Modern Status of Accelerators in R&D of Structural Materials for Nuclear Reactors

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Abstract. 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 and electrons that generate the processes of radiation damage which are similar to that expected in reactor of next generation. Authors publication in a previous Conference of Accelerators application was devoted to KIPT experience in simulation of radiation damage in materials by charge particles. This presentation show modern status of accelerators using for such purposes is determined by main task – to ensure safe and economic work of nuclear energetic.

1. Inroduction

Today nuclear power is the more real in the world that possesses the humanity for the production and supply of low cost electrical and thermal power for distant prospective with guarantee of nuclear, physical, ecological and technical safety in amounts corresponding with society needs. According to the data of IAEA more than 500 nuclear power units of research and other reactors are in operation. Nuclear energy "renaissance" demands to perform research and development of materials which determine the safe and economical operation of running and developed nuclear facilities. Unfortunately structural materials in the nuclear power plants undergo degradation due to irradiation influence. Rate of degradation of nuclear materials tends to accelerate due to irradiation.

Behaviour of materials under irradiation has been studied for more than 50 years. Most experience has arisen from the area of "thermal reactors" where core structural materials are subject to temperatures up to 400ºC and damages up to 60 dpa for near-core austenitic internals and 10 dpa for typical LWR fuel cladding at a burn up of 40 GWd/tU. Fast reactor experience reaches ~200 dpa and temperatures approaching 600 ºC. Looking forward, it is projected that structural components of Generation IV fission reactors will operate at 500ºC–1000ºC and reach damage levels of up to 100–200 dpa. For projected fusion devices with greatly differing neutron spectra arising from 14 MeV neutrons there will also be a wide range of dpa and temperature. ITER will experience approximately only 100ºC–300ºC and damages of about 3 dpa, the prototype fusion power reactor DEMO is expected to operate in the range of 500ºC–1000ºC and reach ~150 dpa at the end of five years of full power operation. Radiation doses in future commercial fusion power reactors might be significantly higher. High irradiation doses and temperatures planned for advanced fission and future fusion reactors will most certainly require the development of new improved materials. Solving of such ambitious programs, which are suggested by world society for (INPRO, GEN IV, GNEP etc) need in a first raw to solve materials problems.

The cost of material testing under neutron irradiation for these advanced nuclear systems is continuously increasing while availability of test reactors is steadily decreasing. Last time irradiation possibilities were strongly decreased also due to the shut down of all spectrums of nuclear facilities.

In 1969 Mazey and Nelson [1] performed research of specimens of M316 steel, which were irradiated by ions of C, O and Fe and showed that structure of ion-irradiated specimens is very similar to structure of neutron-irradiated specimens. During the 1980s processes of swelling and structure-phase changes were investigated by simulation experiments. This experiment served as a basis of new scientific direction of radiation material science – simulation of reactor damage with irradiation by charge particles [2].

2. 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/sec) in comparison with rates of displacements in the different reactors (10^{-6}-10^{-8} dpa/sec) allow to achieve necessary doses much faster, for few hours (Fig. 1).

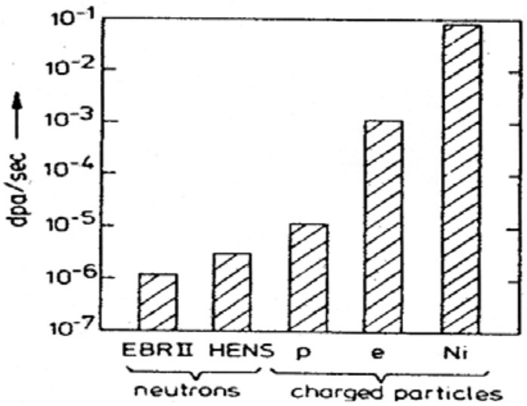


FIG. 1. Damage rate of fast reactor neutron and charged particles.

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.

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.2) [3].

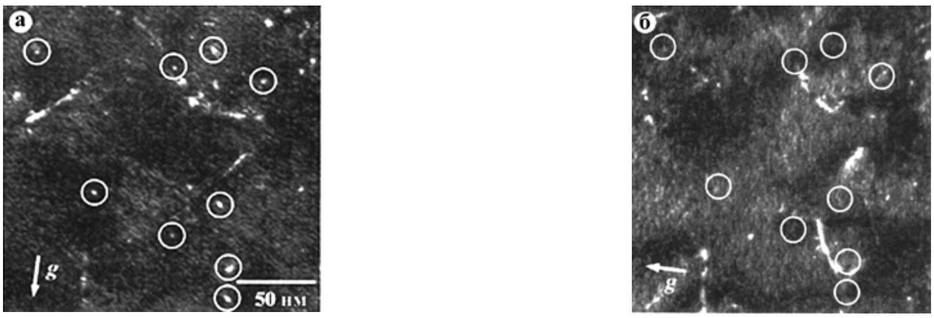


FIG. 2. Dislocation loops images in different diffraction conditions a) g=020, b) g=200 [3].

Total number of point defects produced under irradiation to the dose 1 dpa is $8.10^{28}m^{-3}$ and the concentration of point defects contained in visible dislocation loops represents only low fraction of the total number) ~$2.10^{-5}m^{-3}$. 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. 3) 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 (see Fig. 3 (a) and (b)).

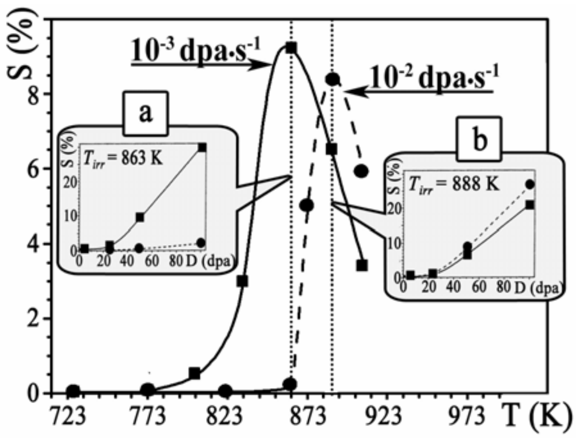


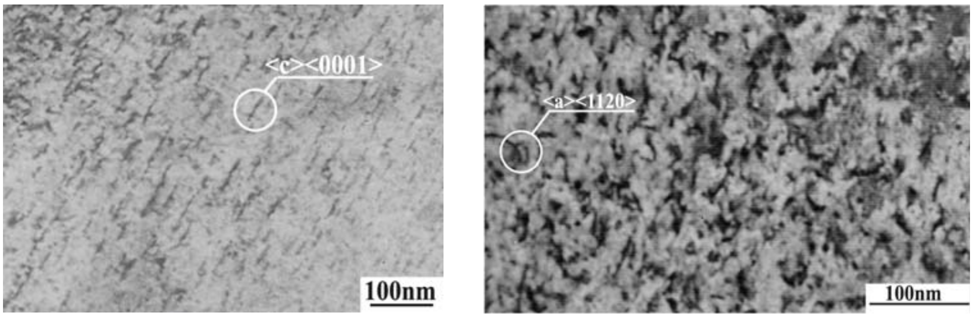
FIG. 3. Temperature dependence of swelling of solution-annealed stainless steel 18Cr-10Ni-Ti (Dose = 50 dpa.) The dose rates are: 1×10^{-3} dpa·s^{-1} (■) and 1×10^{-2} dpa·s^{-1} (●); a) and b) – dose dependence of swelling at T_{irr} = 863 and T_{irr} = 888 K, respectively.

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 [4].

3.3. Zr-base alloys

Nucleation, growth, and interaction of dislocation loops is one of the main components of the structure evolution. They control the variation of physical and mechanical properties under irradiation and more, they are in the base of some phenomena: irradiation hardening, embrittlement and irradiation growth. The influence of oxygen on these processes is studied weakly now.

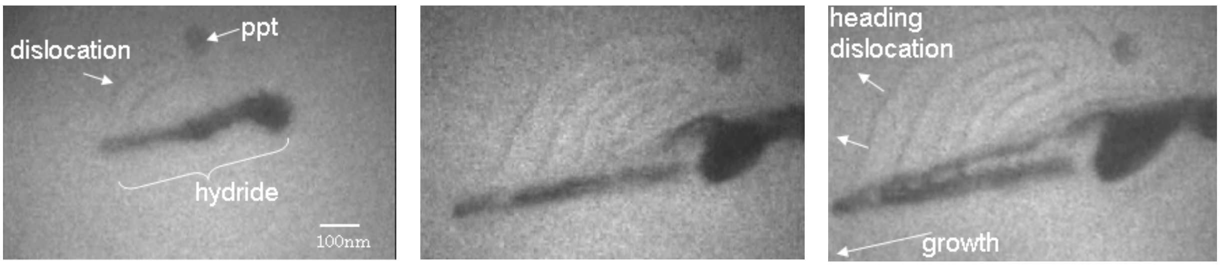
Influence of heavy ions Zr^{6+} irradiation on parameters of dislocation loops in alloy Zr1%Nb with oxygen content 0.08, 0.14 and 0.19 wt.% is studied by methods of transmission electron microscopy. Performed simulation experiments allowed to investigate evolution features of dislocation structure in E-110 alloy and showed strong influence of oxygen on suppression of number density of c-type loops, which are responsible for radiation growth (fig. 4).



a b

FIG. 4. Strong influence of oxygen on suppression of number density of c-type loops; oxygen content: 0.08 (a) 0.19 wt.% (b).

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 [5] (fig.5).



a b c

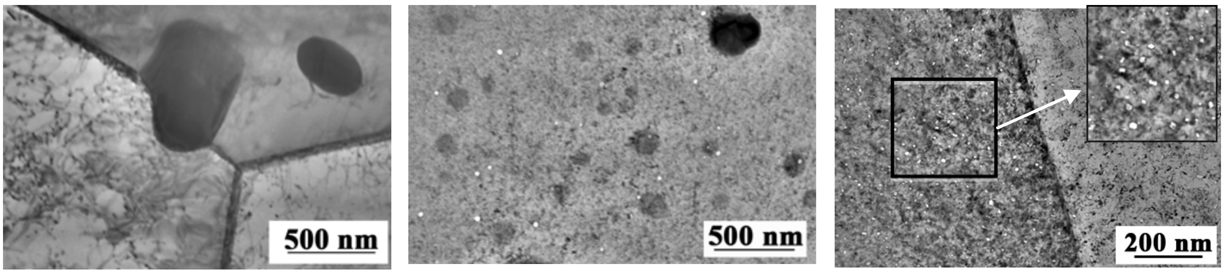
FIG. 5. Growth of a intra-granular Zr hydride in Zircaloy-4 specimen under 150 keV H_2^+ irradiation, B=z=[0001], g =$10\bar{1}0$ up to 1.5×10^{15} (a), 4.0×10^{15} (b) and 4.8×10^{15} ions/cm^2 (c) [5].

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. Irradiation of industrial 12% chromium steel EI-852 of Russian production was carried out (C-0.13; Si-1.19; Cr-13.15; Ni-0.27; Mo-1.69; Mn-0.31 wt. %).

The initial microstructure of steel EI-852 represent ferrite where precipitates of the type $M_{23}C_6$ are distributed homogeneously (fig.6.a). After irradiation by heavy ions of chromium at temperature 480°C and dose 100 dpa the ferritic structure of steel EI-852 remains. Formation of fine voids with dimension 20 nm and of large voids ~60 nm is observed (concentration $4.0\cdot10^{14}cm^{-3}$); these voids are homogeneously distributed by the grain volume. In this case the swelling was 0.01% (fig.6.b). Under triple-beam irradiation (ions of Cr^{+5}, He^{+} and H^{+}) of steel EI-852 formation of large voids ~50 nm and of number of fine gaseous voids with dimension 10-15 nm with concentration $2.2\cdot10^{16}cm^{-3}$ was observed; this formation causes the increase of swelling to ~0.4% (fig.6,c).



a b c

Fig.6. Microstructure of steel EI-852 before and after irradiation: a) initial microstructure; b) after irradiation with Cr^{+5}, c) after irradiation by Cr^{+5}, 4800 appm He, 6000 appm He.

Processes of accumulation and of trapping of hydrogen and helium in defects nucleated in low energy cascades and at preliminary production of damage on the level of 100…2000 displacement per atom are investigated in ferritic/martensitic steel EI-852 under irradiation by ions of deuterium with energy 6 keV, of helium -12 KeV, of argon – 1400 KeV. The measuring system "ESU-2" is used for study the retention, accumulation and distribution of hydrogen (deuterium) and displacement damage production under high energy heavy particles (He^{+}, Ar^{+}, Kr^{+}, Xe^{+}) irradiation. Measuring system allow to use the sets of the ion beam analysis techniques including Rutherford backscattering spectroscopy (RBS), channeling, nuclear reaction analysis (NRA). The ion beam technique and methodologies for the analysis of experimental data provide a comprehensive tool for studying crystal defects.

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. 7. 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.

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 [6].

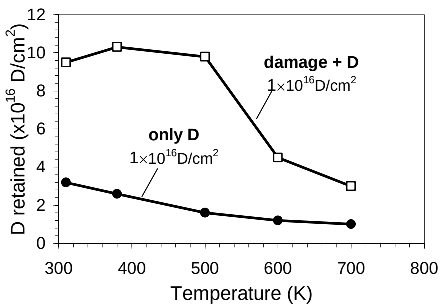


Fig. 7 Quantity of deuterium trapped in steels EI-852 with deuterium implanted to the dose $1.10^{16}cm^{-2}$ without and with preliminary implanted to dose $5.10^{16}cm^{-2}$ argon.

3.5. Test of materials for MSR and ADS

A key problem of the G-IV program is R&D of construction materials compatible with molten salts and Pb and PBE melts at high temperatures. Ni-Mo alloys (Hastelloys) were used in MSR and ferritic/martensitic chromium steels were applied in reactors with Pb and PBE. Pilot experiments have performed which have to be continued in larger scale but they allowed to get an important information on impact of electron and γ -irradiation on corrosion, compositional and mechanical properties of the tested materials [7]. REMMA microanalysis and SIMS were used to investigate compositional changes of the surface layers of specimens. Results show considerable compositional heterogeneities in the surface layers (Fig. 8).

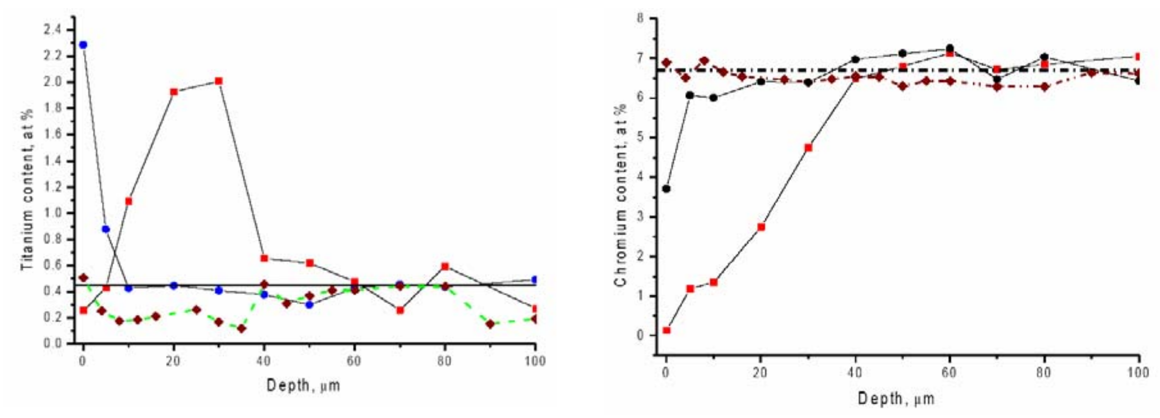


FIG. 8. Redistribution of a) Ti content and b) Cr content in surface layers of irradiated and non-irradiated samples after 700 h exposure in salt (♦) – no irradiation; (•) – E_{dep} =64 eV/at; (■) - E_{dep} =5066 eV/at. (E_{dep}-energy deposited on specimens surface) [7]

5. Conclusions

Effective radiation effects experiments can be performed using ion-beam facilities, because world nuclear society is essential to evaluate and qualify materials for Generation IV systems. Ion-beam facilities are good for studying microstructural and microchemical changes during irradiation as well as corrosion and mechanical properties in many circumstances.

Charged particles irradiations can provide a low-cost method for conducting valuable radiation effects research in absence of, or as a precursor to verification experiments in reactors.
Modern status of using accelerators demand by such main tasks:
• Understanding of radiation damage mechanism of nuclear materials; achievement of better knowledge of the nature of point defects and interaction between them;
• Set up the correlation between radiation-induced defects, structure phase evolution and material degradation mechanism;
• Investigation of stability of systems which have nanoscale features. It is especially important for development and prediction of radiation behavior at high irradiation doses of nano-precipitates in ODS steels, which are the most pronounce materials for of next generation.
• Combining irradiation (reactor+accelerator). In spite on experimental difficulties this method can give the best result in predicting of radiation behavior up to very high doses. Creation of primary defect structure which is typical for reactor irradiation allows to receive on second stage of accelerator irradiation.
• Development of technology forestimating and predicting radiation damage up to doses, needed for reactors of future generations.
Model predictions must be validated with advanced experimental techniques which are able to determine materials properties in a multiscale approach.
A strong cooperation between modelers, experimentalists and designers and the acceptance of a considerable development time is necessary to achieve goal – development of materials which determine the safe and economical operation of running and developed nuclear facilities.

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