Simulation of radiation damage in tungsten, molybdenum and iron using high energy protons, fission and fusion neutrons

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Outline

Damage caused by fusion neutrons is one of the key DEMO lifetime limiting issues for in-vessel components of any material used in a fusion reactor. Power fusion neutron source does not exist yet.

What is the best approach to simulate fusion neutron-induced damage in materials?

- **Charged particles**
  - **Protons**
    - **<21.4 MeV>**
  - **Heavy ions**
    - **<20 MeV>**
- **Neutrons**
  - **Fission (LVR-15)**
    - **<0.2 MeV>**
  - **high energy (p(35 MeV)-Be)**
    - **<8.7 MeV>**

Outline

Damage occurs by two main mechanisms

Displacement of atoms

Transmutation

from one element to another element
from one isotope to another isotope

☐ **PALS is a powerful tool for primary defect study: it allows us to detect defect distribution on size and defect concentration**

☐ **For long-term irradiation: combination of PALS, TEM, APT (atom probe tomography) and TDS (thermal desorption spectroscopy) is essential**

☐ **γ-ray energy spectra of irradiated samples**

Primary defect detection

Res (electrical Resistivity)	TEM (transmission electron microscopy)	PALS (positron annihilation lifetime spectroscopy)	x-ray diffraction
+ low density	+ low density	+ low density	- high density
- no small defect	- no small defect (>2 nm)	+ small def (0.1 nm)	- no small def (>10nm)
- No defect size	+ Defect size	+ Defect size	- No defect size

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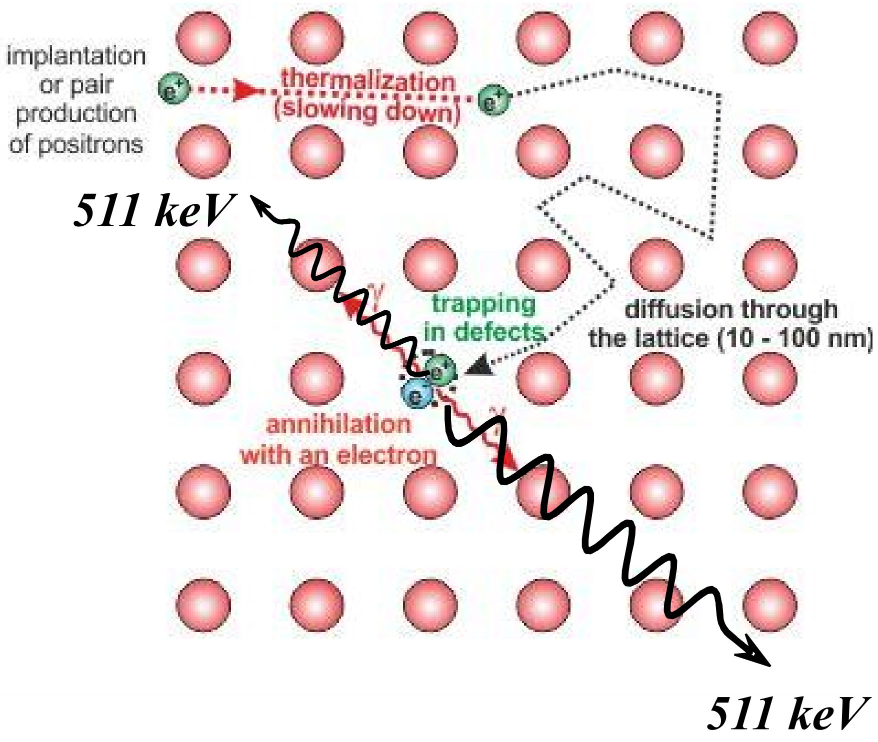
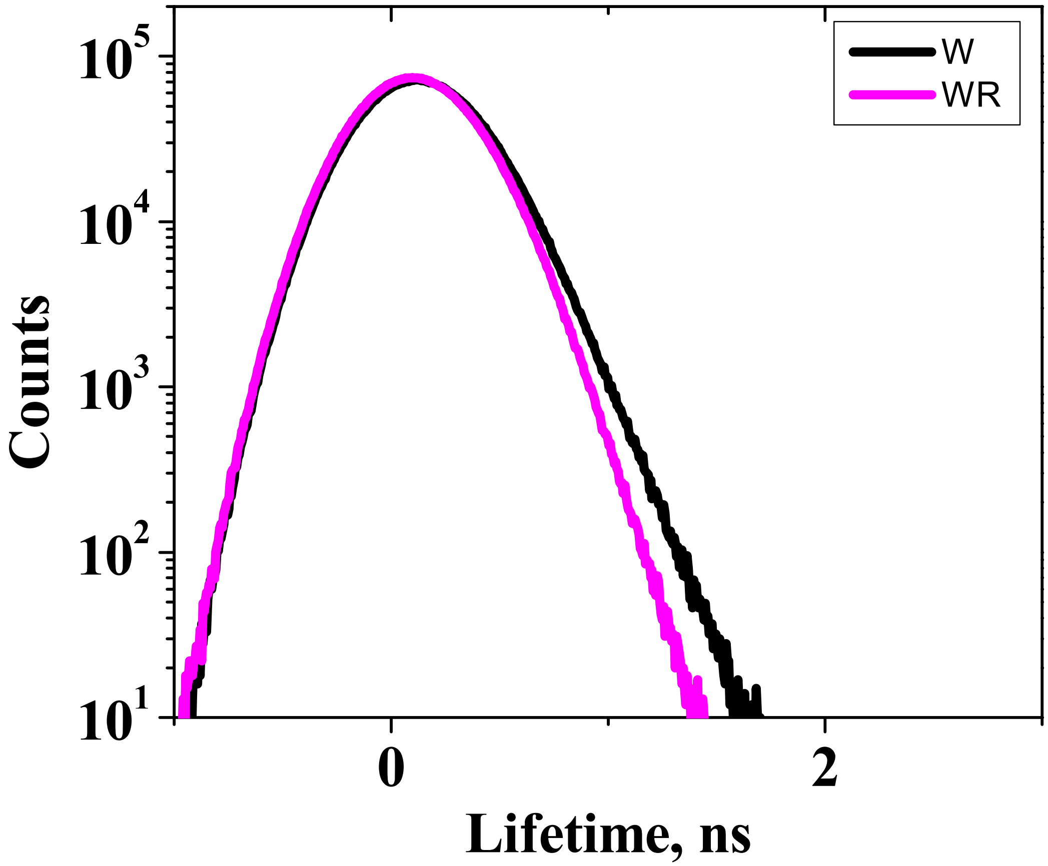
Positron annihilation lifetime spectroscopy: initial defects

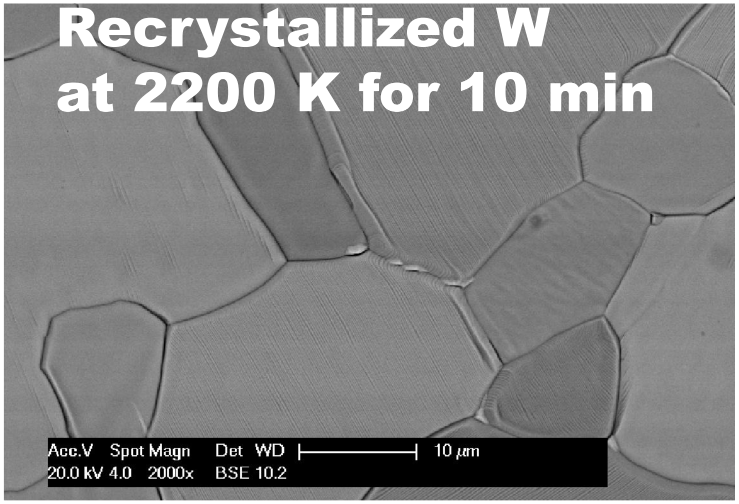


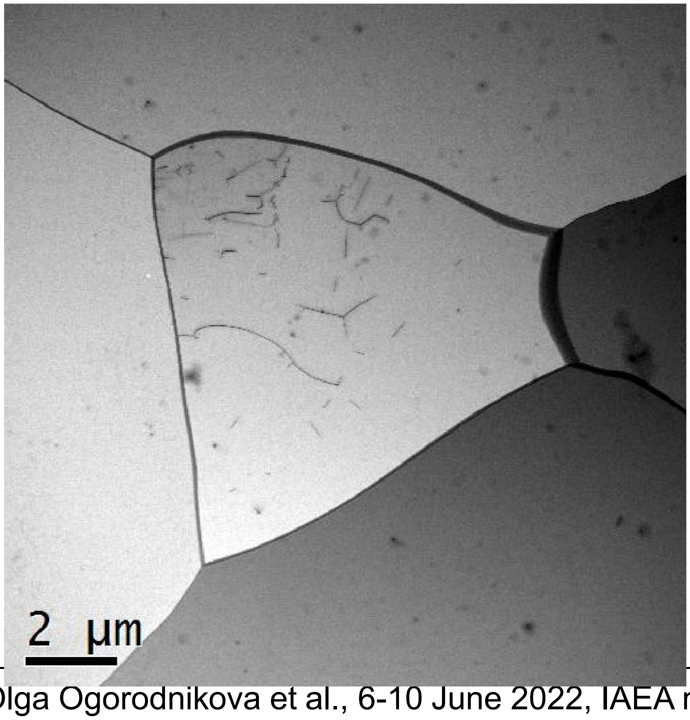
$e^{+}+e^{-} \rightarrow 2\gamma$ (511 keV)



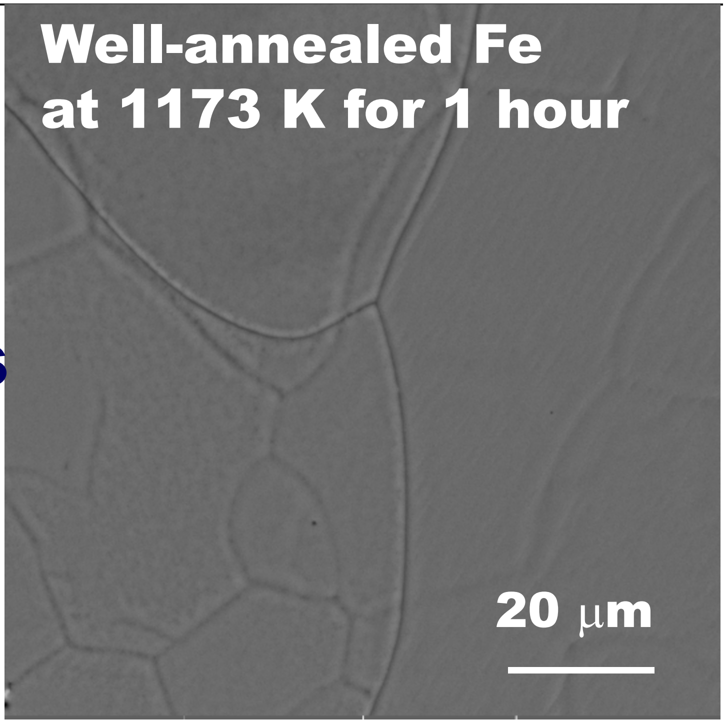
- Defect-free sample: lifetime 105 ps was found for recrystallized W and 110 ps for well-annealed Fe samples

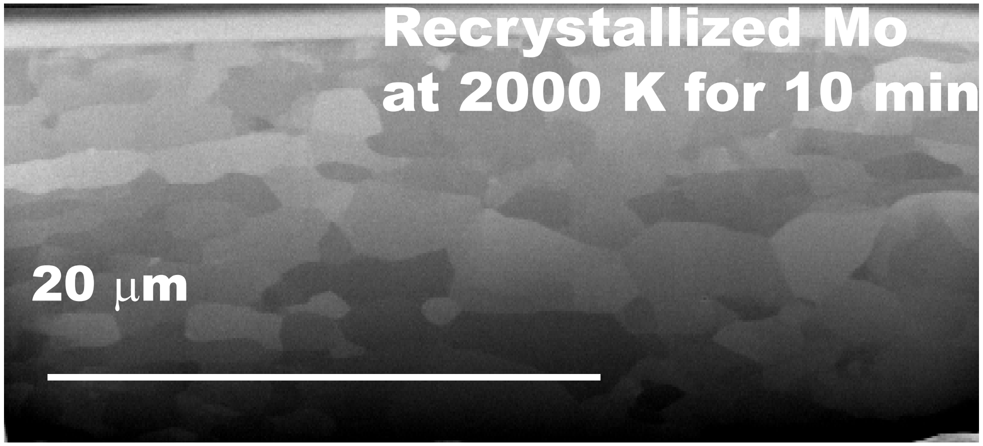
Well-annealed samples: W, Fe and Mo


Recrystallized W at 2200 K for 10 min

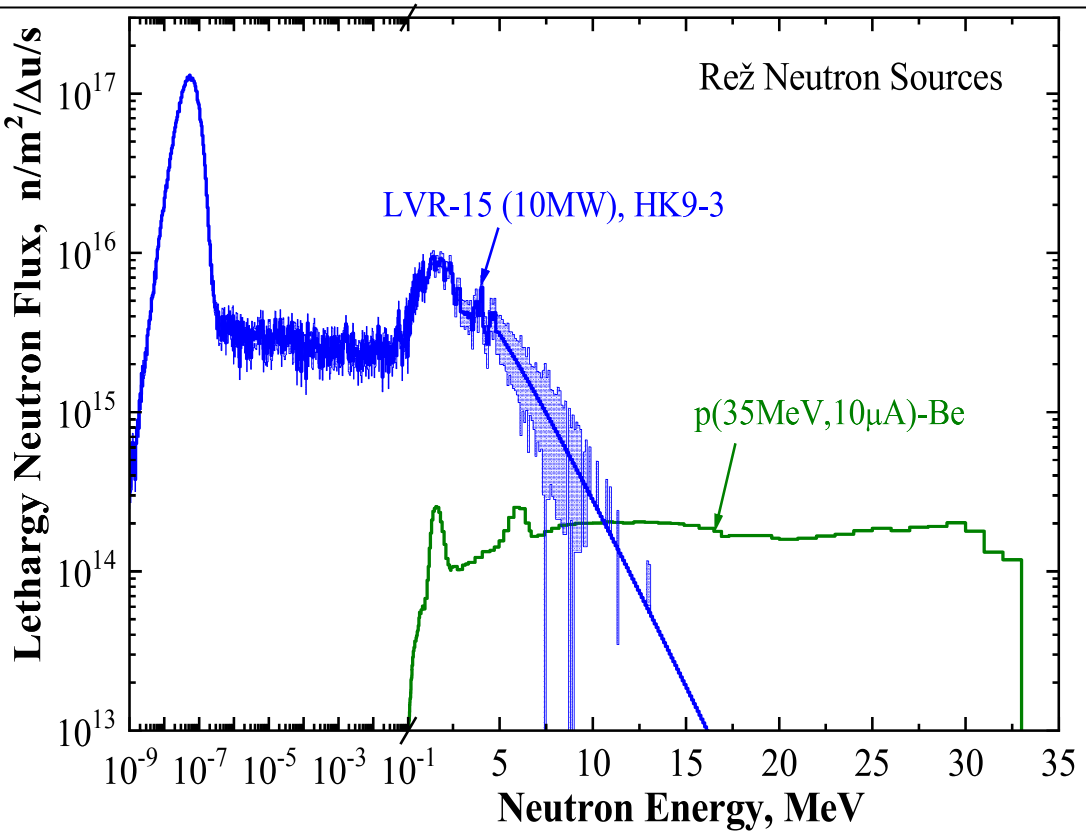


Dislocations $< 10^{13}$ m^{-2}


Well-annealed Fe at 1173 K for 1 hour


Recrystallized Mo at 2000 K for 10 min

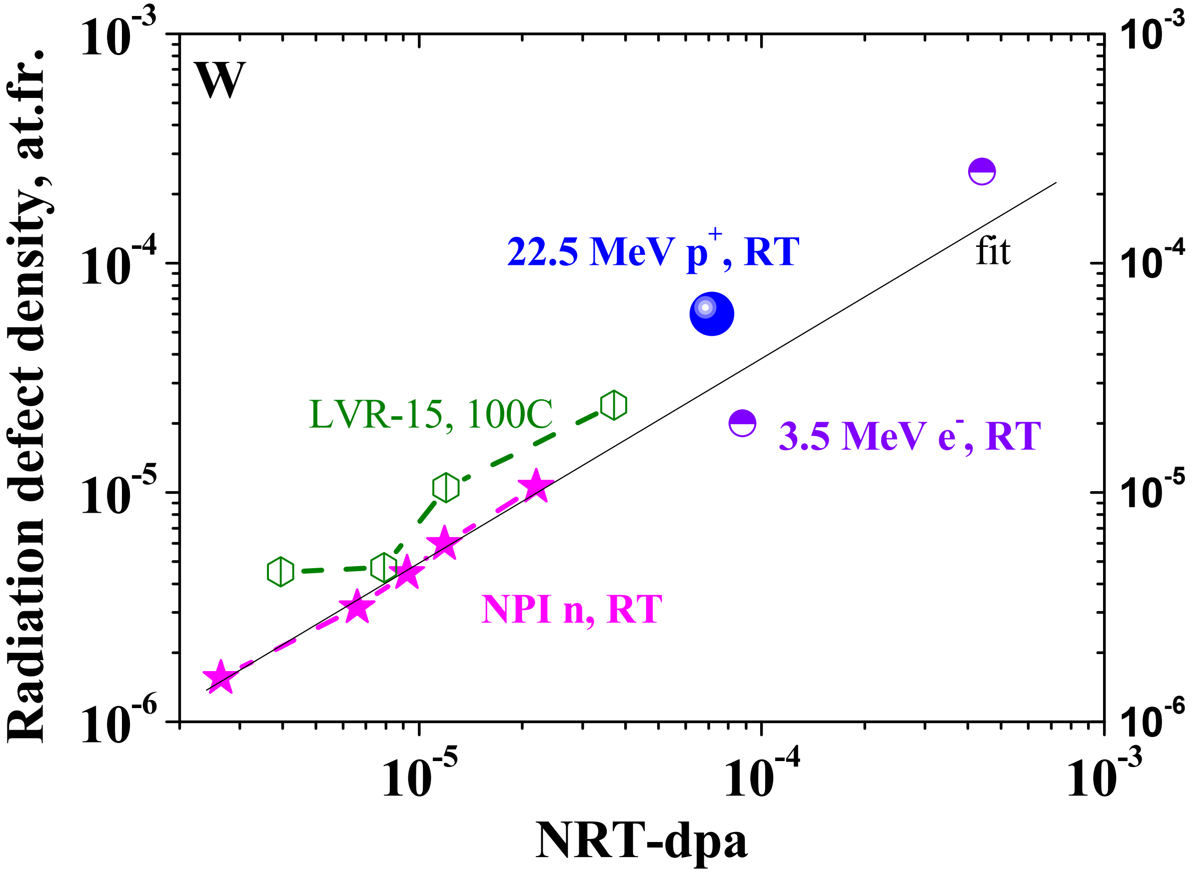
Irradiation of ‘defect-free’ samples



Wide-spectrum high energy neutron irradiation was carried out at a source of fast neutrons with continuous neutron spectra up to 33 MeV generated by a p(35 MeV) beam incident on a thick Be target at the U-120M cyclotron of Nuclear Physics Institute (NPI CAS) in Řež.

The energy spectrum of neutrons in the reactor and neutrons produced by the p(35 MeV)-Be source are different.
There is a significant fraction of thermal and epithermal neutrons in the reactor neutron spectrum. Thermal and epithermal neutrons do not have sufficient energy to produce a considerable concentration of vacancy-type defects.

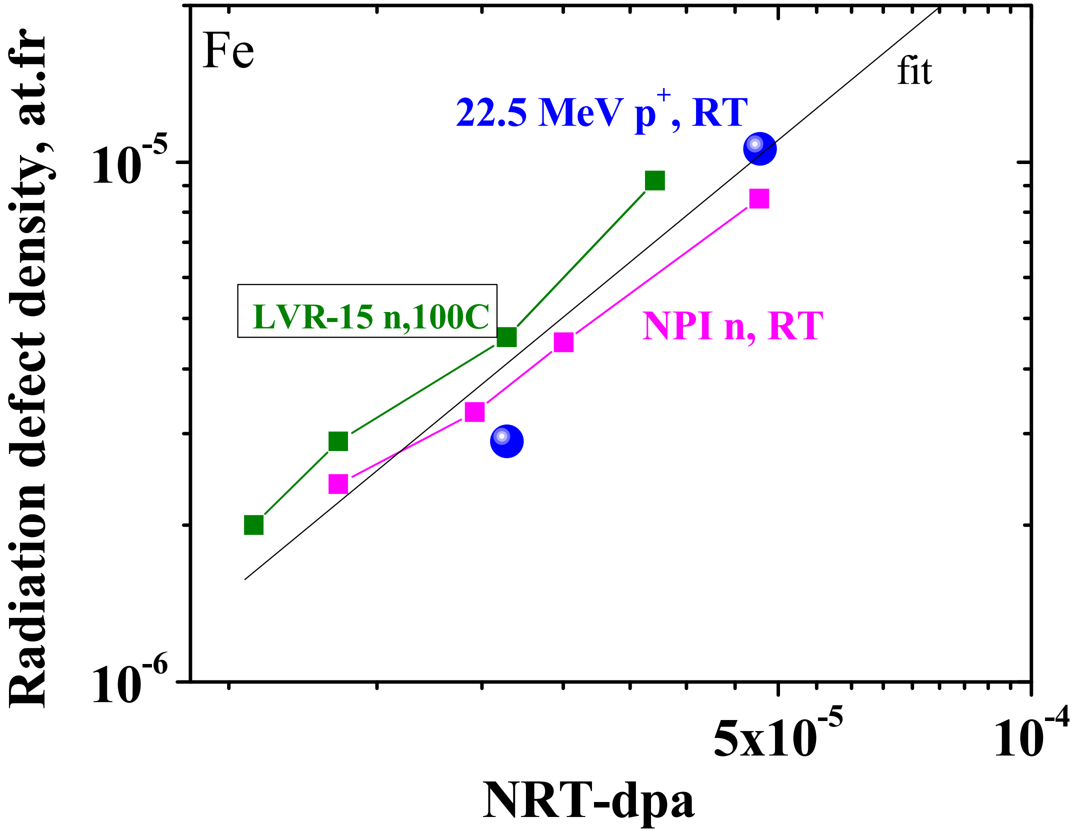
Irradiation of ‘defect-free’ samples: W



❑ Defect density in W irradiated with p+ , fission and energetic neutrons at low doses is close to each other and is well described by a linear fit (~dpa$^{0.56}$).

❑ The dpa is a good parameter for quantification of the total number of **primary defects** in a material caused by different types of irradiation. (In this case, the vacancies are immobile)

Irradiation of ‘defect-free’ samples: Fe

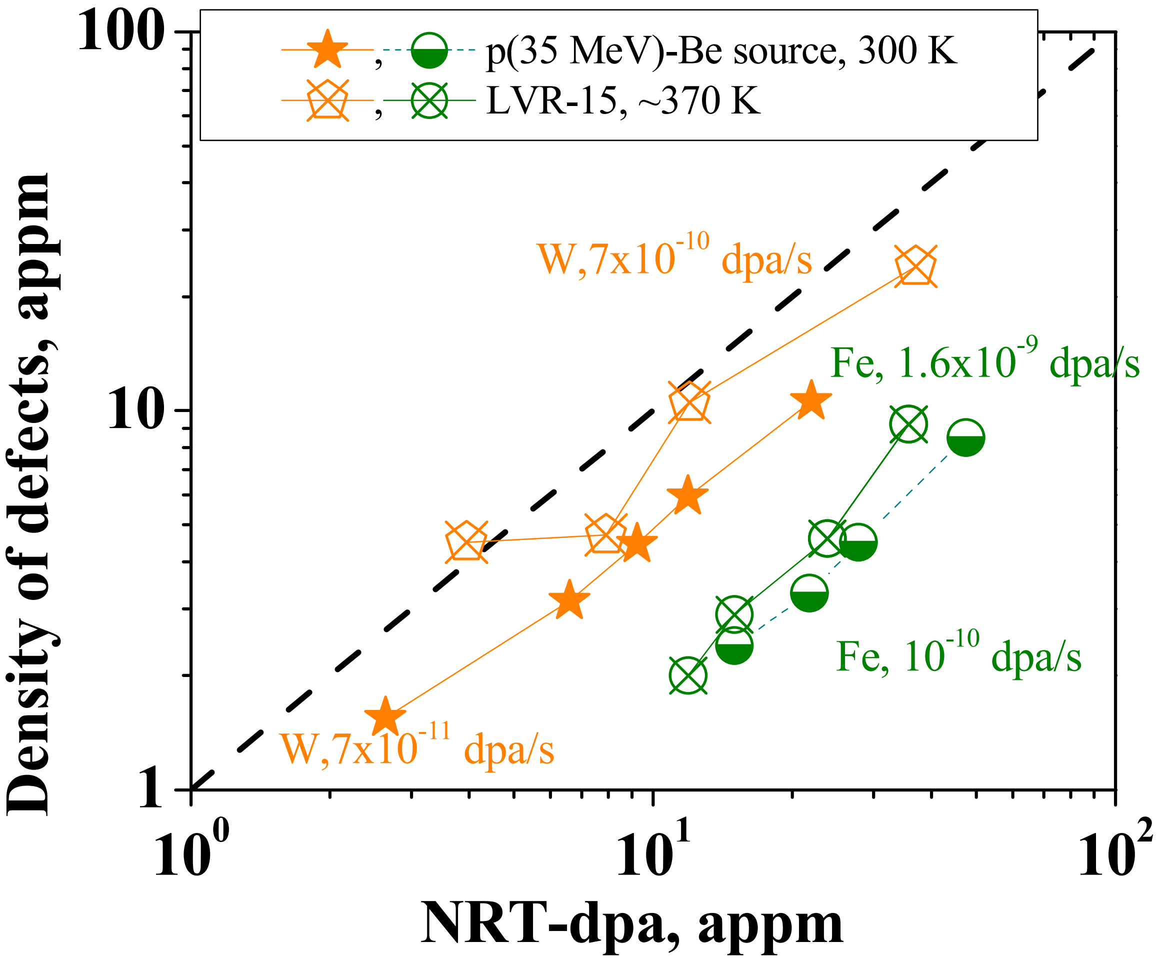


❑ Defect density in Fe irradiated with p+ , fission and energetic neutrons at low doses is close to each other and is well described by a linear fit (~$dpa^{0.2}$).

❑ Irradiation of Fe and W with fission neutrons leads to slightly higher density of defects compared to fast neutrons

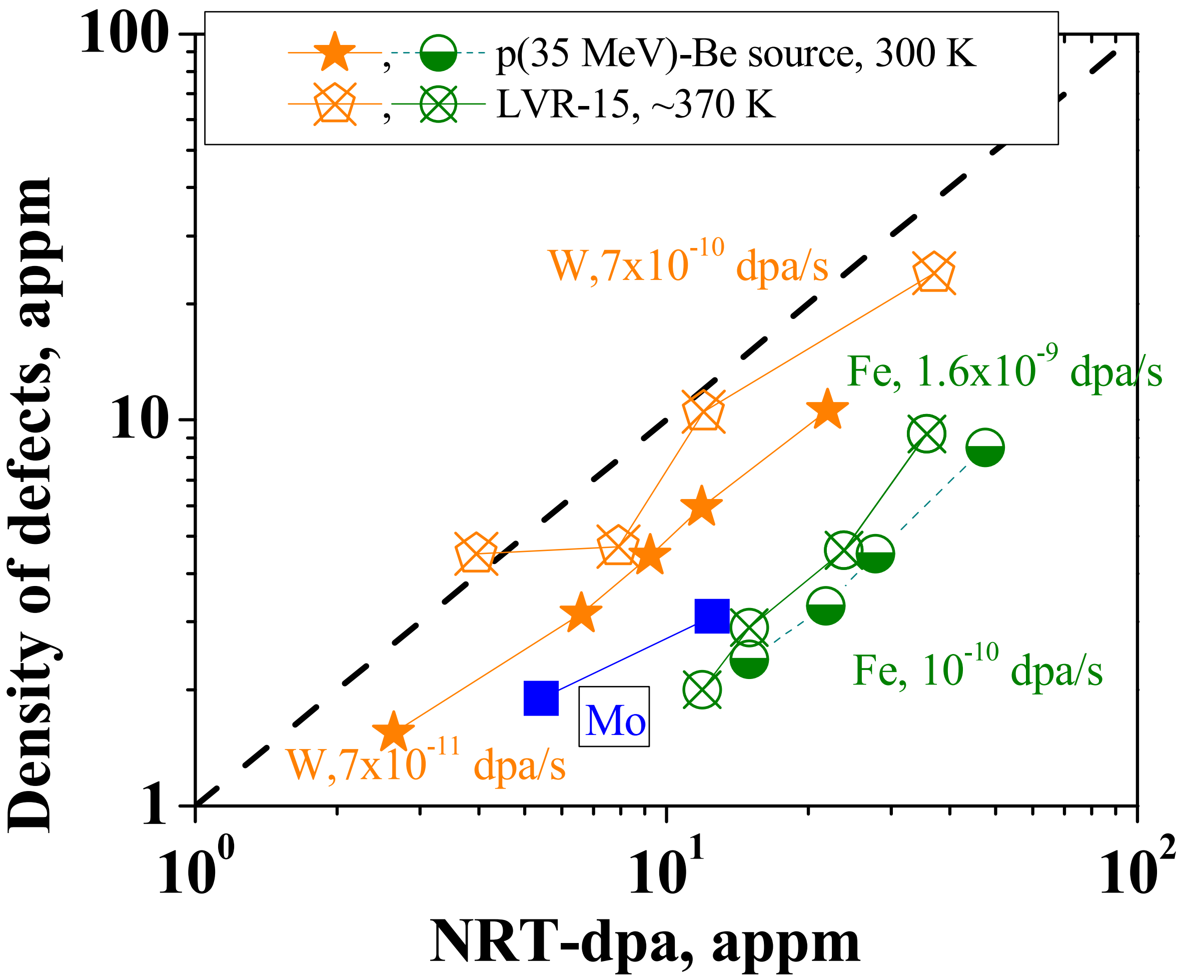
❑ The dpa is a good parameter for quantification of the total number of **primary defects** in a material caused by different types of irradiation. (In this case, the vacancies are mobile)

PALS measurements of the density of radiation-induced defects



No noticeable influence of rate of defect production varied by one order of magnitude at low dpa of the orders of several appm: slightly lower density at lower rate.

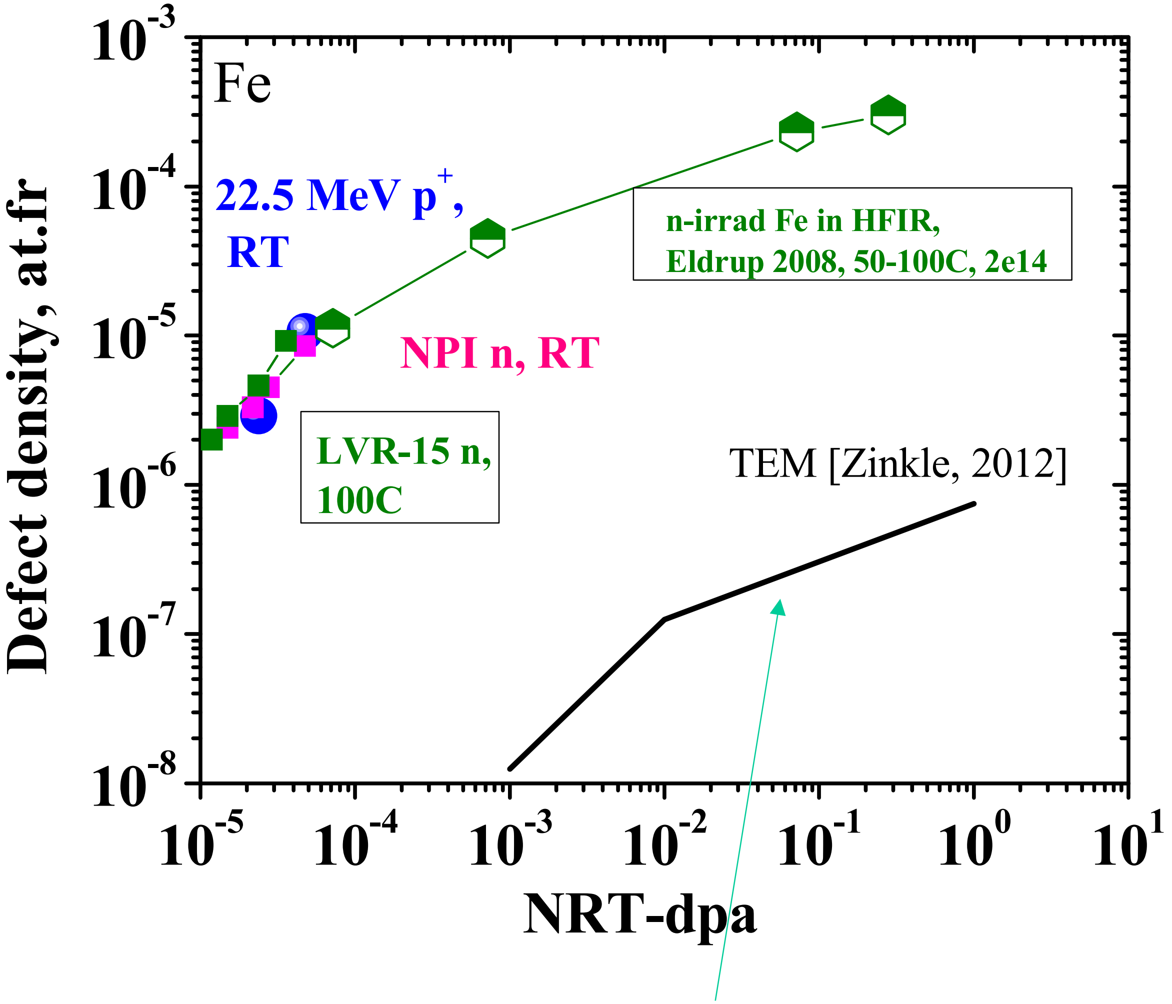
PALS measurements of the density of radiation-induced defects



The radiation-induced defect density is highest for W and lowest for Fe for the same irradiation temperature.

The radiation-induced defect density for Mo is between W and Fe.

PALS versus TEM measurements



TEM (295-340 K) significantly underestimates the defect density

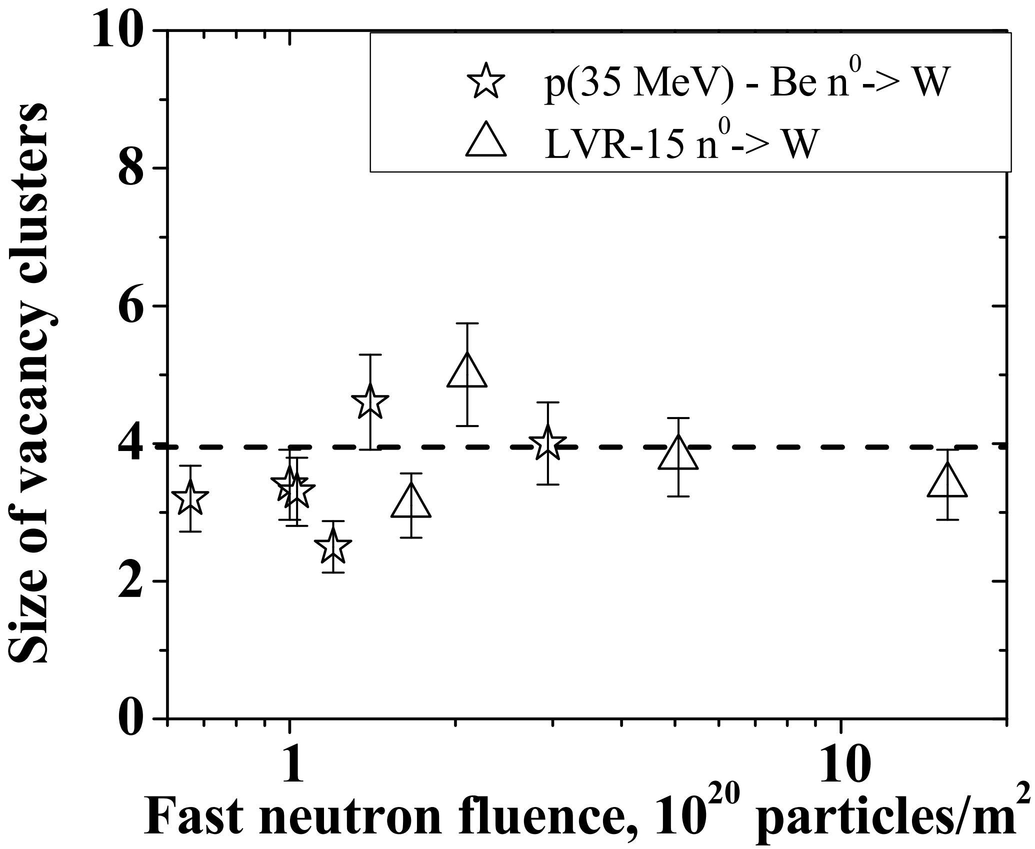
The radiation-induced defects of small size (less than 2 nm) , namely, vacancies and small vacancy clusters, dominate in n-irradiated W and Fe

Size distribution of radiation defects

The dpa models are not taken into account the size of defects.

Different cluster sizes can invariably contribute to changes in the microstructure and, consequently, the physical properties of irradiated materials, and influence the fuel retention.

W: Fission versus fusion neutrons

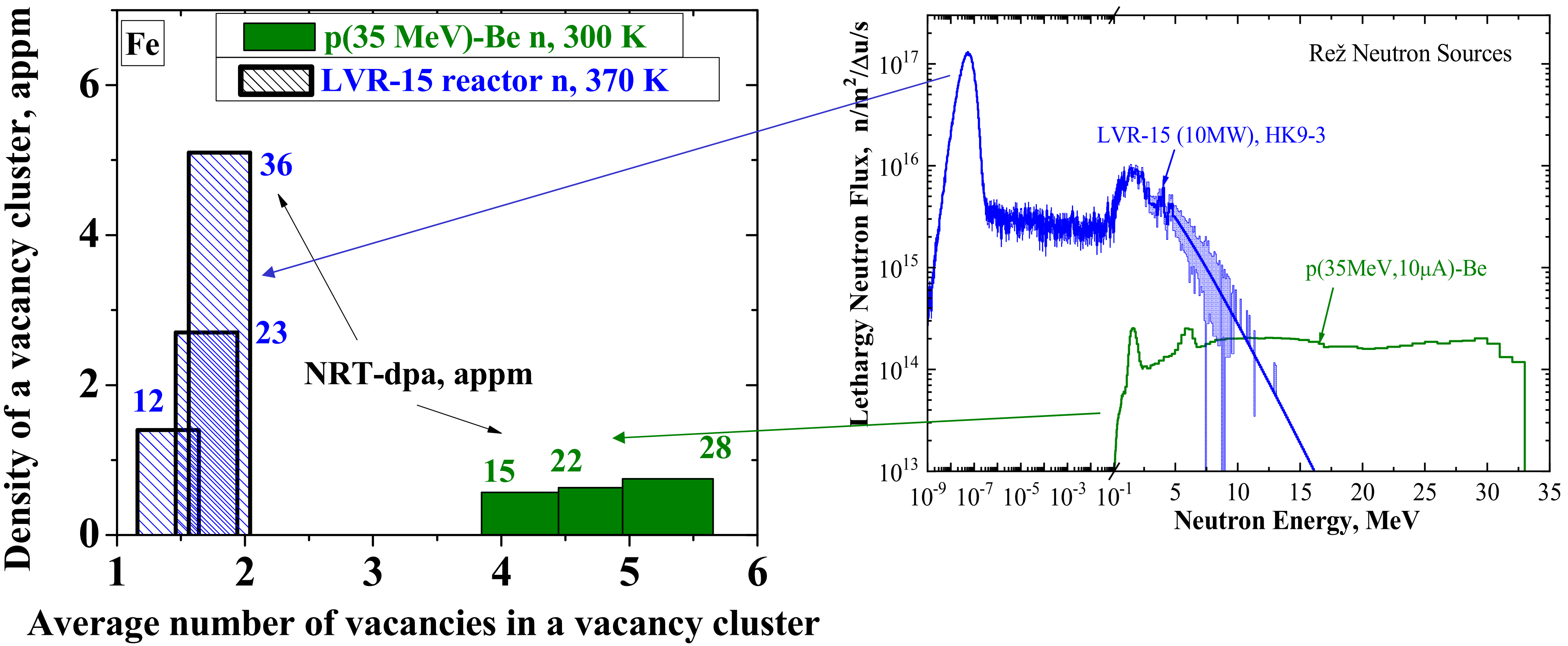


Irradiation of W in the fission reactor and by high-energy neutrons from the p(35 MeV)-Be generator leads to the formation of small radiation-induced vacancy clusters with comparable mean size of 3-4 vacancies in a vacancy cluster.

This is because vacancies are immobile at the irradiation temperature 300-370 K and the fluences are too low for cascade overlap events. So, flux would not have any effect on in-cascade local vacancy concentration

O.V. Ogorodnikova et al., Sci.Reports, 2020

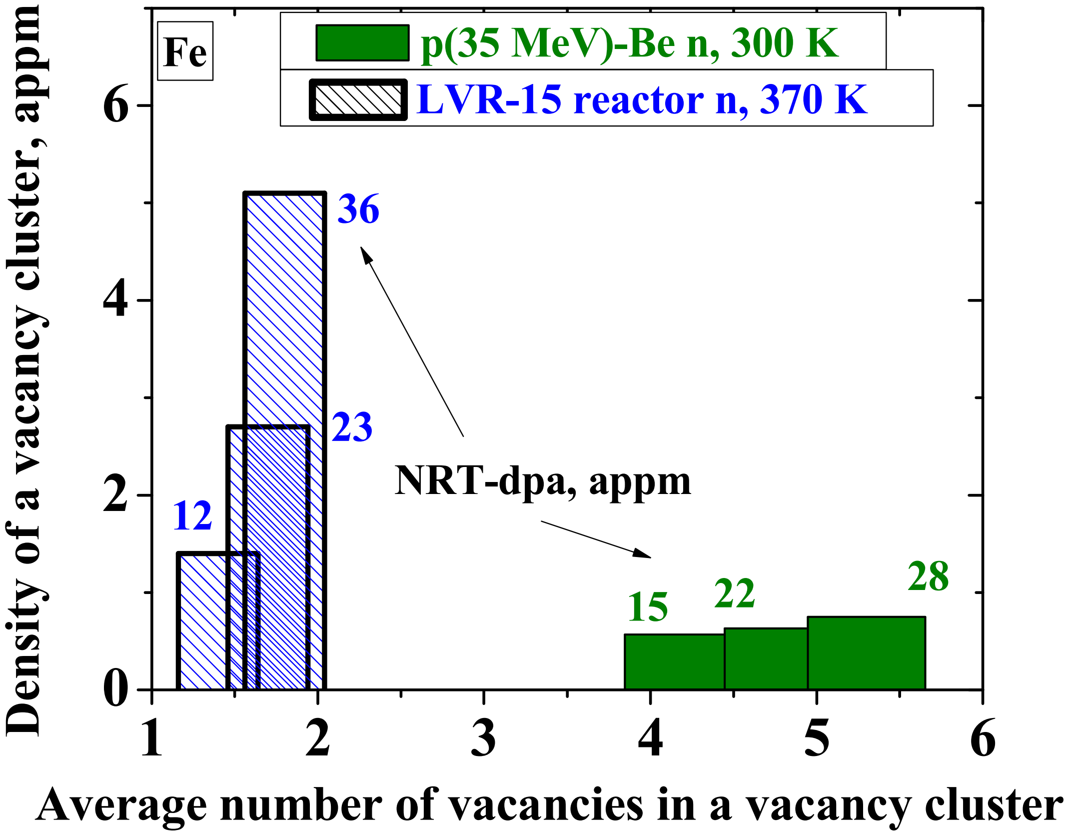
Fe: Fission versus fusion neutrons



the vacancy clusters in Fe appear to increase in the size and the density with increasing fluence. The increase in the size is evidence for vacancy flow to vacancy clusters produced in earlier stages of the irradiation.

O.V. Ogorodnikova et al., Sci.Reports, 2020

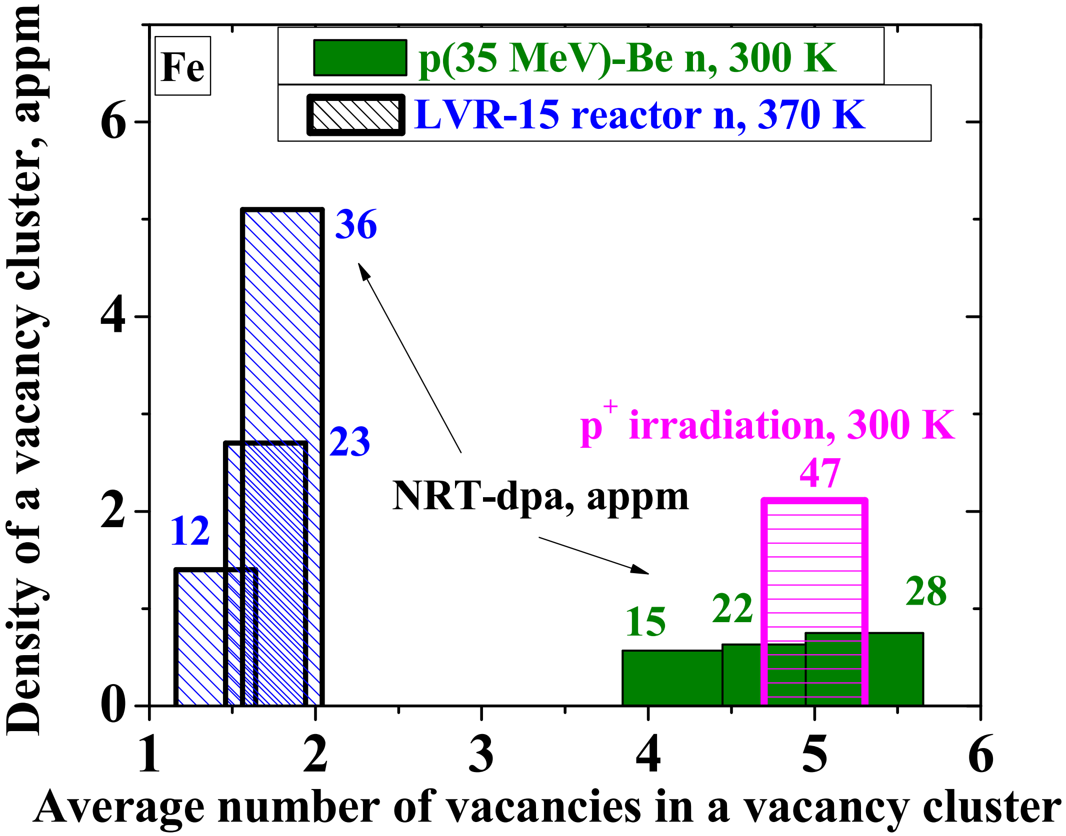
Fe: Fission versus fusion neutrons



the vacancy clusters in Fe appear to increase in the size and the density with increasing fluence. The increase in the size is evidence for vacancy flow to vacancy clusters produced in earlier stages of the irradiation.

- ☐ **Two reasons of the formation of the larger size of the defects with lower density in the case of irradiation with high-energy neutrons from the p(35 MeV)-Be source are: (i) a high fraction of high-energy neutrons in the neutron spectra from the p(35 MeV)-Be source (pronounced defect agglomeration and recombination during the cascade quench phase take place) and (ii) flux effect caused by thermally activated migration of radiation-induced vacancies (a higher density of vacancy clusters with a smaller size is formed with increasing the flux).**

Fe: Proton- versus neutron- irradiation



the vacancy clusters in Fe appear to increase in the size and the density with increasing fluence. The increase in the size is evidence for vacancy flow to vacancy clusters produced in earlier stages of the irradiation.

Proton irradiation produces vacancy clusters in Fe due to the vacancy migration even at 300 K. Proton irradiation could lead to similar size of vacancy clusters in Fe as in the case of wide-spectrum high energy neutrons.

❑ **Two reasons of the formation of the larger size of the defects with lower density in the case of irradiation with high-energy neutrons from the p(35 MeV)-Be source are: (i) a high fraction of high-energy neutrons in the neutron spectra from the p(35 MeV)-Be source (pronounced defect agglomeration and recombination during the cascade quench phase take place) and (ii) flux effect caused by thermally activated migration of radiation-induced vacancies (a higher density of vacancy clusters with a smaller size is formed with increasing the flux).**

Conclusions

- ❑ **Fission neutrons do not appear to be a good surrogate for simulating radiation damage caused by thermonuclear neutrons concerning to defect size distribution and transmutation gases. Fast neutrons from p-Be source or other accelerator source can be a good surrogate to simulate radiation damage caused by fusion neutrons.**

- ❑ **Energetic protons can be a surrogate to simulate fusion neutron damage in certain materials over a certain temperature range (further research is needed to establish these conditions).**

Future work

- PALS measurement of our samples after JET DT campaign
- PALS measurement of our samples after irradiation in BR-2 (within IAEA CRP)
- Irradiation at various temperature
- Effect of initial dislocations, vacancies, and impurities/doping (WCrY, Eurofer97) on the radiation defect development

Outline

Damage occurs by two main mechanisms

Displacement of atoms

Transmutation

from one element to another element
from one isotope to another isotope

- γ-ray energy spectra of irradiated samples
- gaseous and solid transmutation products can be simulated

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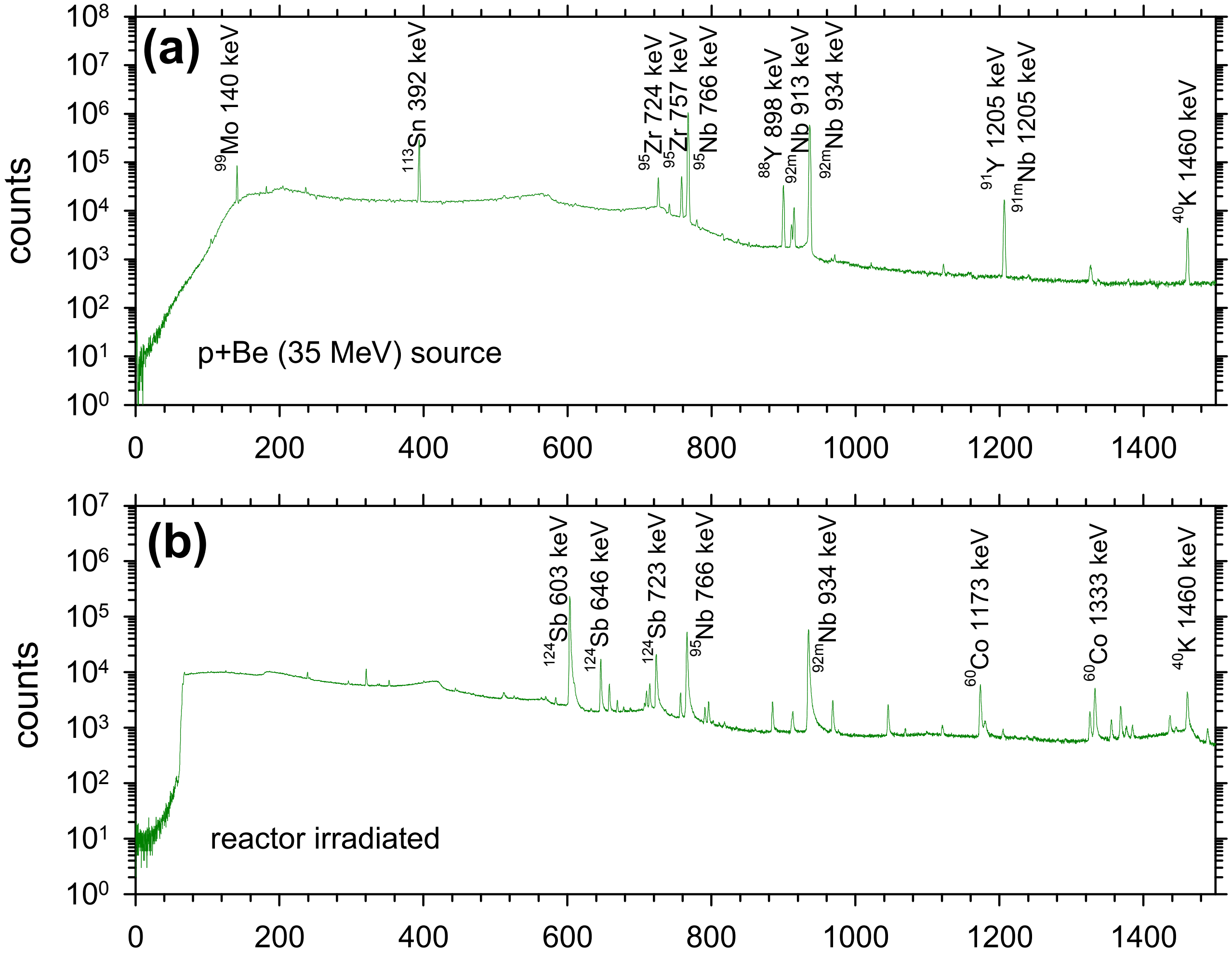
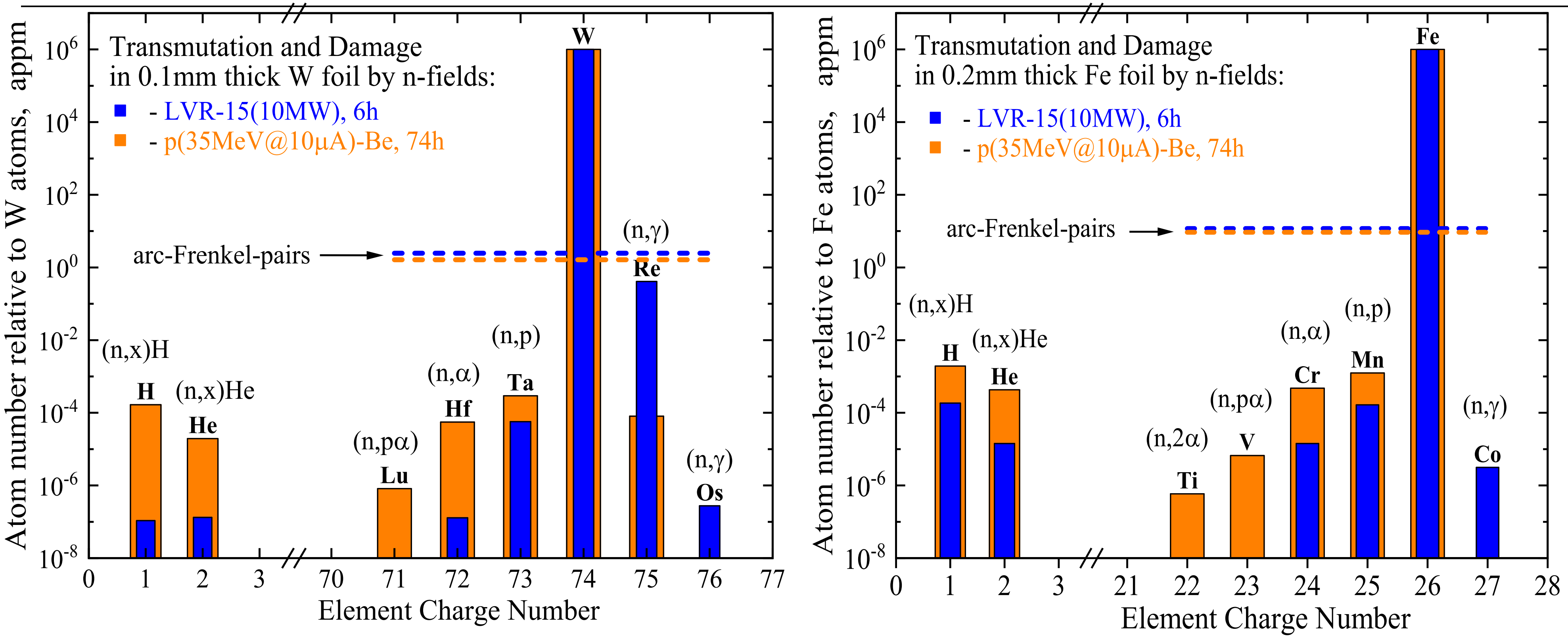
Impact the T retention in a material

Transmutation product measurements



Calculations of transmutation products in W and Fe



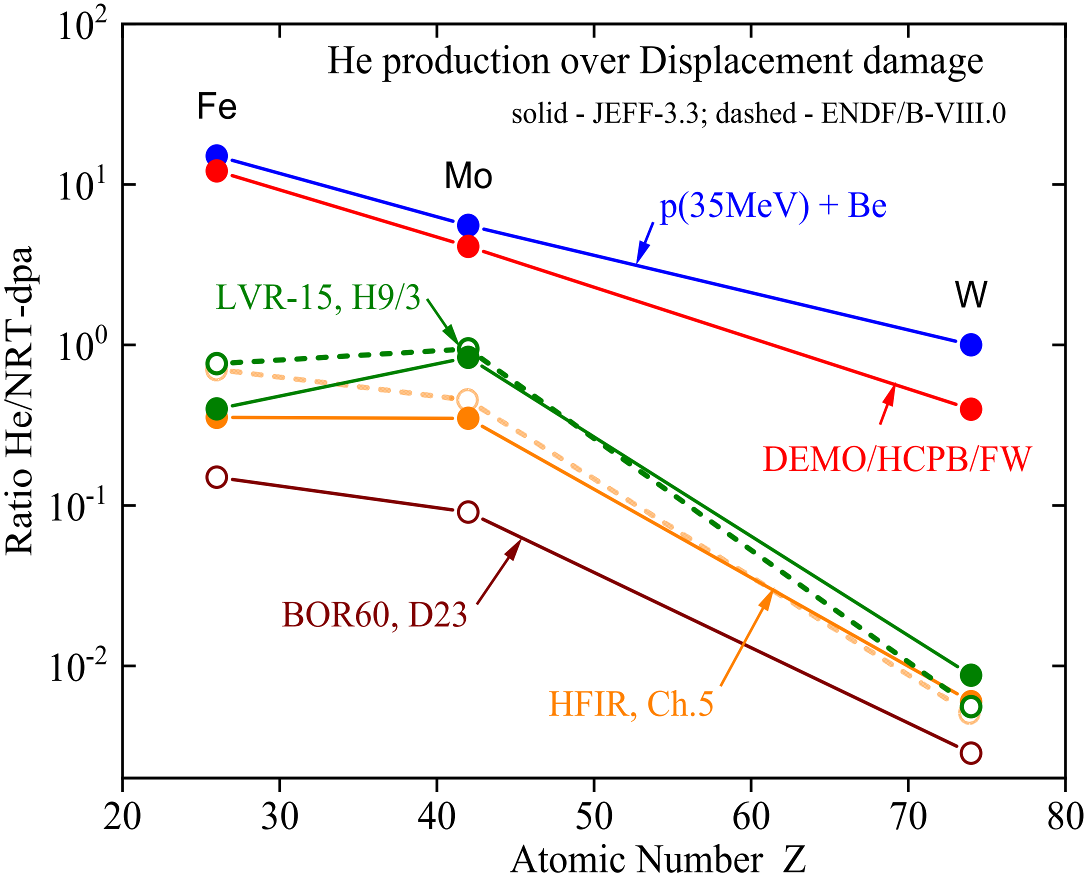
FISPACT-II code and JEFF-3.3 library

- **For both W and Fe, the softer reactor neutron spectrum results in a higher concentration of high Z-elements by the (n,γ) reactions than the accelerator driven neutron source. In contrast, the neutrons from the p(35 MeV)-Be source produce more low-Z elements and hydrogen or helium gases than the neutrons from the LVR-15 reactor. The ratio of the concentration of solid build-up**

W: $\mathrm{trans}^{\mathrm{reactor}}/\ \mathrm{trans}^{\mathrm{p\text{-}Be}} = 4.1\mathrm{x}10^{-1}/4.3\mathrm{x}10^{-4} = 10^{3}$

Fe: $\mathrm{trans}^{\mathrm{reactor}}/\ \mathrm{trans}^{\mathrm{p\text{-}Be}} = 1.8\mathrm{x}10^{-4}/1.7\mathrm{x}10^{-3} = 10^{-1}$

Calculations He/dpa



JEFF-3.3 (solid curve with close symbols) and ENDF/B-VIII.0 (dashed curve with open symbols).

- **He/dpa ratios in the facilities with the fast energy spectra (fusion like) p-Be and DEMO are one-two orders of magnitude higher than in the fission ones LVR-15, HFIR and BOR60;**

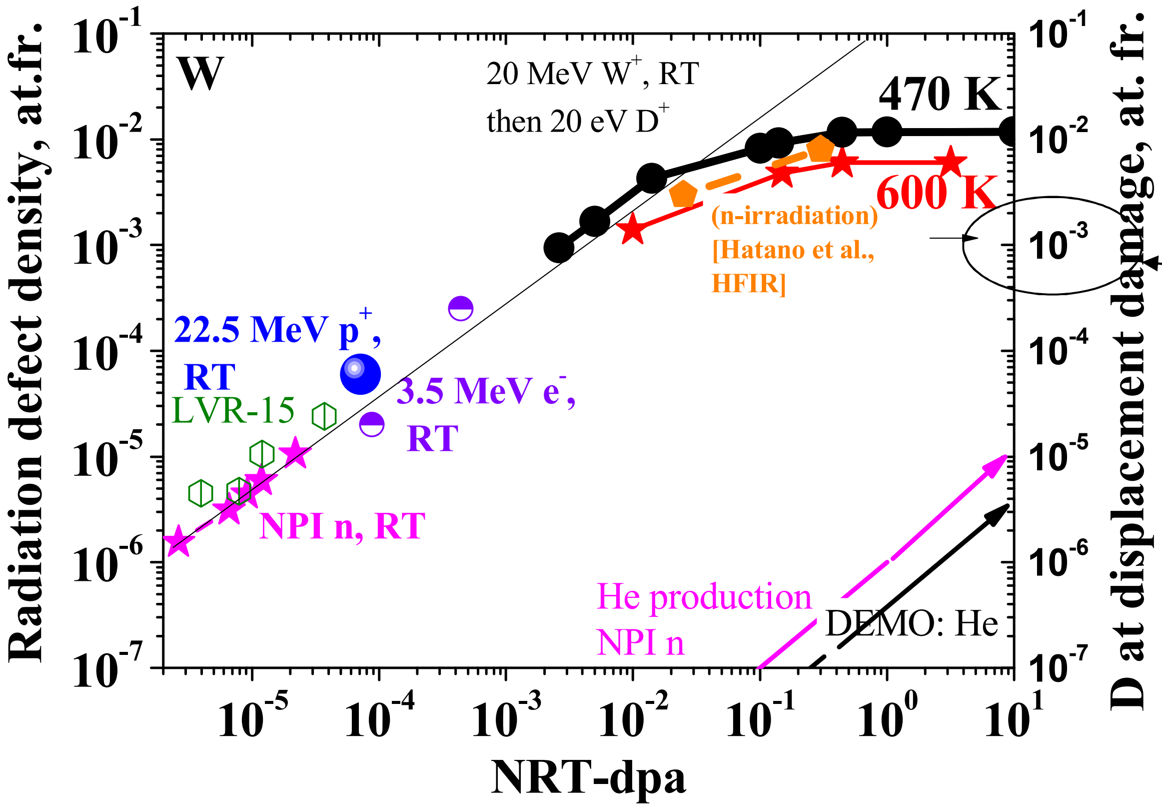
- **He/dpa ratio substantially decreases with increasing the target nucleus charge Z;**

- **Facilities with fusion-like spectra show monotonic decrease of He/dpa ratio versus Z, that is not a case for the fission spectra. The more smooth trend for irradiation in BOR60, which has the fast fission spectrum, likely points to the relative large contribution to the He production of the thermal neutrons, which are present in the thermal reactors LVR-15 and HFIR.**

- Transmutation products would be better simulated using the p(35 MeV)-Be source compared to fission source

Total density of radiation defects and decoration with Deuterium + He effect

What is the He concentration to influence the T retention?



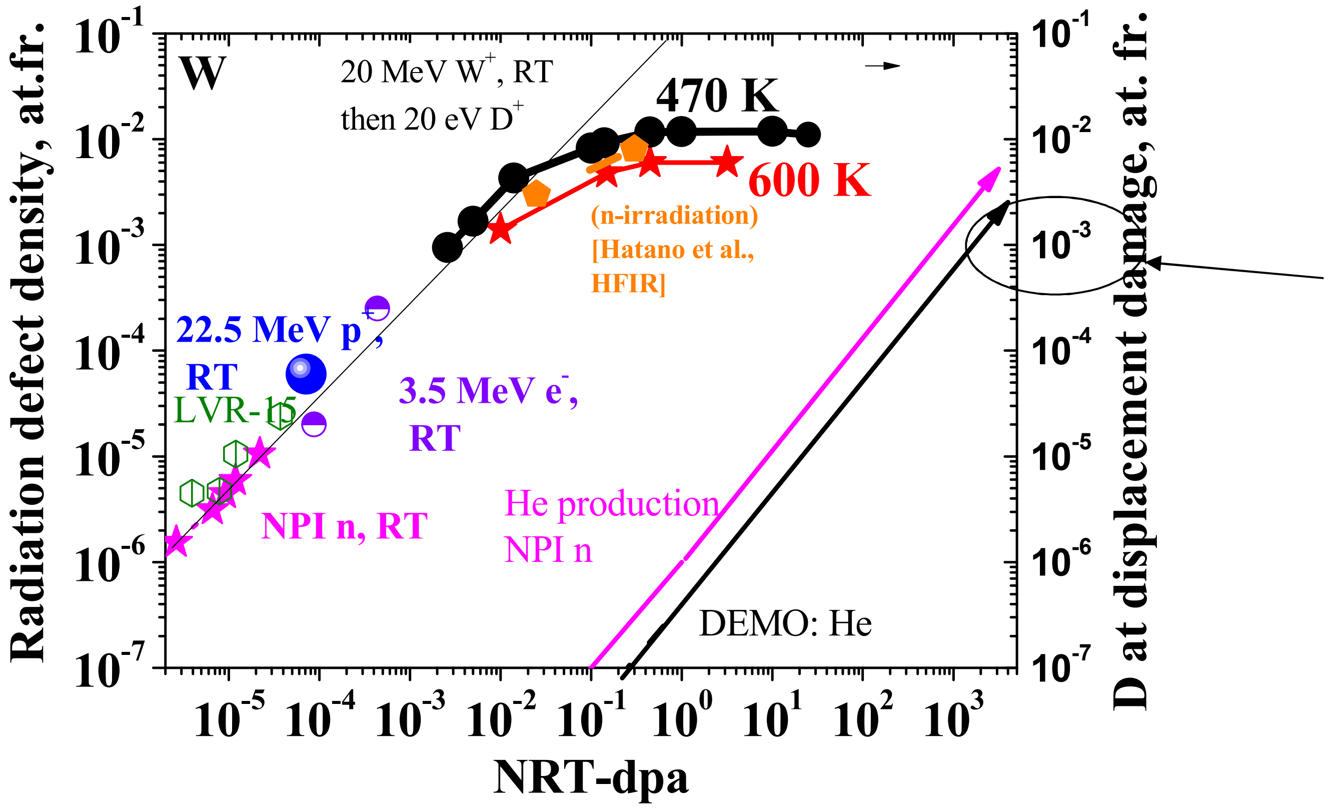
Above 10^{-3} at.fr. He influences the T retention in W.

No effect for ITER (0.7 dpa)

He production is essential at high dpa in W

Total density of radiation defects and decoration with Deuterium + He effect

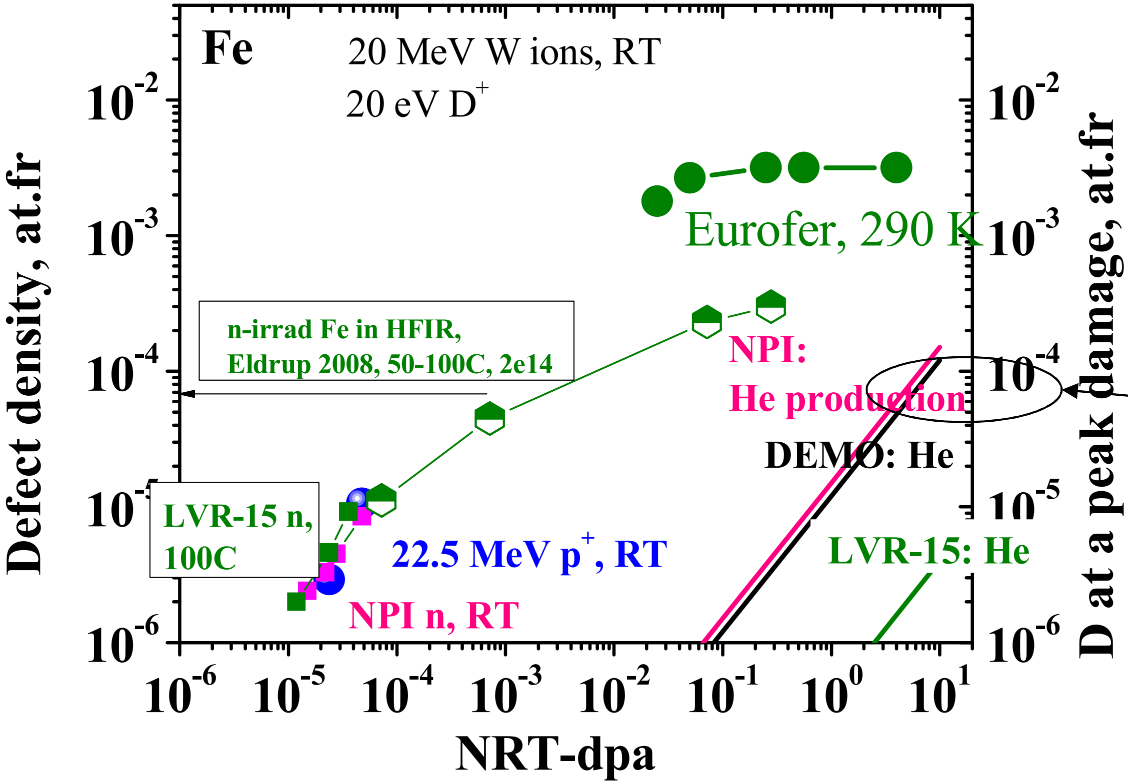
What is the He concentration to influence the T retention?



Above 10^{-3} at.fr. He influences the T retention in W

He production is essential for T retention at above 100 dpa

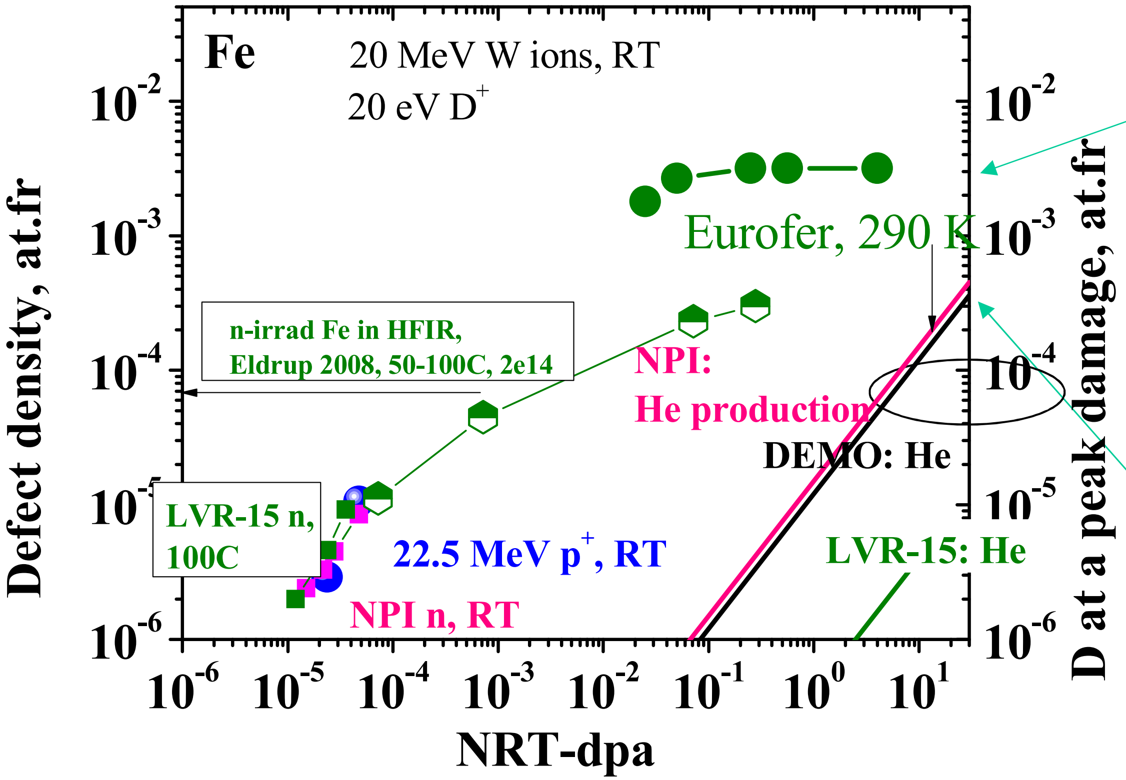
Total density of radiation defects and decoration with Deuterium + He effect

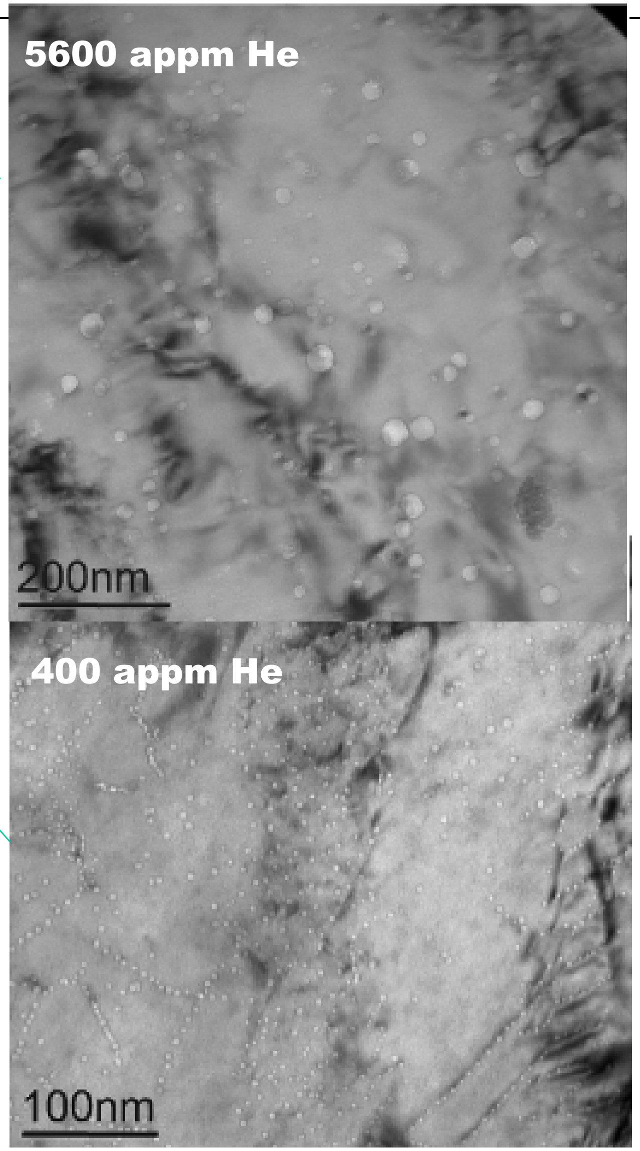


Above $5x10^{-5}$ at.fr. He influences the T retention in Fe-based materials

He production is essential at low dpa in Fe-based materials (above 1 dpa)

Total density of radiation defects and decoration with Deuterium + He effect





B-alloyed Eurofer97, 400C, 16 dpa, [Coppola et al., 2016, Nuclear Mat. and Energy]

Conclusions

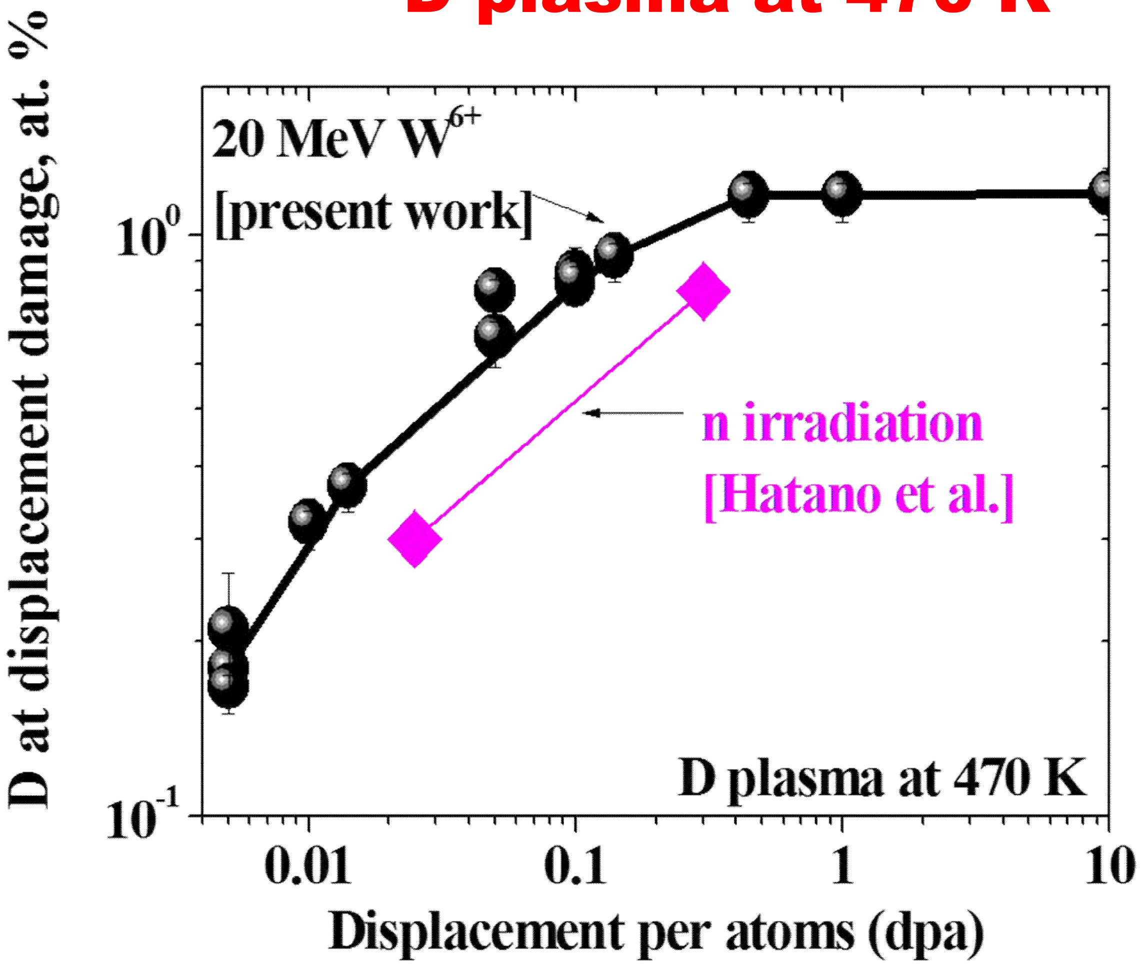
- He/dpa ratios in the facilities with the fast energy spectra (fusion like) from p-Be source and DEMO are one-two orders of magnitude higher than in the fission ones LVR-15, HFIR and BOR60;

- Transmutation products by fusion neutrons would be better simulated using neutrons from the p(35 MeV)-Be source compared to fission source

- Critical He concentration influenced the T retention is about 10^{-3} at.fr. He for W and $5x10^{-5}$ at.fr. He for Fe.

Ion irradiation & fission neutron irradiaiton

D plasma at 470 K



K_{exp}=0.65

This means that being irradiated up to the same dose (in dpa) neutron-irradiated W retains only ~65% of the deuterium that retains in self-ion irradiated W

O.V. Ogorodnikova and V. Gann, JNM, 2015