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Investigation of samples of F/M and ODS steels irradiated in the spallation source SINQ by Positron Annihilation

Outline

- BACKGROUND – HELIUM VS. DISPLACEMENT DAMAGE
- POSITRON ANNIHILATION – MODELS AND EXPERIMENTS
- WHAT WE KNOW SO FAR
  - POSITRON TRAPPING RATES
  - HE/V RATIO
  - STAGES OF MICROSTRUCTURAL EVOLUTION
  - DPA VS. TEMPERATURE
  - EFFECT OF OXIDE DISPERSOIDS
- SUMMARY AND CONCLUSIONS

INTRODUCTION

HELIUM VS. DISPLACEMENT DAMAGE

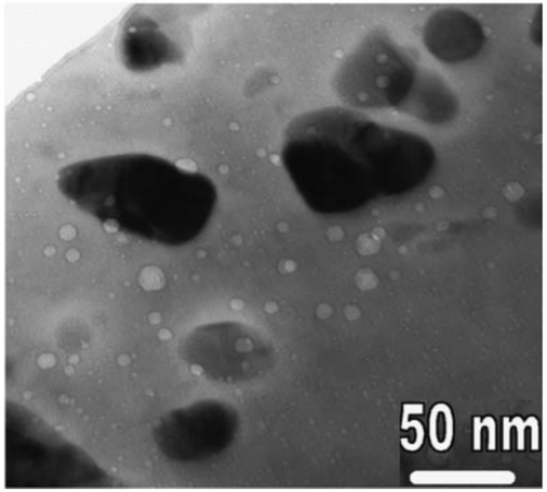
Helium in nuclear structural materials

Helium is practically **insoluble in all metals** and tends to cluster with various defects and to precipitate as gas bubbles.

Accumulation of He can lead to accelerated growth of voids and formation of He bubbles → **embrittlement, hardening, void swelling** …

Pronounced He effects for He concentrations **> 500 appm**

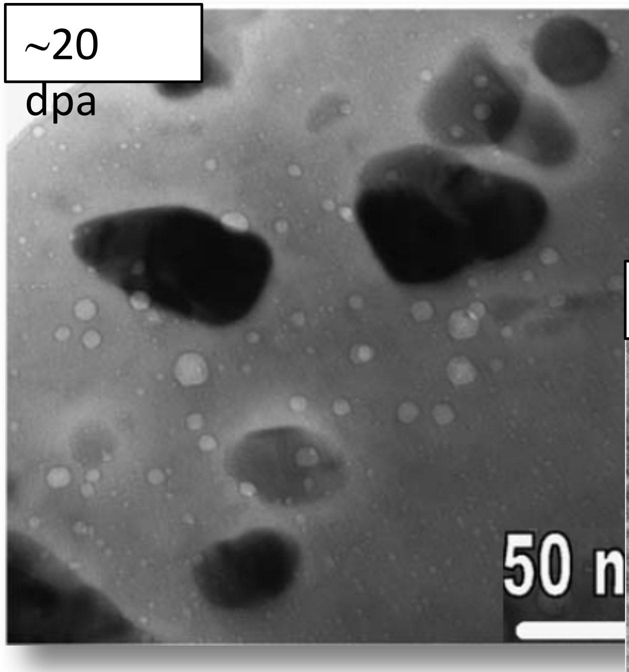
Typical production route is via interactions of metals with high energy neutrons or protons. Another significant production route is via interaction of thermal neutrons with some elements e.g. nickel, boron.



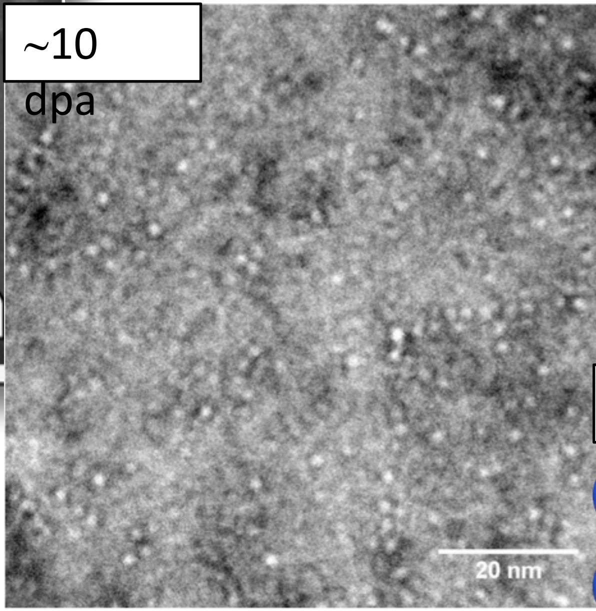
Typical helium production rates in nuclear technology		
(n, α)	Fast breeder reactors	5-15 ppm/y
(n, α)	Fusion devices	50-300 ppm/y
(p, α)	Spallation sources	up to 100 ppm/d
α	Ion implantation	up to several %/h

H. Ullmaier Radiation Effects 78 (1983

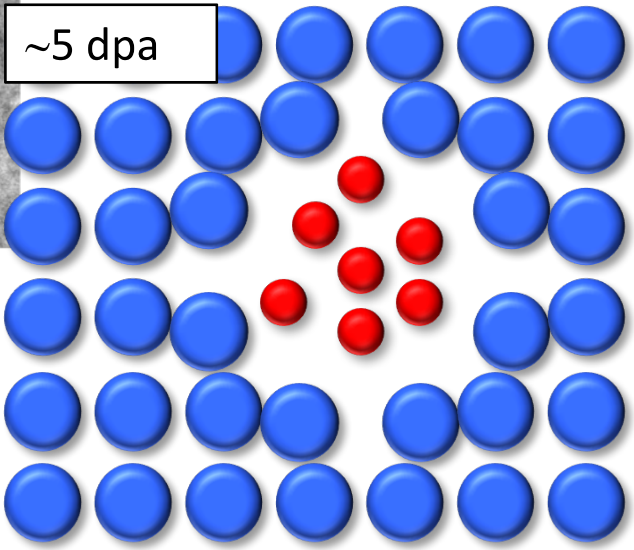
Imaging techniques limitations



Severe displacement damage, large He bubbles; He bubbles can be well characterized by TEM and STEM/EELS; Lot of experimental data – good level of understanding



Intermediate damage, small helium bubbles; quantitative characterization by TEM; limited information on helium – good level of understanding



Early stage displacement damage, He-V clusters; almost no experimental data from electron microscopy techniques

Limitations of different experimental approaches:

EELS: Limited by TEM resolution

MD: Limited by computing capacity (large defects) and the transition from gaseous to solid-like state of helium (small defects)

PAS: Lifetime of positron in empty clusters saturates at >30 vacancies; PAS characterization limited by positronium formation (not observed) and/or saturated trapping (observed in few cases)

POSITRON ANNIHILATION – MODELS AND EXPERIMENTS

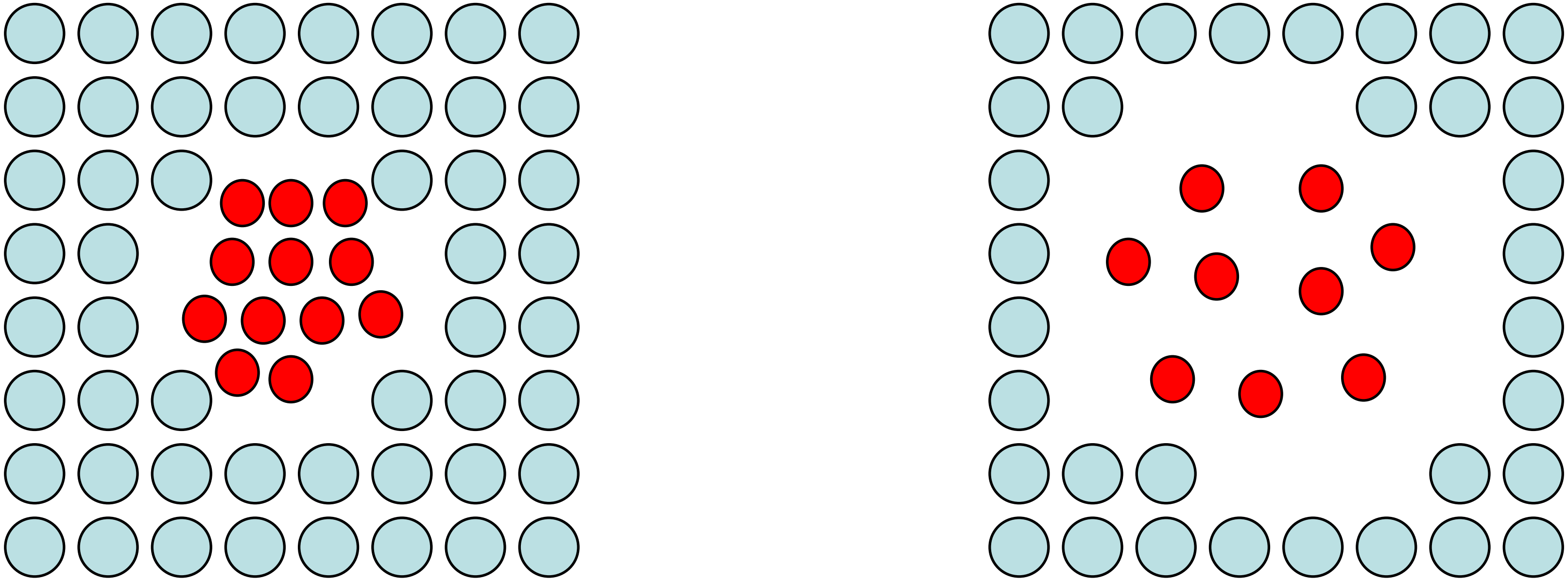
Characterization of vacancy-type defects

In most spallation samples - represented by two groups of defects, in addition to the bulk component:

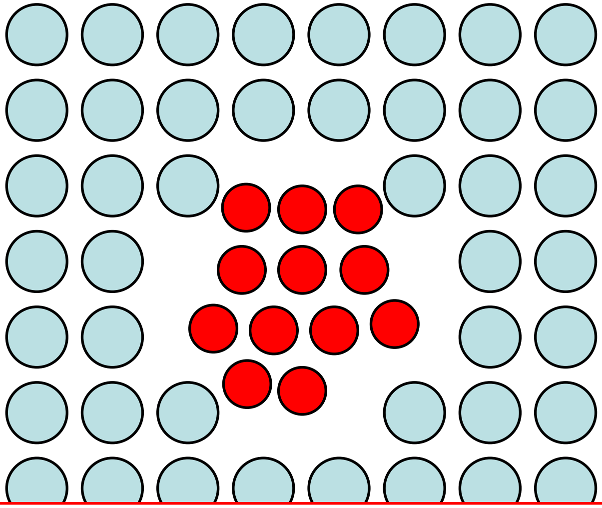
Defect 1: τ_2, I_2 is attributed to small VC

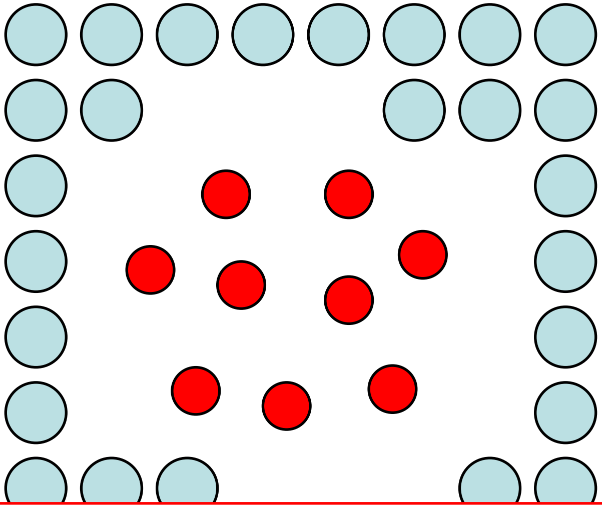
Defect 2: τ_3, I_3 is attributed to clusters/bubbles up to 1nm

At low-dpa samples these two defects represent cluster of about dozen vacancies and clusters of about 30 vacancies respectively.



PALS ANALYSIS OF LOW DPA SAMPLE





Situation gets more complicated at high dpa’s (i.e. large bubbles cannot be quantitatively characterized by means of PALS; can be hardly modeled by two defect components)

helium.

$V_{12}He_{12}$ (He/V=1) gives a good agreement with MD calculations

Clusters ≤30 with only a small amount of helium (He/V < 0.3)

THE RATE OF POSITRON TRAPPING AT DEFECTS

$$\kappa_1 = \frac{I_2 I_3(\lambda_{D1} - \lambda_{D2}) + I_2(\lambda_b - \lambda_{D1})}{I_1} = \boxed{\mu_{D1}} N_{D1}$$

$$\kappa_2 = \frac{I_2 I_3(\lambda_{D2} - \lambda_{D1}) + I_3(\lambda_b - \lambda_{D2})}{I_1} = \boxed{\mu_{D2}} N_{D2}$$

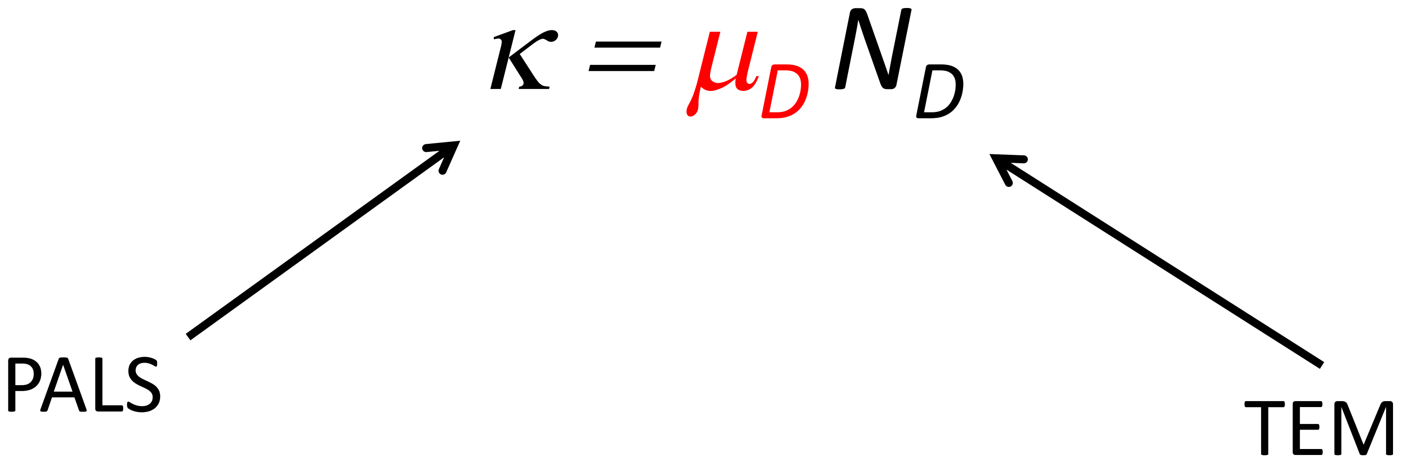
The Specific positron trapping rate (Trapping coefficient) is constant of proportionality between positron trapping rate and number density of given defects.

Quite accurate values available for simple defects; only limited data published on complex defects (helium nano-bubbles)

WHAT DO WE KNOW SO FAR

Positron trapping coefficient

Empirical determination of positron trapping coefficient at nano-scale “helium bubbles”



$$\kappa = \mu_D N_D$$

PALS

TEM

Vacancy clusters/helium bubbles of **~ 1nm** are “visible” for both **TEM** and **PALS**. Mutual comparison helps to estimate the positron trapping coefficient.

Positron trapping coefficient

Decomposition of the PALS spectra measured on studied materials

Material	dpa/T[°C]	τ_1 [ps]	τ_2 [ps]	τ_3 [ps]	I_2 [%]	I_3 [%]	MLT [ps]	FV
F82H	17.5/296	**24**	233	**500**	72.3	13.9	241.5	0.95
F82H	20.3/344	**21**	245	**500**	69.5	18.6	265.9	1.05
CLAM	12.2/263	**14**	245	490	77.8	14.0	260.4	0.97
CLAM	18.3/406	**10**	271	**500**	84.0	9.3	274.8	0.84

Trapping coefficient as determined for helium bubbles in F/M steels

Material	dpa*	T* [°C]	Bubble size [nm]	Bubble density [$\times 10^{23}$ m^{-3}]	κ_b [$\times 10^{9}$ s^{-1}]	μ_b [10^{-14} m^{3}s^{-1}]
F82H	17.5	296	1.04	7.53	5.5	0.7
F82H	20.3	344	1.19	3.84	8.9	2.3
CLAM	12.2 (12.8)	263 (204)	1.1	16.9	9.3	0.6
CLAM	18.3 (19.2)	406 (308)	1.75	5.9	7.9	1.3
					Average	1.2 ± 0.8

TEM

$$\kappa = \mu_D N_D$$

PALS

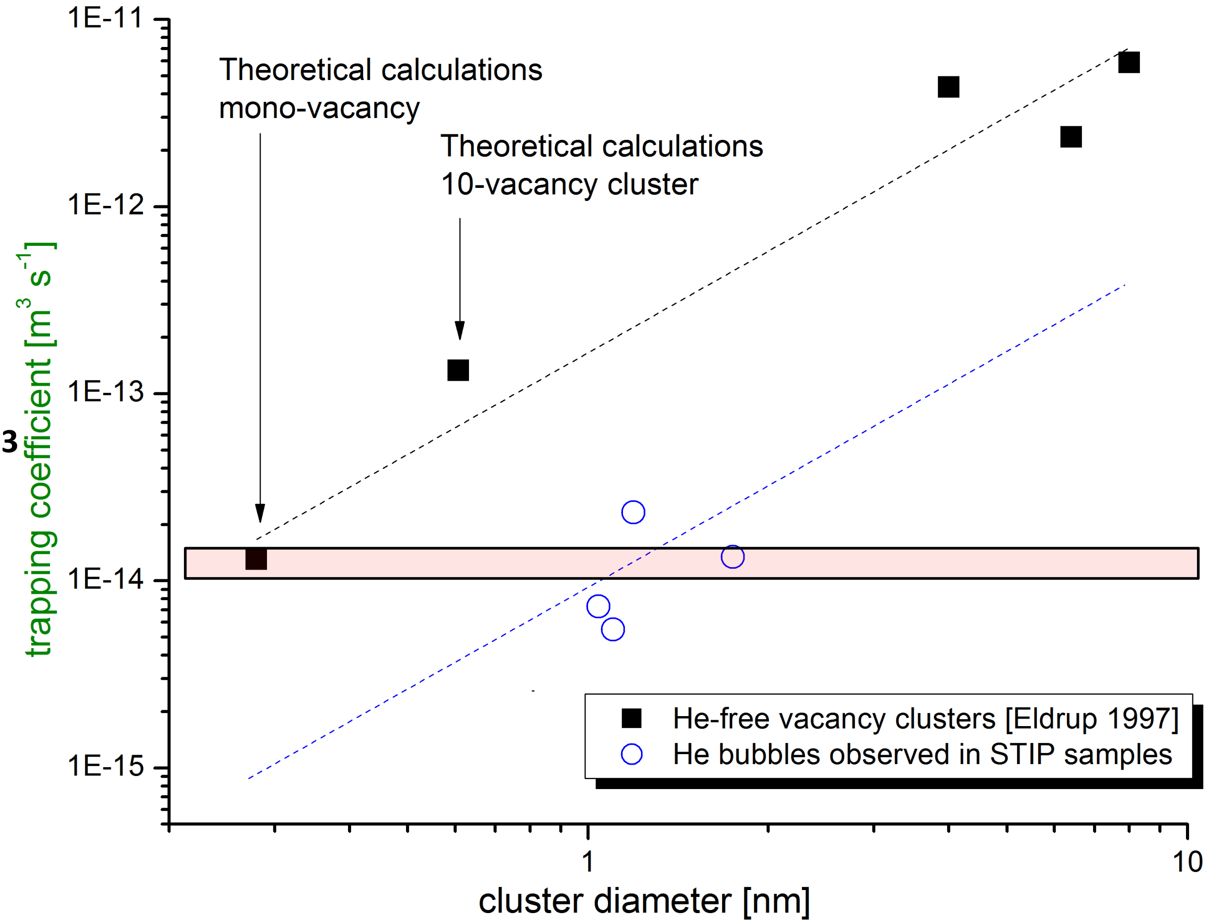
$$\kappa$$

Positron trapping coefficient

Empirical determination of positron trapping coefficient at nano-scale "helium bubbles"

$$\kappa = \mu_D N_D$$

PALS	TEM
~ 1nm	1-1.5 nm
~ 5×10^{9} s^{-1}	~ $5\text{-}10\times10^{23}$ m^{-3}

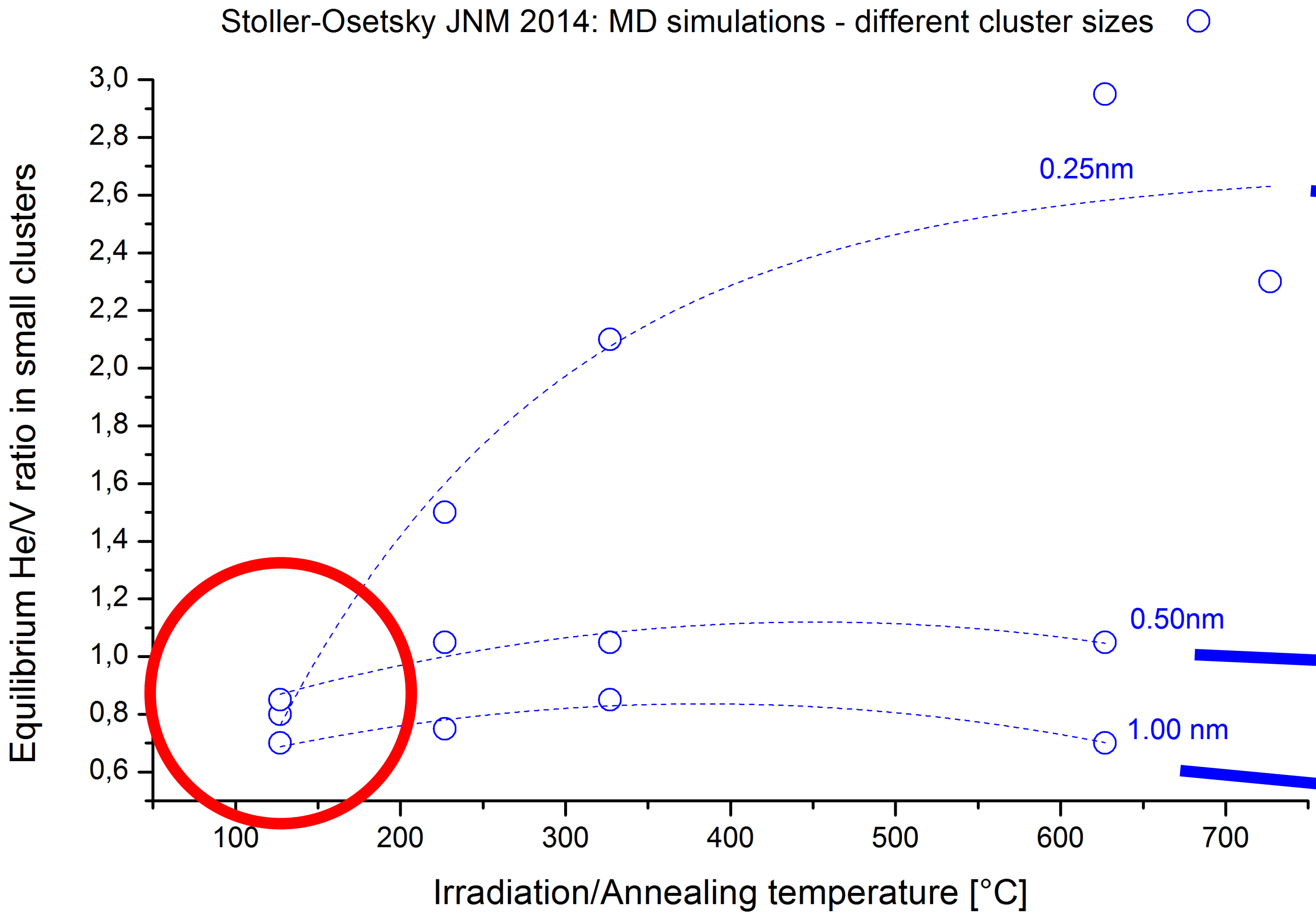


Positron trapping coefficient for small He bubbles in f/m steels $1.2 \pm 0.8 \times 10^{-14}$ m^3s^{-1}

Similar to trapping coefficient of $He_{12}V_{12}$ cluster and empty monovacancy (1.3×10^{-14})

He/V ratio – small clusters

MD simulation BCC Fe system 128 000 atoms and



9 vacancies

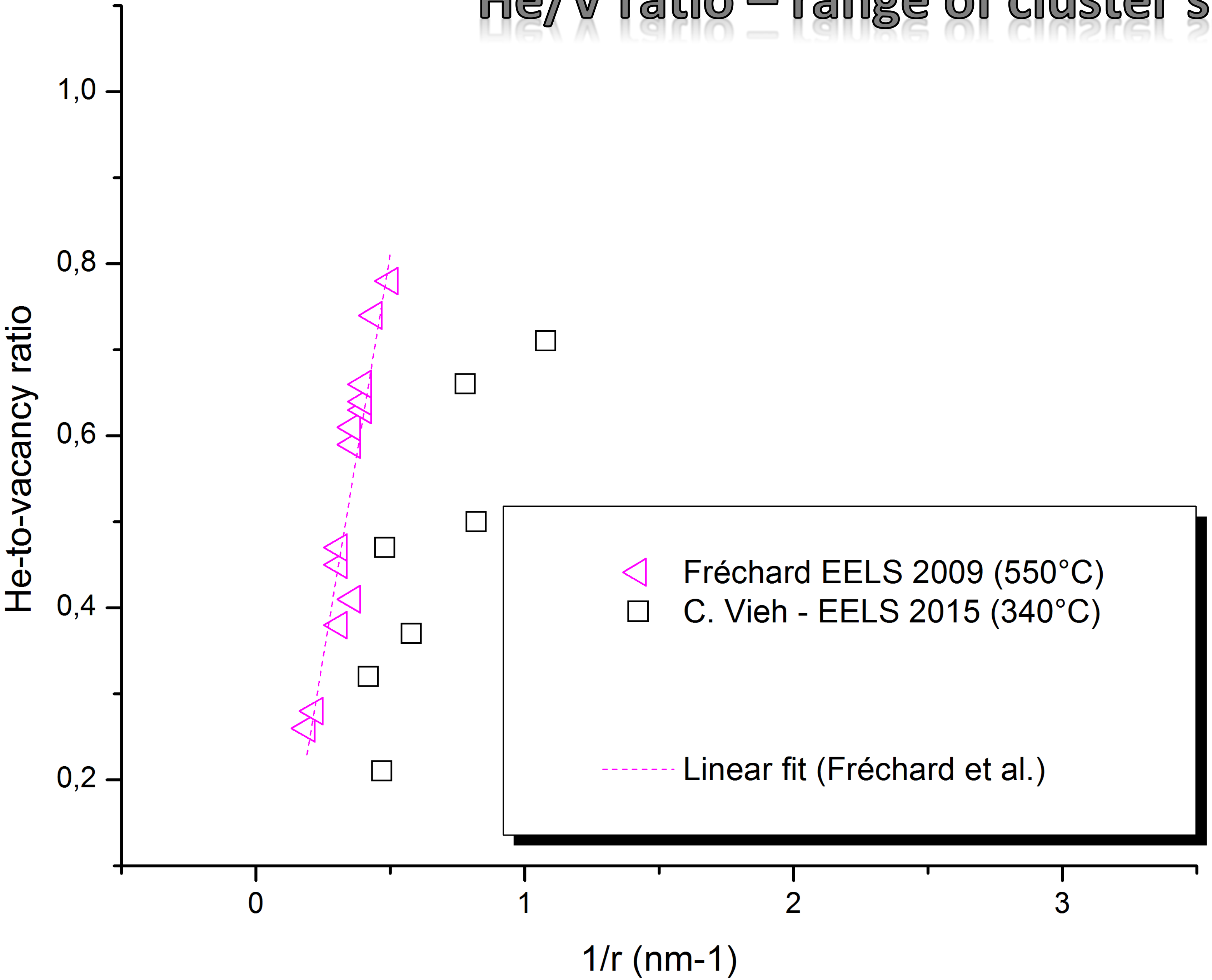
12 vacancies

45 vacancies

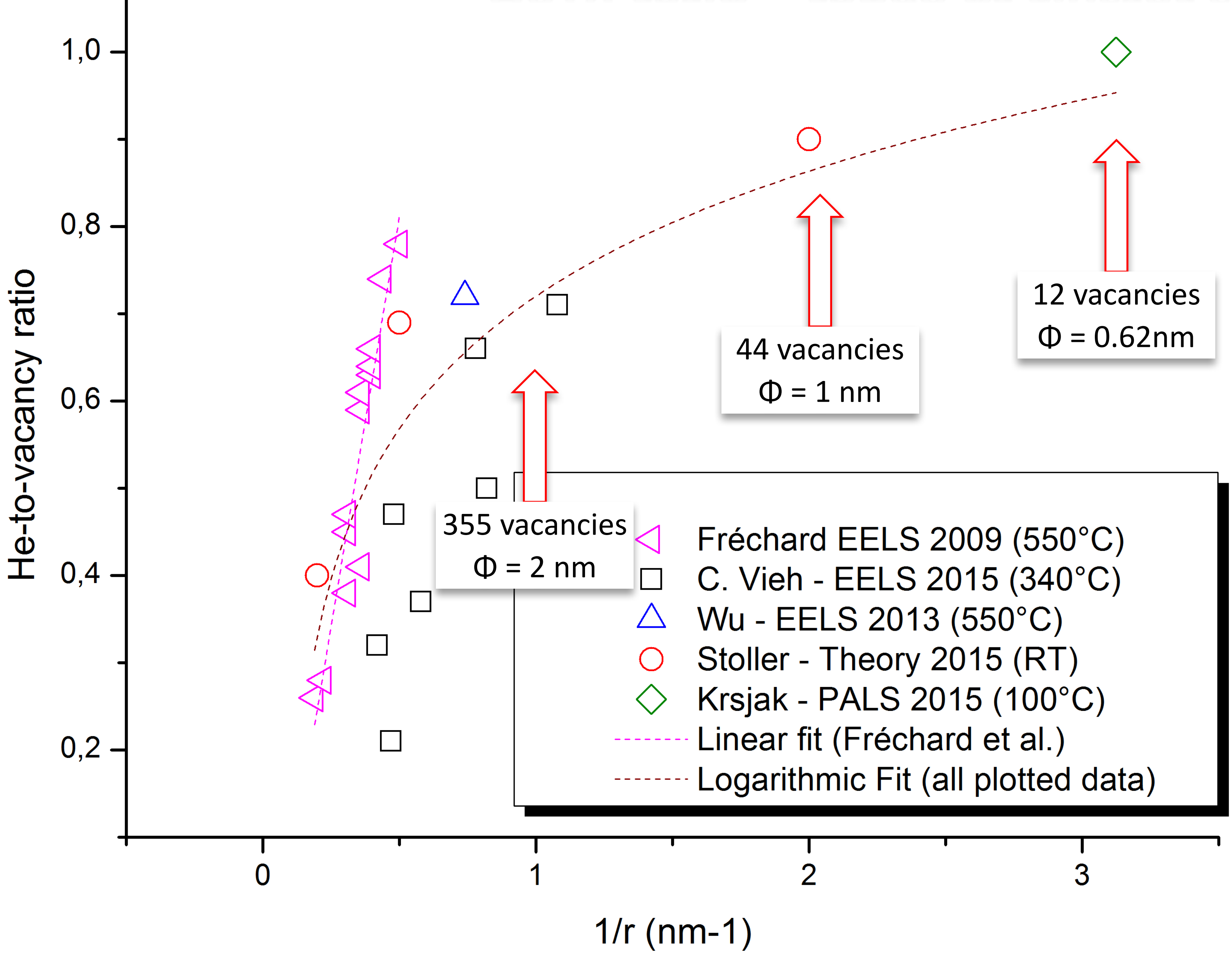
355 vacancies

PALS measurements show, that substantial amount of helium is trapped in **clusters** of mean size **12 vacancies**. Considering experimentally obtained number density, we can estimate **how much He can be captured by these clusters**.

He/V ratio – range of cluster sizes



He/V ratio – range of cluster sizes

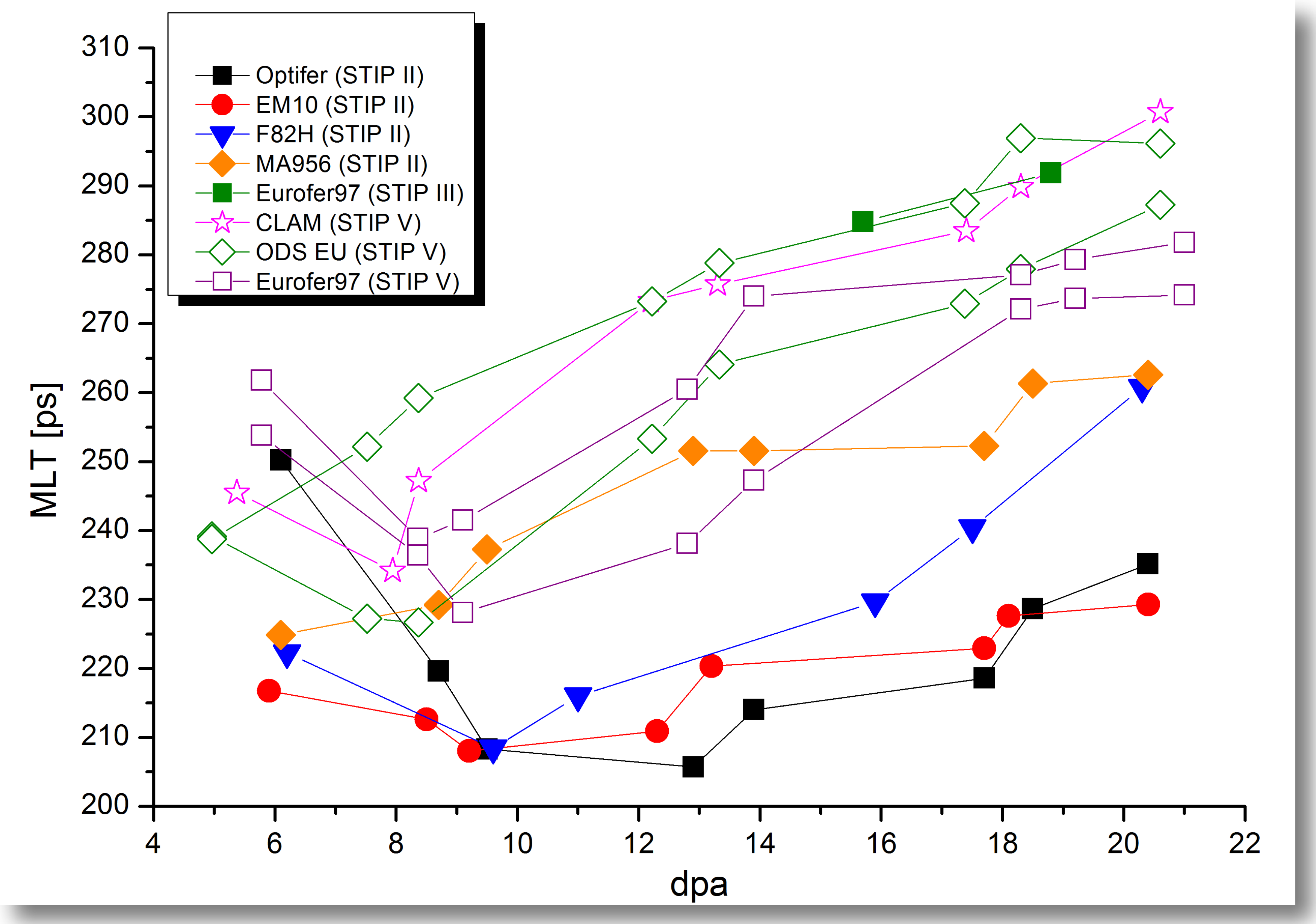


355 vacancies
Φ = 2 nm

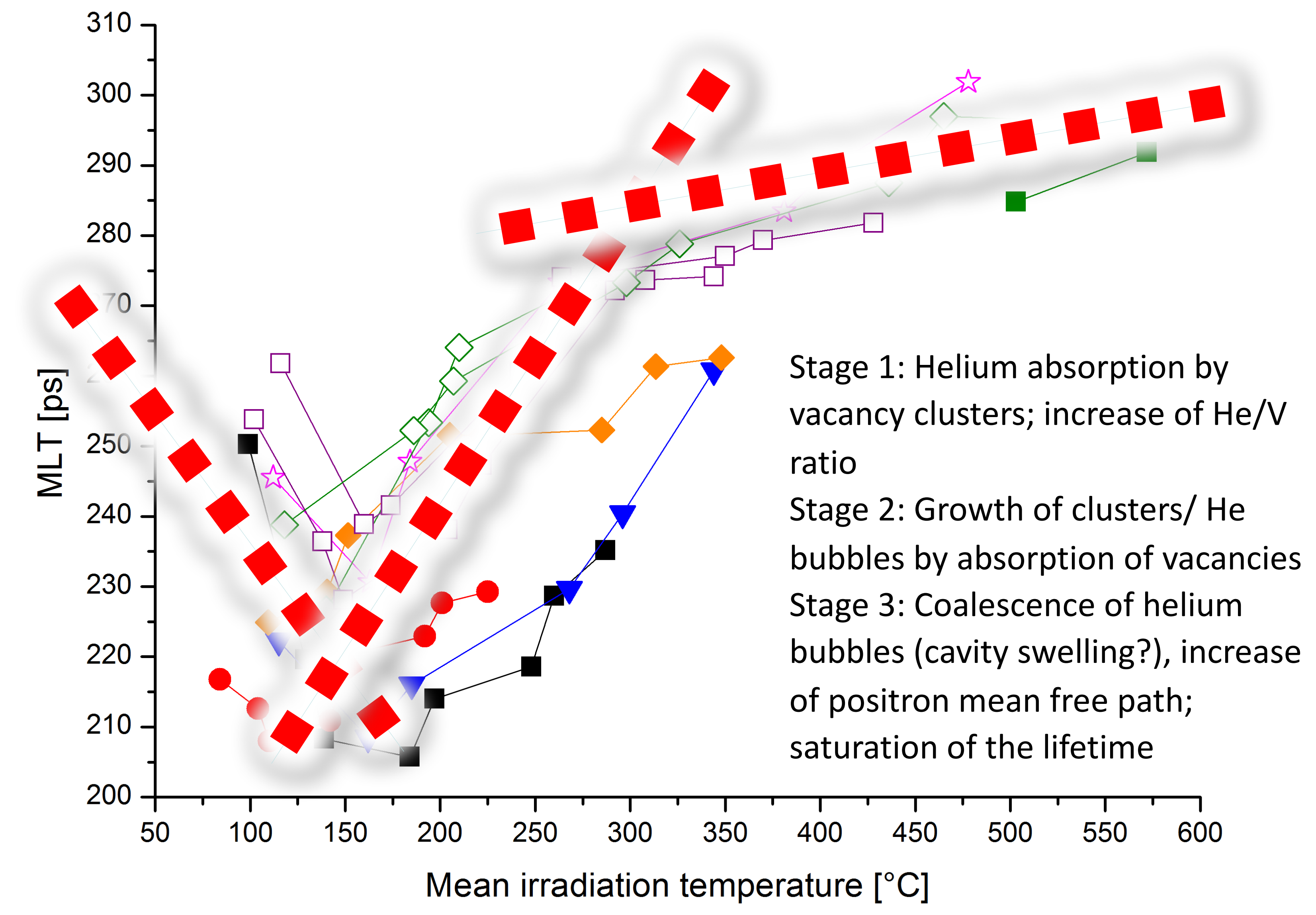
44 vacancies
Φ = 1 nm

12 vacancies
Φ = 0.62nm

Stages of microstructural evolution



Stages of microstructural evolution

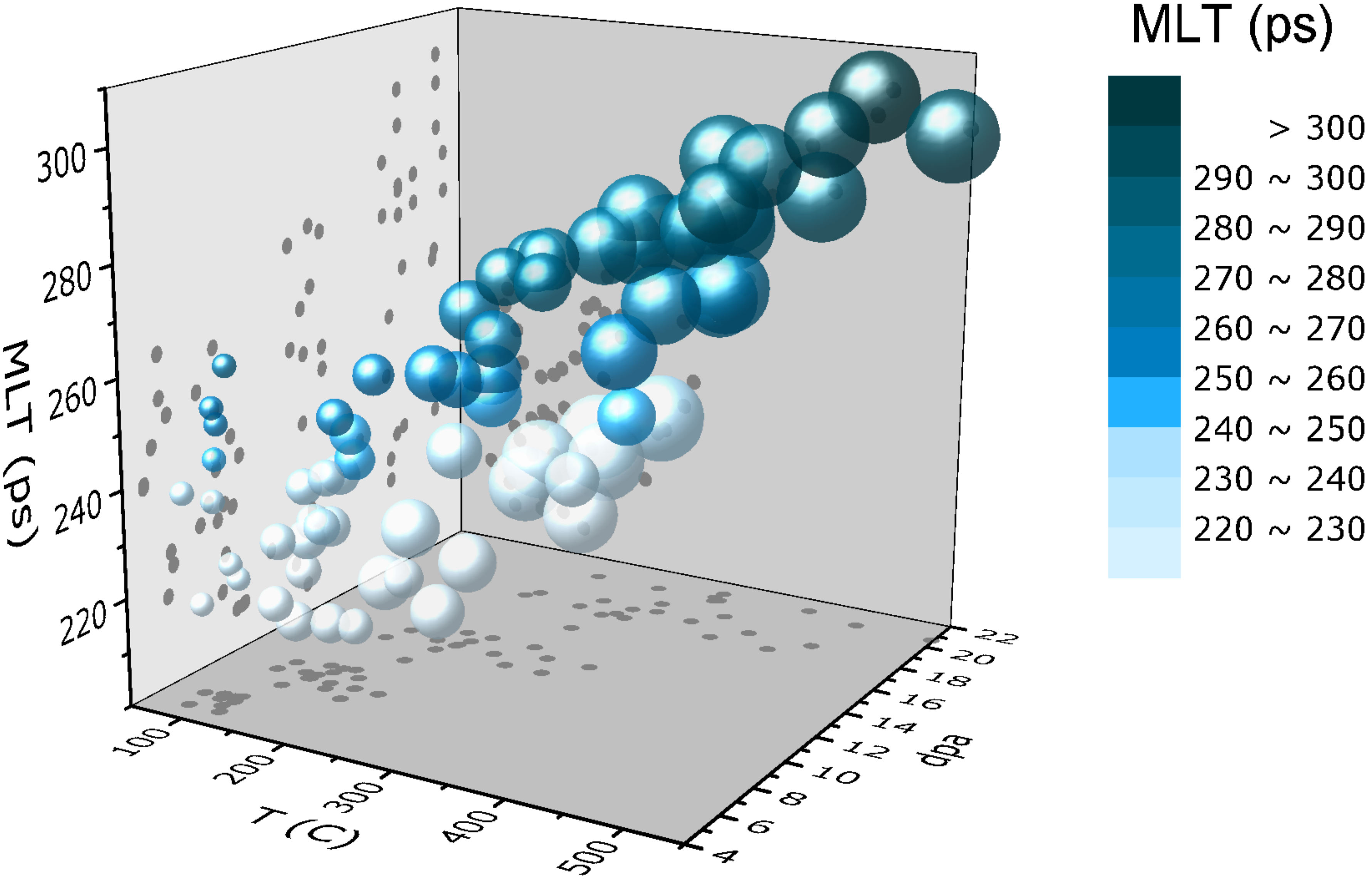


Stage 1: Helium absorption by vacancy clusters; increase of He/V ratio

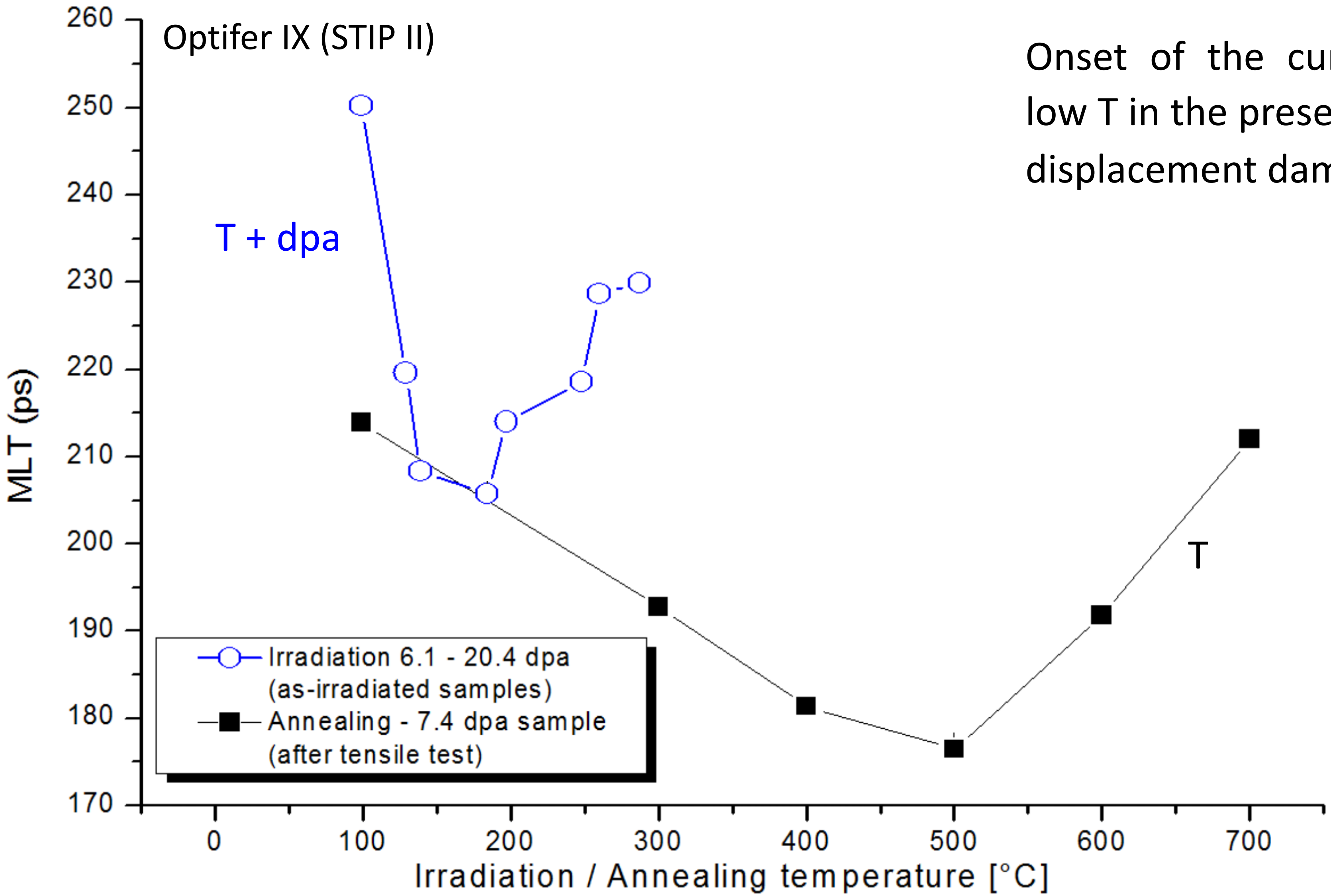
Stage 2: Growth of clusters/ He bubbles by absorption of vacancies

Stage 3: Coalescence of helium bubbles (cavity swelling?), increase of positron mean free path; saturation of the lifetime

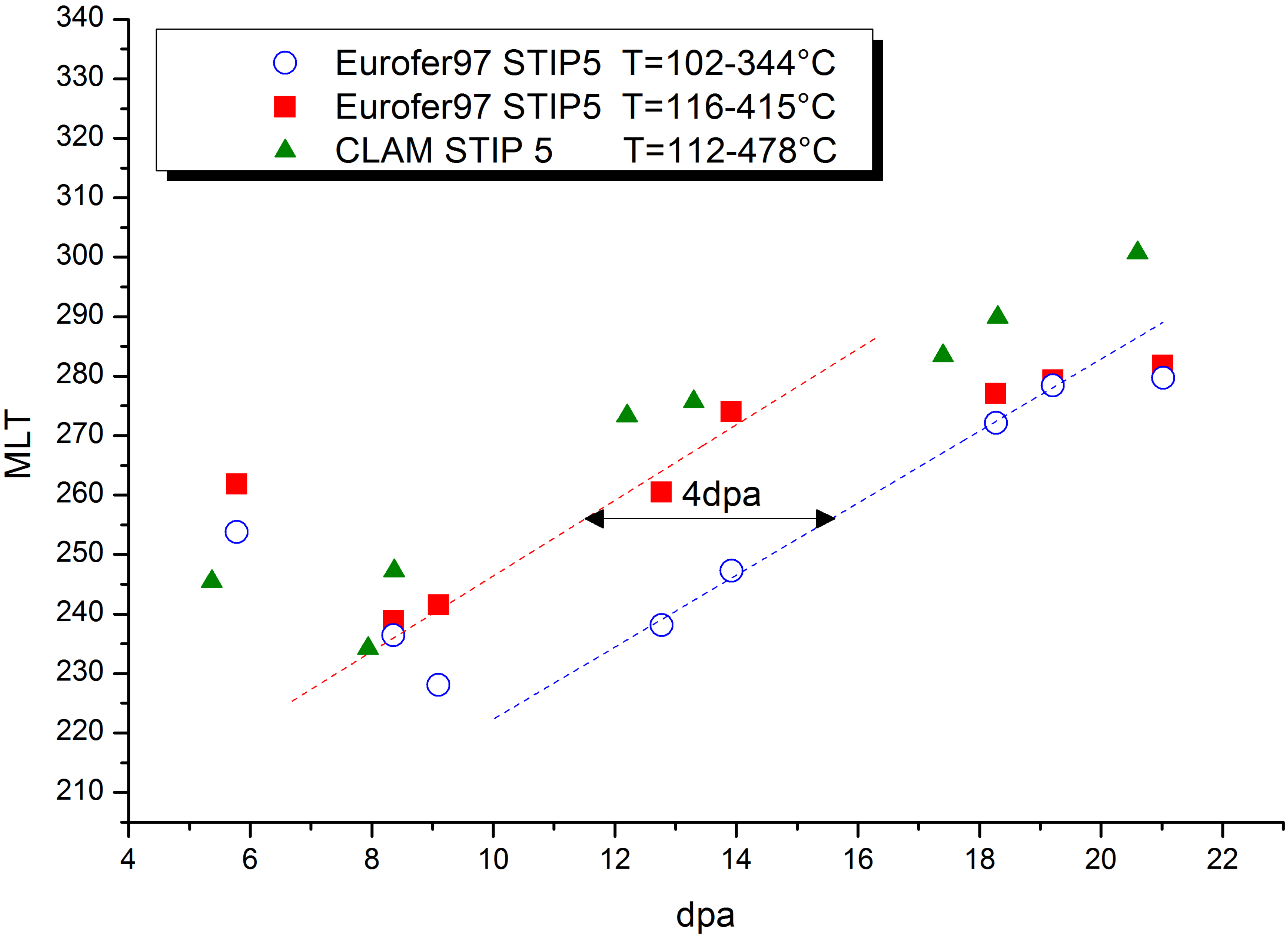
Positron mean lifetime in various STIP FM samples

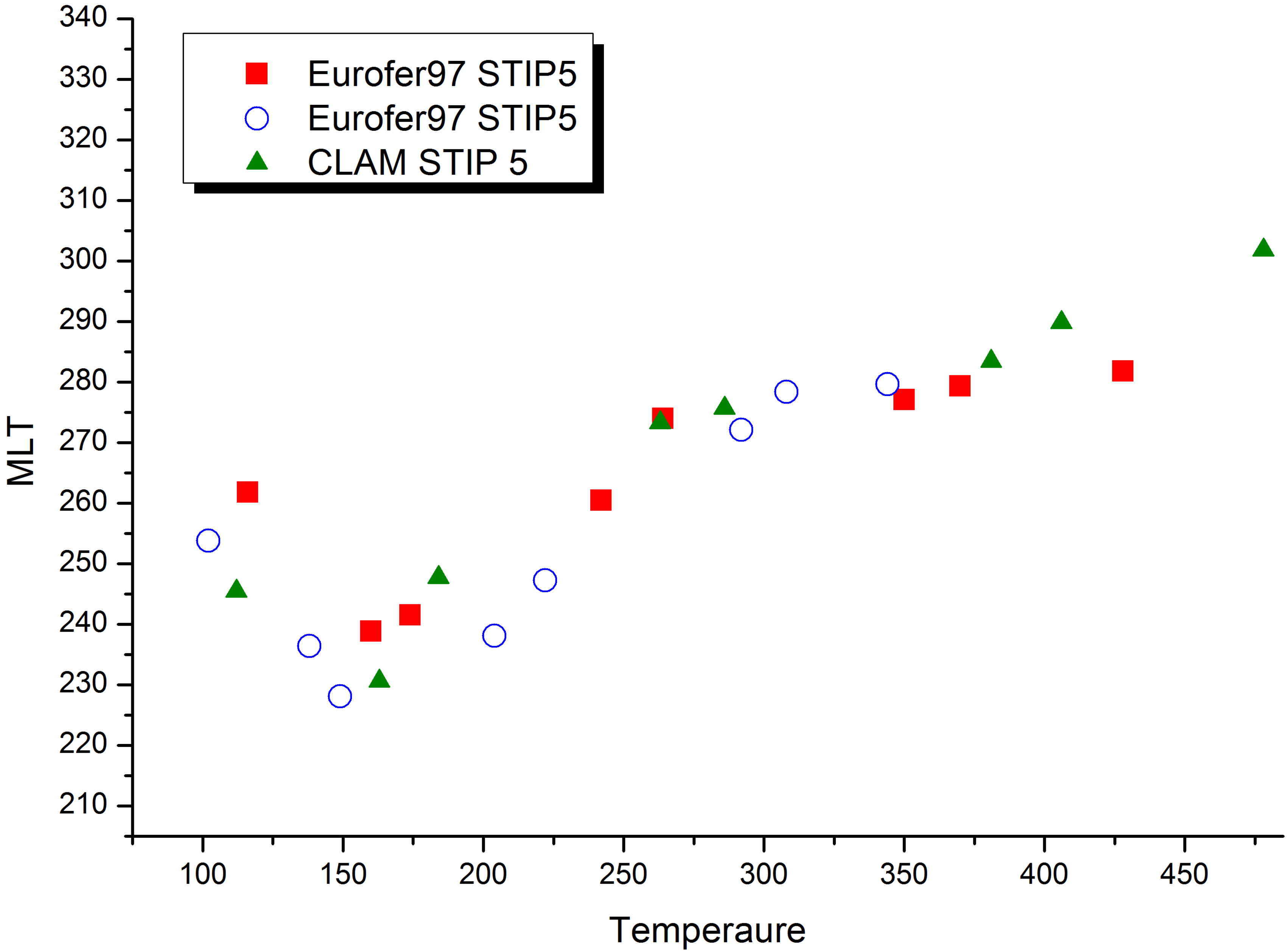


Temperature vs. dpa

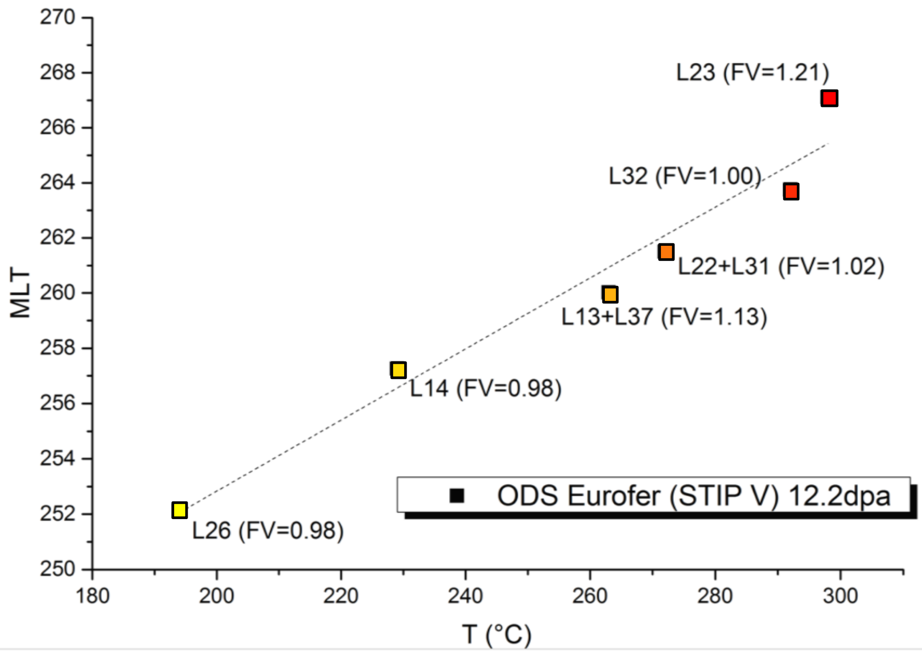


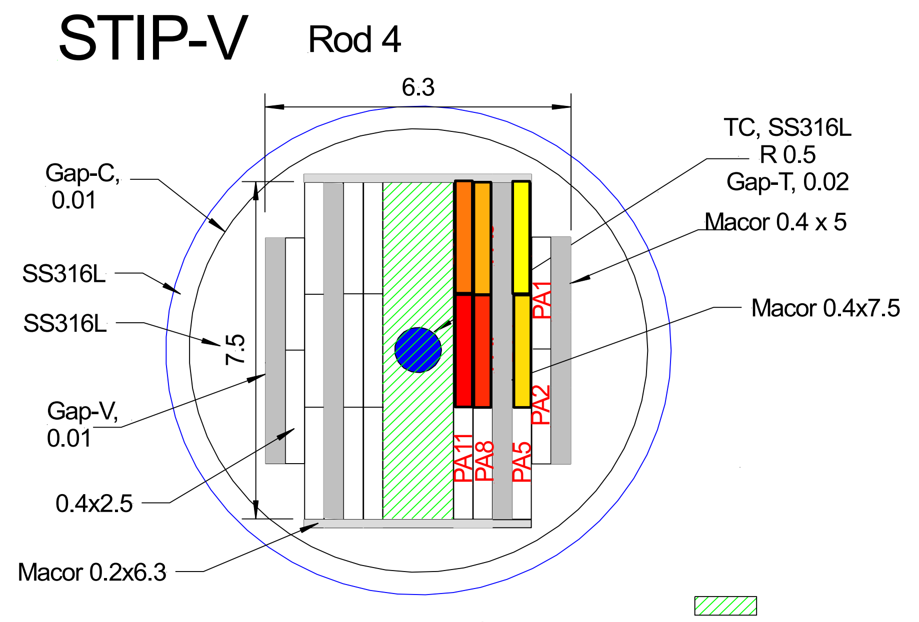
Onset of the curve to low T in the presence of displacement damage

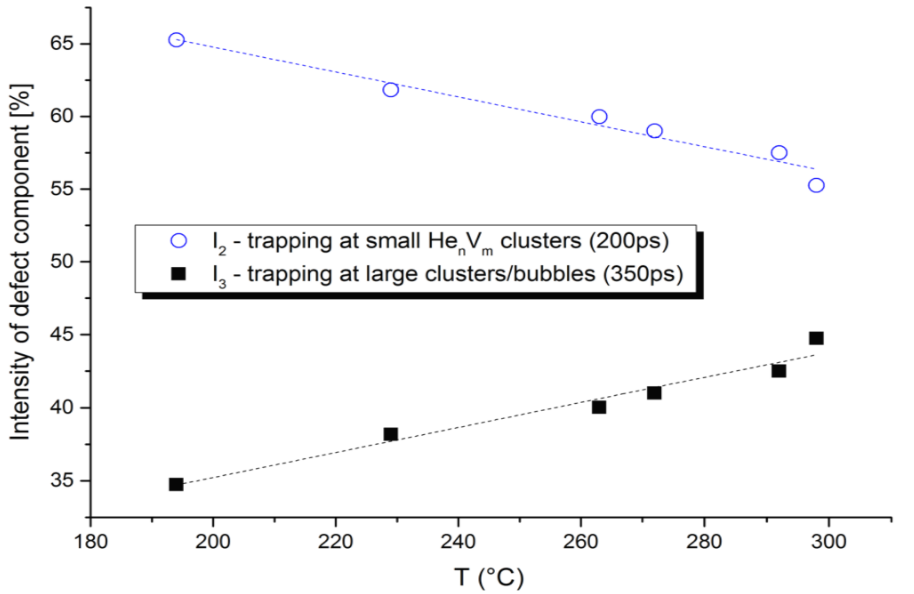

○ Eurofer97 STIP5 T=102-344°C
■ Eurofer97 STIP5 T=116-415°C
▲ CLAM STIP 5 T=112-478°C
4dpa
MLT
dpa
340
330
320
310
300
290
280
270
260
250
240
230
220
210
4
6
8
10
12
14
16
18
20
22

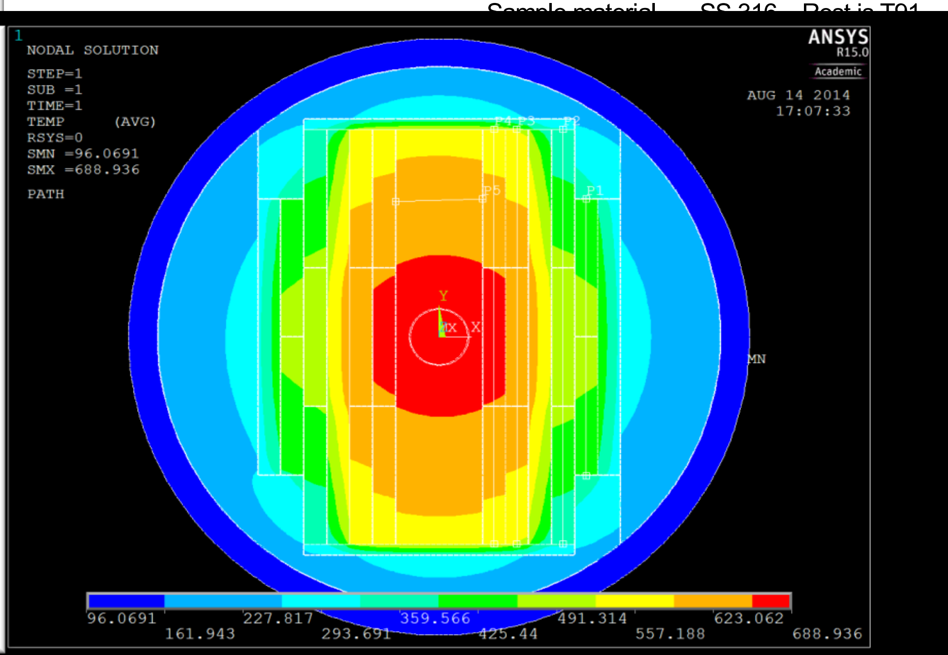

340
330
320
310
300
290
280
270
260
250
240
230
220
210
MLT
100
150
200
250
300
350
400
450
Temperaure
Eurofer97 STIP5
Eurofer97 STIP5
CLAM STIP 5

Temperature effect at 12dpa



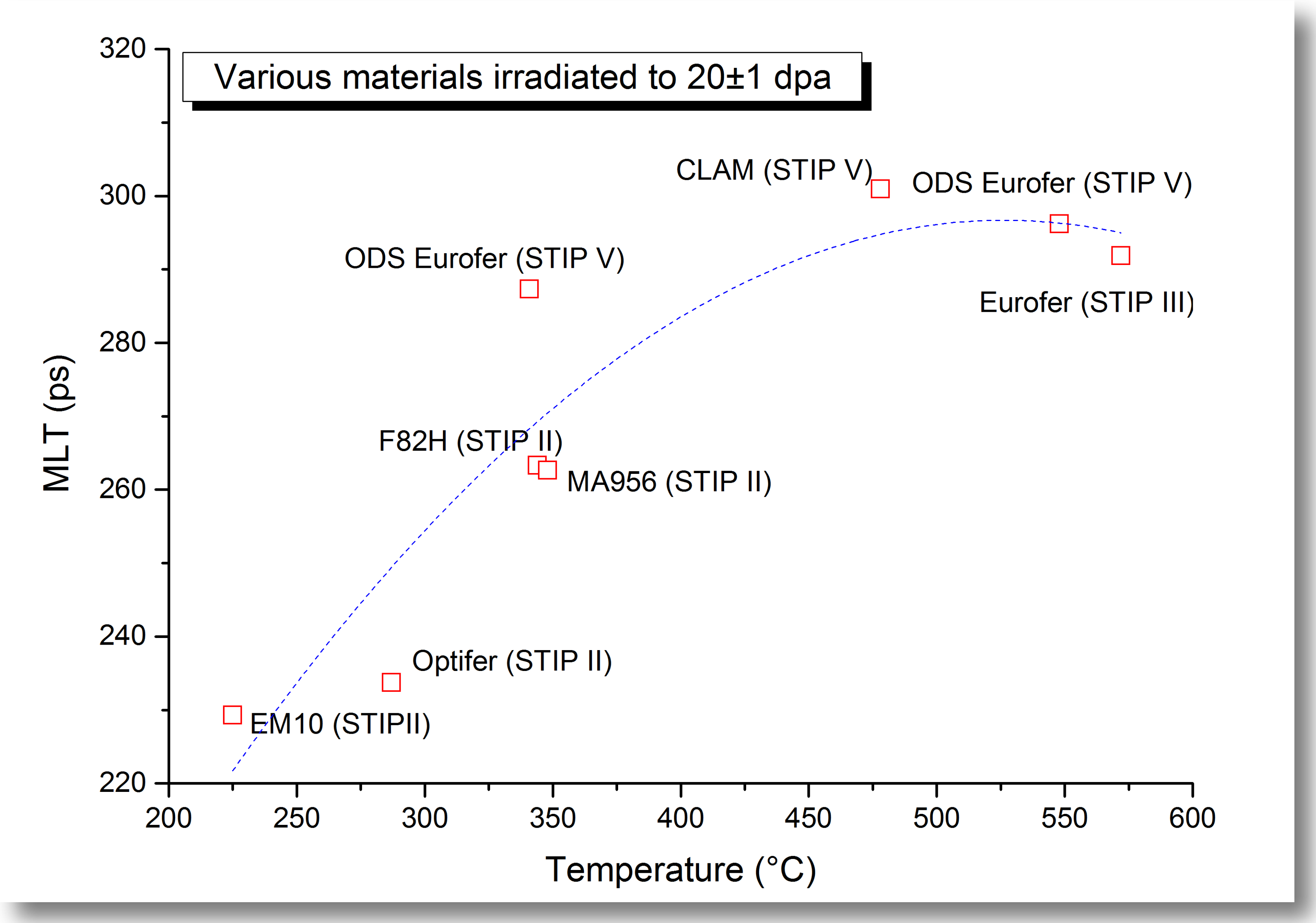




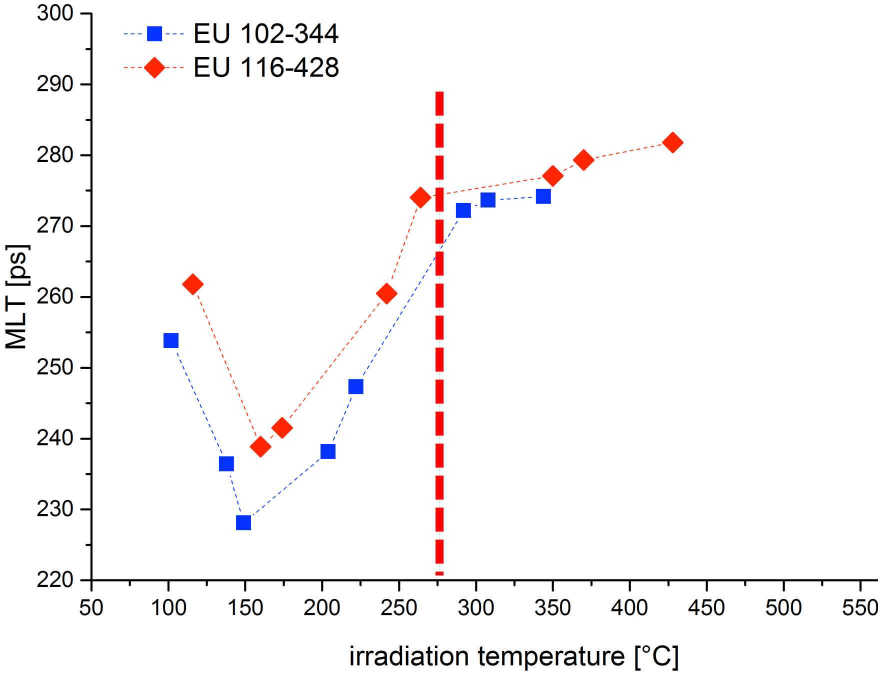


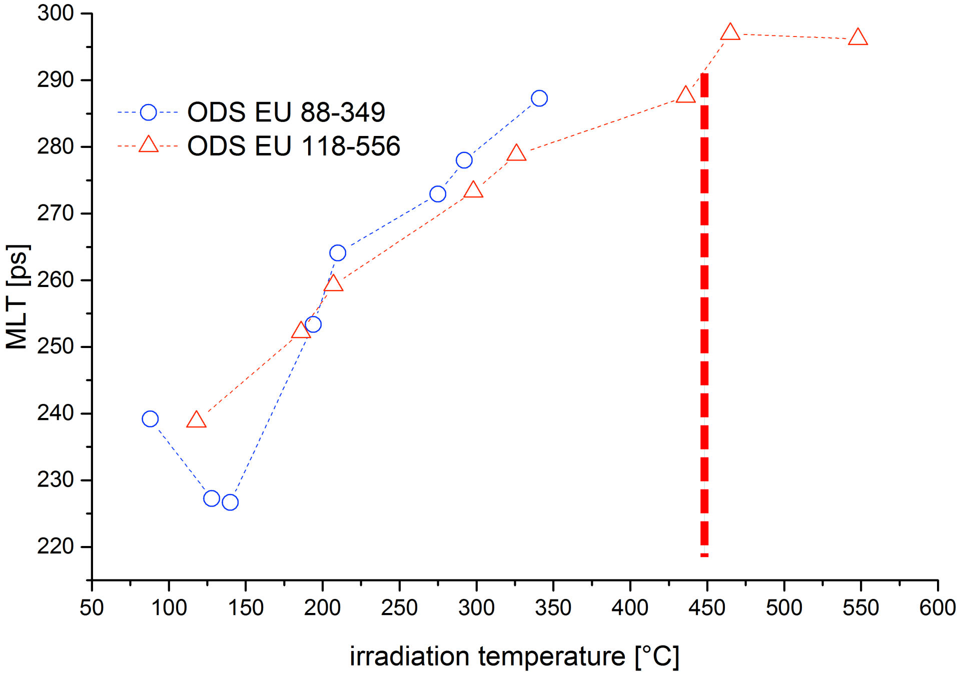
```
1
NODAL SOLUTION

STEP=1
SUB =1
TIME=1
TEMP        (AVG)
RSYS=0
SMN =96.0691
SMX =688.936

PATH
```

Temperature effect at 20dpa



Effect of oxide dispersoids





Saturation of MLT due to an enhanced coalescence of helium bubbles, decrease of their number density and the related increase of positron mean free path (i.e. decrease of positron trapping rate).

ODS Eurofer does not show the saturation od positron lifetime above 250°C. Results on ODS Eurofer incicate saturation above 450°C. This resistance against He bubbles coalescence may be due to presence of small oxide dispersoids, which act as sinks for radiation induced defects and/or helium.

SUMMARY AND CONCLUSIONS

SUMMARY

- **Specific trapping rate** of positron at nm helium bubbles (attractiveness to positron trapping) is **similar** to those of empty vacancy or **small clusters densely filled with helium**
- **Most of helium is trapped at small VC**, invisible to TEM
- Equilibrium He/V ratios at “low” irradiation temperatures obtained from PALS is in **good agreements with available MD results**
- **Three stages** of the evolution of He-V clusters can be distinguished in PALS data
- Both **temperature** and **dpa** can **accelerate growth of He bubbles**
- Presence of **oxide dispersoids** in ODS steels **postpone the formation of helium bubbles**

THANK YOU FOR YOUR ATTENTION

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