ACCELERATORS SIMULATION OF STRUCTURE-PHASE EVOLUTION AND RADIATION RESISTANCE OF MATERIALS FOR NUCLEAR REACTORS

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At this presentation activity of KIPT which during many years was involved into the field of simulation and investigation of Radiation Damage in reactor materials, is described. Well known that charge particle accelerators are widely used for the purpose of obtaining express information on radiation resistance and investigating physical nature of the radiation. During irradiation with charged particle beams one could reproduce and examine under well-controlled conditions practically all the known radiation effects and investigate physical nature of these effects in more detail. Simulation experiments together with results of reactor investigation much contribute to radiation physics phenomena, radiation and ion-beam technologies as well as solving problems of creation low-activated materials with good radiation resistance. Modern status of simulation experiments in investigation of structure-phase transformations in reactor materials during irradiation is discussed.

INTRODUCTION

Problems of life extension for exploitation nuclear reactors and development of new type reactors demand to receive a lot of data for properties of structure and fuel materials under irradiation, that practically impossible without using of accelerators. The problem of material development for operation in unique conditions of irradiation and evaluation of their radiation resistance consists in the use of existing irradiation facilities for determination of mechanisms of radiation damage and selection of materials with high radiation resistance.

Irradiation of structural materials at temperature of reactor operation creates the unprecedented possibility of microstructure change, of mechanical properties and even of external dimensions of structural components. Theses changes are caused by radiation-induced evolution of microstructure and micro composition [1–3]. Radiation-induced phenomena determine the safety, economics and term of safe operation of reactors of each type. From all the number of known and investigated widely phenomena of radiation damage occurring in the materials during irradiation with fast particles and radiations, in the matter of fission and fusion reactor core structural steels and alloys the principal ones are:

- dimensional changes (swelling, radiation growth, radiation creep, surface relief changing);
- loss in ductility and increase of ductile-brittle transition temperature;
- oxidation and corrosion process acceleration during irradiation and under interaction between the material and heat-transfer agent, nuclear fuel, transmutation products;
- erosion of the fusion and fission core materials surface (blistering, flaking, sputtering, arcing);
- local and bulk change in chemical composition of the initial material (radiation-enhanced segregation of alloy components, nuclear reactions and fast ion implantation).

Up to now a very big amount of theoretical and experimental research work has been devoted to investigation of physical mechanisms of radiation effects occurring in materials under irradiation, enormous number of data on different structural materials behaviour in thermal and fast reactor cores and in the installation simulating the environment of fission and fusion reactors has been accumulated. However, it is still impossible to explain unambiguously the nature and regularities of even the principal radiation phenomena and estimate materials behaviour under irradiation, since, as distinguished from ordinary machine-building materials, the reactor core ones undergo to a great extent more complex and intensive changes of their properties as a consequence of radiation influence.

Now it is evident, that only on the base of physical nature of interaction between radiation and materials, mechanisms of radiation damage in solids one may give scientifically substantiated recommendations both on development of new materials or improvement of existing ones, on evaluation of their behaviour in reactor core and for choice of the optimal conditions of operation of nuclear power systems and the environment, and as a way of obtaining new materials.

In this paper data are presented on investigation carried out in KIPT during last years in the field of simulation of structure-phase evolution and radiation resistance of materials with the use of accelerators of charged particles.

1. METHODOLOGY: ADVANTAGES AND DISADVANTAGES

For investigations of radiation effects such as strengthening, embrittlement, creep and growth of materials one uses high energy beams of light ions (protons, d-particles, ions of carbon or nitrogen, etc.), electrons and gammas to be able to produce homogeneous defect structure along all the thickness of irradiated samples. The grain sizes in austenitic stainless steels are of 20…30 µm. The maximum thickness of the samples for mechanical tests must be of 100...250 µm. Therefore, for these purposes it is necessary to use charged particle beams with the energy providing zone of homogeneous damage through all the irradiated specimen thickness. For radiation damage physics studies of solids the high-energy protons and a-particle beams in cyclotron are used too. The field of accelerator technology is exciting and dynamic. As a result the accelerator community is able to provide brighter light sources, higher collision rates in particle generation, and more precise measurements of physical properties.

Higher damage rate as a result of higher cross-section of charged particles interaction with materials in accelerators (10^{-2}–10^{-4} dpa/s) in comparison with rates of displacements in the different reactors (10^{-6}-10^{-8} dpa/s) allow to achieve necessary doses much faster, for few hours (Fig. 1).

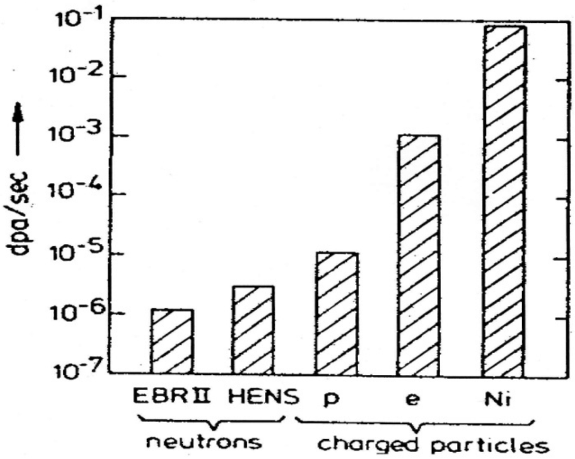


Fig. 1. Damage rate of fast reactor neutron and charged particles

Simulation experiments in investigations of radiation damage of materials have the few **advantages** in comparison with reactor tests; these are:

- precise and good continuous control of experimental parameters of irradiation (temperature, flux, etc.);
- possibility of differential and direct investigation of different factors influence on structure-phase evolution under irradiation; ideally suited for optimizing alloying composition;
- practically absence of induced radioactivity; specimens can be handled in conventional conditions;
- relative cheapness of experiments realization.

Simulation experiments together with advantages have substantial **disadvantages and limitations:**

- difference in recoil spectra and the structure of primary radiation damage;
- phase stability at high dpa rate- and increased temperatures - changing of typical for reactor experiment conditions for nucleation and growth of voids;
- injected interstitial effect leads for typical in simulation experiments decreasing of void size;

- difficulties in simulation of trasmutants accumulation (mainly He and H). This problem can be solved only with multibeam accelerators;
- stress induced by irradiation – surface proximity can go to abnormal evolution of radiation-induced structure.

2. CHARACTERISTICS OF SOME RADIATION SOURCES AND EXPERIMENTAL PROCEDURES

The problem of material development for operation in unique conditions of irradiation and evaluation of their radiation resistance consists in the use of existing irradiation facilities for determination of mechanisms of radiation damage and selection of materials with high radiation resistance.

These experiments may be carried out under neutron irradiation in existing nuclear reactors or by irradiation with ions that generate the processes of radiation damage which are similar to that expected in reactor of next generation.

The most of experimental data on physical nature of radiation effects and radiation resistance are material examinations in commercial and research reactors, charged particle accelerators, and various ion-plasma machines. Characteristics of some radiation sources used for studies of radiation effects and radiation resistance are given in Tabl. 1.

Table 1

The main characteristics of irradiation conditions in the reactors

Reactors and their locations	E, MeV	Flux density, Particles/cm^2·s	appm He, dpa	T, °C
BOR-60 (Dimitrovgrad)	>0.1	$3 \cdot 10^{15}$	0.6	360-600
SM-3 (Dimitrovgrad)	<0.1 >0.1	$5 \cdot 10^{15}$ $2 \cdot 10^{14}$	300	200-500
BN-350 (Aktau, closed)	>0.1	$4 \cdot 10^{15}$	0.5	300-650
BN-600 (Sverdlovsk)	>0.1	$3 \cdot 10^{15}$	0.5	350-670

Material development programmes needs high fluence irradiation facility.

Unfortunately now some of intensively used facilities are shut down (FFTF, DFR, PFR, EBR-II, ORNL triple beam facility, ANL 1 MeV electron microscope and others). This was the reason of the situation that some research programs oriented on the solution of material science problems were not solved. Now part of some basic material science program is realized on accelerators of charged particles.

KIPT possesses a wide choice of accelerators (protons, heavy ions, electrons with different energies spectrum (Fig. 2).

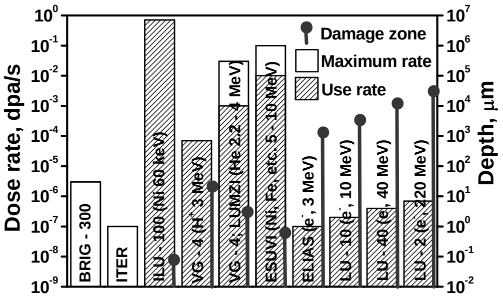


Fig. 2. KIPT ion and electron accelerators

Damage efficiency of irradiated particles is quite difference, that's why choice of irradiated particles is very important.

Heavy ions "generate" the highest defect production rate. But these possess a very short path lengths (Fig. 3). Therefore, heavy ion accelerators with beam energies from the hundreds of keV to a few MeV are mainly used for producing high levels of defectivity in thin layers of the irradiated material.

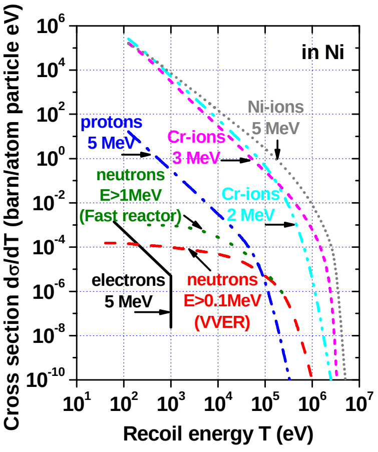


Fig. 3. Damage Efficiency of different particles

The special research electrostatic accelerator with external injector (the ESUVT), which allowed to irradiate the specimens of mateials by monochrom beams of metals Cr, Ni, Fe ions with beam energy 2–5 MeV at. T_{irr}= 60 to 625 °C [4, 5].

In the KIPT the method of investigation and evaluation of mechanical properties of the reactor core materials with the high-energy electrons and gammas has been proposed.

For investigations of radiation effects such as strengthening, embrittlement, creep and growth of materials one uses high energy beams of light ions (protons, d-particles, ions of carbon or nitrogen, etc.), electrons and gammas to be able to produce homogeneous defect structure along all the thickness of irradiated samples. The grain sizes in austenitic stainless steels are of 20…30 µm. The maximum thickness of the samples for mechanical tests must be of 100...250 µm. Therefore, for these purposes it is necessary to use charged particle beams with the energy providing zone of homogeneous damage through all the irradiated specimen thickness. The used (e,γ)-beams in displacement production rate do not exceed the neutron fluxes in reactors but in respect of helium accumulation the high energy electron- and γ-beams are more effective than fast neutrons by two orders of magnitude approximately. This fact makes it possible to simulate expressly high temperature radiation (Fig. 4). Experimental procedures were described elsewhere [6, 7].

The changes in structure and properties of solids under *irradiation with* high-energy particles and γ-quanta are due to proceeding of the interconnected physical processes. They may be subdivided conditionally into:

- the nuclear processes resulting in a primary knock-on atoms (PKA) generation and transmutation products (TP) appearance;
- the atomic ones which lead to displacement cascades development and emergence of primary regions of point defect agglomeration;
- "sub structural" processes conducting to formation of clusters, loops, and nucleation of voids and new phases;
- the diffusion ones which are responsible for microstructural evolution, and in the long run for changing of physical-mechanical properties of the solids.

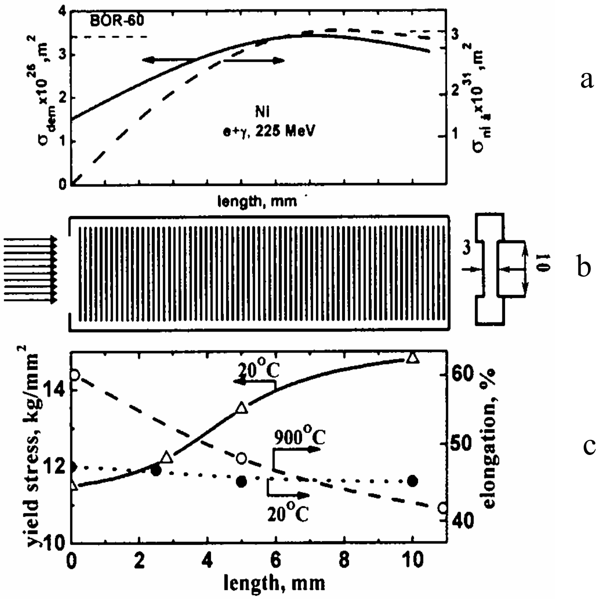


Fig. 4. Profiles of primary damages and segregation of helium (a), along the fabrication (b) and evalution of the yelding stress and elongarion (c) in nickel samples, irradiated by electrons and γ-quants with energy 225 MeV

The surface effects under irradiation of first wall .diaphragms, diverter plates and other units of fusion reactor (sputtering, blistering, flaking, chemical processes, etc.) have been predicted. And now they are investigated by means of (H, D, T, He)-ion beams with energies ranging from a few keV to a few MeV, and by the use of various type plasma machines. The characteristics of the plasma machines used in the KIPT are given in Tabl. 2.

It is very difficult during irradiation with different particles reproduce the conditions completely even under the use of radiation sources of the same type. Now for comparison of irradiation results" criterions of similarity are used; validity of these conditions is confirmed by the results of experiments with irradiation in reactors and in accelerators.

Table 2

Facilities for simulation of interaction processes between plasma and materials

Facility	Kind of ions in plasma	E, eV	Density, $cm^{-1} \cdot s^{-1}$	T,°C	Specific power, wt/cm^2	Purpose
Stellarator "URAGAN-2M	H_2	100	10^{13} (during one impulse)	20	10-1000	Investigations of dynamics of impurities increase in plasma and mass transport in first wall materials
Plasma facility "DRAKON"	H^+_2, He^+ , H^+_2+ He^+	100 500-1700	10^{17} 10^{15}	100-700	0.1-20	Investigations of sputtering of materials, hydrogen permeability. Treatment of cutting tools
Coaxial accelerator of plasma "PROSVET-1"	H^+, He^+, Ar^+, (H^++0.25 Ar^++ 0.5He^+)	0.2-2000	$5 \cdot 10^{15}$- $5 \cdot 10^{16}$ cm^{-2}	20-1200	$2 \cdot 10^4$-10^7	Modeling of energetic spectrum of fusion reactors; Investigations of materials surface erosion
QSPA-Kh-50	H_2	200-900	$(0.1–7) \cdot 10^{16}$ cm^{-3}		10^7	Investigations of materials behavior under influence of high density plasma

3. MODERN STATUS OF SIMULATION EXPERIMENTS

The modern status of simulation experiments is determined by the use of new types of accelerators (of two and three beams), of new methods of preparation and study of specimens (FIB (Focus Ion Beam), Nano-Indentation Tester, EXAFS (X-ray Absorption Fine Structure, positron annihilation, nuclear-physical methods), mathematical modeling methodology. The use of modern methods allows remove the restrictions in the use of results of simulation experiments caused by low depth of damaged layer.

Briefly main tasks, which are needed in accelerators using are such:

- Investigation of fundamental processes. (Simulation of particle collisions; quantification of kinetic properties of radiation defects; simulation of formation & growth of defects; defect characteristics depending on radiation dose (type, size, density, etc.).

- R&D materials for fast reactors (swelling and embrittlement). Observation of radiation-induced microstructure such as segregation and hardening.

- Microstructural predicting for possibilities of life extension for operation reactors; RPV steels (dpa rate), RVI (low temperature embrittlement).

- Gases influence on mechanisms of radiation damage. Synergetic effect of helium and hydrogen in fusion and spallation systems.

3.1. Pressure vessel steels

Accelerators using for experiments of radiation resistance and life extension for pressure vessel steels and pressure vessel internals are now in progress. On the base of the results of investigation of contrast of electron-microscopic images of dislocation loops formed under irradiation in matrix of pressure vessel steel A533B irradiated by Ni ions (3 MeV) to the dose of 1 dpa at 290 °C it was detected that majority of loops has Burgers vector b=a <100> (Fig. 5) [8].

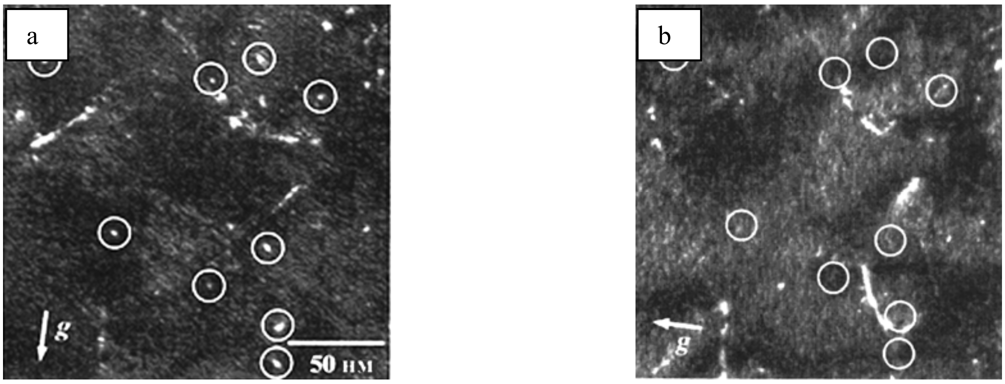


Fig. 5. Dislocation loops images in different diffraction conditions: a - g=020; b - g=200 [8]

Total number of point defects produced under irradiation to the dose 1 dpa is $8\cdot10^{28}$ m^{-3} and the concentration of point defects contained in visible dislocation loops represents only low fraction of the total number) ~$2\cdot10^{-5}$ m^{-3}. It means that the recombination between vacancies and interstitial is the dominating process in steels A533 irradiated by ions with a dose of 10^{-4} dpa/s.

Using of simulation experiments for investigation of radiation behavior of pressure vessel steels was limited but now application of modern experimental facilities (HREM, 3D tomographe) gave a chance to study behavior of matrix defects and distribution of elements and impurities during irradiation.

3.2. Influence of dose rate on void swelling

The temperature dependency of steel swelling (Fig. 6) has the characteristic bell-like appearance and reveals a displacement by 25 K to higher temperatures during the variation of dose rate from 10^{-3} to 10^{-2} dpa·s^{-1}. It must be noted that the behaviour of the swelling curve in the rising area (low temperature range of swelling) is more extended at a dose rate of 10^{-3} dpa·s^{-1} in comparison with 10^{-2} dpa·s^{-1}. The influence of dose rate is manifested in the duration of the transient period of swelling and is more defined at lower temperatures. Irradiation at 888 K leads to a considerably lower influence of dose rate.

Comparison of typical dose rates of 10^{-3}–10^{-2} dpa·s^{-1} used in the presented investigation and 10^{-9}–10^{-8} dpa·s^{-1} which are typical for VVER reactor internals allows concluding that the damage level corresponding to the start of swelling will not exceed 20 dpa. This assumption confirms the results which are obtained in experiments on the BOR-60 reactor [9].

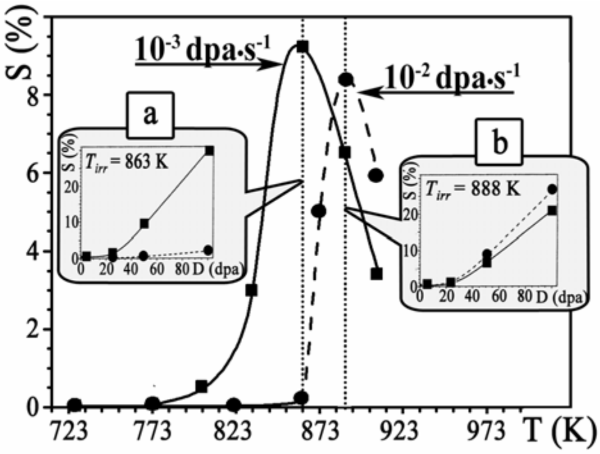
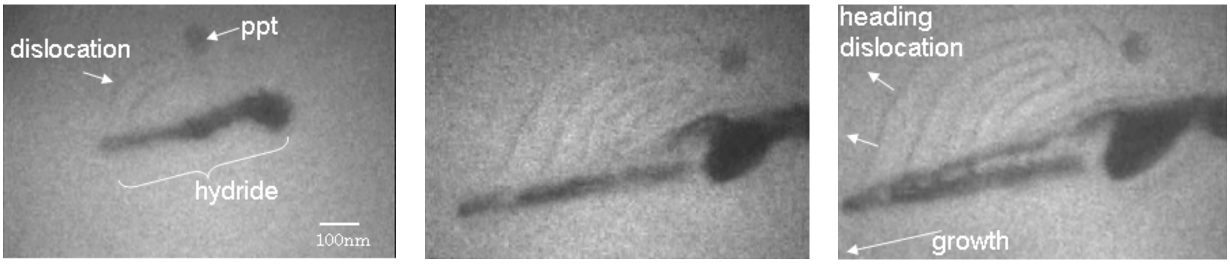


Fig. 6. Temperature dependence of swelling of solution-annealed stainless steel 18Cr-10Ni-Ti (Dose = 50 dpa). The dose rates are: $1\cdot10^{-3}$ dpa·s^{-1} (■) and $1\cdot10^{-2}$ dpa·s^{-1} (●); a - and b – dose dependence of swelling at T_{irr} = 863 and T_{irr} = 888 K, respectively

3.3. Zr-base alloys

The necessity of provision of fuel elements burn up to 75–80 GVt d/t U is related with the increase of the temperature of FE claddings to 358 °C and with vapor content in coolant to 13% mass. Here the main mechanisms of degradation will be the radiation growth and hydride formation. First report on microscopic evolution of Zr hydride in Zircalloy-4 was done as result of simulation experiments, which was performed in Tokyo University [10] (Fig.7).



a b c

Fig. 7. Growth of a intra-granular Zr hydride in Zircaloy-4 specimen under 150 keV H_2^+ *irradiation, B=z=[0001], g =*$10\bar{1}0$ *up to* $1.5\cdot10^{15}$ *(a),* $4.0\cdot10^{15}$ *(b) and* $4.8\cdot10^{15}$ *ions/cm*^{2} *(c) [10]*

The precipitation process of Zr hydride in Zircaloy-4 investigated by in-situ TEM observation under hydrogen ion irradiation. The dynamic process of the formation of Zr hydrides accompanied with dislocations around hydrides was observed. The observation was conducted on (0001) basal plane, which is usually the habit plane of Zr hydrides, and the hydride was the γ-hydride phase with fct structure and the orientation relationship was <110>γ || <1120>α as reported previously. As the hydride grew, the dislocation was generated gradually.

3.4. The synergistic effect of radiation damage and helium + hydrogen

Last time it is shown very complex and synergetic influence of radiation damage, hydrogen and helium (for different reactor conditions are) on materials of PVI internals of VVER and PWR-type reactors. Comparatively low dose rate and swelling temperature shift to area of low temperatures together with production of He and H as result of transmutation reactors can be responsible for low temperature swelling and connected with it low temperature embrittlement. Influence of helium and hydrogen on nanostructural changes in steels of ferritic/martemsitic class was investigated on three-beam accelerator, based on the currently operating electrostatic heavy ion accelerator ESUVI, located in the Kharkov Institute of Physics and Technology.

The substitution of deuterium for protium allows the use of nuclear reactions to determine the depth distribution and concentration of hydrogen isotopes. By processing depth distribution profiles the values of deuterium retention were obtained. The temperature dependences of deuterium retention are shown in Fig. 8. With preliminary implantation of inert gases of helium or argon, formation of radiation defects practically all deuterium implanted at T_{room} is trapped in the investigated steels. Temperature of deuterium detrapping from the specimen shifts by 200 K to higher temperature.

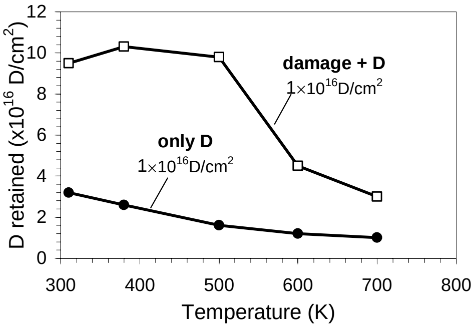
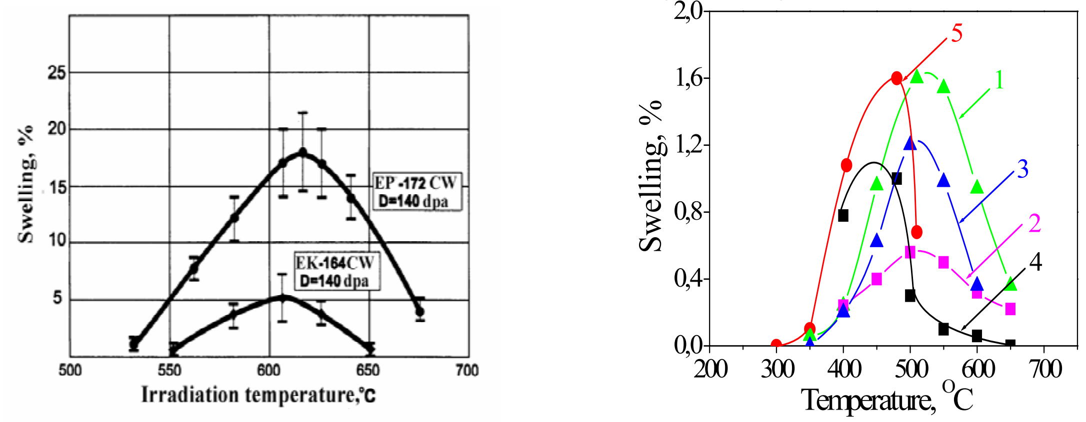


Fig. 8. Quantity of deuterium trapped in steels EI-852 with deuterium implanted to the dose $1\cdot10^{16}cm^{-2}$ *without and with preliminary implanted to dose* $5\cdot10^{16}cm^{-2}$ *argon*

So, the synergistic effect of displacement damage, helium and hydrogen atoms can enhance the irradiation-induced degradation. Such effects were observed in a number of investigations. The highest swelling has been observed in ferritic model alloys of Fe-Cr under triple ion irradiation. In vanadium alloys, simultaneous irradiation of Ni, He and H ions enhanced cavity formation and swelling. The synergistic effect of He and H irradiation in these alloys and F82H martensitic steel was confirmed by the occurrence of larger cavities and higher swelling under triple ion irradiation as compared with dual ion irradiations [11].

3.5. The investigation of prospective structural materials

Achievement of high burn-out (20–25 % of h.a.) in fast reactors demands to solve swelling problem of cladding's and wrapper's materials. Void swelling up to now is the main limiting factor for using structural materials for fast reactors and reactors of future generations [12]. At this area using advantages of accelerators is much more productive because allow methodically investigate the input of different factors, which influence on voids nucleation and growth. Many of them were investigated-role of structure-phase evolution, influence of crystal lattice, gaseous impurities etc. [1, 3]. Role of different alloying elements in radiation behaviour of cladding for fuel elements was investigated (Fig. 9) [13].



a b

Fig. 9. Temperature dependence of swelling for: austenitic steels (EP-172, EK-164) (a); some iron based materials (b): 1 - α-Fe (D=100 dpa); 2 - EP-450 (D=150 dpa); 3 - Fe-12%Cr (D=100dpa); 4 - 01X13M4 Cr^{3+} (D=100 dpa); 5 - 01X13M4 Ar^{3+} (D=100 dpa)

In nuclear power technology of following generation (Generation 4) the using of charged particles accelerators (electrons or protons) for energy from 100 to 1000 MeV is supposed for generation of neutrons. Energy spectrum of emitted neutrons reaches energies 100 and more MeV and cross-sections of nuclear reactions of transmutation (n,2n), (n, p) and (n, α) increases; due to this fact the rate of transmutation variation of steels element composition and level of transmutation formation of gaseous transmutants increases.

Damage of structural materials of electronuclear systems develops as result of irradiation by high-energy protons and neutrons and increases under the influence of liquid metals and possibly of other coolants. Such damage is similar to the damage in (d, t) fusion reactors but transmutation effects (production of H and He) are more pronounced.

Various international programs are now underway to develop advanced reactor concepts under the umbrella of the Generation 4 and INPRO efforts. Each of these programs envisions the use of ferritic-martensitic steels with exposure levels of ~100–200 dpa and temperatures reaching as high as 650–750 ºC, and in many cases with concurrent generation of exceptionally high levels of helium and hydrogen.

4. THE THREE-ION SINGLE BEAM DEVICE

Based on the availability of a currently-existing accelerator STCU project “Evaluation of the performance of ferritic-martensitic steel s under gas conditions relevant to advanced reactor concepts” will first complete the development and improvement of a special ion irradiation system that will provide a state-of-art irradiation test facility. This facility will be used to determine the full parametric performance of all ferritic-martensitic steels currently being developed in the world community for advanced reactor applications (STSU Project #3663). This facility will allow irradiation to proceed under gas generation conditions specific to each reactor concept, using iron ions to generate radiation damage without gas, but also allowing co-implantation of both helium and hydrogen at reactor-specific levels, using a new, novel concept of a three-ion single beam rather than the usual three-accelerator (Fig. 10 a, 10 b).

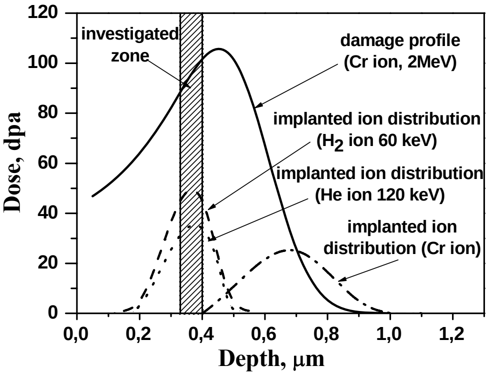


Fig. 10 a. Ions distribution of He, H, Cr and damage profile by Cr ions under irradiation of Fe (TRIM-92)

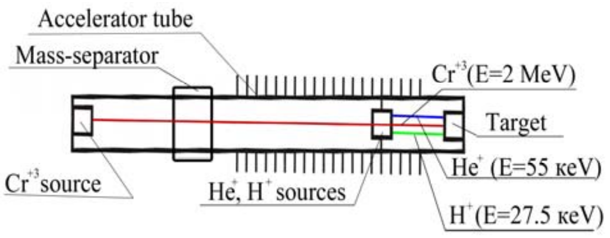


Fig. 10 b. Schematic illustration of the three-ion single beam device to be used in the proposed project

5. CONCLUSIONS

Nuclear energy renaissance demands renaissance in research and development of nuclear materials which determine the safe and economical operation of exploited and developed nuclear facilities.

Material development programmes needs high fluence irradiation facilities. Unfortunately last time irradiation possibilities were strongly decreased due to shut down of few nuclear facilities. Now simulation experiments are very useful for evaluation of radiation behaviour of materials, which work in different facilities with different spectral conditions.

KIPT during many years was involved into the program of simulation of radiation damage in reactor materials. Results received during its performing show that irradiation with charged particle beams allows reproduce and investigate in controlled conditions practically all the known radiation effects in fission and fusion reactor materials and investigate their physical nature more completely and reliably.

Carrying out of research programs in charged particle accelerators together with reactor tests results allow to speed up significantly the studies of mechanisms of radiation phenomena, their connection with irradiation and structural parameters, to reveal many of their regularities, to make selection and development of radiation-resistant materials.

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ИМИТАЦИЯ СТРУКТУРНО-ФАЗОВОЙ ЭВОЛЮЦИИ ПРИ ПОМОЩИ УСКОРИТЕЛЕЙ И РАДИАЦИОННОЕ СОПРОТИВЛЕНИЕ МАТЕРИАЛОВ ДЛЯ ЯДЕРНЫХ РЕАКТОРОВ

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В предлагаемой работе приводится описание деятельности ХФТИ на протяжении многих лет в области имитации и исследования радиационного повреждения реакторных материалов. Хорошо известно, что ускорители заряженных частиц широко применяются с целью получения экспресс-информации о радиационной стойкости и для исследования физической природы излучения. В процессе облучения пучком заряженных частиц можно воспроизвести и исследовать в хорошо контролируемых условиях практически все известные радиационные эффекты, а также исследовать физическую природу этих эффектов более подробно. Имитационные эксперименты наряду с реакторными испытаниями вносят весомый вклад в явления радиационной физики, радиационные и ионные технологии, а также в решение проблем создания слабоактивируемых материалов с высокой радиационной стойкостью. Обсуждается современный статус имитационных экспериментов в исследовании структурно-фазовых превращений в реакторных материалах под облучением.

ІМІТАЦІЯ СТРУКТУРНО-ФАЗОВОЇ ЕВОЛЮЦІЇ ЗА ДОПОМОГОЮ ПРИСКОРЮВАЧІВ ТА РАДІАЦІЙНА СТІЙКІСТЬ МАТЕРІАЛІВ ДЛЯ ЯДЕРНИХ РЕАКТОРІВ

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В представленій роботі описано діяльність ХФТІ протягом багатьох років в галузі імітації та дослідженні радіаційного пошкодження реакторних матеріалів. Добре відомо, що прискорювачі заряджених часток широко використовуються з метою отримання експрес-інформації щодо радіаційної стійкості і для вивчення фізичної природи випромінювання. У процесі опромінення пучком заряджених часток можна відтворити та дослідити в добре контрольованих умовах практично всі відомі радіаційні ефекти, а також дослідити фізичну природу цих ефектів більш докладно. Імітаційні експерименти нарівні з реакторними випробуваннями вносять вагомий вклад в явища радіаційної фізики, радіаційні та іонні технології, а також в рішення проблем слабкоактивуємих матеріалів з високою радіаційною стійкістю. Обговорюється сучасний статус імітаційних експериментів в дослідженні структурно-фазових перетворень в реакторних матеріалах під опроміненням.