STU FEI
SLOVAK UNIVERSITY OF TECHNOLOGY IN BRATISLAVA
FACULTY OF ELECTRICAL ENGINEERING AND INFORMATION TECHNOLOGY

INPE
INSTITUTE OF NUCLEAR AND PHYSICAL ENGINEERING

VACANCY TYPE DEFECTS BEHAVIOUR IN MATERIALS FORESEEN FOR LIQUID METAL COOLED FAST REACTORS

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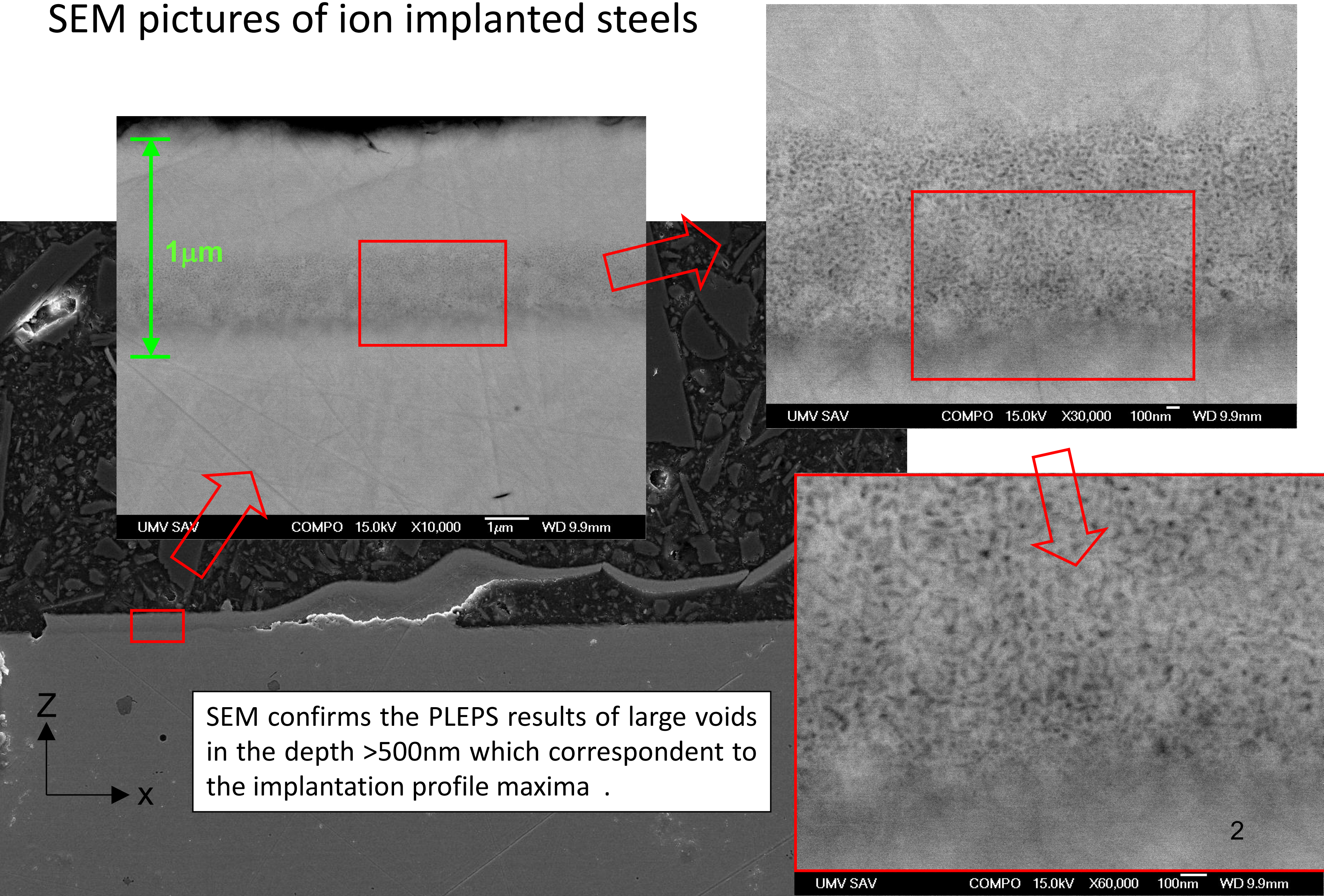
SEM pictures of ion implanted steels



SEM confirms the PLEPS results of large voids in the depth >500nm which correspondent to the implantation profile maxima .

Available techniques for material studies:

Positron Annihilation Spectroscopy: Conventional PALS 2-det. or 3-det. Set-ups (for irradiated materials), digital Doppler Broadening set-up

Moessbauer spectroscopy,

Atomic force microscopy,

X-ray diffraction, Nano-indetor,

Barkhausen Noises measurements,

Alfa, beta, gamma spectroscopy

including low/background chamber,

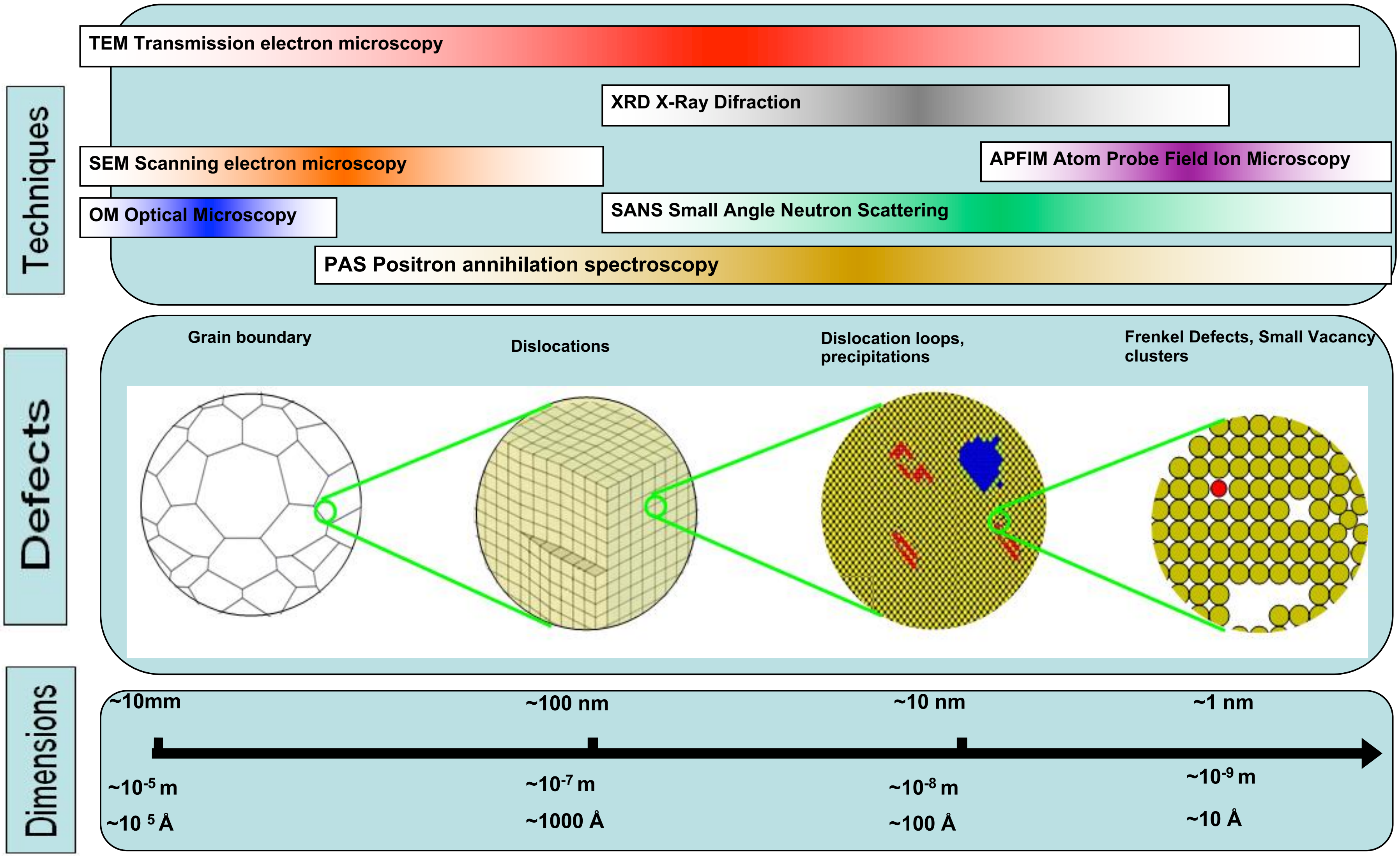
In collaboration with other institutes:

TEM, SEM, Auger spectroscopy and

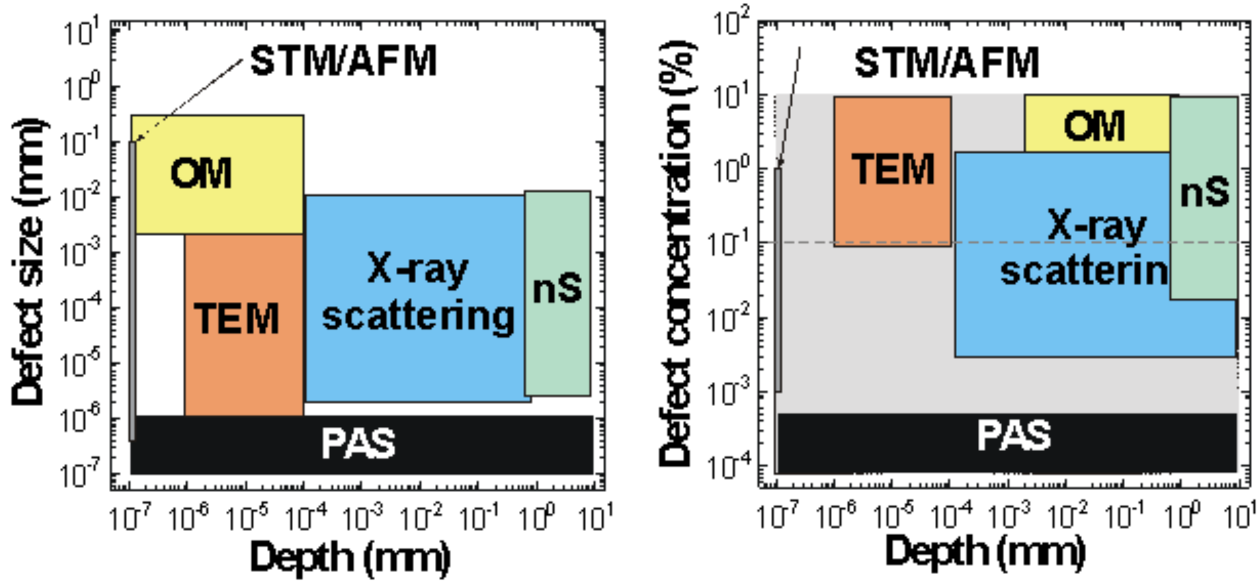
access to tandentor for ion implantation

Slovak scientific-technical team 2014

Overview


Techniques
TEM Transmission electron microscopy
XRD X-Ray Difraction
SEM Scanning electron microscopy
APFIM Atom Probe Field Ion Microscopy
OM Optical Microscopy
SANS Small Angle Neutron Scattering
PAS Positron annihilation spectroscopy
Defects
Grain boundary
Dislocations
Dislocation loops, precipitations
Frenkel Defects, Small Vacancy clusters
Dimensions
~10mm
~100 nm
~10 nm
~1 nm
~10^{-5} m
~10^{-7} m
~10^{-8} m
~10^{-9} m
~10^{5} Å
~1000 Å
~100 Å
~10 Å

Non-destructive techniques for defects investigation



- General overview of applicability range for some spectroscopy techniques [Howell, R., Physics Space Technology, 1987]. *(OM – optical microscopy, TEM - transmission electron microscopy, nS – neutron scattering, STM/AFM – scanning tunnelling microscopy/atom probe ion microscopy).*

Irradiation-induced changes

✓ Neutron-irradiation

- Defect production
  - Self-interstitial atom (SIA) & vacancy (V) rich regions
- Matrix damage
  - SIA-clusters, SIA-loops
  - Micro voids
- Solute atom diffusion
  - Precipitates
  - Complex defect-solute configurations
  - GB segregation

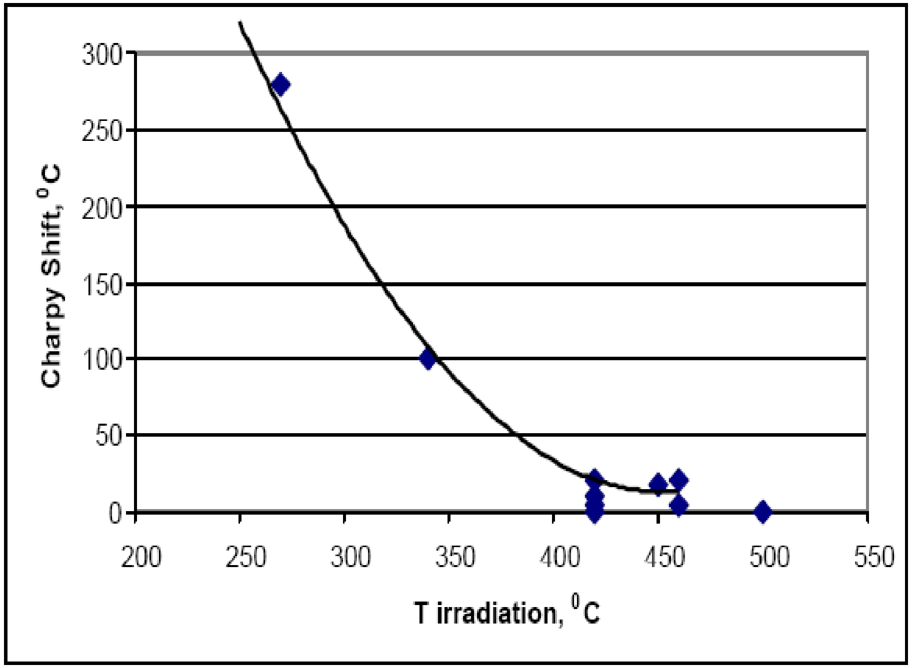
PAS can give additional information about:

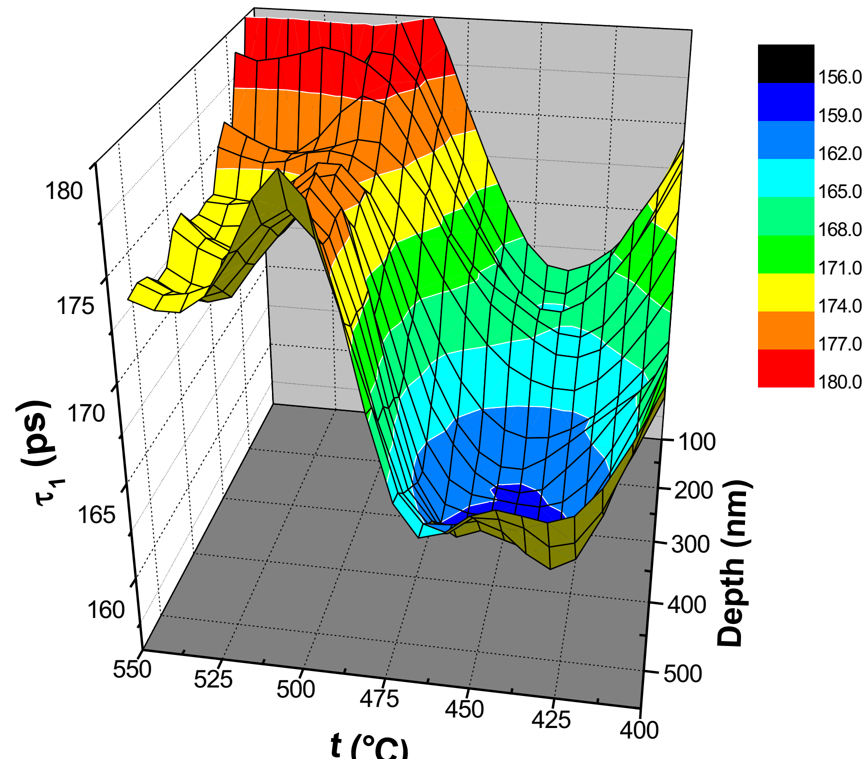
- radiation induced defects
- thermal annealing of these defects.

[15] EURAD, European Raioactive Waste Management. Project of the European atomic energy community. (H2020 program EU) 52 spoluriešiteľov. (2019-2023)

Slugeň, V., Safety of VVER-440 Reactors – Barriers Against Fission Products Release, Springer, 2011

PAS Results - Annealing temperature for WWER-steels at 475 °C is acceptable, but PAS gives more information.



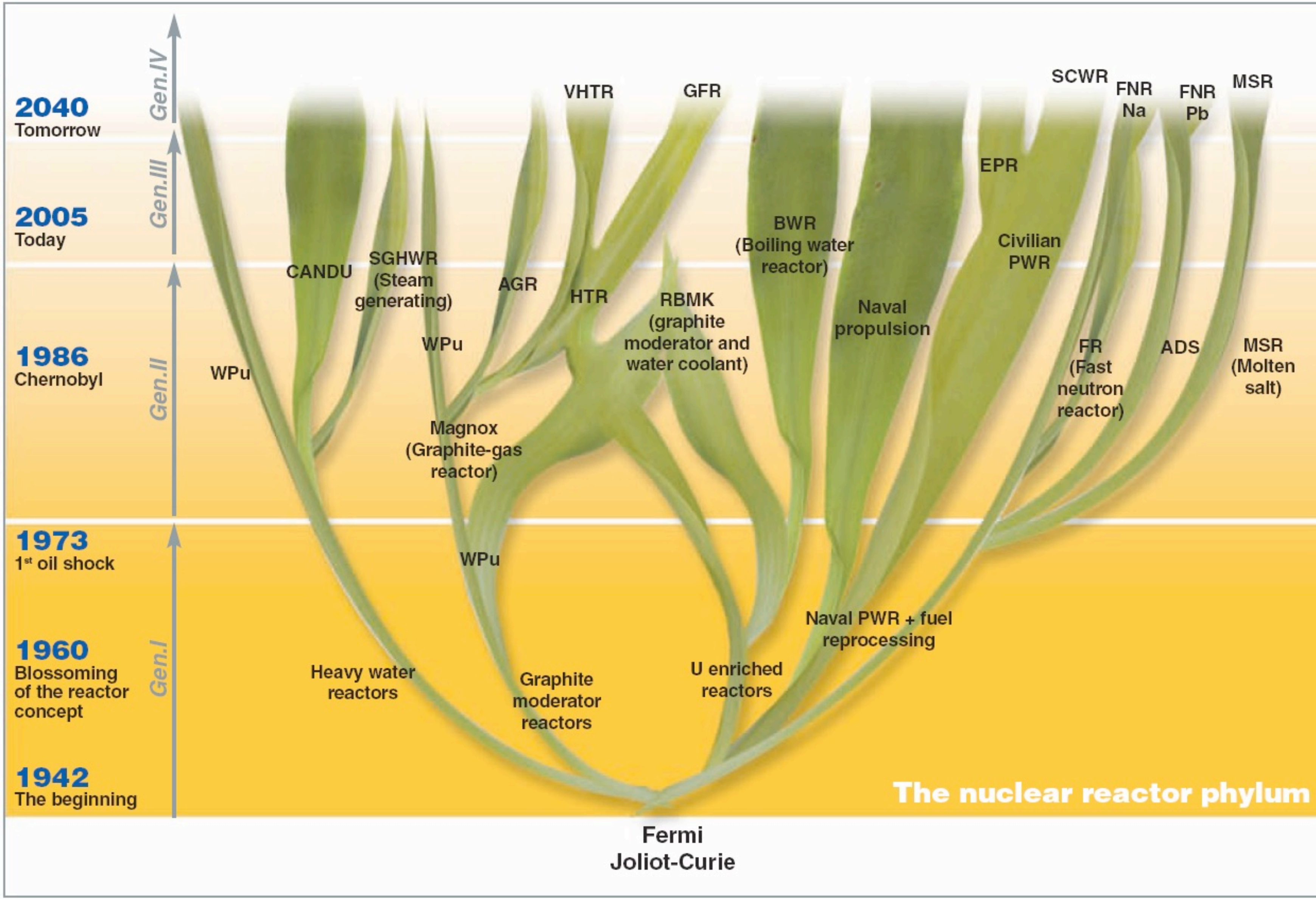


The 3D presentation of PLEPS results (τ_1) of irradiated ($1.25x10^{24}m^{-2}$) and annealed Sv-10KhMFT steel (WWER-440 weld).

The effectiveness of the annealing process to removing of small defects (mono/di-vacancies or Frenkel pairs) can be followed via significant decrease of parameter tau1. This figure also shows rapid increase of mentioned small defects in WWER type of RPV steels after about 480 ºC.

Slugen, Kryukov, NED 263 (2013) 308 *Slugen et al: NTD&E Int. 37 (2004) 651*

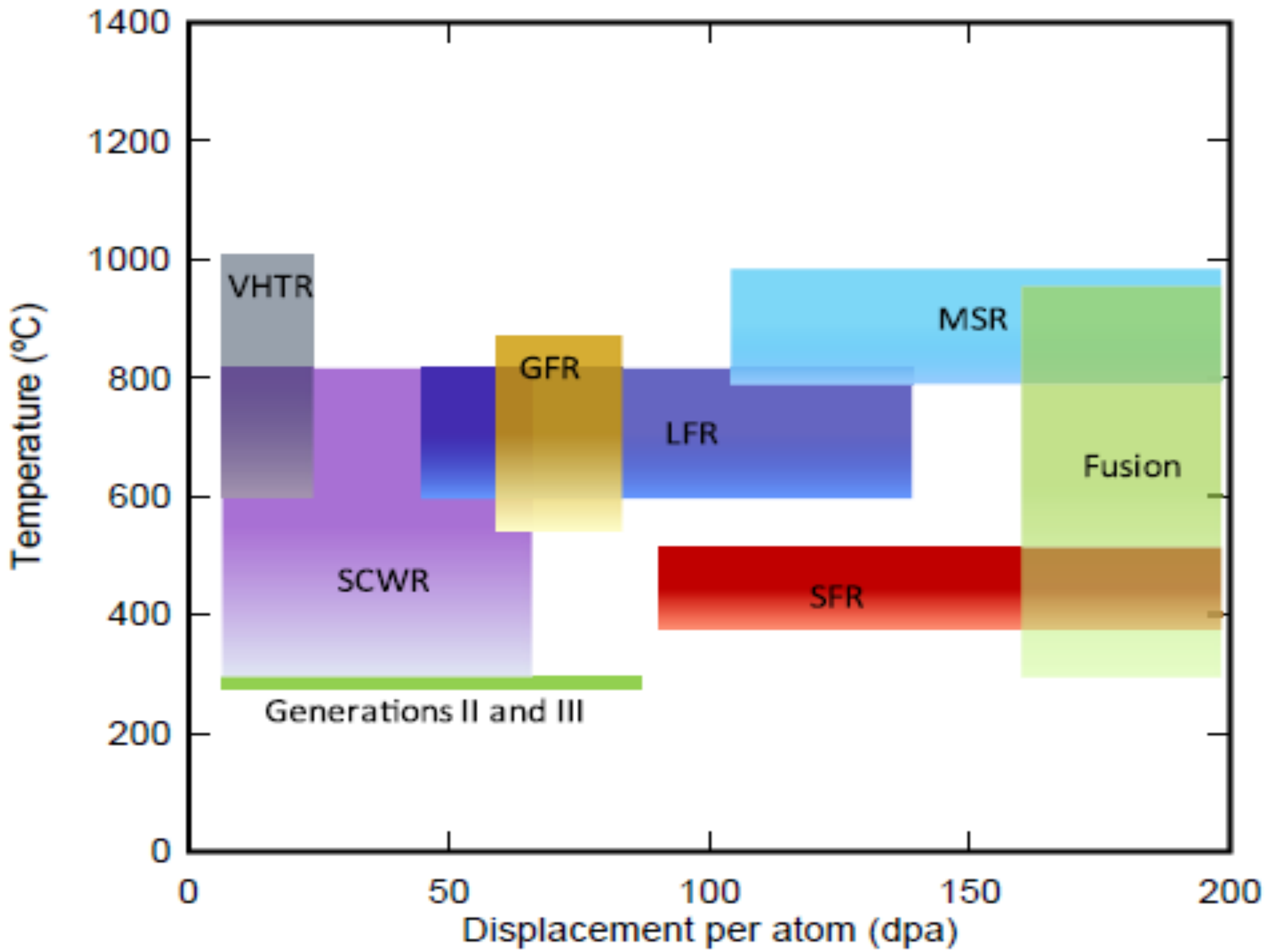
PAST – application of PALS in reactor pressure vessel studies



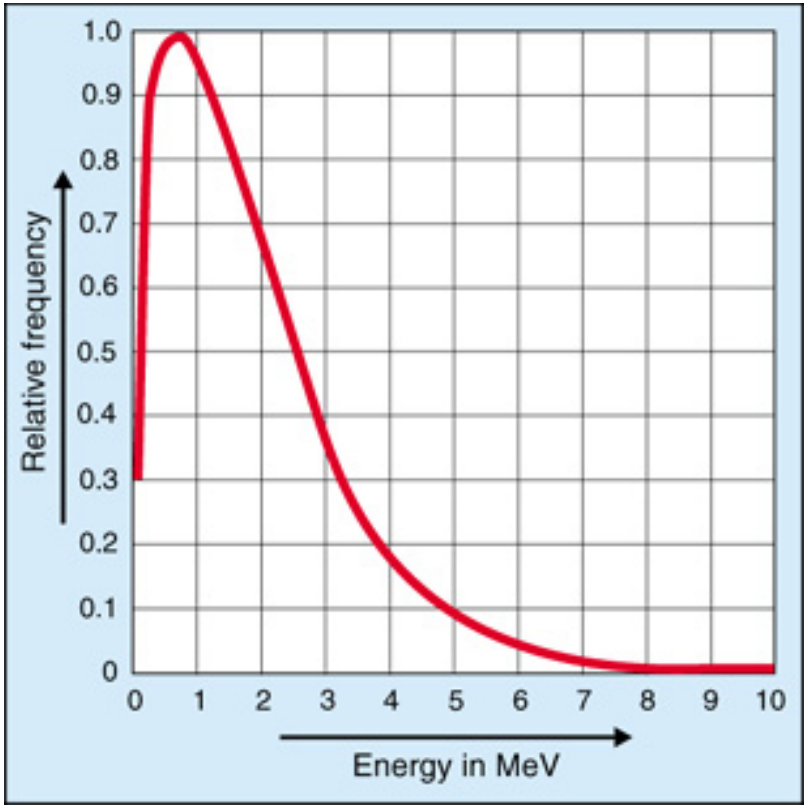
First fast reactor

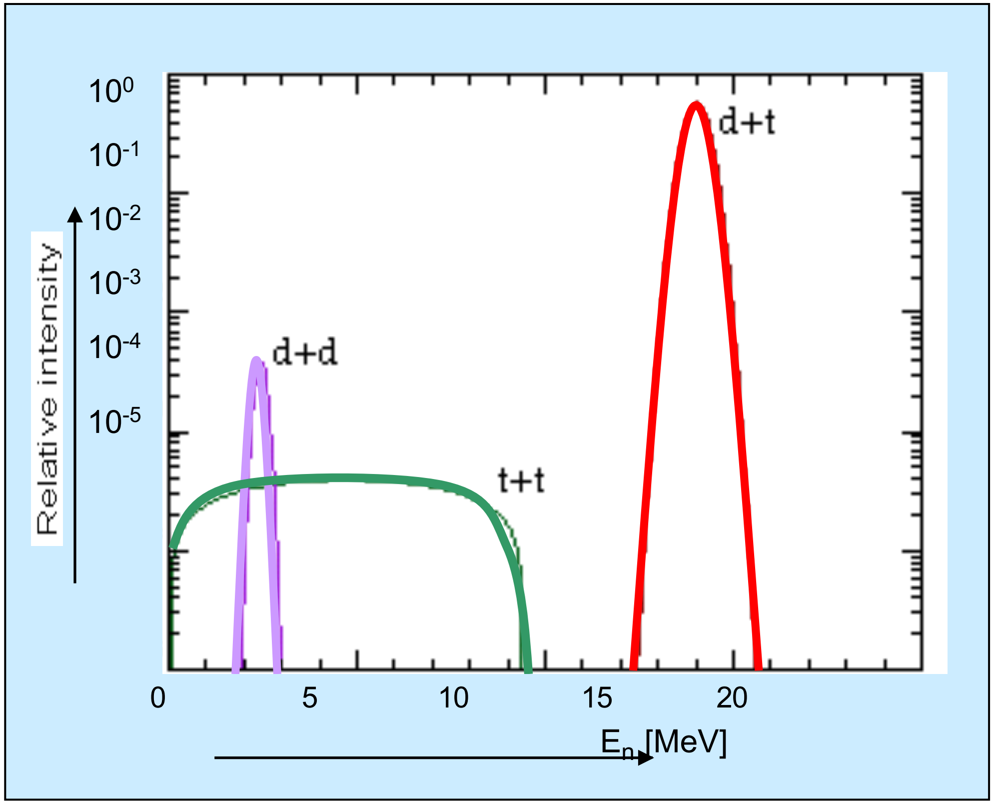
- **EBR-I** (Experimental Breeder Reactor I) operated by U.S. Atomic Energy Commission (Idaho – USA), since 20.12.1951.
- U-235 enrichm. > 90%,
- coolant - eutektikum Na-K

Temperature and dose requirements for in-core structural materials for the operation of the six proposed Generation IV advanced reactor concepts and fusion reactor concepts.
Adapted from Zinkle and Was (2013).



Fission and fusion neutrons



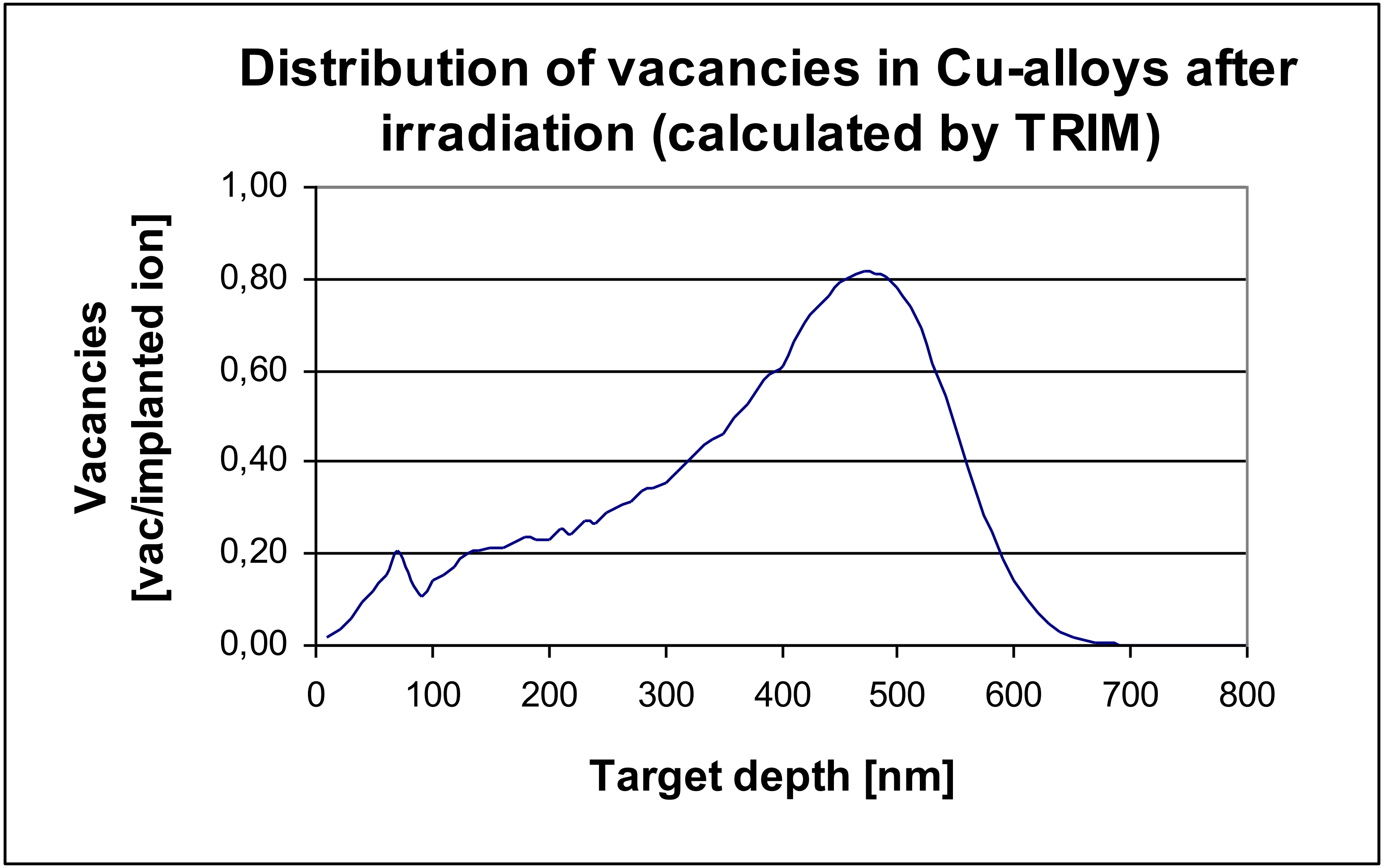


Studies Cu alloys Chemical composition and implantation doses.

Specimen	**Material**	**Implantation dose [C/cm^2]**
SS	CuCrZr – 98.95w.%Cu; 0.6-0.9w.%Cr; 0.07-0.15w.%Zr	None
SA	CuCrZr	0.4
SM	CuCrZr	1.1
TT	CuAl25 – 99.5w.%Cu; 0.25w.%Al as Al_2O_3; 0.22w.%O as Al_2O_3; 0.025w.%B as B_2O_3	None
TA	CuAl25	0.4
TM	CuAl25	1.1

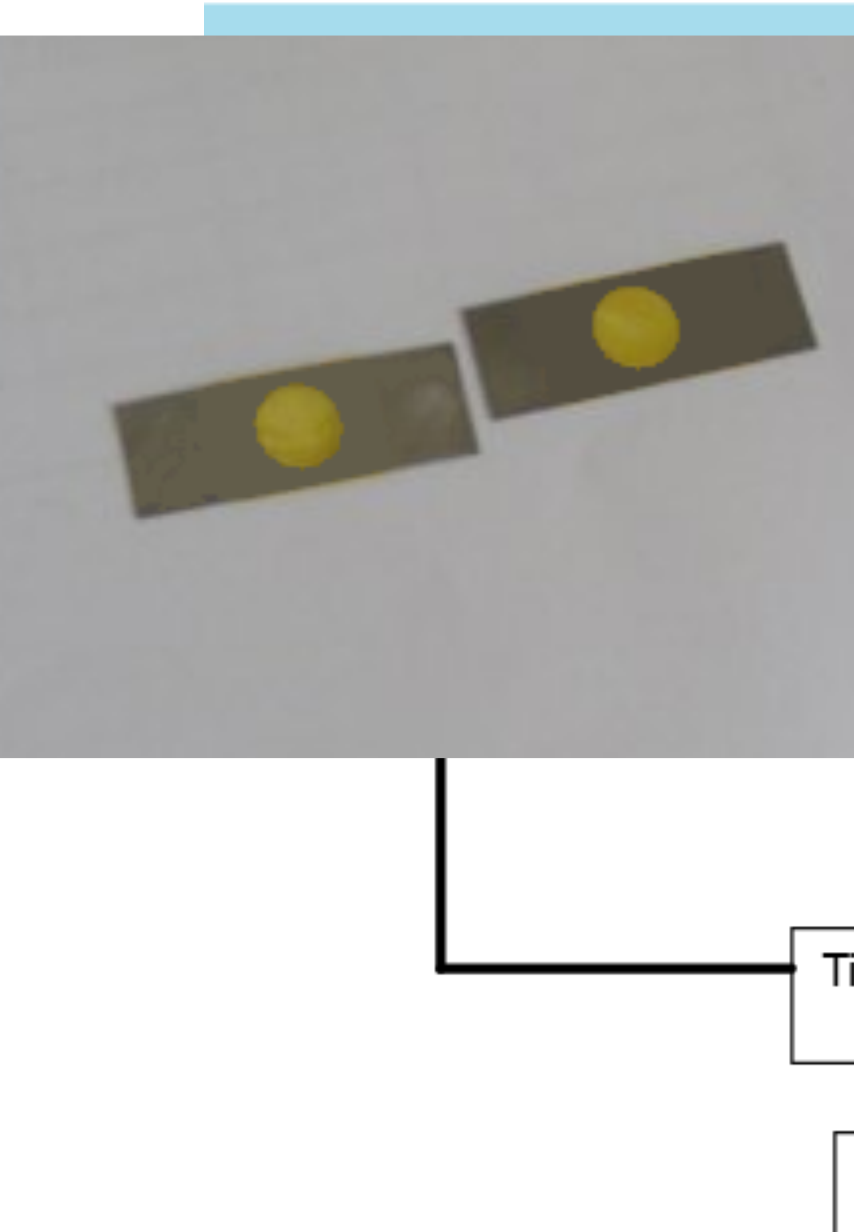
Irradiation

- The selected specimens were implanted in Ion beam laboratory of FEI STU Bratislava.
- The energy of implantation was E_H=2x95 keV for the molecular H_2^+ ion beam.
- Two implantation doses were chosen for both of the alloys: 1.3×10^{19} ions/cm^2 (1.1 C/cm^2) and 5×10^{18} ions/cm^2 (0.4 C/cm^2).
- For the protons with the energy of 95 keV, the range of 480 nm was found by TRIM calculations. This range was chosen with regard to the scope of the PLEPS equipment. The PAS-PLEPS spectra were evaluated using program POSWIN.

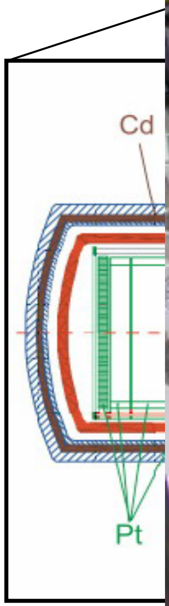


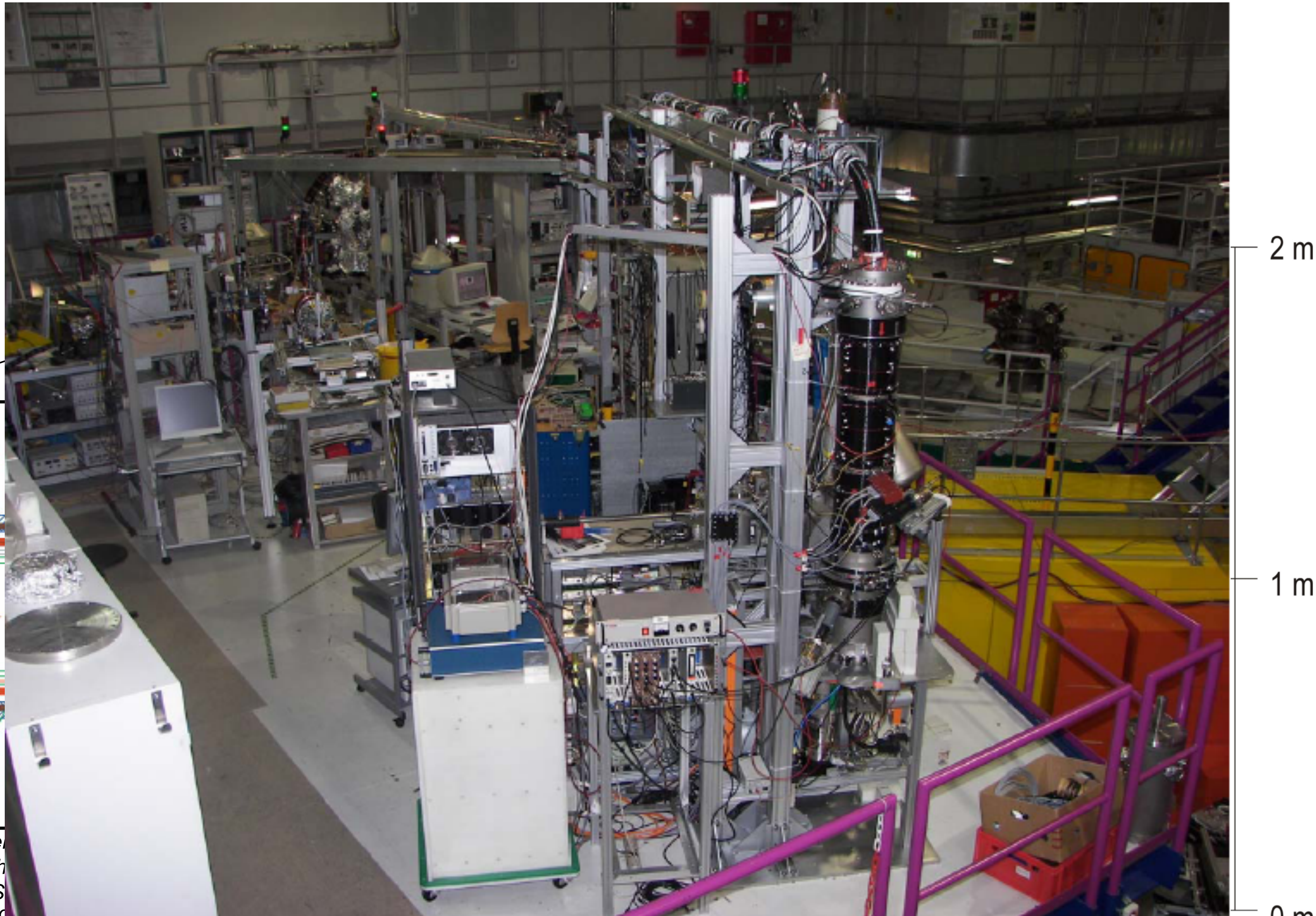
Distribution of vacancies in Cu-alloys after irradiation (calculated by TRIM)
Vacancies [vac/implanted ion]
1,00
0,80
0,60
0,40
0,20
0,00
0
100
200
300
400
500
600
700
800
Target depth [nm]

PALS equipment

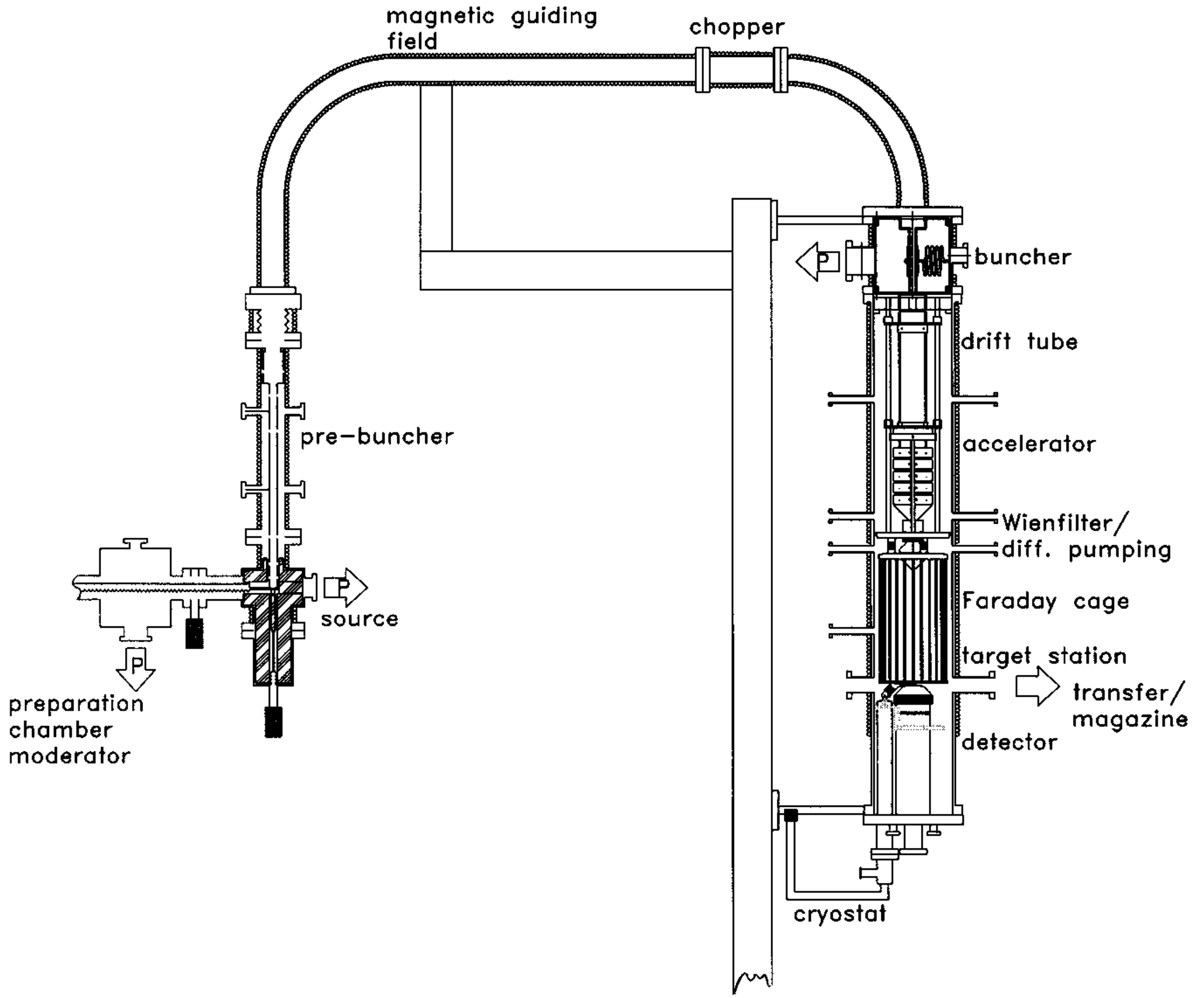


Pulsed low energy positron system (PLEPS)



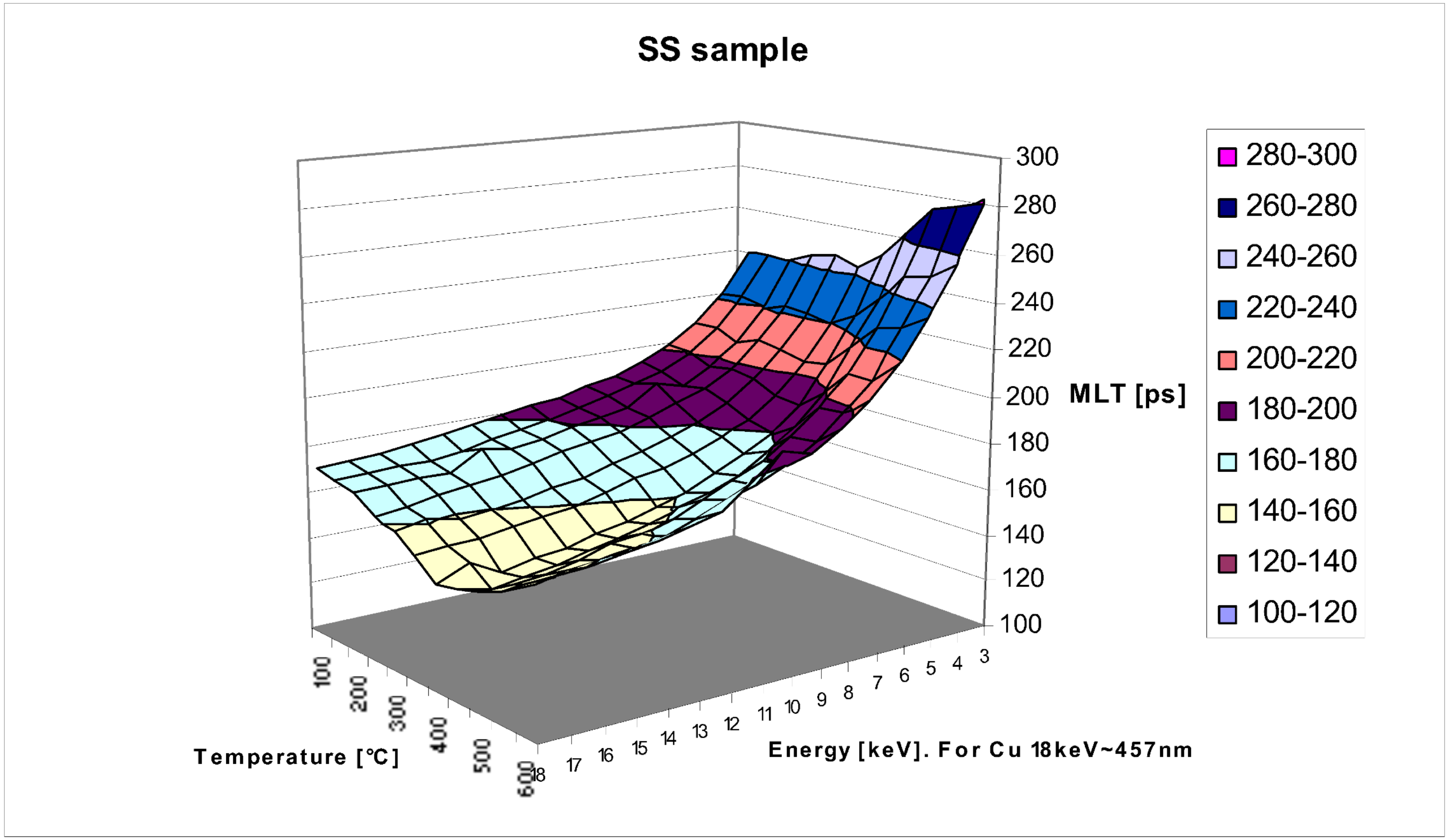


Surface Scie
[2] Hugensch
Repper R., S
Issue 1, p. 29



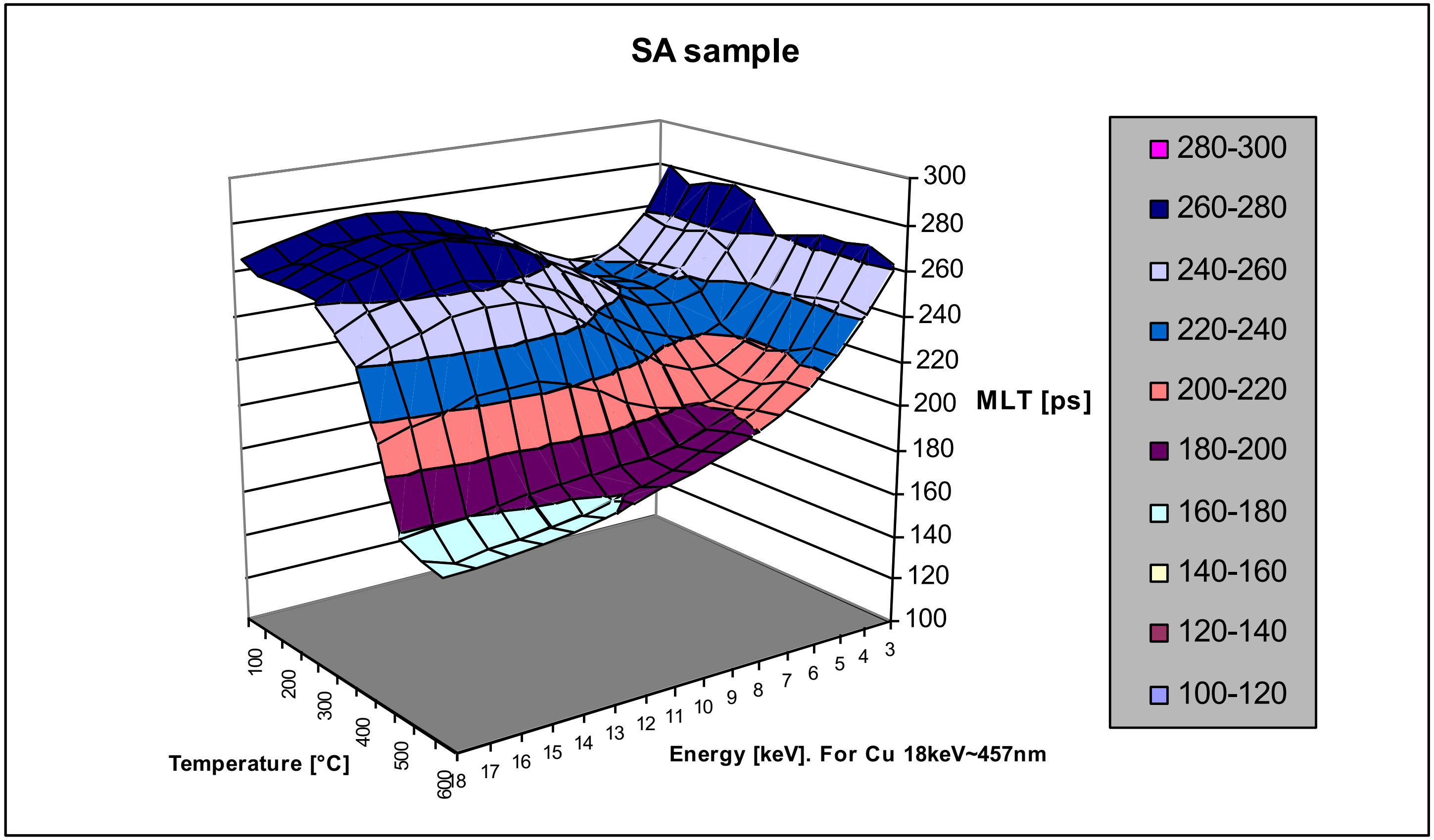
- *Schematic view of the latest version of the low energy pulsed positron system.*

PLEPS Results for ITER



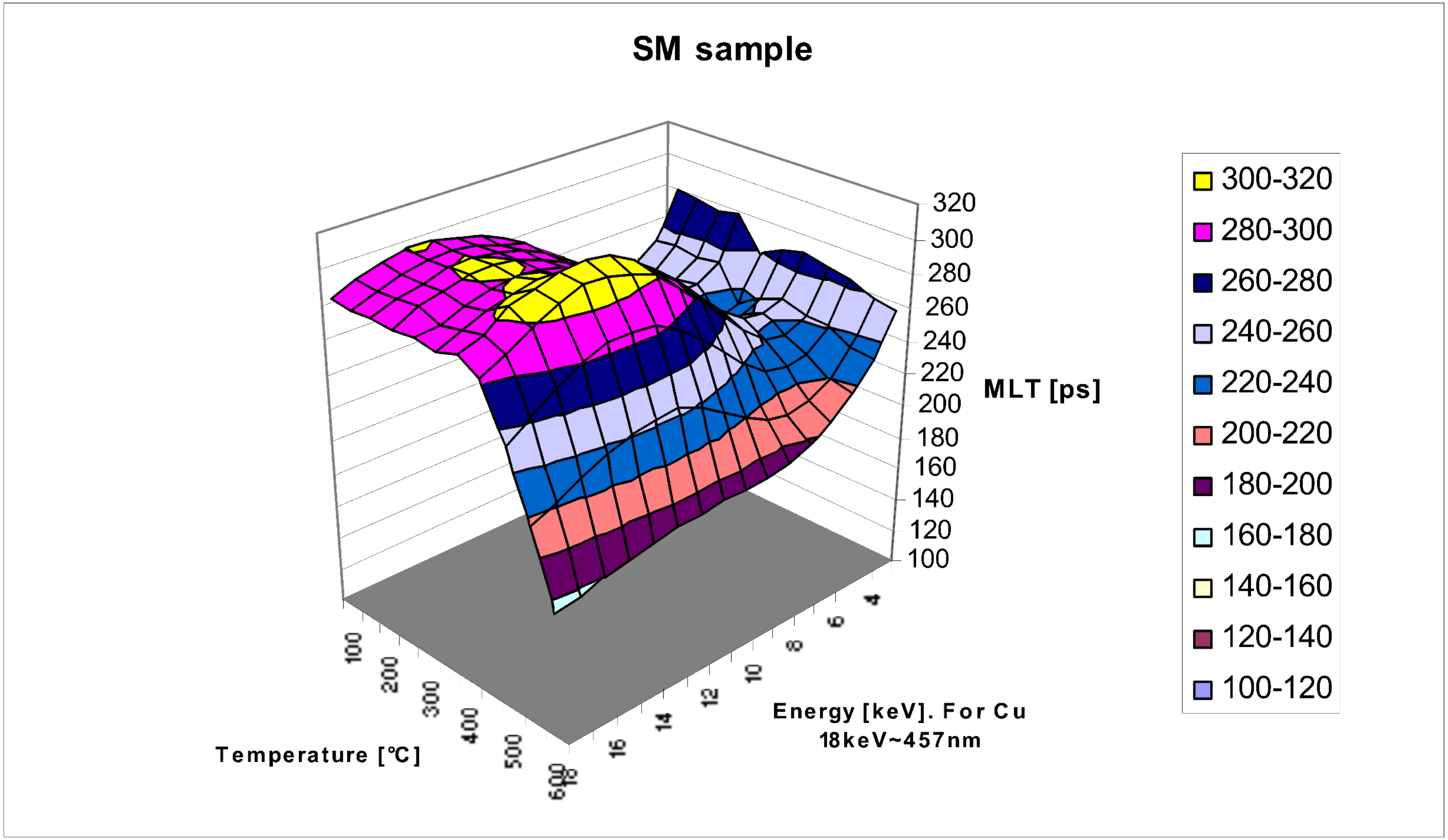
Annealing behaviour of not-irradiated CuCrZr-sample (SS)

PLEPS Results for ITER



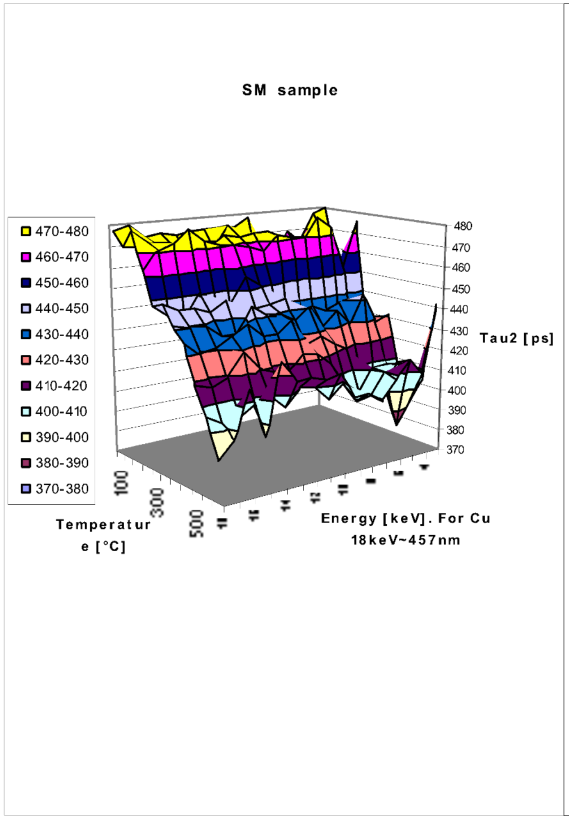
Annealing behaviour of irradiated CuCrZr-sample (SA)

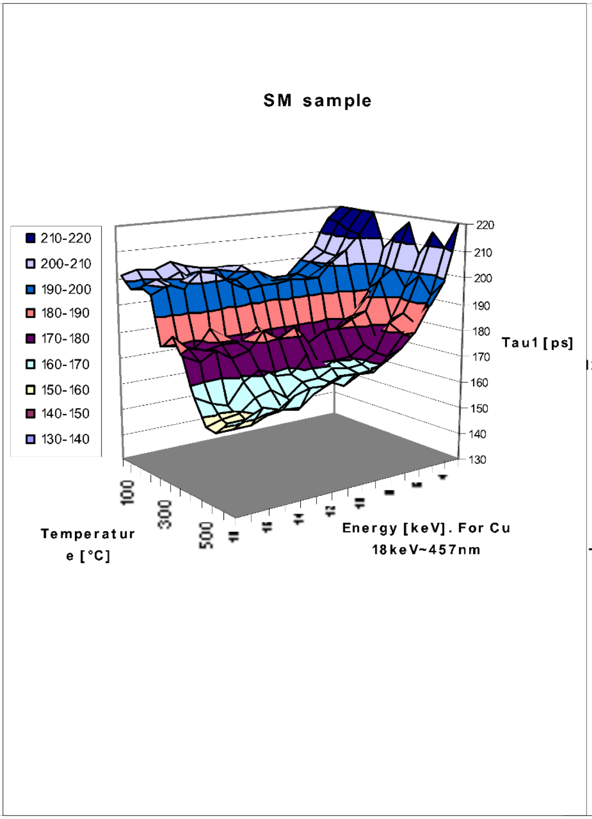
PLEPS Results for ITER

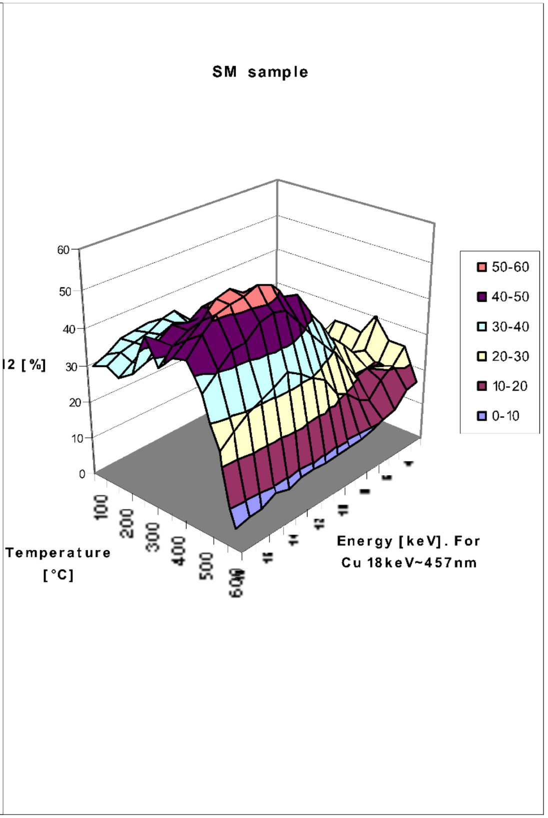


Annealing behaviour of irradiated CuCrZr-sample (SM)

PLEPS Results for ITER

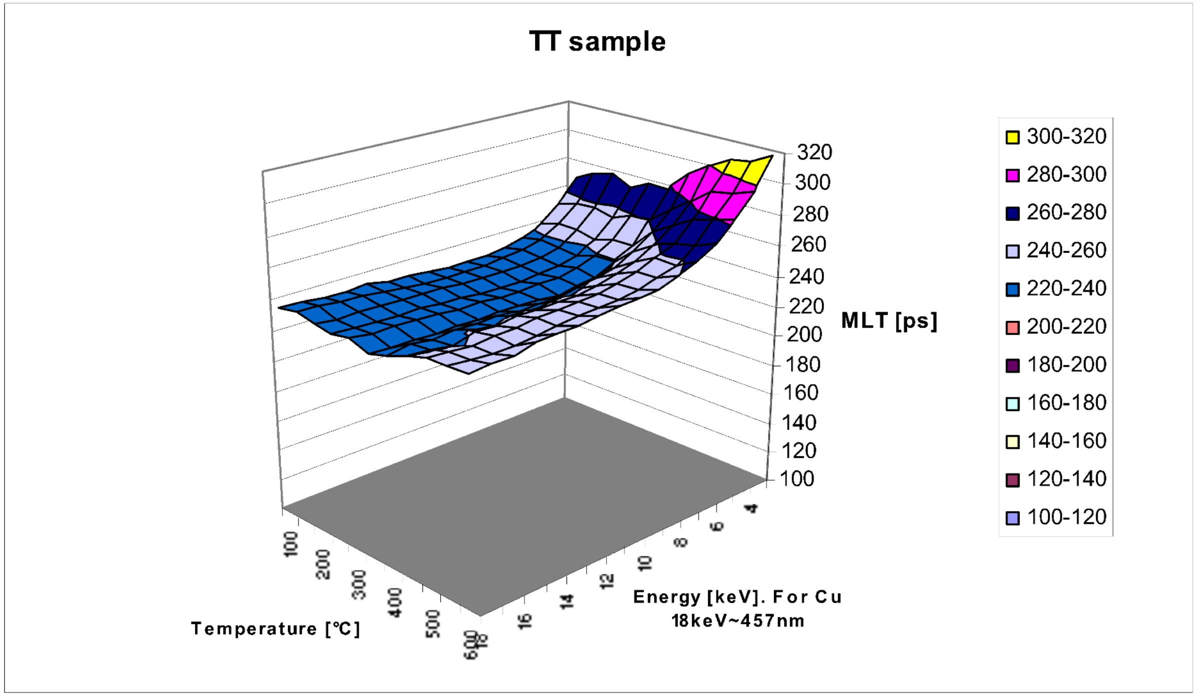






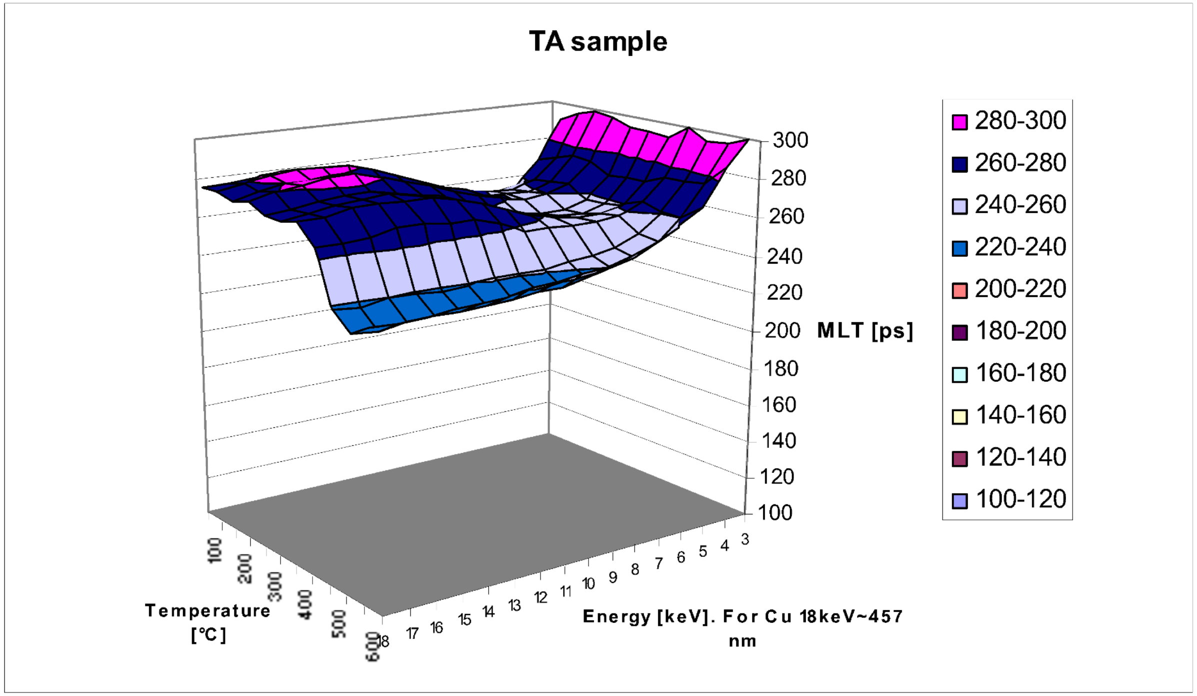
Decomposition of PASLT spectrum in two components

PLEPS Results for ITER



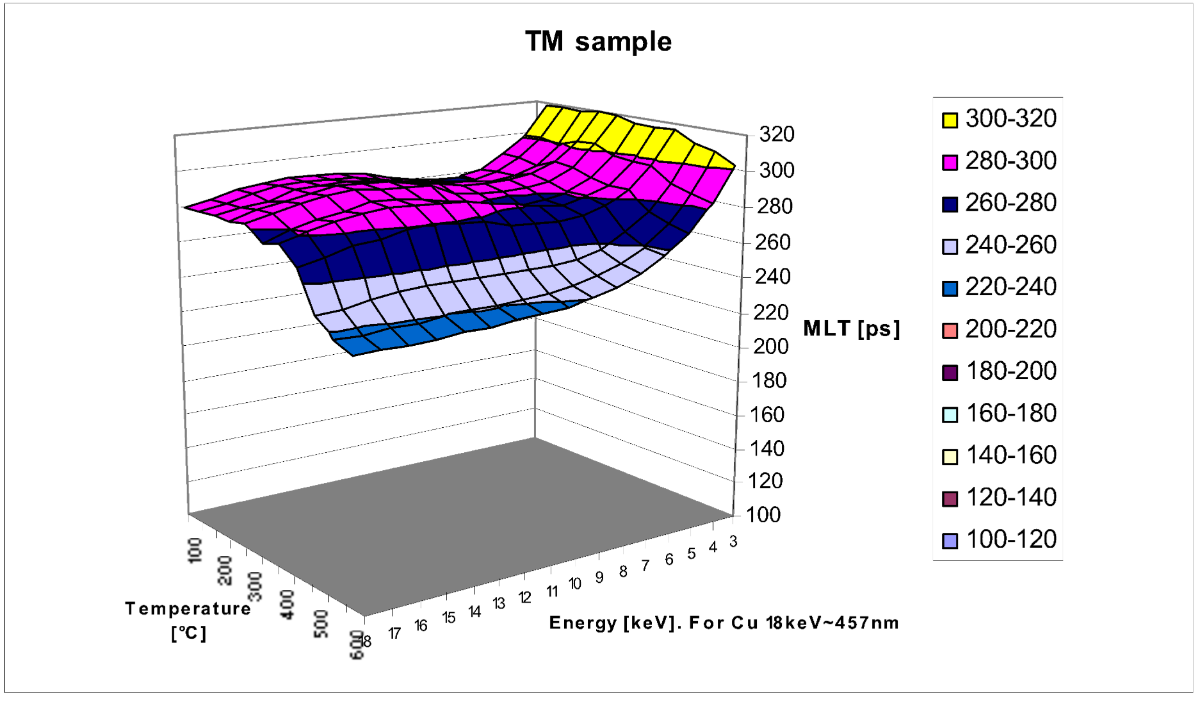
Annealing behaviour of irradiated CuAl25-sample (TT)

PLEPS Results for ITER



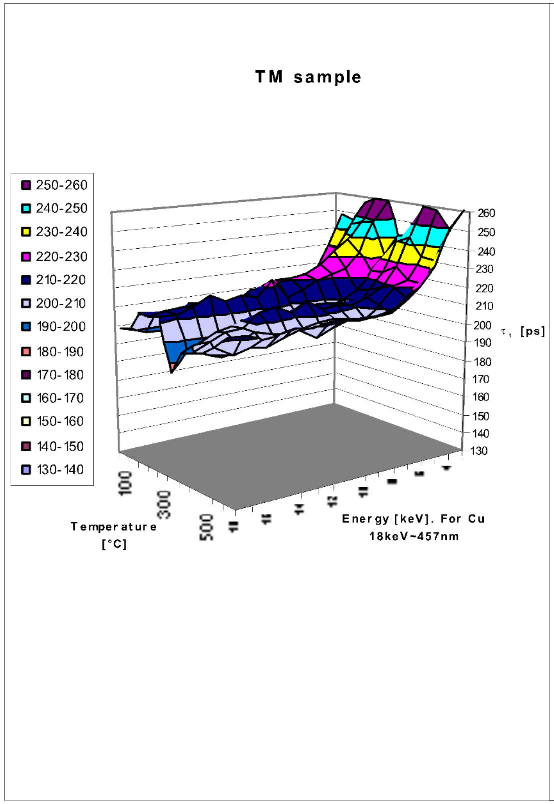
Annealing behaviour of irradiated CuAl25-sample (TA)

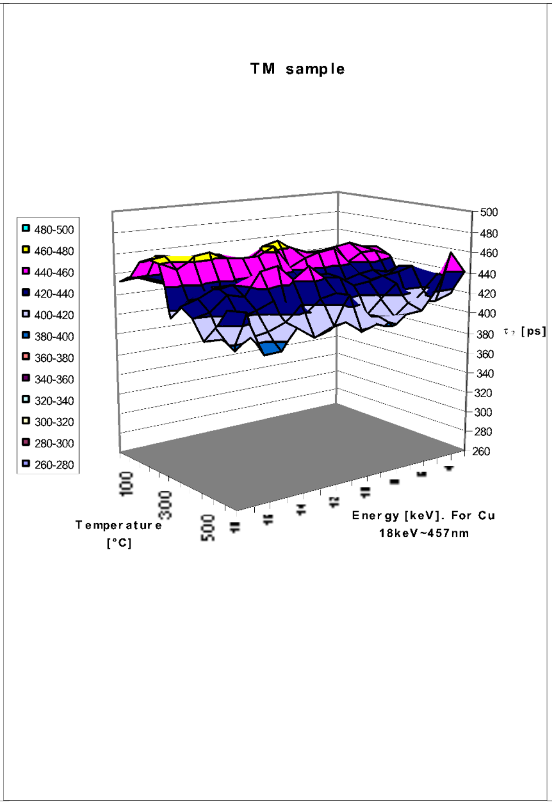
PLEPS Results for ITER

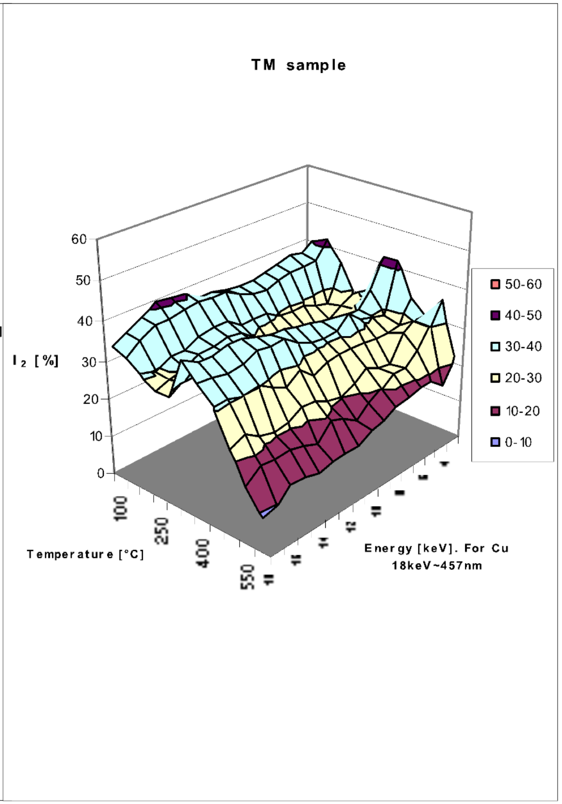


Annealing behaviour of irradiated CuAl25-sample (TM)

PLEPS Results for ITER

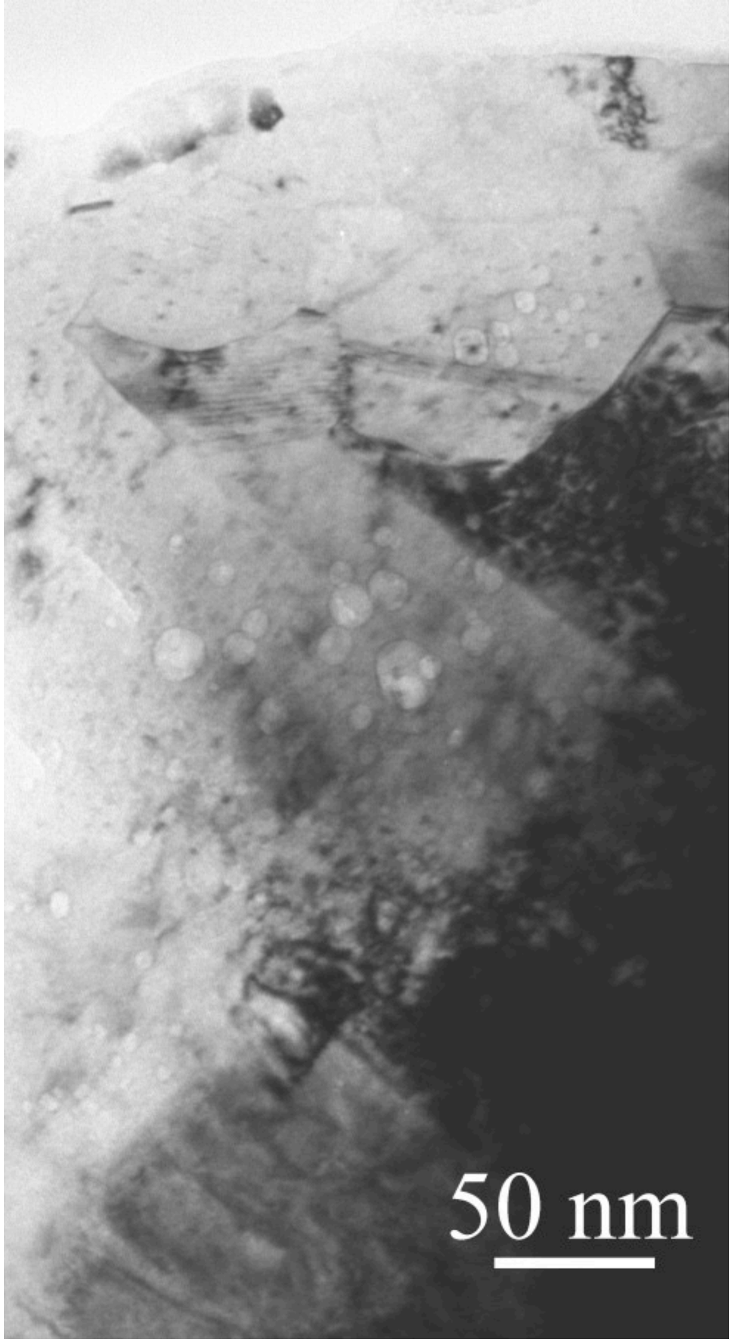




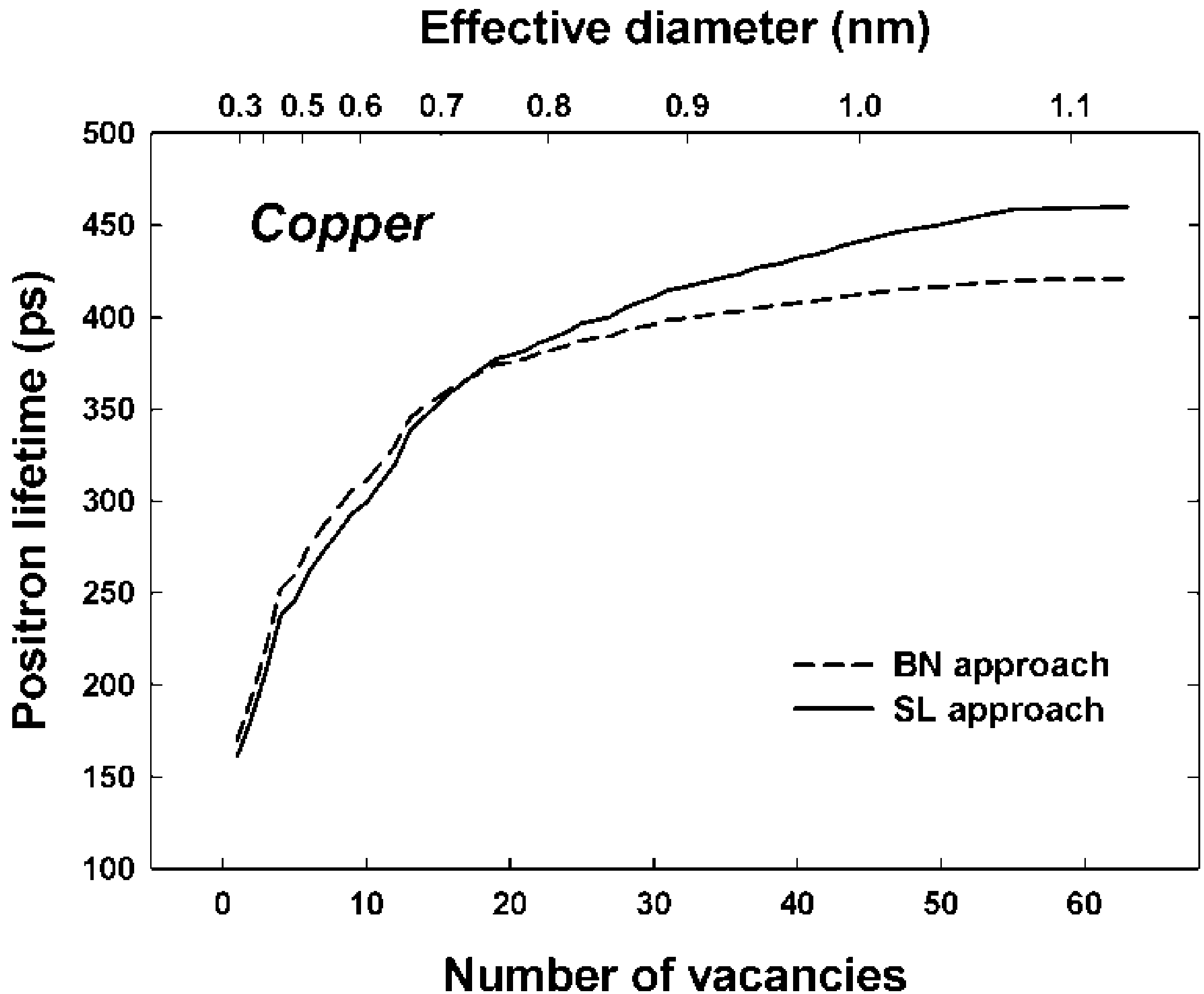


Decomposition of PASLT spectrum in two components

TEM Results



Cross-sectional TEM micrograph of the irradiated polycrystalline CuCrZr alloy (SM specimen).

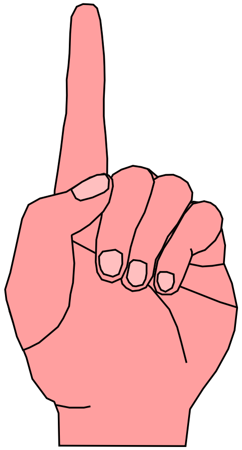


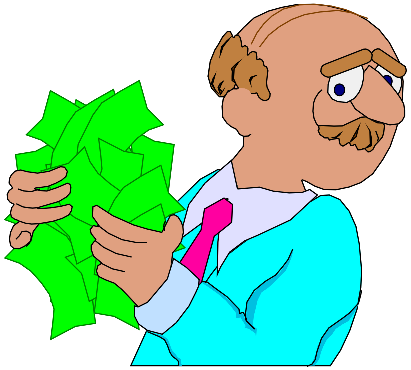
The calculated dependence of the positron lifetime on the size of the vacancy cluster in Cu.

Discussion

- Protons were used instaed on neutrons for the investigation of the changes in the surface of selected copper alloys.

- It is important **to be very precise by sample preparation** and by excluding of contributions from oxides, source, back-diffusion, etc.



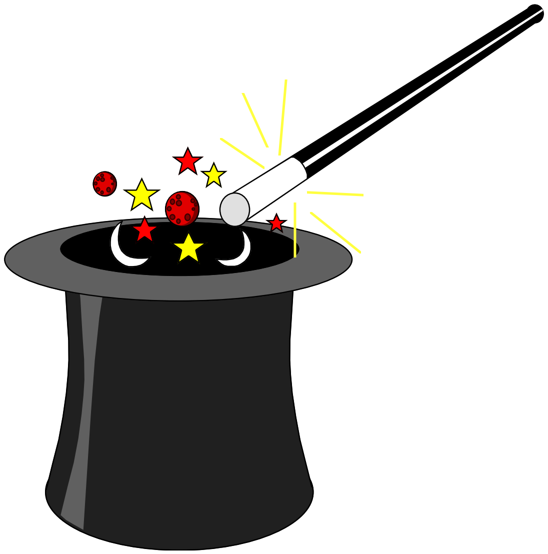


Conclusions

- According to PAS results, the CuAl25 material appeared not to be very sensitive to the change of microstructure upon proton irradiation. The only indication of irradiation is the larger intensity *I*2 corresponding to larger free volumes.
- Contrary, in CuCrZr material positron annihilation characteristics are apparently changed due to irradiation.
- Annealing up to 600 °C seem to result to a state similar to annealed non-irradiated sample.

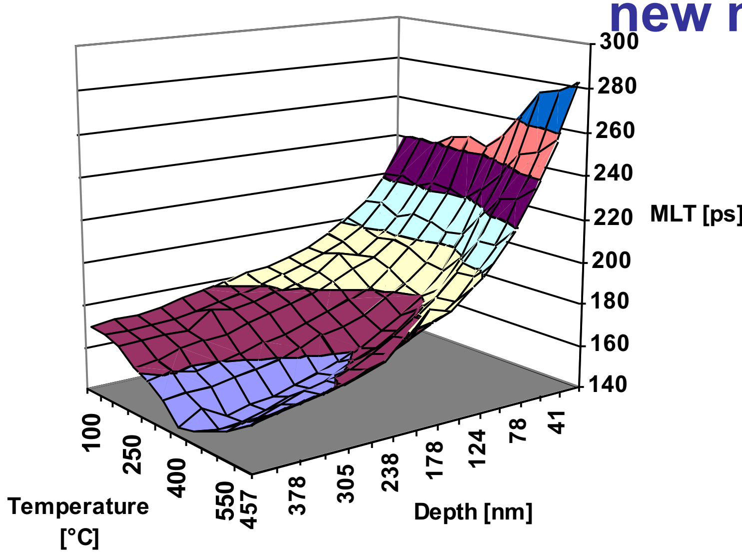
Conclusions

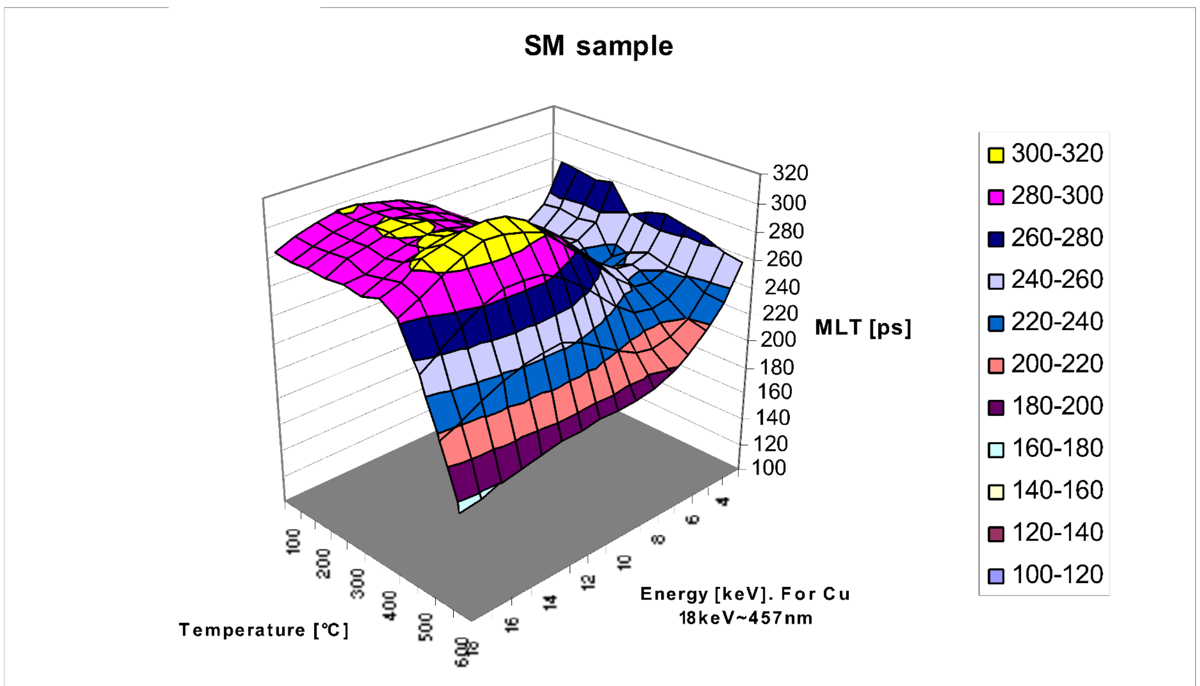
- The distinction between the two studied materials consists – in accordance with TEM observations – in the fact that the microstructure of CuAl25 material is almost intact to irradiation contrary to CuCrZr.
- Larger free volumes seem to play slightly more important role in CuAl25 material in comparison with CuCrZr one.

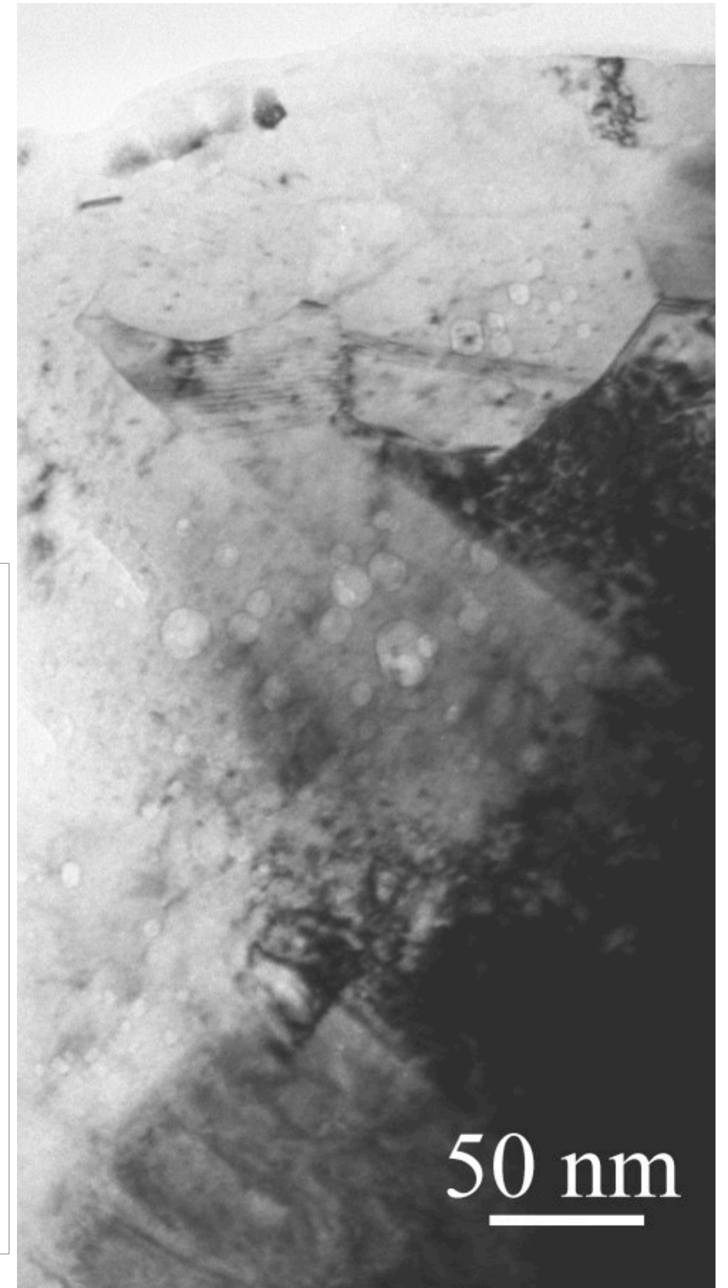


Defects depth profiling study and studies of near-surface region.

PAS and TEM results are useable for microstructural evaluation of new materials

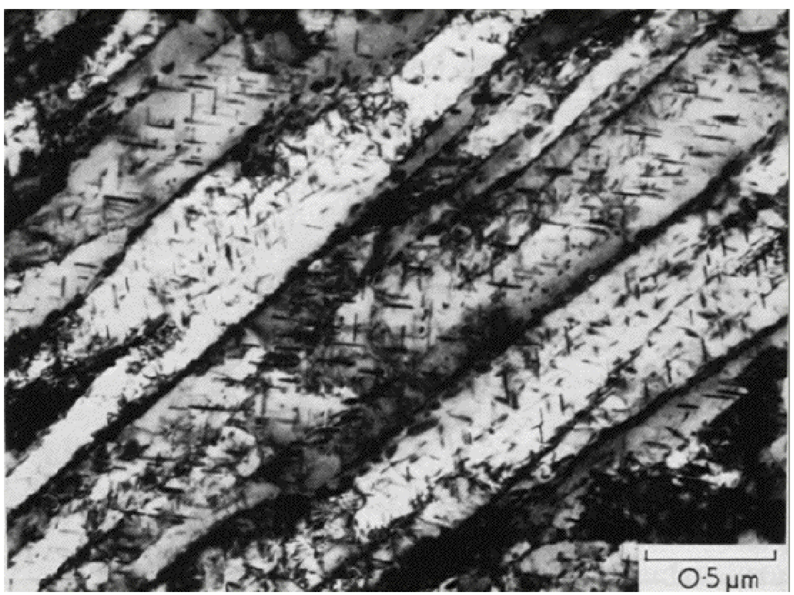




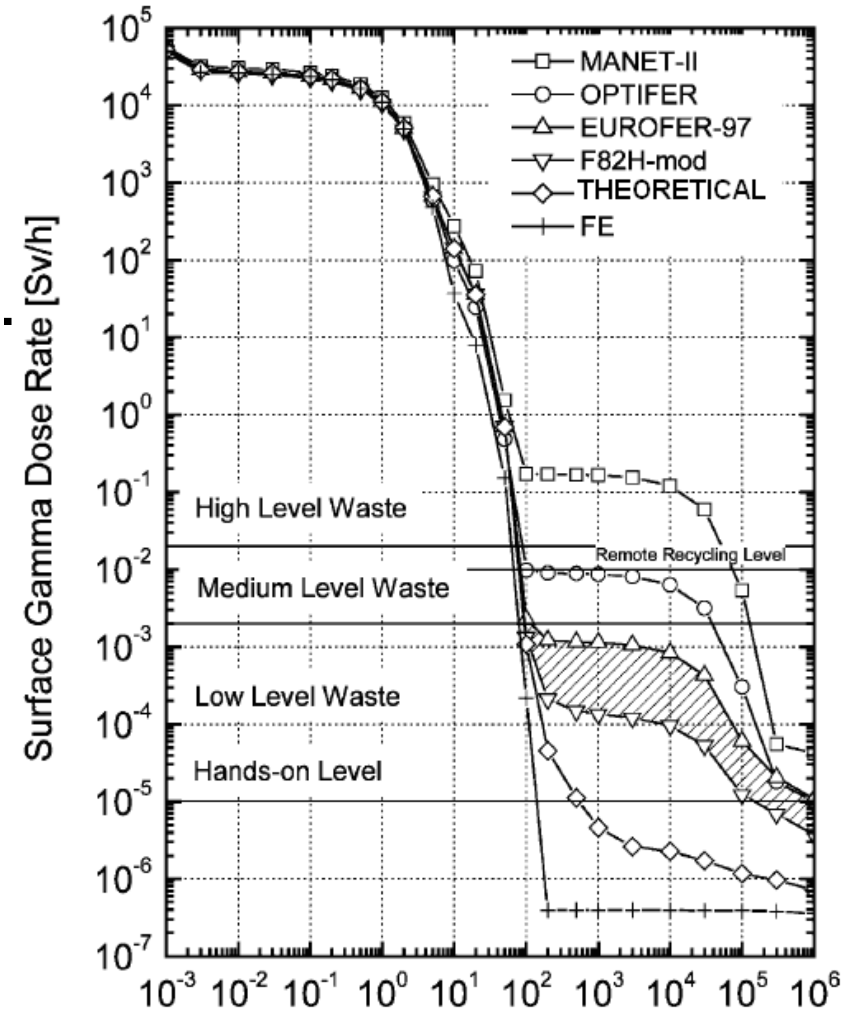


Feritic/martensitic (FM) steels

- Temperature limit - 650°C (ductility loss),
- Appropriate values of strength (martensite) and ductility (ferrite),
- Lower content of nickel and cobalt – reduced activated FM steels (RAFM).



FM steel – martensite looks like needles.

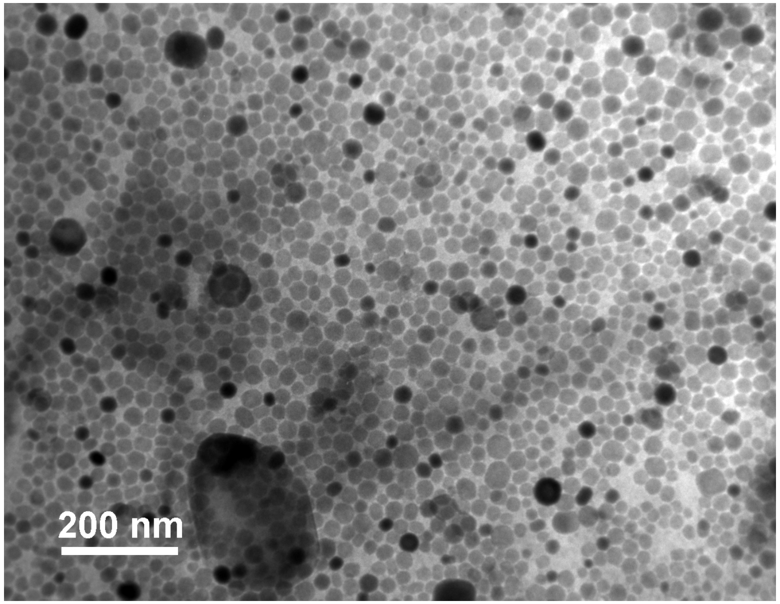


Chemical composition of studied ODS steels (in % wt.).

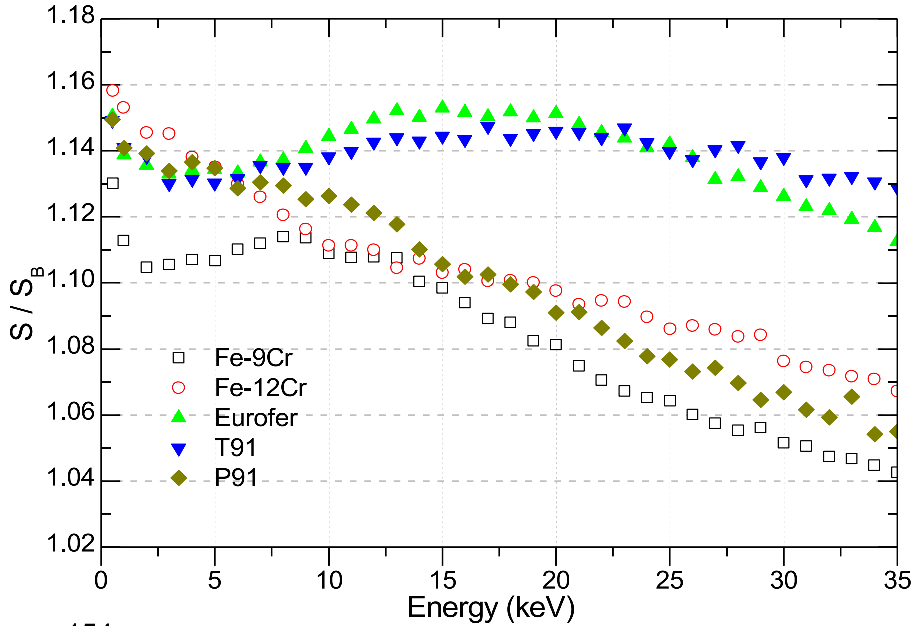
	C	Mn	Ni	Cr	Mo	Ti	Al	Si	W	Y_2O_3
MA 956	0.07	0.12	0.07	20	0.1	0.3	3.4	0.04	-	0.5
ODM 751	0.07	0.07	0.02	16	1.74	0.7	3.8	0.06	-	0.5
ODS Eurofer	0.1	0.44	-	9	0.01	-	-	0.01	1.1	0.3

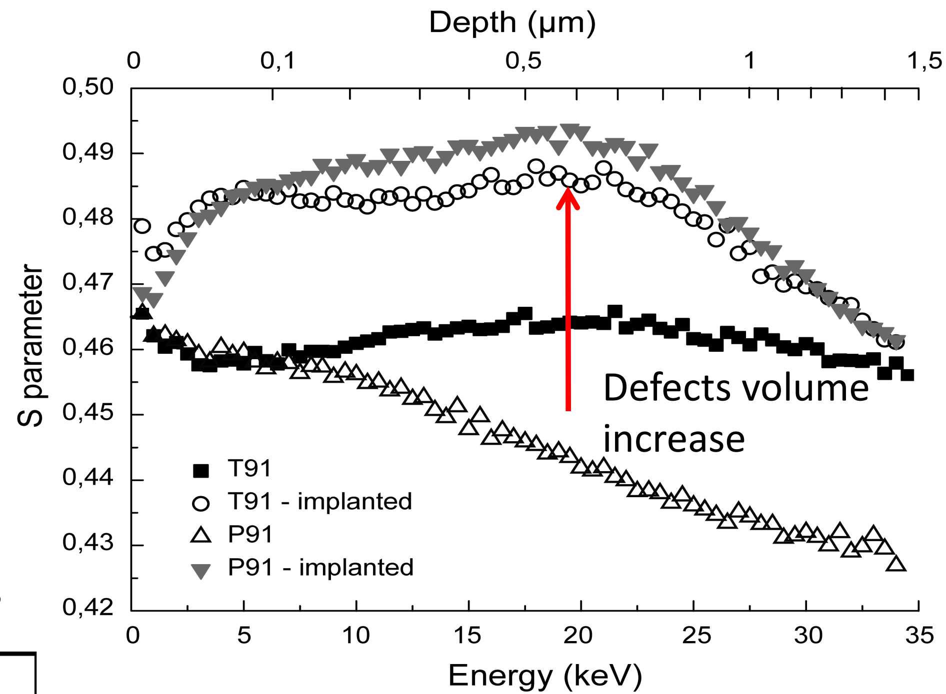
- Addition of stable oxides (Al_2O_3, Cr_2O_3, Y_2O_3)
- Better mechanical properties – strength, toughness
- Better corrosion resistance and resistance to thermal loading
- Candidate materials for fuel cladding in new reactors (fast reactors)

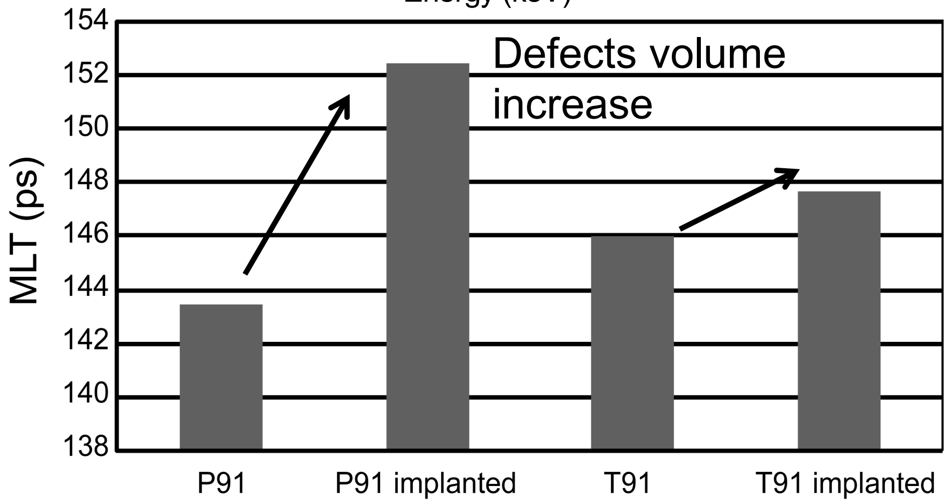
Oxide Dispersion Strengthening might improve the swelling resistance of F/M steels



Study of implanted *9Cr RAFM* steels (PALS & DBS)







- Accumulation of vacancies due to helium implantation – 500 keV, 1.8×10^{18} ions.cm^{-2} (~ 20 dpa).

- T91 with tungsten has smaller change of positron data after implantation.

J. Veterníková, J. Degmová, S. Kilpeläinen, V. Slugeň, F. Tuomisto, J. Räisänen, V. Kršjak, M. Petriska, S. Sojak, R. Hinca, M. Stacho (2012) ***Nucl. Eng. Des.*** *251, 354.*

Conclusion

- Defect concentration, defet size (PALS)
- Defect concentration, defet size (DBS)
- Hardness = residual stress (MBN)

Same or similar results

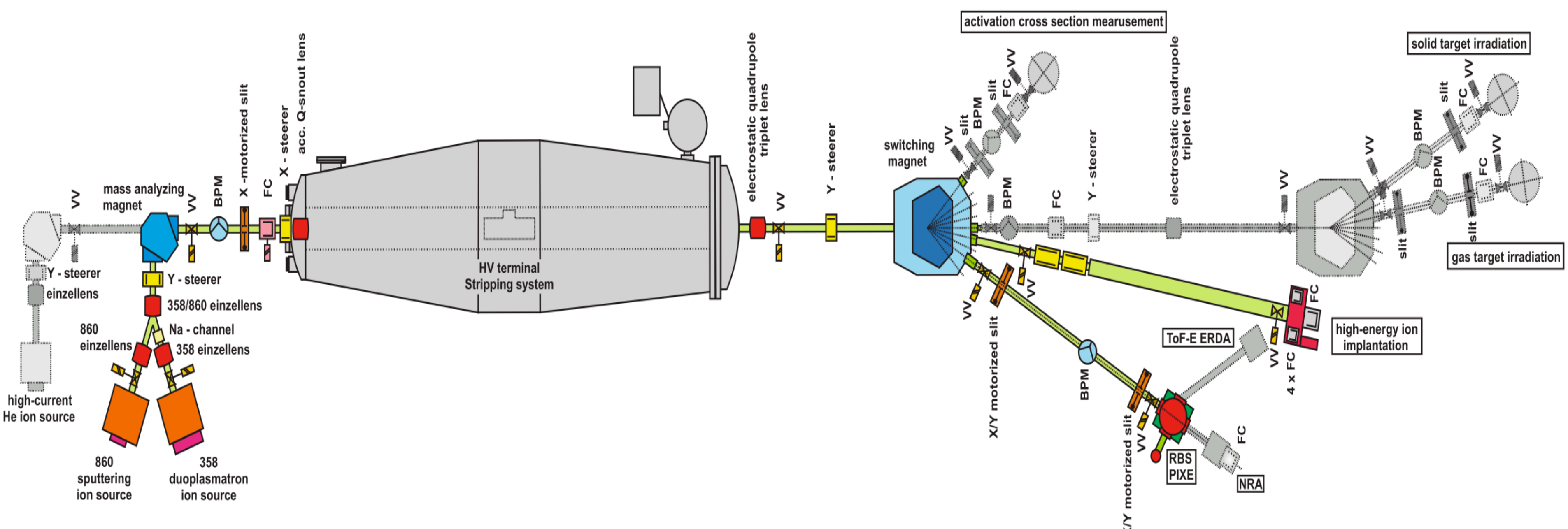
Hardness (residual stress) increases with defect concentration growth, no with defect size growth (precipitation vs dislocation)

- No relation to chromium content as was assumed
- Influence of Cr + Mo + W and aslo Al on hardness (creation of precipitates

Ion implantations for conventional PAS and PLEPS

New 6MV Tandetron system at STU enable to implant He to 70μm in Fe

- 6MV Tandetron system
- He^{+} or He^{2+} - max. 18MeV = implantation depth little more than 70μm
- He beam current of up to 5 emA
- possibility sample heating up to 800°C and cooling down to LN2 temperature
- Sample holder on a four-axis goniometer 0-45°
- More at *P. Noga et al, Nuclear Instruments and Methods in Physics Research, 2017* Section B



Thank you!

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