SIMULATION OF RADIATION DAMAGE IN MATERIALS BY CHARGED PARTICLES (KIPT EXPERIENCE)

V.Zelenskij I.Neklyudov, L.Ozhigov, V.Voyevodin, A.Dovbnya, G.Tolstolutskaya

NSC "Kharkov Institute of Physics and Technology", 1, Akademicheskaya Str, 61108, Kharkov, Ukraine
(voyev@kipt.kharkov.ua)

In this paper, the activity of Kharkov Institute of Physics and Technology (KIPT) which during many years was involved into the field of simulation and investigation of radiation damage in reactor materials, is described.

It is well known that charged particle accelerators are widely used for obtaining express information on radiation resistance and investigating physical nature of the radiation.

Using irradiation with charged particle beams one could reproduce and examine practically all the known radiation effects and investigate physical nature of these effects in more details under well-controlled conditions.

The characteristics of accelerators and plasma machines for simulation and investigation of radiation effects used in the KIPT are given in this paper.

The results of simulation and reactor experiments on different materials are analyzed. Most representative results concerning fission and fusion reactor structural materials behaviour may be obtained in real conditions of exploitation of the relevant reactors.

Simulation experiments together with results of reactor investigation contribute much to radiation physics phenomena, radiation and ion-beam technologies as well as to creation of low-activated materials with good radiation resistance.

Modern status of simulation experiments is discussed.

I. INTRODUCTION

Now Ukraine exploits 15 nuclear power units used for production of more than fifty percent of total electric power

Problems of life extension for the working nuclear reactors and development of new-type reactors demand to obtain a lot of data for structural properties of fuel materials under irradiation, that is practically impossible without using accelerators.

In this paper, data are presented on investigations carried out in KIPT during last years in the field of simulation and studies of radiation damage in materials with the use of accelerators of charged particles.

The problem of development of materials 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 and electrons that generate the processes of radiation damage similar to those expected in a reactor of the next generation. High rates of dose accumulation (10^{-2} 10^{-3} dpa/sec) under ion irradiation that are much higher than doses in existing test and power reactors (10^{-6} – 10^{-8} dpa/sec) permit the obtaining of express information on radiation resistance of materials. Simulation experiments on investigations of the basic phenomena of material damage in reactors has the advantages in comparison with reactor tests; these are:

- precise and continuous control of irradiation parameters;
- possibility of differential and direct investigation of different factors influence on defected structure and composition evolution under irradiation;
- absence of induced radioactivity;
- relative cheapness of experiments realization.

One of the novel features of the KIPT simulation program was application of high-energy electrons and γ-quanta together with heavy ions for simulation purposes.

It is well known that the complex structural-phase transformations in materials under irradiation lead to significant changes of their initial properties and, as a rule, to their degradation [1–3].

In the case of fission and fusion reactor core structural steels, the principal phenomena of radiation damage 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 the choice of the optimal conditions of operation of nuclear power systems.

II. CHARACTERISTICS OF SOME RADIATION SOURCES AND EXPERIMENTAL PROCEDURES

The most of experimental data on physical nature of radiation effects and radiation resistance come from 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 Table1.

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, ^{o}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 programs need high fluence irradiation facility.

Unfortunately, now some of the 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 to the solution of material science problems were not realized. Now a part of some basic material science programs is realized using accelerators of charged particles.

KIPT possesses a wide choice of accelerators (protons, heavy ions, electrons with different energies spectrum (Fig 1).

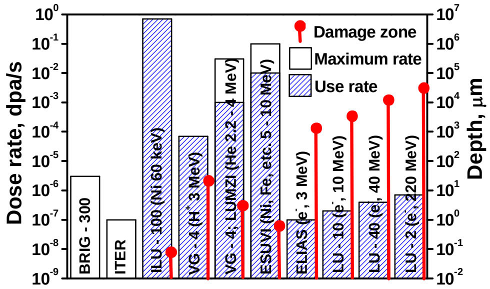


Fig. 1. KIPT ion and electron accelerators

Damage efficiency of irradiated particles is quite different that’s why the choice of particles for irradiation is very important.

Heavy ions "generate" the highest defect production rate. But they possess very short path lengths (Fig. 2). Therefore, heavy ion accelerators with the beam energies from the hundreds keV to a few Mev are mainly used for producing high levels of defect concentration in thin layers of the irradiated material.

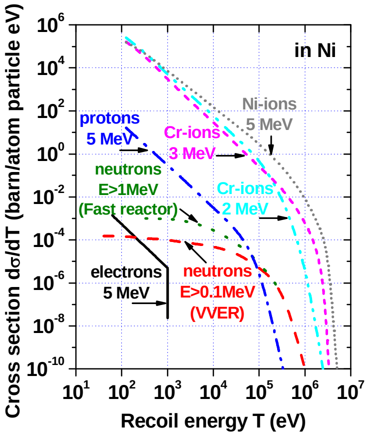


Fig. 2. Damage Efficiency of different particles

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^{o}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 high energy beams of light ions are used (protons, d-particles, ions of carbon or nitrogen, etc.); electrons and gammas can 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 a zone of homogeneous damage through all the thickness of the irradiated specimen. The used (e,γ)-beams do not exceed the reactor neutron fluxes in displacement production rate 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. 3). Experimental procedures were described elsewhere [6, 7].

The surface effects under irradiation of first wall diaphragms, diverter plates and other units of the fusion reactor (sputtering, blistering, flaking, chemical processes, etc.) have been predicted. 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 means of the various types of plasma machines. The characteristics of the plasma machines used in the KIPT are given in Tab.2.

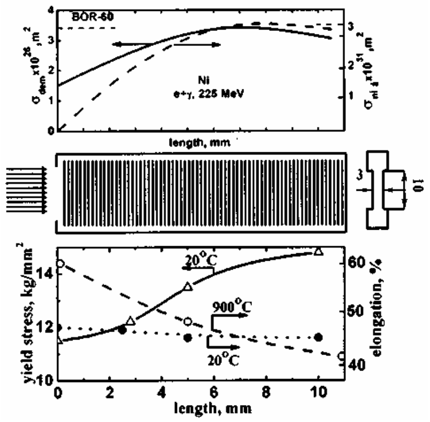


Fig. 3. Profiles of primary damages and segregation of helium (a), along the fabrication (b) and evalution of the yelding stress and elongation (c) in nickel samples, irradiated by electrons and γ-quants with energy 225 MeV.

TABLE 2. Facilities for simulation of interaction of plasma and materials

Facility	Kind of ions in plasma	E, eV	Density, cm^{-1}·s^{-1}	T,°C	Specific power, wt/cm^2	Purpose
Stellarator "URAGAN-2M	H_2,	100	10^{13} (during one pulse)	20	10…1000	Investigations of dynamics of impurities increase in plasma and mass transport in the 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, *and* hydrogen permeability. Treatment of cutting tools
Coaxial accelerator of plasma "PROSVET-1"	H^+, He^+ , Ar^+, (H^++0.25 Ar^++ 0.5He^+)	0.2-2000	$5\cdot10^{15}$-$5\cdot10^{16}$ 1/cm^2	20-1200	$2\cdot10^4$-10^7	Modeling of energy spectrum of fusion reactors; investigations of materials surface erosion
QSPA-Kh-50	H_2,	200…900	(0.1–7)· 10^{16} cm^{-3}		10^7	Investigations of materials behavior under influence of high density plasma

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 conventionally subdivided into:

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

It is very difficult to completely reproduce the conditions of irradiation with different particles even if radiation sources of the same type are used. Now, for comparison of irradiation results, criteria of similarity are used; the validity of these conditions is confirmed by the results of irradiation experiments using reactors and accelerators. These conditions are the following [8, 9]:

the condition of equality of irradiation doses expressed as the number of displacements per atom (dpa)

$$D_j = \sigma_d \varphi_j t, \tag{1}$$

where $\varphi_j = \int_0^\infty dE \varphi_j(E)$ – beam density for particles of sort j_i, σ_d -total cross-section of defect production;

the condition of structural similarity of primary radiation damage (Z=1; Z-quantity criterion characterizing the number of defects formed by the same manner);

the condition of equality of relations of the production rate of PNR (products of nuclear reactions) to the rate of radiation defects production

$$\frac{K_{pn}}{K_n} = \frac{K_{pi}}{K_i}, \tag{2}$$

This is especially important for high irradiation doses; in this case, it is necessary to calculate and to compare PNR accumulation in the materials under irradiation by neutrons, charged particles and γ-quanta.

the condition of equality of the relations of point defect generation rate to the rate of their disappearance at sinks; for vacancies, this condition may be presented as:

$$\frac{K_1}{K_2} = \frac{D_{v1}}{D_{v2}}, \tag{3}$$

where K_1 and K_2 – rates of damage in comparable experimental conditions; D_{v1}, D_{v2} – diffusion coefficient of vacancies under irradiation

$$D_{v(1,2)} = D_0 \exp(-E_{mv} / kT_{1,2}), \tag{4}$$

where E_{mv}- energy of vacancy migration, $T_{1,2}$-irradiation temperature in compared experiments, K;

the condition of similarity of diffusion progress, including diffusion of gaseous atoms, recrystallization, solid solution decay and dissolution of precipitates, redistribution of alloys components.

Apart from the general similarity criterions for simulation irradiation, one should take into account the influence of specific similarity conditions for each phenomenon: proximity of the free surface under heavy ion irradiation; pulse characteristics of high-energy electron-beam irradiation, etc.

III. SOME RESULTS OF SIMULATION INVESTIGATIONS

Data were developed on microstructural evolution for pure metals (Ni, Cr, Fe, Ti, Cu), model alloys (Ni-Sc, Ni-Pr, Ni-Cu, Fe-Cr, Fe-Cr-Ni, Fe-Cr-Mo, Fe-Cr-N) and for steels and alloys of different classes: austenitic—16Cr-15Ni-3Mo-Nb, 18Cr-10Ni-Ti, 16Cr-llNi-2Mo; ferritic and ferritic-martensitic— 13Cr-(l-2) Mo-NbVB, 9Cr-6Mo-Nb, etc.; and high-nickel alloys.

III.A. Mechanisms of radiation damage

The simulation experiments made a significant contribution to knowledge of radiation damage of materials. The majority of modern results on defect production processes, defect annealing, structural-phase state evolution during irradiation, influence of gaseous and other impurities on radiation phenomena were established in experiments with use of accelerators and mathematical modeling methodology.[6] The basic part of experimental data on energetic and geometric parameters of point defects and their complexes was obtained for metals irradiated with electrons and ions.

Using the method of low-temperature (~80 K) irradiation with electron energy of 2 MeV and measurements of electrical resistance, annealing of radiation point defects were investigated in low-doped alloys Zr-Gd, Zr-Dy, and Zr-La.

It was experimentally established that impurity atoms such as La, Dy and Gd interact effectively with point defects in zirconium matrix. Such interaction can result in formation of interstitial-impurity and vacancy-impurity complexes. This influences considerably on the processes of annihilation and redistribution of radiation defects and must be taken into account in development and modification of alloys on the base of zirconium for the core of nuclear power reactors (fig. 4) [10].

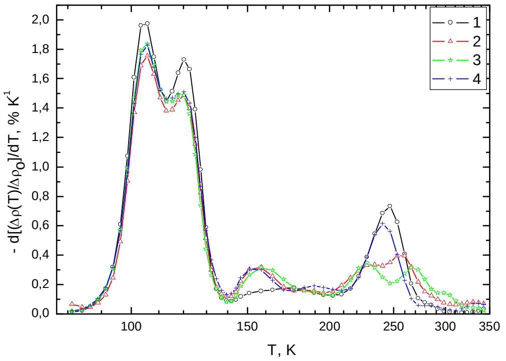


Fig. 4. Spectrums of isochronous annealing of Zr and Zr-Dy, Zr-Gd, and Zr-La alloys irradiated with electrons (E= 2 MeV ,$T_{irr=}$ 82 K ,D= $1{,}4\cdot10^{19}$ $e^-/cм^2$: 1- pure Zr; 2- Zr-Dy alloy; 3 –Zr-Gd alloy; 4 –Zr-La alloy.

Irradiation with high-energy electrons (E>8 Mev) leads not only to generation of Frenkel pairs but also to formation of complex radiation defects and nuclear reactions products. It allowed to separate the mechanisms of radiation embrittlement. High temperature embrittlement exists under irradiation by electrons with γ-quantas with energy higher than the level of nuclear reactions (Fig.5) [7].

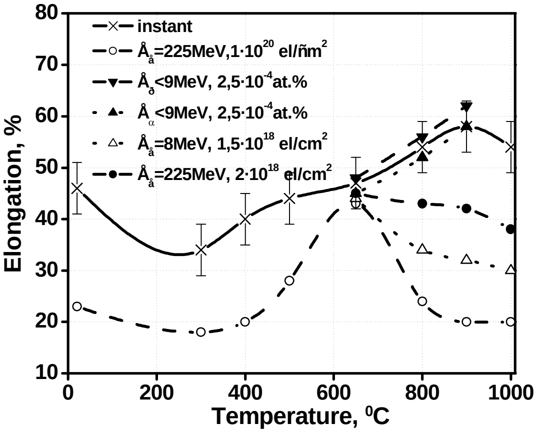


Fig. 5. Effect of radiation on mechanical properties of nickel.

The use of electron accelerators has allowed detailed studies of the first stage of radiation creep of Cr-Ni-Sc steel. It is shown that the initial deformation value and the initial duration of creep in the Cr-Ni-Sc steel are less than for the prototype steel. The rate of radiation creep for the Cr-Ni-Sc steel is several times less than that for the prototype steel. The results obtained from the studies using accelerators have been confirmed by the reactor tests of Cr-Ni-Sc steel at Nuclear Physics Institute of Ukrainian Academy of Sciences (Fig. 6).

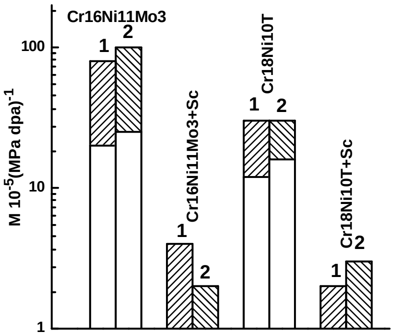


Fig. 6. Radiation creep modulus of steel; dose up 0.5dpa, 1 – accelerator, 2 – reactor [7].

Comparison of the dose dependencies of swelling for different types of irradiation showed considerable differences both in the duration of incubation period and in the swelling rate, that can be explained by the nature of irradiation. Formation of vacancy loops in cascades during reactor and ion irradiation decreases the level of vacancy super saturation and moderates the processes of nucleation and growth of voids (Fig. 7) [4].

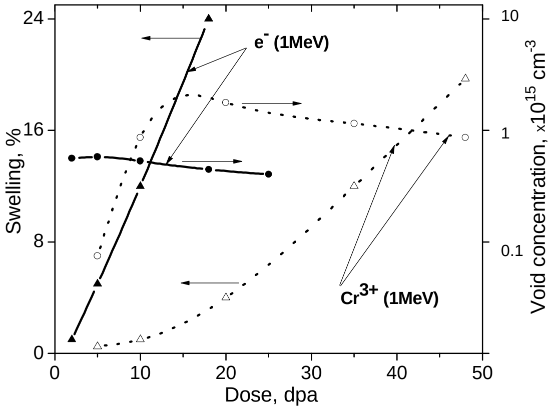
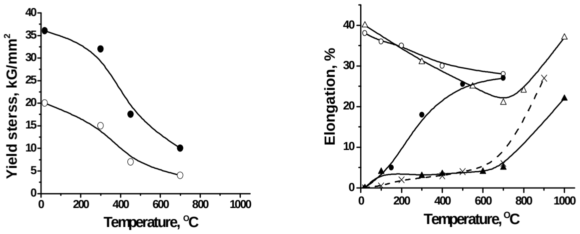


Fig.7. Dose dependence of swelling for steels 16Cr-15Ni-3Mo-Nb under electron and Cr-ion irradiation (T_{irr} = 650ºC).

The simulation experiments have been used to investigate regularities of radiation damage as well as swelling and embrittlement resistance of the low-doped Cr-alloys [11]. The data of reactor examination of these materials obtained after several years showed a good fit to the data of simulation experiments (Fig.8).



a) b)

Fig.8. Temperature dependences of yield stress (a) and relative elongation (b) for Cr+0.3 La+0.4 Ta+0.3 V alloy in original (○, Δ) and irradiated (●, ▲, ×) samples :
○, ● - gaseous impurities content up to 0.001 wt%;
Δ, ▲, × -impurities content is higher than 0.02 wt%;
○, ●, × -irradiation by electrons with energy of 225 MeV (T_{irr}=150°C) up to 10^{21} electron/cm^2;
Δ, ▲ -irradiation in the BOR-60 fast reactor up to $3.7 \cdot 10^{22}$ n/cm^2, [11].

Simulation experiments gave possibility to discover the effect of reduction of void swelling and embrittlement in Ni doped with rare-earth elements (Sc, Y, Ce, La, Pr, etc (REE) (Fig.9) [1]. Swelling data for some basic steels doped with the REE show that presence of the REE up to 0.1 wt% in the steels reduces the swelling value by 2-5 times under irradiation in accelerators and reactor environment.

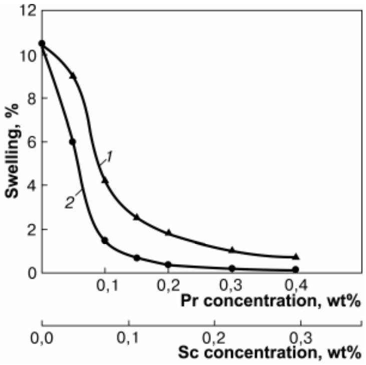


Fig. 9. Swelling versus concentration of REE elements in Ni. T_{irr}=600ºC, D=40dpa, E=2.8MeV.

The main effects in the first wall materials of fusion reactors are surface erosion and embrittlement of the materials under influence of neutron fluxes and plasma beams. We have investigated the effect of hydrogen, helium and hydrogen-helium plasma on surface erosion and mechanical properties of the austenitic and ferritic/martensitic steels. As a result of irradiation of the Cr13 type steel by hydrogen plasma ions (0.6-1.2 kev) to fluence levels as large as $5\cdot10^{18}$ cm^{-2} in near-surface layer gas-filled cavities are formed, the depth of their lying exceeds the hydrogen ion free path by 4 orders of magnitude. Such cavities-cracks enhance essentially the surface erosion and tendency to brittle fracture of the ferritic/martensitic steel [24]. The effect of pulsing plasma beams with specific power of 3 Mw/cm^2 , duration of 2 μsec, and average particle energy of 2 keV on austenitic steel specimens is accompanied by their strengthening and embrittlement. The "layer by layer" electron microscopy analysis has shown that as result of influence of plasma beams on the near-surface layers down to 25 μm, a cellular structure with pseudo-amorphous cell boundaries is produced (Fig. 10) [12].

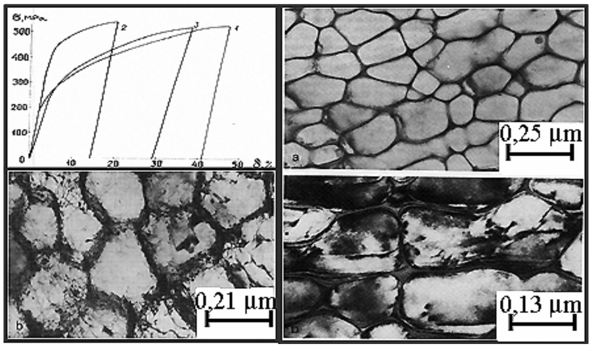


Fig. 10. Typical cellular structure formed in the Cr16Ni15Mo3Nb steel as a result of plasma influence at different distances from the irradiated surface.

III.B. Modern status of simulation experiments

Now KIPT is working directly for Ukrainian nuclear energetic and also for the world nuclear community.

Experiments on simulation of radiation resistance and life extension for pressure vessel steels and pressure vessel internals, claddings from Zr-base alloys are now in progress Simulation experiments on pressure vessels suggest the unique possibility for investigation of flux effects mechanisms on pressure vessel embrittlement, which are very important from technological point of view.

Radiation behavior of Zr-based alloys is studied. Now investigations of influence of oxygen and dose rate on defect structure of Zr-1Nb alloy are performed in frame of STCU project P-174.

It is shown that influence of radiation damage, hydrogen and helium (for different reactor conditions are) on materials of PVI internals of VVER-type reactors is very complex and synergetic. Comparatively low dose rate and swelling temperature shift to the area of low temperatures together with the production of He while H can be responsible for low temperature swelling and connected with its low temperature embrittlement.

It was shown that the process of steel fracture (radiation embrittlement) may be separated into two components: matrix and the grain boundary. The crack propagation into the grains proceeds due to the localization of gliding along the voids line (under the stresses the growth of the voids is observed) (Fig. 11) [13].

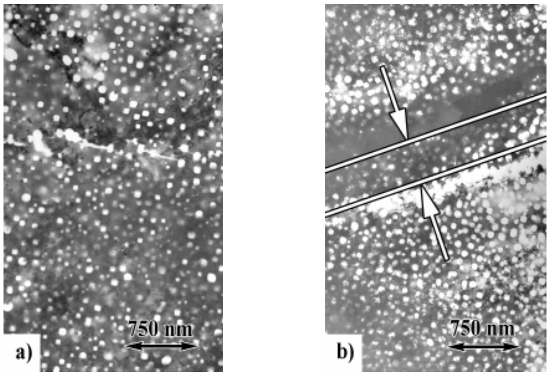
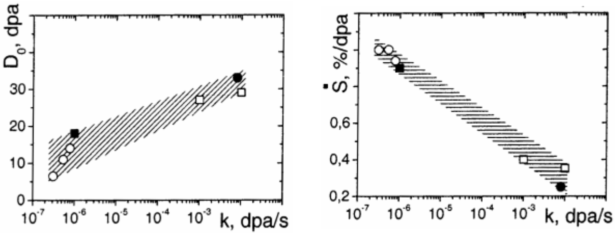


Fig. 11. TEM microstructures of 18Cr-10Ni-Ti SS irradiated at 635°C to 100 dpa (damage rate 10^{-2} dpa/s) illustrate fracture modes during room temperature deformation: (a) matrix area and (b) grain boundary denuded zone.

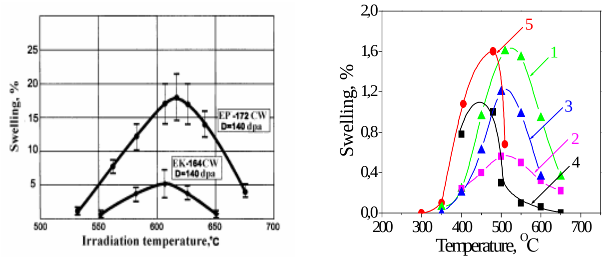
Dependences of transient period and rates of swelling for austenitic steels in the temperature range of maximum swelling (fig. 12) demonstrate that the dose rate influences on the duration of transient period of swelling, namely, with the increasing of damage rate the value of transient period increases [14].



a) b)

Fig. 12. Influence of dose rate (k) on: a) value of transient period (D_o) and b) rate of swelling ($\dot{S}$) in the temperature maximum of steel swelling:
(□) – 18Cr10NiTi, T_{irr} = 615°C at 10^{-2}dpa/s and 590°C at 10^{-3}dpa/s;
(■) – Cr18Ni10Ti $T_{irr} \approx 480°C$ (BOR-60);
(○) – Fe-15Cr-16Ni $T_{irr} \approx 410°C$ (FFTF);
(●) – Cr1615Ni3MB T_{irr} = 650°C (heavy ions).

Achievement of high burn-out (20-25% of h.a) in fast reactors demands to solve swelling problem of cladding and wrapper materials. Up to now, void swelling is the main limiting factor for using structural materials for the fast reactors and the reactors of future generations [15]. In this field, advantages of accelerators are much more considerable because allow comprehensive studies of the input of different factors, which influence on voids nucleation and growth. Many of these factors, such as the role of structure-phase evolution, influence of crystal lattice, gaseous impurities etc. [1, 3] were studied. The role of different alloying elements in radiation behaviour of cladding for fuel elements was investigated (Fig. 13) [16].



a) b)

Fig. 13. Temperature dependence of swelling for:
a) austenitic steels (EP-172, EK-164);
b) some iron based materials: 1- α-Fe (D=100dpa), 2- EP-450 (D=150dpa), 3- Fe-12%Cr (D=100dpa), 4- 01X13M4 Cr^{3+} (D=100dpa), 5- 01X13M4 Ar^{3+} (D=100dpa).

In nuclear power technology of the following generation (Generation 4), charged particles accelerators (electrons or protons) for energies from 100 to 1000 MeV are used for neutron generation (Fig.22). Energies of the emitted neutrons reach 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 the 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 650–750ºC, and in many cases with concurrent generation of exceptionally high levels of helium and hydrogen.

Basing on (the availability of) the current STCU accelerator project “Evaluation of the performance of ferritic-martensitic steels under gas conditions relevant to advanced reactor concepts”, development and improvement of a special ion irradiation system will be completed; 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 under gas generation conditions specific for each reactor concept. Iron ions will be used to generate radiation damage without gas. It will also allow 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 approach (fig. 14).

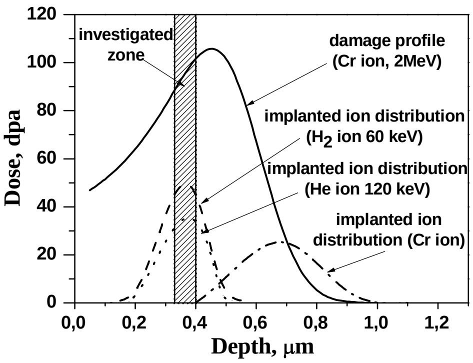


Fig. 14a. Ions distribution of He, H, Cr and damage profile by Cr ions under irradiation of Fe (TRIM-92).

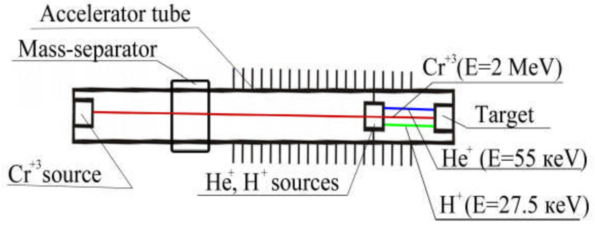


Fig. 14b. Schematic illustration of the three-ion single beam device to be used in the proposed project.

IV. CONCLUSIONS

Nuclear energy renaissance demands to pursue research and development of materials which determine the safe and economical operation of operational and developed nuclear facilities.

Material development programs need high-fluence irradiation facilities. Unfortunately, irradiation possibilities of the former times strongly decreased due to the shut down of several nuclear facilities. Simulation experiments are very useful for evaluation of radiation behavior of materials used in different facilities with different spectral conditions.

During many years, KIPT was involved into the Program of Simulation of Radiation Damage in the reactor materials. The results obtained show that irradiation with charged particle beams allows reproduction and investigation of practically all the known radiation effects in fission and fusion reactor materials in controlled conditions and more detailed and reliable studies of their physical nature.

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

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