specified goal, the following research methods and techniques were used:

– the high-dose irradiation with ^{57}Fe ions in the rechargeable accelerator of heavy ions UKP-2-1;

– the conversion Mössbauer spectroscopy in the electronic channel (CEMS) and in the absorption mode (MS);

– the computer package of the STRIM–2008 software;

– the computer package of the SpectrRelax software;

– the EXAFS – spectroscopy.

Provisions for Defense

– The results of modeling the interaction of implanted iron ions (projective range, number of vacancies/ion, number of displacements per atom) using the SRIM-2008 software and their dependence on the properties of molybdenum and tantalum;

– Structure of the Mössbauer spectra of conversion electrons (CEMS) and MS for transmission on ^{57}Fe nuclei, the results of X-ray diffractometry and EXAFS spectroscopy to determine the state and defectiveness of molybdenum and tantalum matrices;

– Structure of the Mössbauer spectrum of conversion electrons (CEMS) on ^{57}Fe nuclei in stainless austenitic industrial steels 12Cr18Ni10Ti, AISI 316 and AISI 304, depending on the nickel content;

– The results of X-ray diffractometry and scanning electron microscopy of 12Cr18Ni10Ti, AISI 316 and AISI 304 steels in various states - before, after irradiation and after annealing, the formation of "stress martensite";

– The results of electron microscopy of the near-surface layers and measurements of nanohardness for determining the value of radiation hardening of 12Cr18Ni10Ti, AISI 316 and AISI 304 steels, which are in different states, depending on the penetration depth of the indenter.

Main results

– Fabrication and irradiation of the foils of pure metals (molybdenum, tantalum) and austenitic steels of different grades 12Cr18Ni10Ti, AISI 316 and AISI 304 with a high-dose flow of ^{57}Fe Mössbauer iron atoms in the heavy ions accelerator.

– Obtaining of the Mössbauer spectra and calculation of the main characteristics of the implanted iron ions interaction with the crystal lattices of molybdenum, tantalum and austenitic steels of different grades. Determination of the quantitative relationships between the formed radiation defects and the new phases initiated by irradiation.

– <u>For the first time</u>, determination of the structural-orientational arrangement of the implanted iron ions and the type of radiation damages in the form of solid solutions was identified - substitution in molybdenum and introduction in tantalum. It was established for the first time that this process is accompanied by strong grain refinement by the type of primary re-crystallization initiated by irradiation.

– It was found that upon implantation (introduction) of iron ions into the crystal lattices of austenitic steels of three grades, the irradiation initiated $\gamma \rightarrow \alpha$ transformation develops with formation of the defective "stress martensite", the stability and quantity of which is limited by the nickel content.

– It was shown (according to the nanohardness value) that all studied steels undergo radiation hardening, which is almost completely removed in steels with high nickel content at the annealing temperatures close to the operating temperatures of the reactor core, which provides a higher stability of their operational properties.

Based on the obtained results, it can be concluded that irradiation with the high-dose flux of ^{57}Fe ions at the energy of 1 MeV makes it possible to create (to simulate) the radiation damages of the same type (vacancies, interstitial and substituted atoms) in reactor materials, which are formed during radioactive decay in nuclear reactors, and also to initiate the structural-phase transformations due to iron ions implantation into their crystal lattices.

Practical significance of research

The research results obtained as the result of irradiation with the high-dose flow of heavy ions will find application in development and modernization of the simulation methods of radiation exposure to reactor structural materials. The effectiveness and safety of the method is illustrated by obtaining of the high dose of the radiation damages with no induced radioactivity and a short time of its accumulation in comparison with materials, spent in the reactor core. It is shown that irradiation with the Mössbauer iron atoms is universal and can be used not only for steels, but also for materials on a different basis.

The important factor indicating the practical significance of the work is a significant reduction in the service life test period for assessing the reliability and operability of structural materials in the reactor core. The express method of irradiation (hours instead of months) simplifies the issue of their suitability for provision of safe operation of the nuclear facilities, makes it possible to objectively evaluate the operational characteristics and to predict behavior at high damaging radiation doses.

The practical significance of the obtained results also includes the fact that, the possibility of the formation of a highly dispersed structure, which effectively resists to corrosion destruction, is illustrated on the example of refractory molybdenum and tantalum. For this reason, they can be used as piping elements for metallic coolants in the primary loop of the reactor. The fact that steels of the austenitic grade with a high content of γ-stabilizing nickel are more resistant to radiation damage, caused by high-dose irradiation, shows that they are characterized by the undeniable advantage. It is reasonable to use molybdenum as the additional alloying element, as shown on the example of AISI 316 steel.

Approbation of works

The main results of the dissertation thesis were reported and discussed at the local and foreign international conferences: International conference on the applications of the Mössbauer effect, 03-08 September, 2017, Saint-Petersburg, Russia; International Scientific Forum "Nuclear Science and Technologies". September 12-15, 2017, Almaty, Republic of Kazakhstan; XV International Conference "Mössbauer Spectroscopy and its Applications", September 10-16, Sochi, Russia; the Fourth Interdisciplinary Scientific Forum with international participation "New Materials and Advanced Technologies", 27-30 November, 2018, Moscow, Russia; XLIX International Tulin Conference on Physics of Charged Particles Interaction with Crystals, M.V. Lomonosov Moscow State University, 28-30 May, 2019, Moscow, Russia; 12th International Conference "Nuclear and Radiation Physics", June 24-27, 2019, Almaty, Kazakhstan; XXIV International Conference "Interaction of Ions with Surfaces VIP-2019", 19-23 August, 2019, Moscow, Russia; IX International Conference "Modern Problems of Nuclear Physics and Nuclear Technologies" 24-27 September, 2019, Tashkent, Uzbekistan.

Publications: Publication of 19 printed papers on the topic of the dissertation thesis, including 2 articles in the journals peer-reviewed by the Scopus database, 4 articles from the list of scientific journals, recommended by the Committee of Control in the sphere of Education and Science of the Ministry of Education and Science of the Republic of Kazakhstan, 8 Abstracts of reports, 5 articles published in the foreign publications.

Structure and volume of the dissertation thesis. The dissertation thesis consists of "Introduction", 4 Sections, "Conclusion" and 1 "Annex". The work is presented on 127 pages of typewritten text, the paper contains 25 Tables and 57 Figures.