

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

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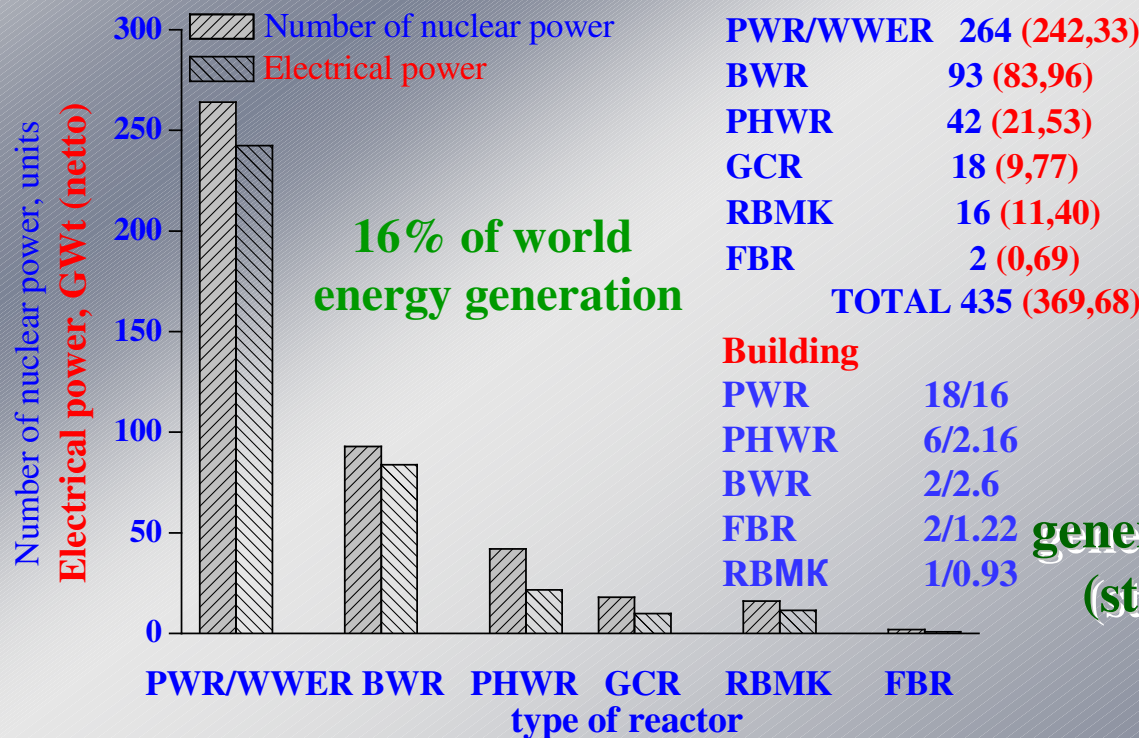
International Topical Meeting on "Nuclear Research  
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## Mission of Nuclear Power

- Today`s nuclear power in the world is the real that possesses humanity for production and supply of low cost electrical and thermal power for long time prospective **with guarantee of nuclear, physical, ecological and technical safety** in amounts corresponding with society needs.

- The key issues of competition with traditional technologies of electrical power production and with alternative (reproducible) sources competition in 21 century are **the safety and economy.**



**Operating nuclear power generating units and electric power (state on 31 of December 2006) (IAEA , 2007)**

## Fuel burn-up and economical characteristics

Now is the one effective - (**real**) way of improvement of technical and economical characteristics of nuclear fuel cycle – the burn-up (in % heavy atoms [h.a.]) increasing (energy, produced from unit quantity of nuclear fuel [GWdays/t]).

### Modern status:

- Light water reactors (LWR) → 45-50 GWtd/t (~5 % h.a.), 8-10dpa
- Fast reactors (FR) → ~75 GWtd/t (10-12 % h.a), 80-90dpa

### Targets:

- Light water reactors (LWR) → 75-80 GWtd/t (~ 8% h.a), 12-15dpa  
→ 100 GWtd/t (~ 10-11% h.a), 18-20dpa
- Fast reactors (FR) → ~ 200 GWtd/t (20-25 % h.a), > 200dpa

## **Radiation stability of structural materials**

- The main problem of achievement of these targets is radiation stability of structural materials.
- The radiation stability (R st) is the ability of the material to resist to the influence of intensive fluxes of radioactive irradiation that causes the structure-phase changes and degradation of initial physical-mechanical properties.
- Creation of radiation resistant materials is very complicated due to: **the insufficiency of our knowledge on nature of radiation-induced phenomena and material damage practically non investigated range of very high irradiation doses.**
- 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, particular *ion and electron irradiation.*

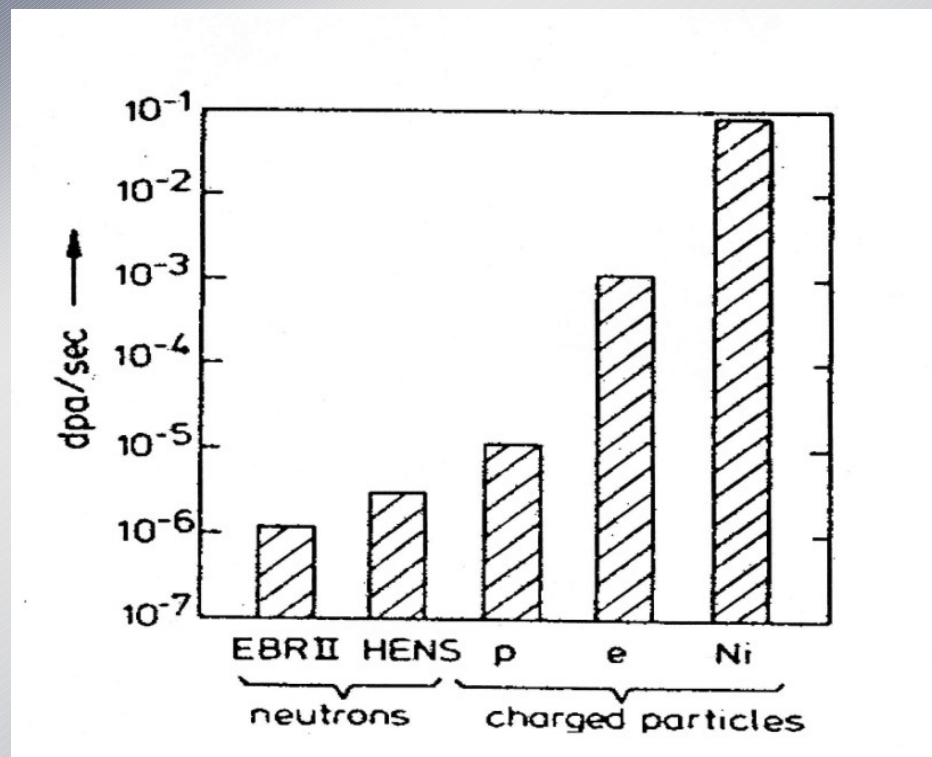
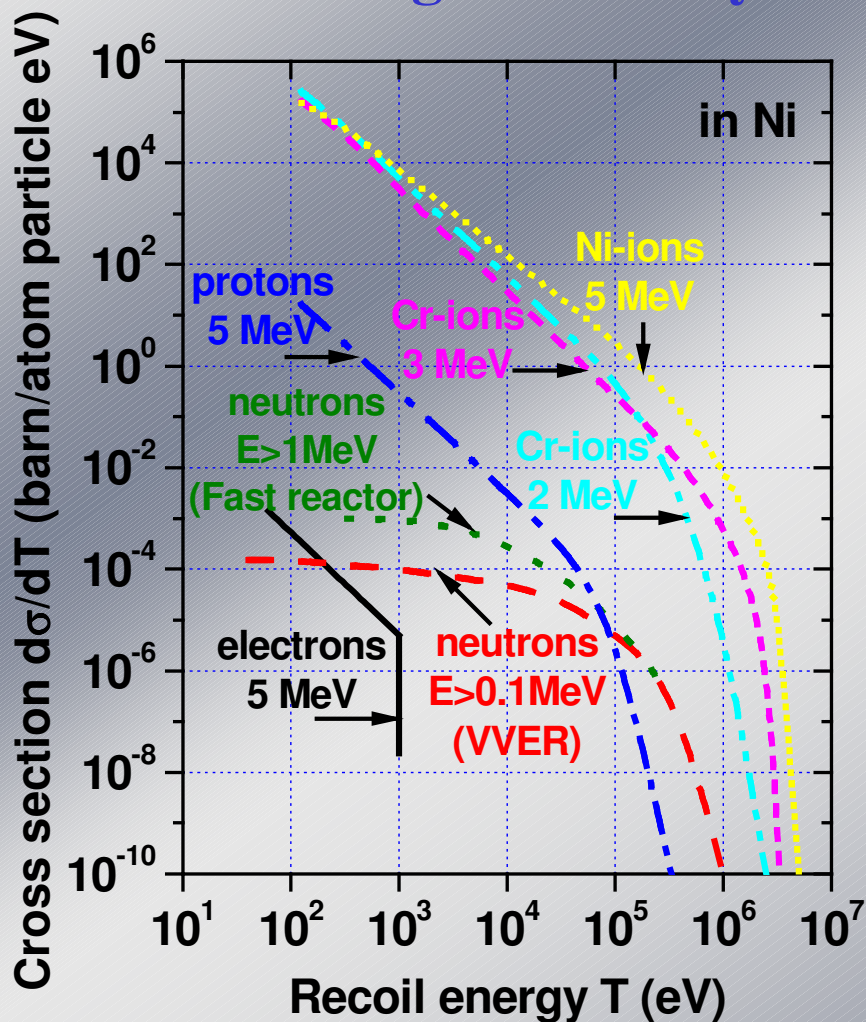
## Advantage of ion and electron simulation

### Why accelerators are needed?

- Higher damage rate ( $10^{-4}$ – $10^{-2}$  /accelerator/ vs  $10^{-6}$ – $10^{-10}$  dpa/s /reactor/)
- Good control of experimental parameters (temperature, flux and environment), possibility of parameters separation
- Ideally suited for optimizing minor alloying composition
- Only one possible choice in the absence of high flux neutron irradiation facility. Many nuclear facilities are shut down now (FFTF, RAPSODIE, DFR, PFR, Superfenix, EBR-II, BR-10, BN-350 etc.)
- Irradiated specimens are not radioactive, unlike reactor specimens which are highly radioactive and may have to be handled only in hot cells

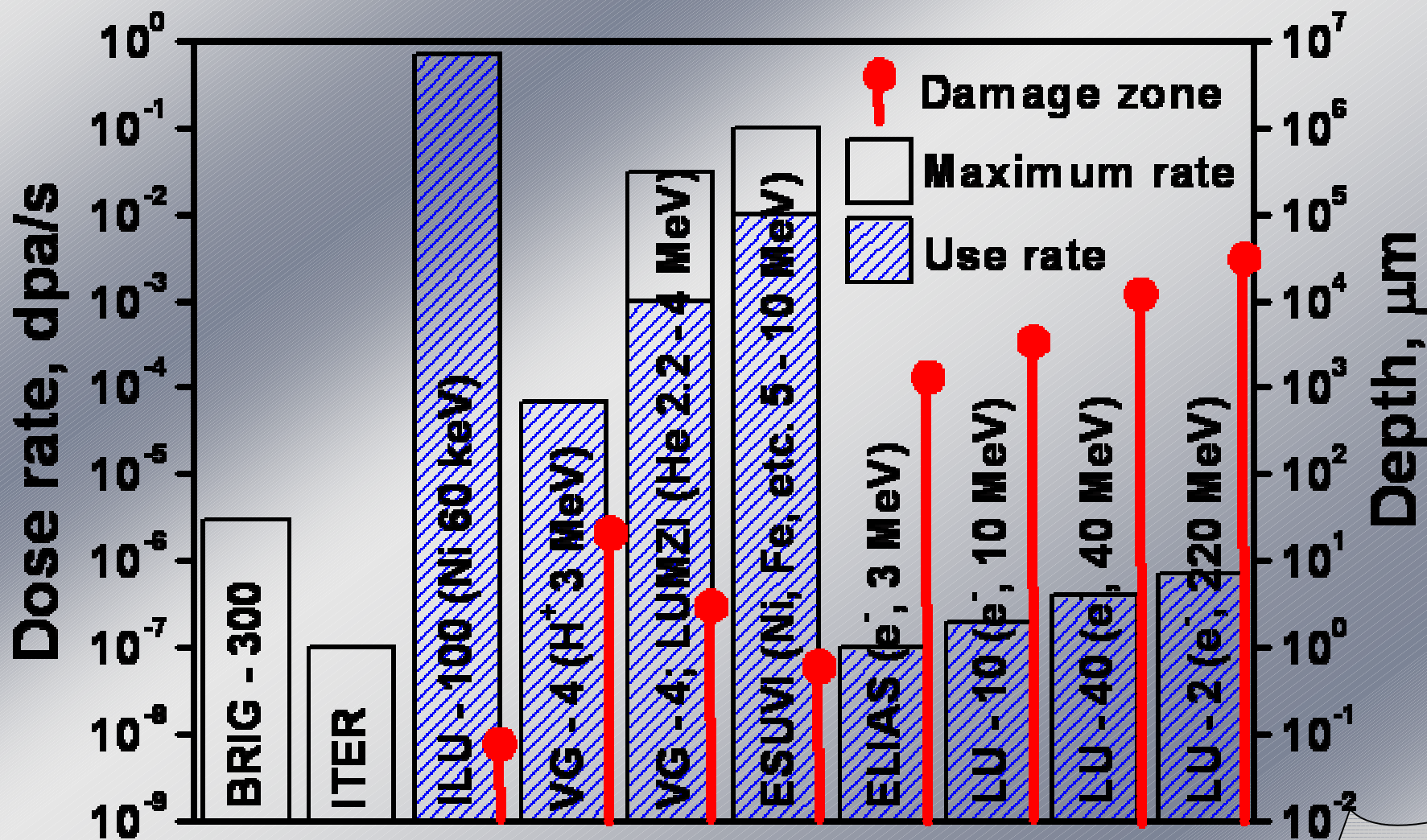
# ION SIMULATION OF NEUTRON DAMAGE

## Damage Efficiency



ION BEAMS CAN PRODUCE  
 DISPLACEMENT DAMAGE AT A MUCH  
 FASTER RATE THAN NEUTRONS

## KIPT ion and electron accelerators



## **Disadvantage of simulation experiments.**

- **Difference in recoil spectra-different primary damage**
- **Problems of phase stability at high dpa rate**
- **Injected interstitial effect**
- **Stress induced by irradiation –surface proximity**



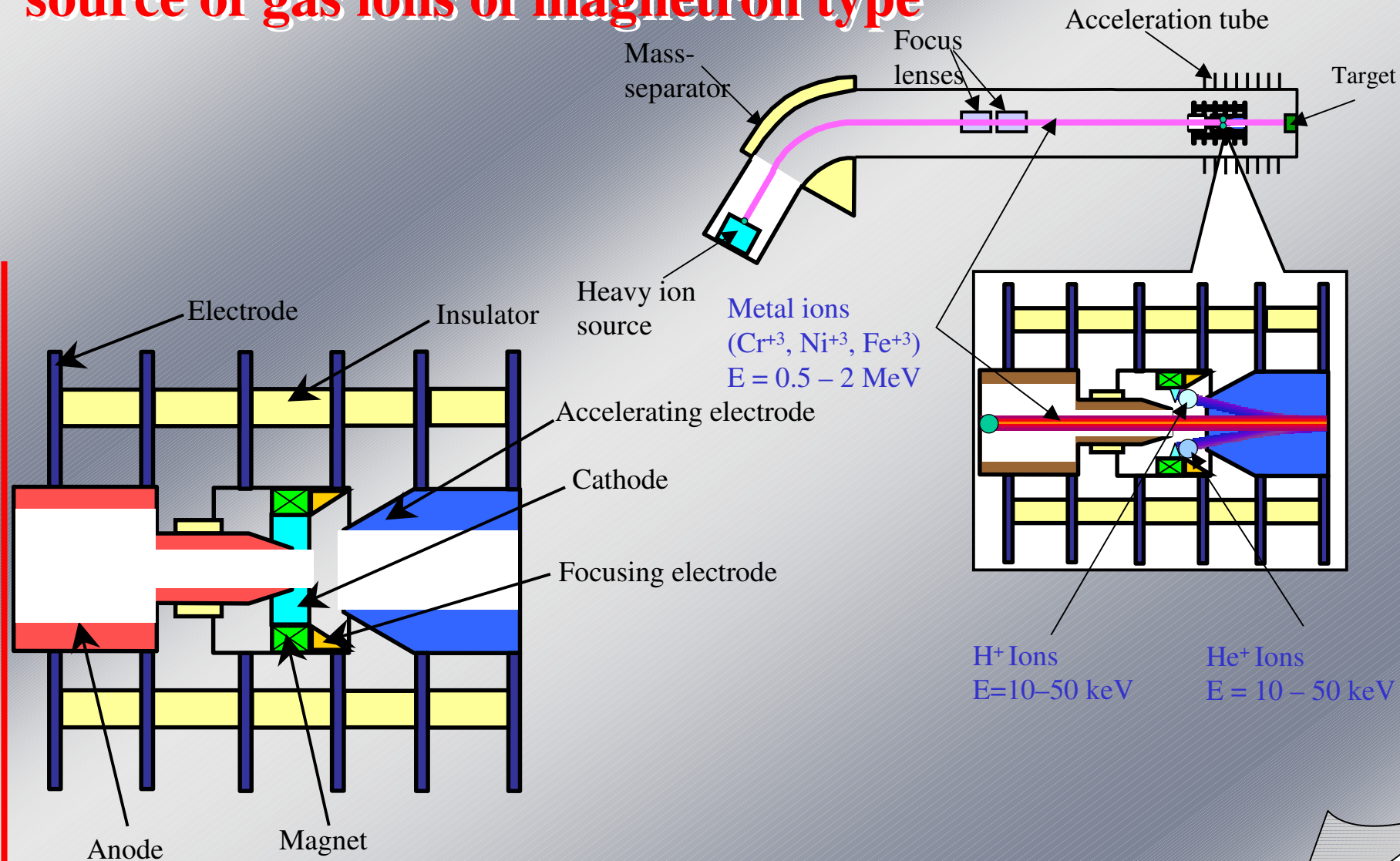
## Modern status of simulation experiments

The using of new types of accelerators (of two and three beams) and of modern methods of research:

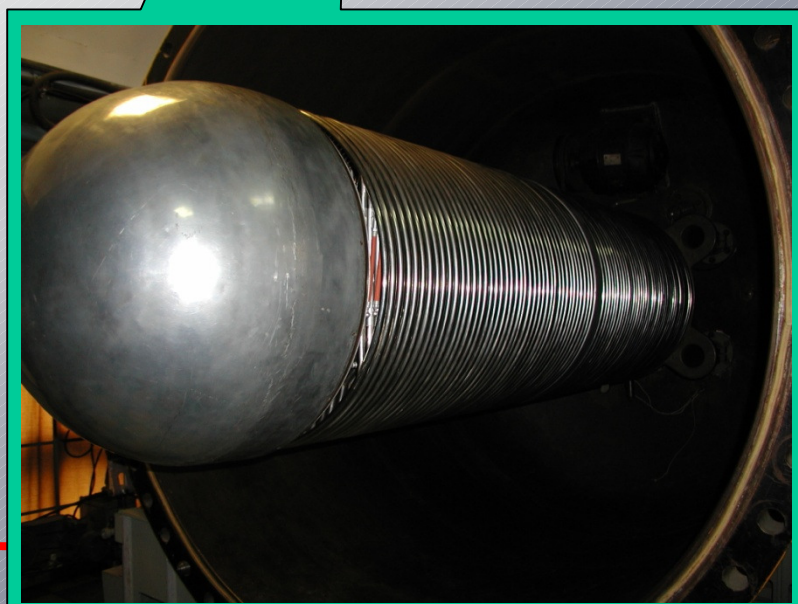
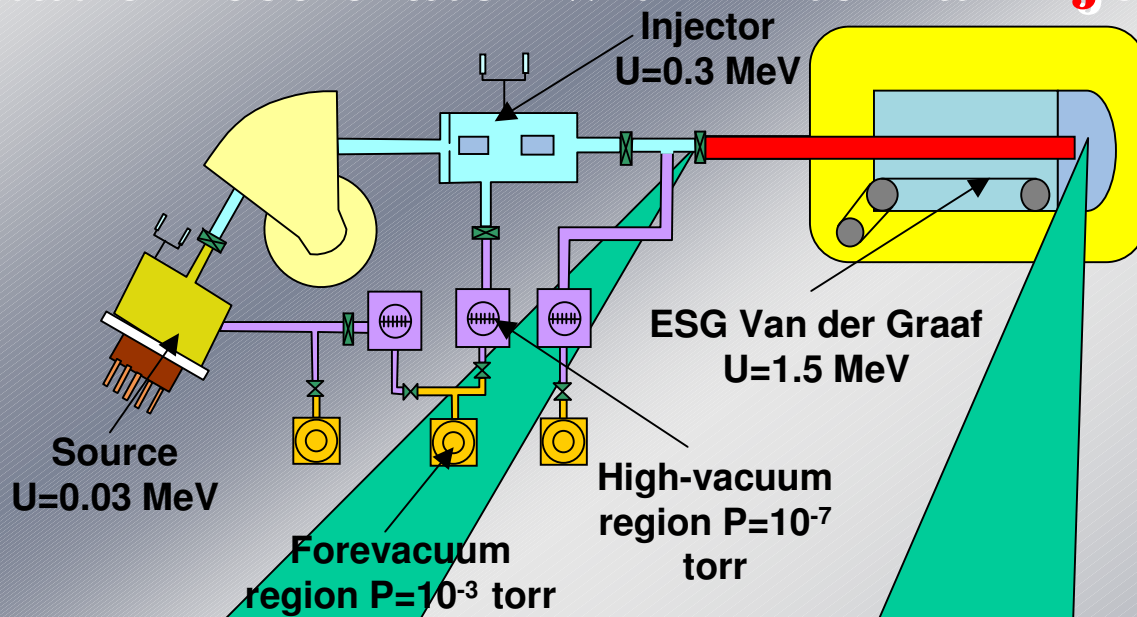
- TEM (Loops, cavities, precipitates)
- STEM + EDS + EELS (Grain boundary segregation)
- TAP (Solute clusters)
- AES, XPS (Surface segregation)
- FIB and nano indenters (microspecimen technologies needed for development of new materials and materials testing for fission and fusions)
- EXAFS, SANS (nano scale structural evolutions)
- nuclear-physical methods (RBS + channeling)

allows minimize the restrictions and disadvantages in the using of results of simulation experiments caused by low depth of damaged layer.

# Accelerator ESUVI ion guide and hollow source of gas ions of magnetron type



# Electrostatic Accelerator with External Injector (ESUVI)

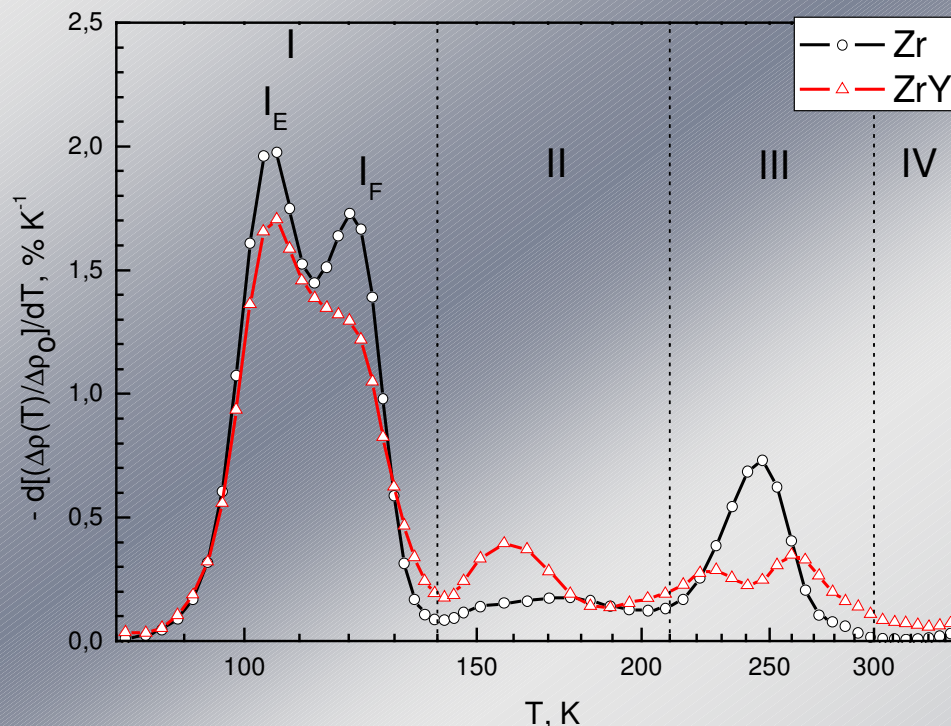


## **Main tasks of simulation experiments:**

- 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 and future reactors (swelling and embrittlement). Observation of radiation-induced microstructure such as segregation and hardening.
- Microstructural predicting for possibilities of Life extension for exploiting 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 (also PVI materials).
- Investigation of stability of systems which have nanoscale features, particular stability of nanoclusters in ODS steels.

## Investigation of fundamental processes

[V. Borisenko, 2009]



Spectra of isochrone annealing of Zr and alloy Zr-Y irradiated by electrons with energy 2 MeV at temperature 82K to fluence  $1,4 \cdot 10^{19} e/cm^2$

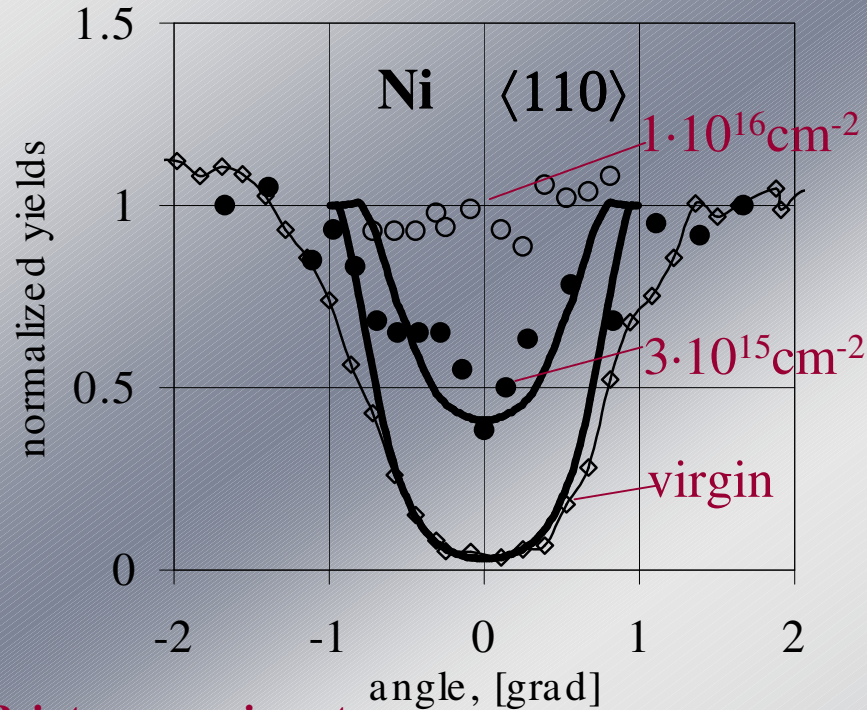
On substages IA, IB and Ic recombination of near pairs interstitial-vacancy occurs  
Annealing in zirconium on substages I and IF occurs at the expense of free migration of interstitials

On stage II processes caused by release of interstitial atoms from impurity traps are observed.

According to the one-interstitial model the annealing on stage III is due to the free migration of vacancies.

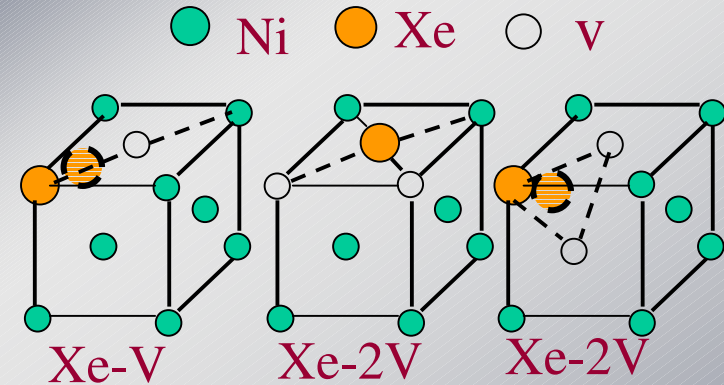
# Lattice location (experiment plus MD simulation)

[G.Tolstolutsкая et.al., 2008]

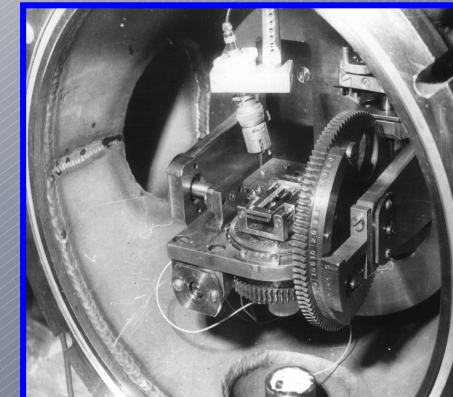
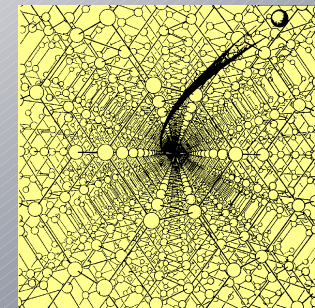
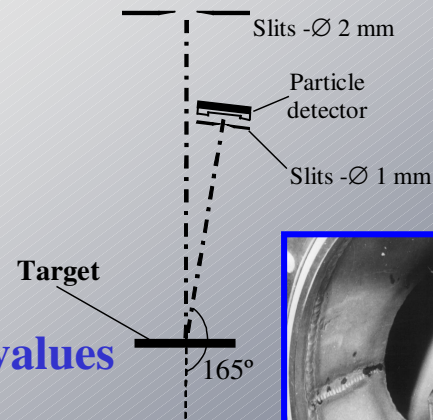


Points – experiment,  
 lines - calculations

The best fitting of experimental and of calculated values of angular dependencies is obtained when under irradiation dose of  $3 \cdot 10^{15} \text{cm}^{-2}$  85% of Xe atoms are in the complexes Xe + v, 15% - in complexes Xe + 2v. Under the dose of  $6 \cdot 10^{15} \text{cm}^{-2}$  60% of Xe atoms form the complexes Xe+2v.



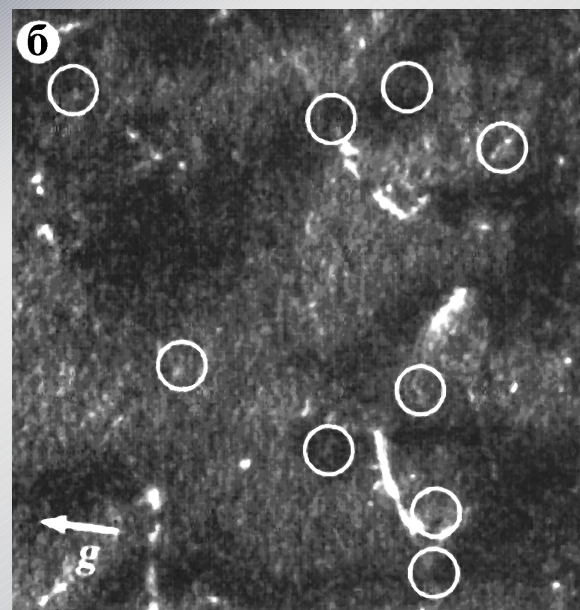
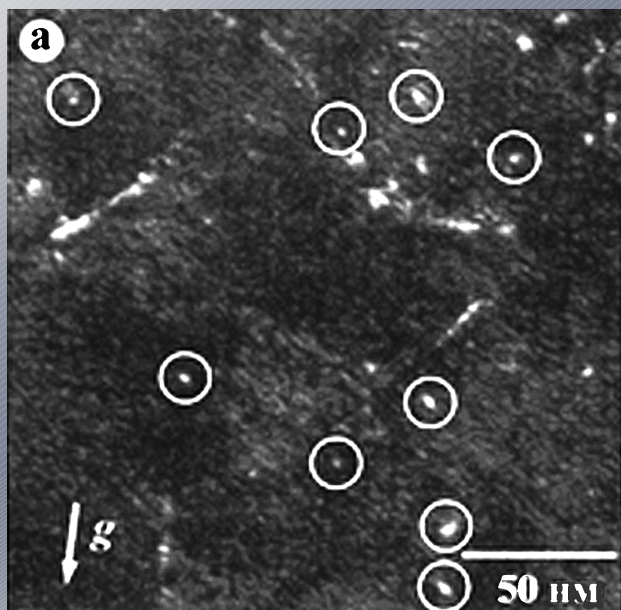
Van de Graaff accelerator  
 $^1\text{H}$ ,  $^4\text{He}^+$ , 1,6 MeV - RBS



## Dislocations behavior in A533 steel

A533 (Ni<sup>+</sup> 3MeV, D=1 dpa, T=290°C)

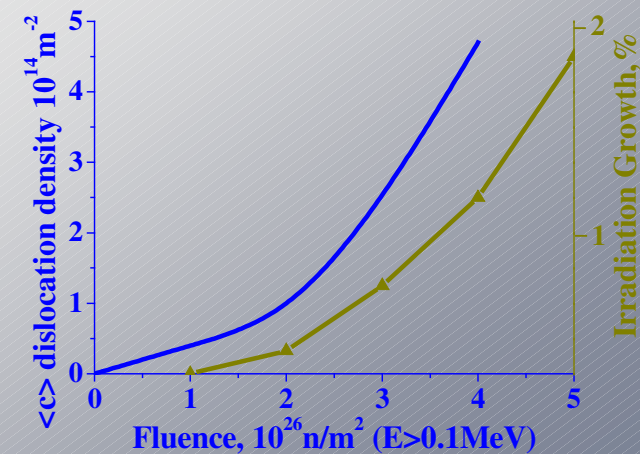
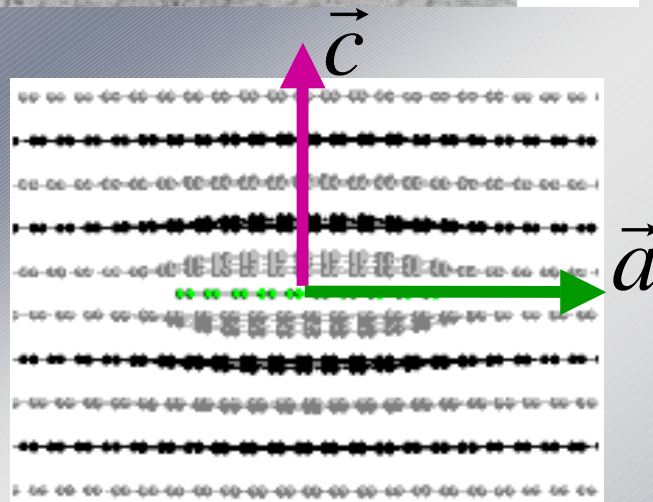
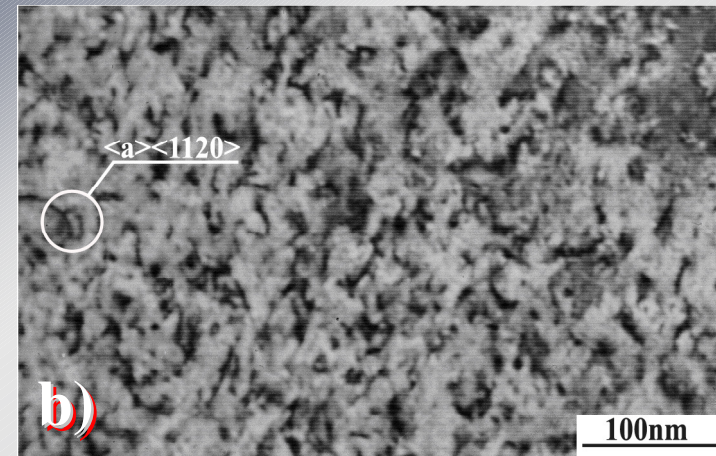
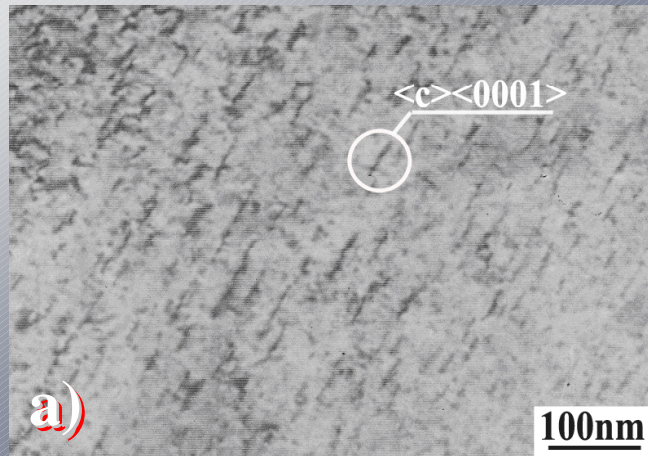
Average diameter of loops  $d=2.5$  nm ,  $\rho=10^{16}$  cm<sup>-3</sup> [K.Fuji, 2004]



Dislocation loops images in different diffraction conditions: a)  $g = 020$ , b)  $g = 200$ .

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

# Radiation behaviour of Zr-base alloys ( $Zr^{6+}$ , 1.8MeV, 15dpa, 560°C) [R.Vasilenko et.al., 2008]

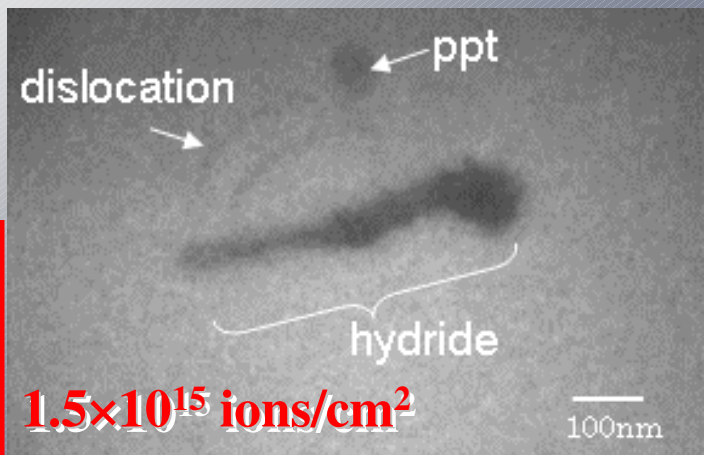


Strong influence of oxygen on suppression of number density of c-type loops; oxygen content: 0.08 (a) 0.19 wt.% (b)

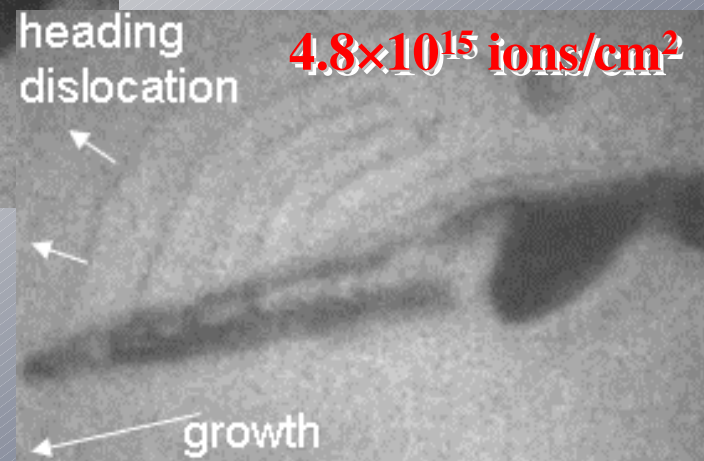


## Microscopic evolution of Zr hydride in Zircalloy-4

[Y.Shinohara et.al., 2007]



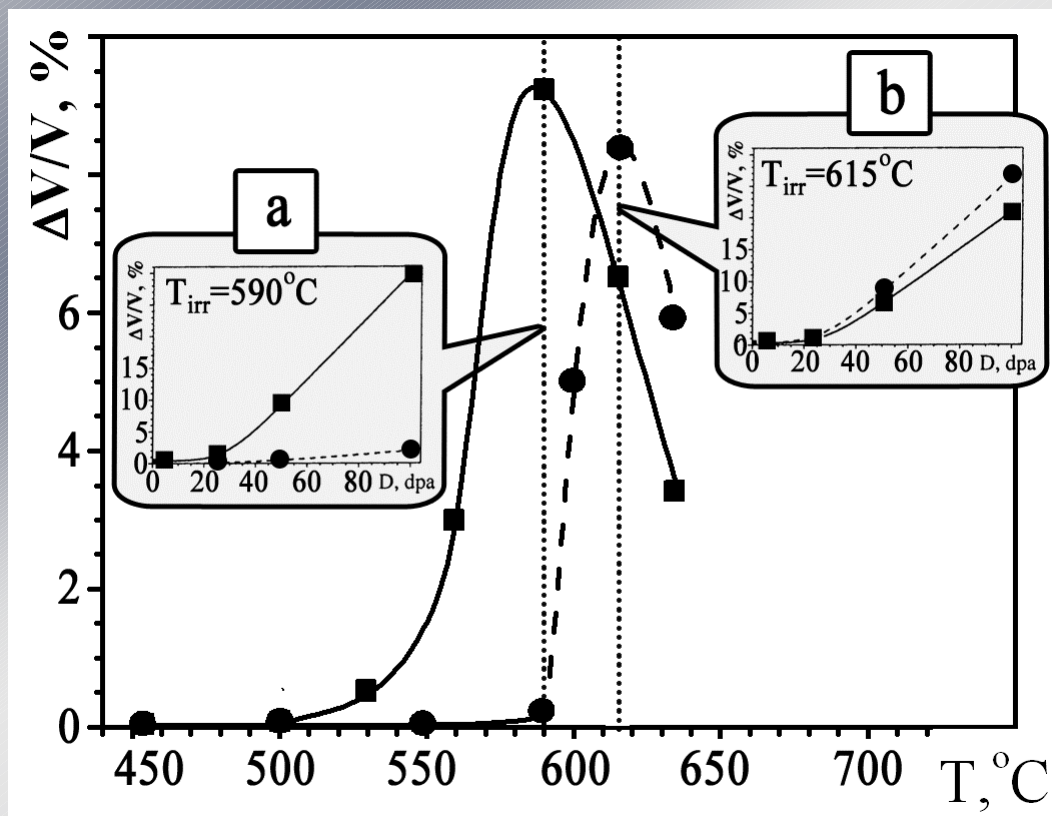
The dynamic process of the formation of Zr hydrides accompanied with dislocations around hydrides was observed.



Growth of a intra-granular Zr hydride in Zircalloy-4 under 150 keV H<sub>2</sub><sup>+</sup> irradiation, B=z=[0001], g = [1010]

## Temperature and dose dependences of swelling of solution-annealed stainless steel 18Cr-10Ni-Ti (D= 50 dpa)

[V.Bryk, A.Kalchenko et.al., 2009]

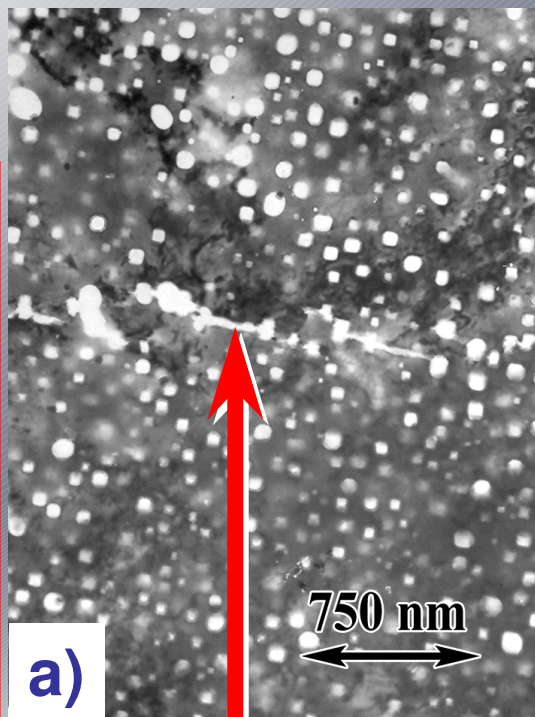


$\text{Cr}^{3+}$ , 2 MeV, the rate doses are:  $1 \times 10^{-3}$  dpa/s (■) and  $1 \times 10^{-2}$  dpa/s (●);  
a)  $T_{irr} = 590^\circ\text{C}$  and b)  $T_{irr} = 615^\circ\text{C}$

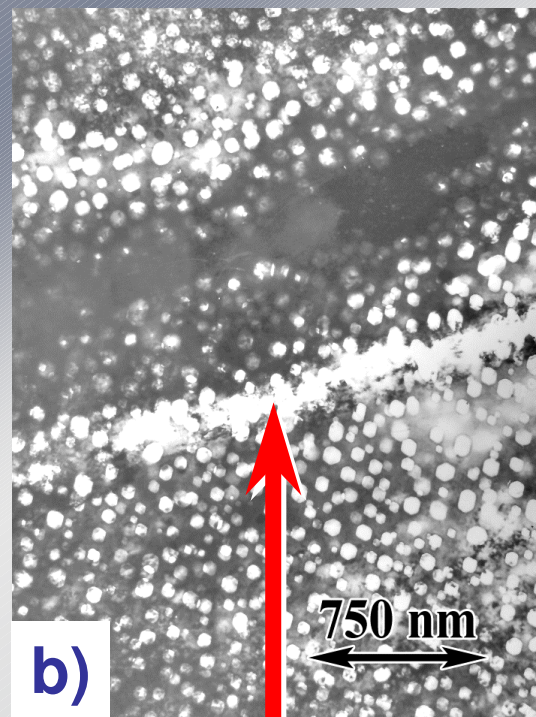
## Failure of material of PVI due to the high swelling

(Cr<sup>3+</sup>, 2 MeV, 100 dpa, 600°C, ε=7%)

[A.Parkhomenko, O.Borodin et.al., 2004]



a) cracks in matrix



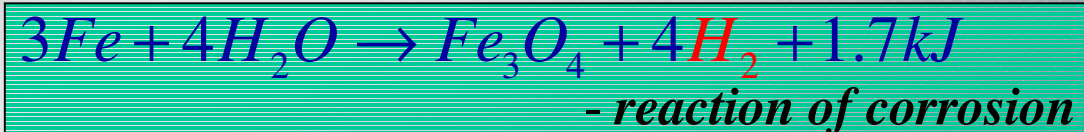
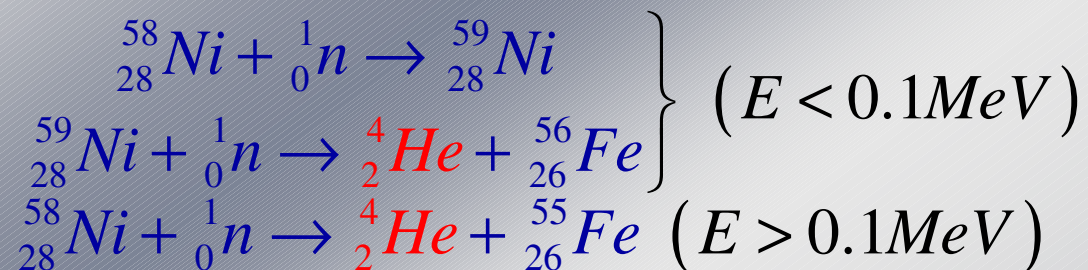
b) failure along the grain boundaries

In the grain body the cracking proceeds due to the localization of sliding on void network (fig. a).

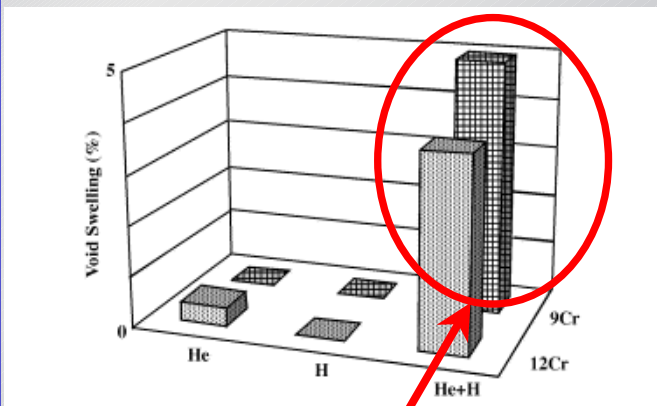
In near boundary sites the failure proceeds on the distance of 0.4-0.5 μm from boundary due to the presence of increased concentration of voids in these sites (fig. b).

## Complex synergies call for aggressive exploratory research

Sources of helium and hydrogen in nuclear reactors are the nuclear reactions in nickel containing materials under neutron influence:

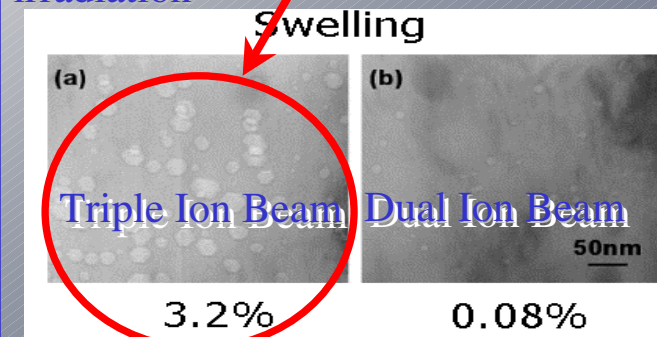


The synergistic effect of He and H was shown clearly in the triple ion ( $\text{Fe}^{3+} \text{He}^+ \text{H}^+$ ) irradiation



T. Tanaka et al. JNM 729-333(2004)294

The average swelling in F82H steel was significantly enhanced by the triple ion irradiation

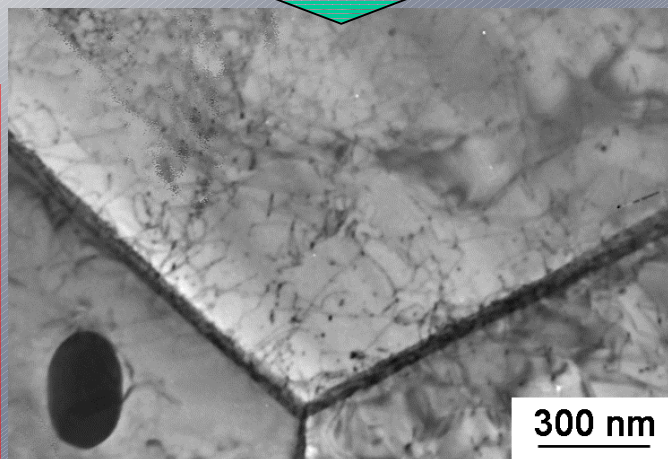


E. Wakai, et al. JNM 307-311(2002)278

# The synergistic effect of radiation damage and helium + hydrogen [J.Fomenko, V.Voyevodin et.al., 2008]

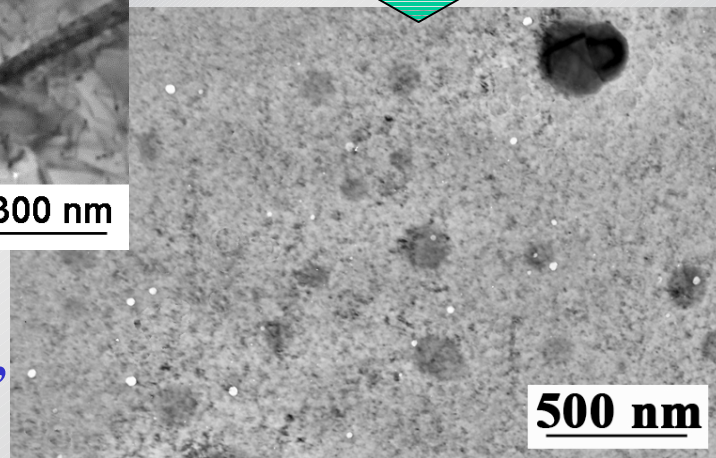
## Microstructure of steel EI-852 before and after irradiation

**Initial  
microstructure**



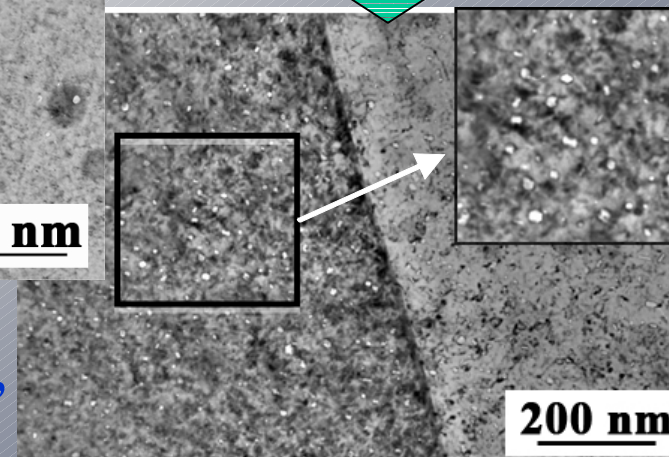
$S=0.01\%$ ,  
 $p=4,0 \cdot 10^{13} \text{cm}^{-3}$ ,  
 $d=15-60 \text{ nm}$

**Cr<sup>+5</sup>  
( $T_{\text{irr}}=480^\circ\text{C}$ ,  
 $D=100\text{dpa}$ )**



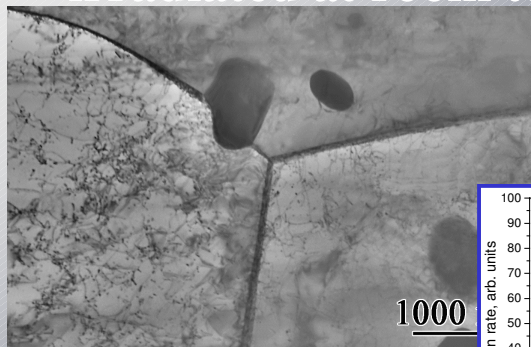
$S=0.4\%$ ,  
 $p=2.2 \cdot 10^{16} \text{cm}^{-3}$ ,  
 $d=10-50 \text{ nm}$

**Cr<sup>+5</sup>  
4800 appm He,  
6000 appm H<sub>2</sub>**

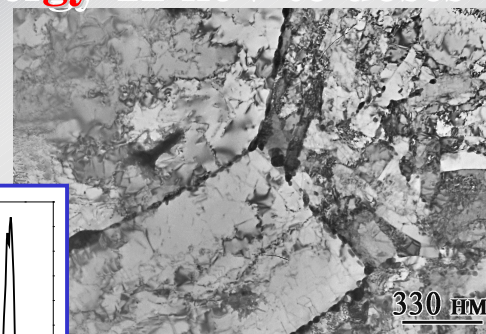
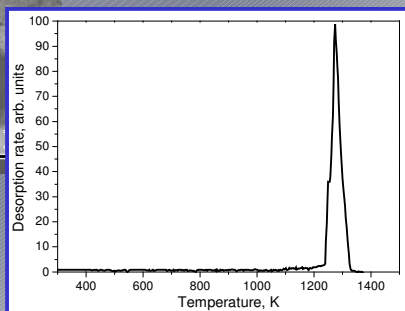


**Spectra of thermal desorption of helium from steels EI-852 and EP-450 irradiated at room temperature by helium ions with energy 12 keV to doses  $5 \cdot 10^{16}$  part./cm<sup>2</sup>**

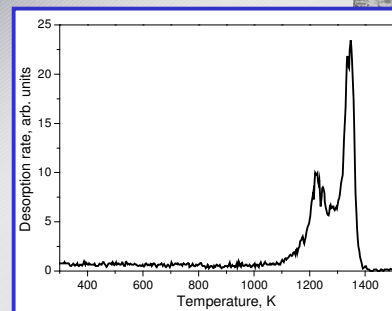
[V. Ruzhytskiy et.al., 2008]



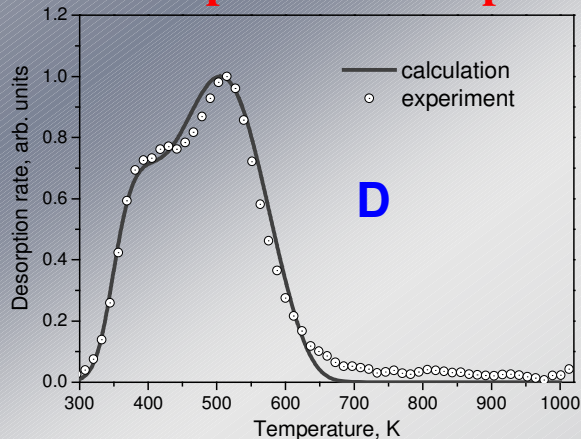
Steel EI-852



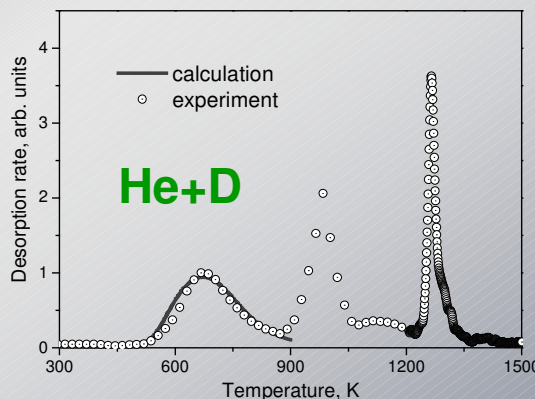
Steel EP-450



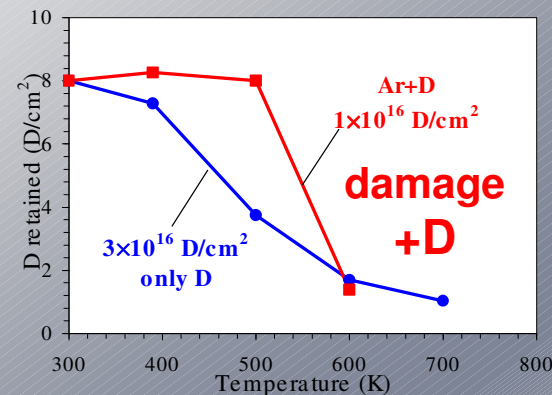
**Linear temperature ramp thermal desorptions: experimental curves with calculation data**



Deuterium (5 keV) is trapped by traps of two types with the binding energies:  $0.57 \pm 0.08$  and  $0.71 \pm 0.1$  eV.

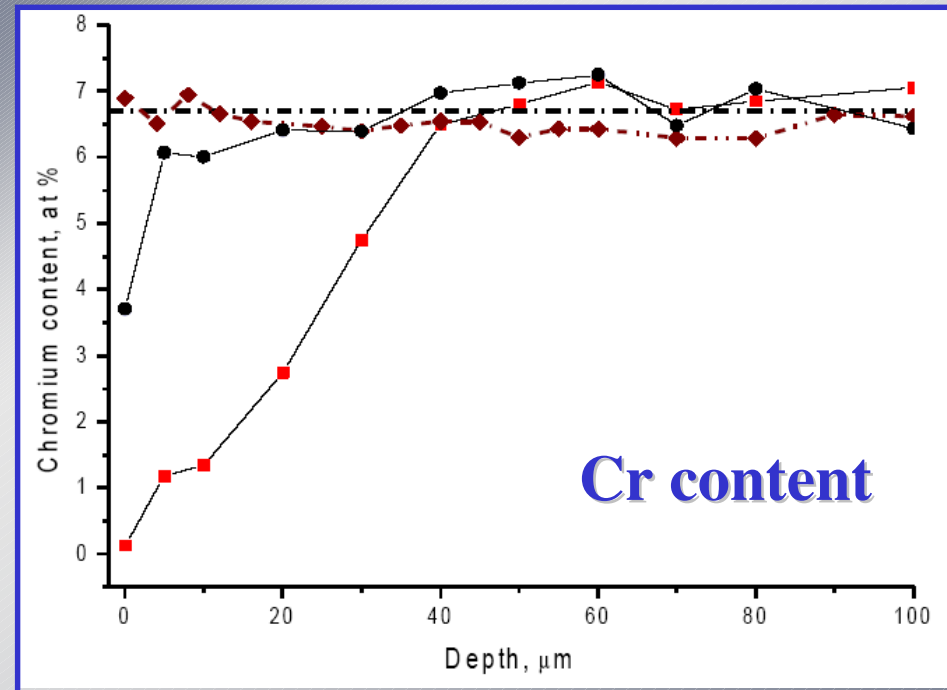
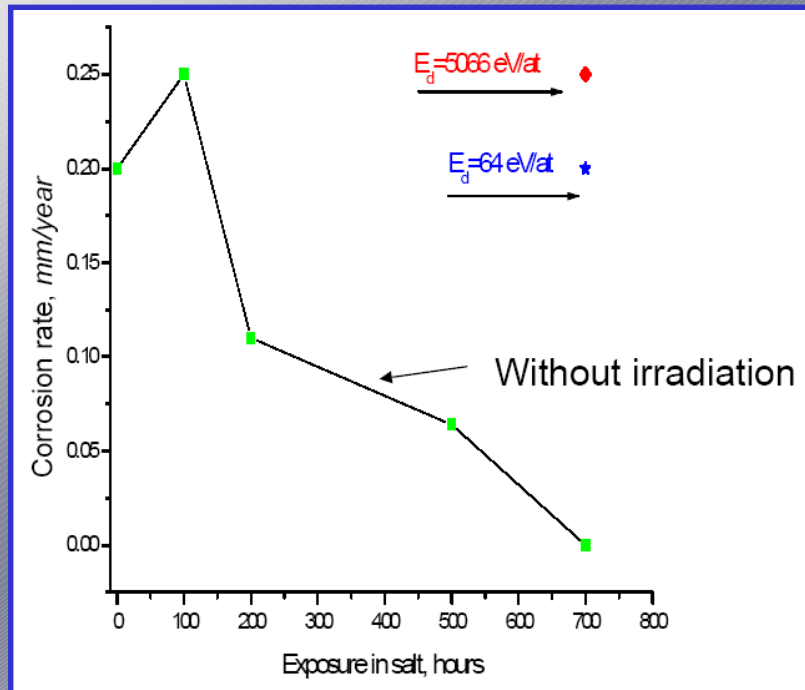


Pre-implantation of helium (12 keV) shifted the temperature range of its release from steel EI-852 from 300-600 to 600-800K.



At argon (1.4 MeV) pre-irradiation deuterium is retained in EP-450 to annealing temperatures of ~500-600 K the level of deuterium retention is strongly increased.

# Corrosion kinetics and composition changes of alloys in molten salts [V. Azhazha et.al., 2005]



Corrosion rates of the alloy depending on the exposure time in salt at 650°C

$e^-$ , 10 MeV

Redistribution of 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)

## 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 the next generation.



## Conclusions (continued)

- 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.**

**Thank you for your attention!**