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Investigation of SINQ-irradiated samples by single detector Doppler-broadening spectroscopy

13th International Workshop on Spallation Materials Technology

30.10. – 4.11.2016

Outline

- Positron annihilation spectroscopy in material science
- Positron source
- Nano-world problems (Helium)
- Experimental part and results
- Summary and conclusions

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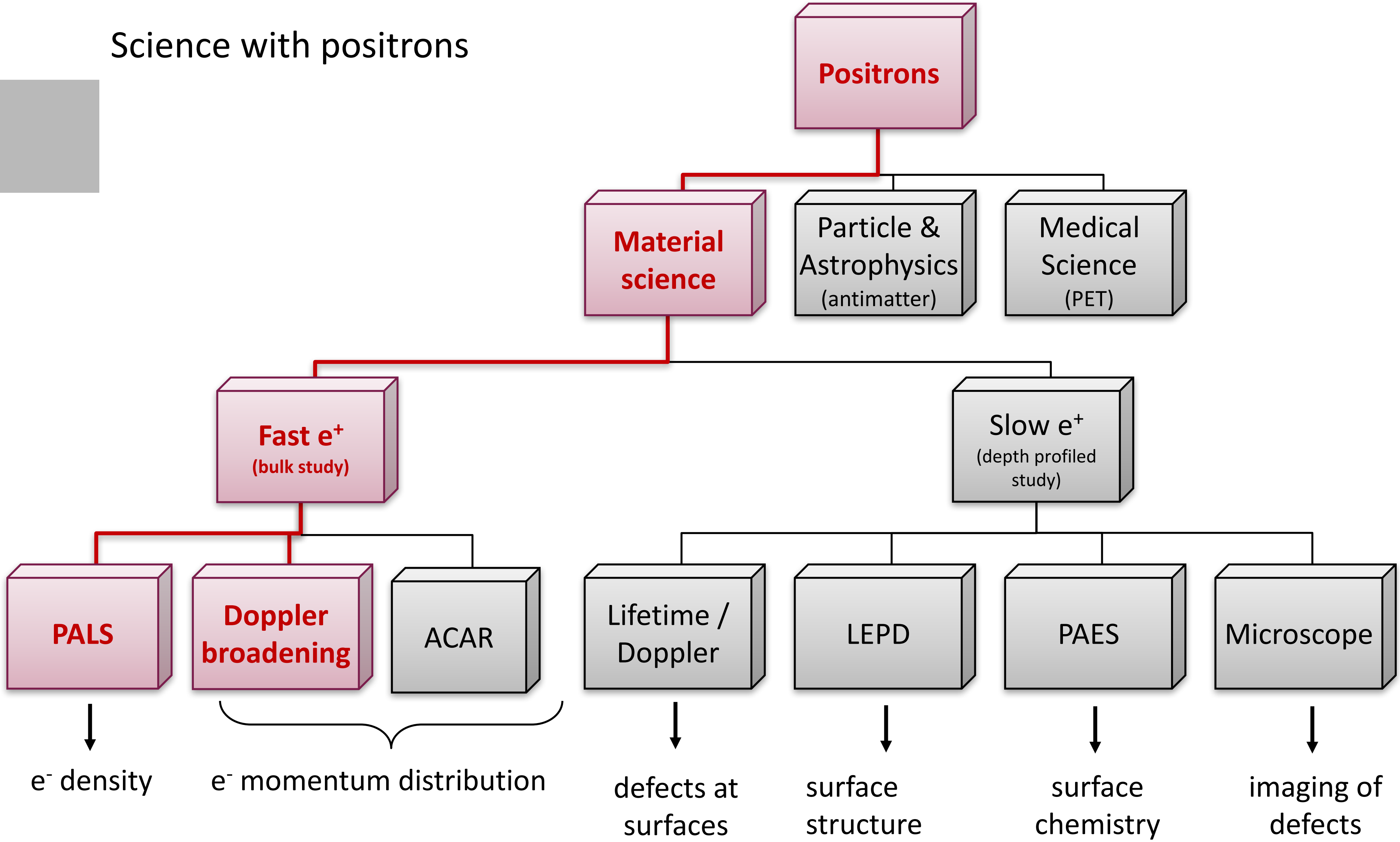
PAS and its place in material science

Science with positrons

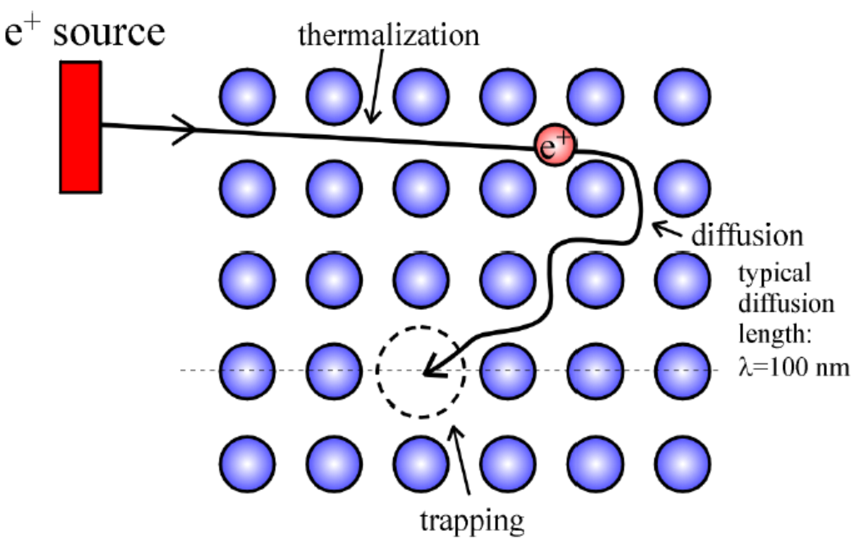


How does it work?

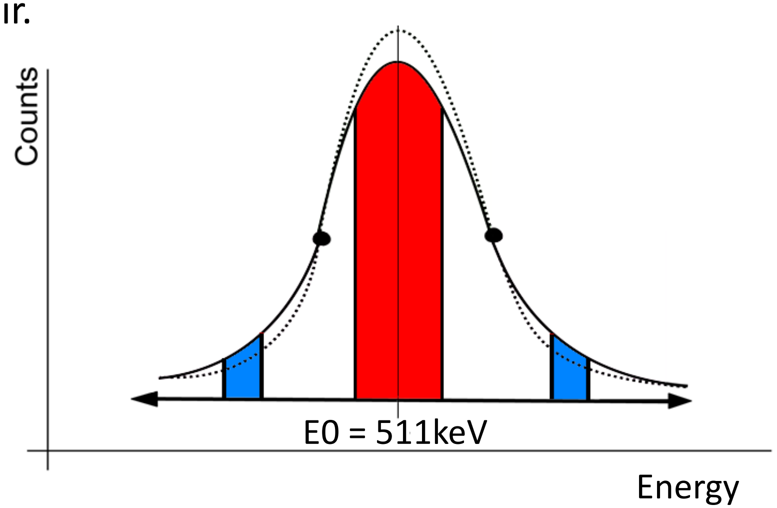
Positron
Annihilation
Lifetime
Spectroscopy

Two techniques of positron annihilation spectroscopy, based on different physical principles, have been widely established in the material research.

Coincidence
Doppler
Broadening
Spectroscopy

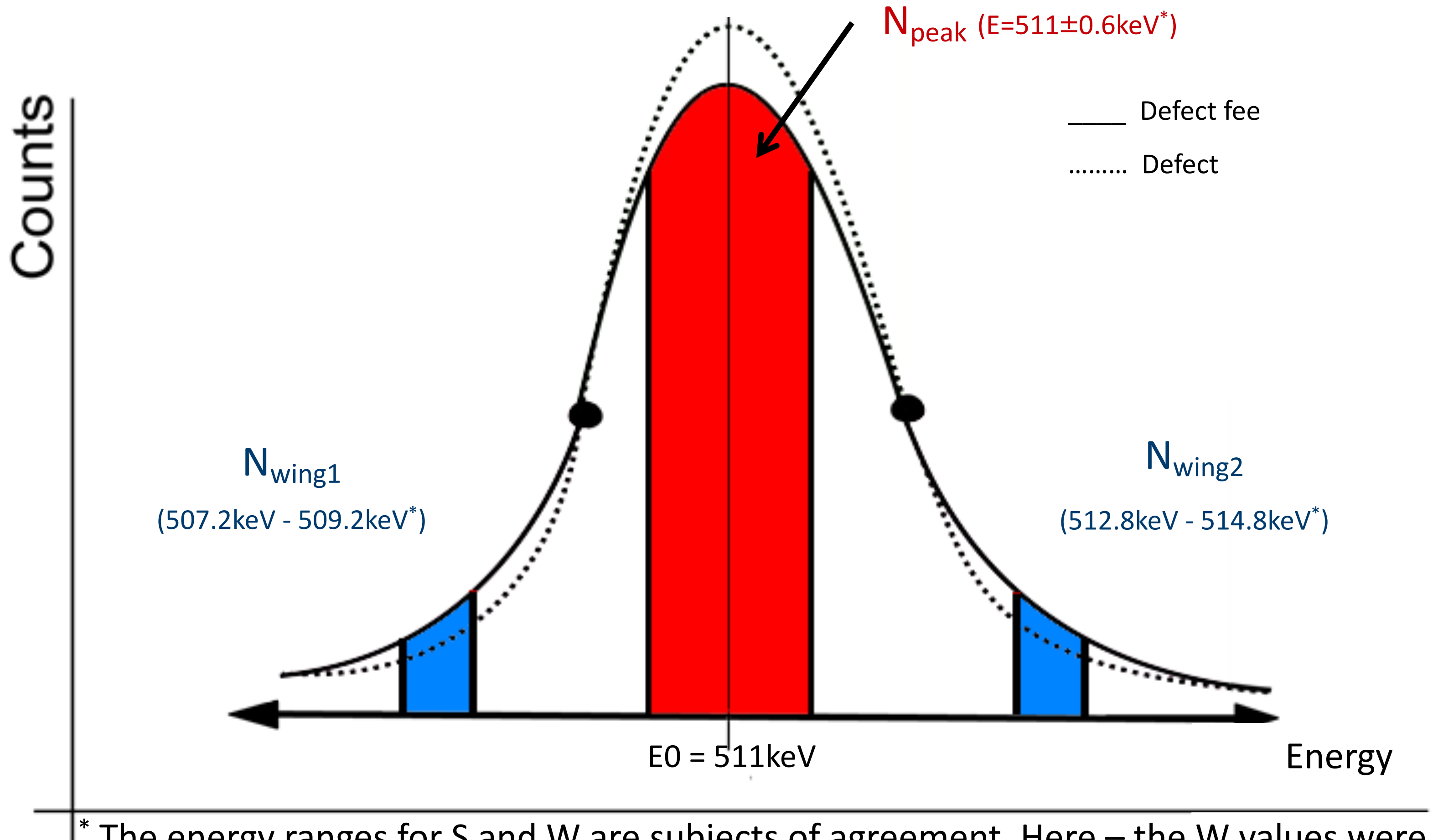


Positrons annihilate mainly with the electrons of the outermost shell due to the repulsion of the nucleus. Such annihilation results in Eg ≅ 511keV. But the annihilation occurs also with core electrons (electrons with higher momentum). Such annihilation leads to **deviation in the energy** of annihilation gamma, which is **proportional to the momentum** of the electron-positron pair.

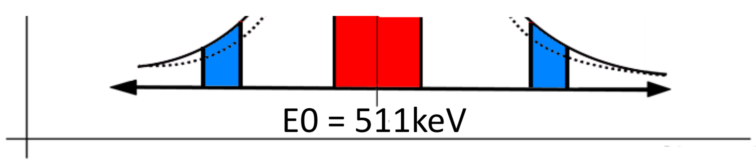


After thermalization (~ 3ps), positron diffuse through the lattice until trapping / annihilation. **Diffusion time and trapping rate are a function of the microstructure** and they can be measured.

Doppler Broadening Spectroscopy



* The energy ranges for S and W are subjects of agreement. Here – the W values were chosen to cover the characteristic He peak



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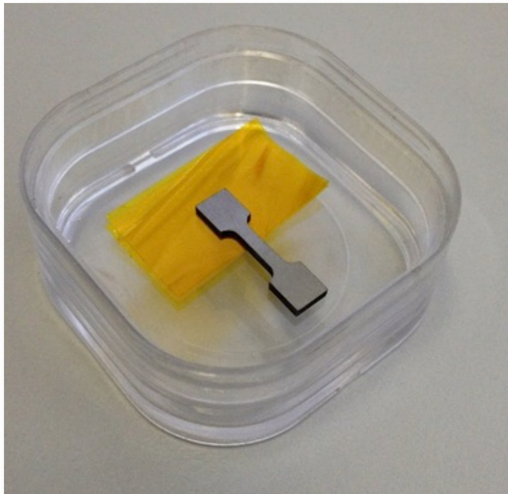
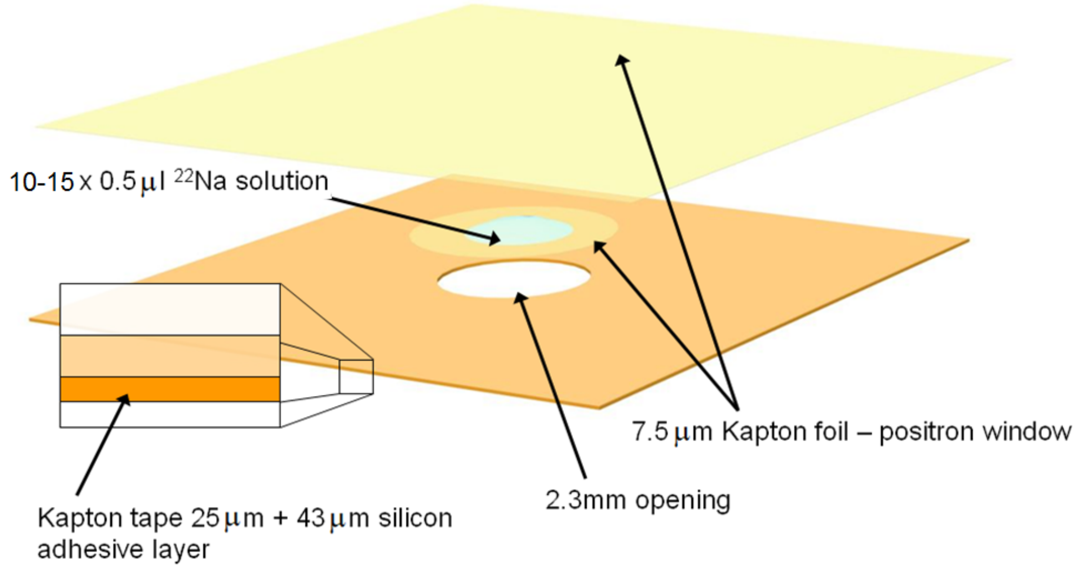
Conventional positron source





Source - emission
(α, β, γ, p, n, μ^- ...)
- Radioisotope sources
- Particle accelerators
- Cathode ray tubes etc.

Sample – interaction
- Nuclear reaction
- Scattering / Diffraction
- Photoelectric effect
- e^- e^+ annihilation etc.

Detector - detection
(β', γ', n', $\mu^{-\prime}$ ...)
- Energy
- Momentum
- Charge
- Time etc.

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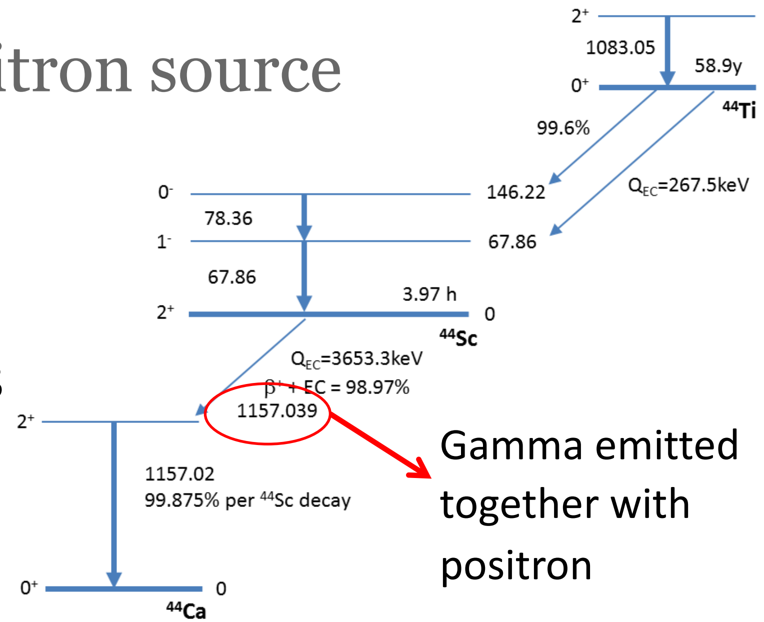
^{44}Ti as an internal positron source



Gamma emitted together with positron

In spallation irradiations, ^{44}Ti is a product of ^{56}Fe(p, x) reaction. It is present in all STIP samples of steels

Irradiated sample containing ^{44}Ti

Scintillation detector

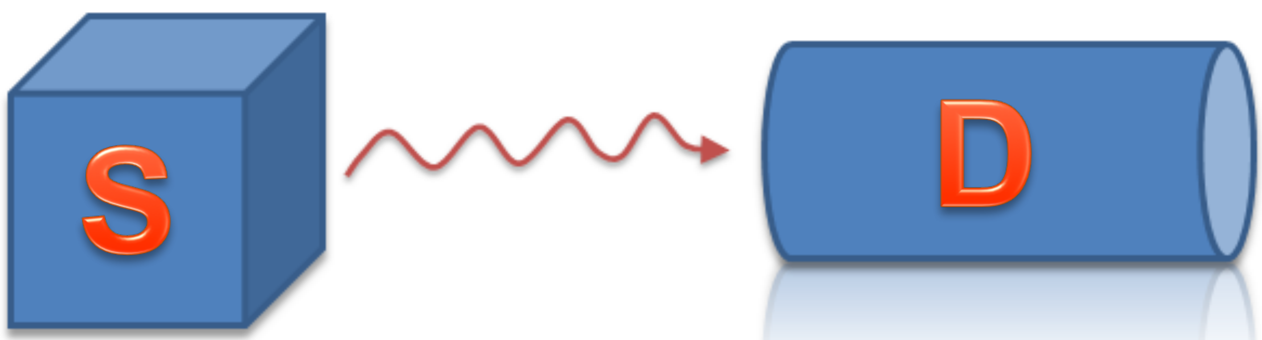
- $^{44}Ti \rightarrow {}^{44}Sc \rightarrow {}^{44}Ca + e^{+} + \nu_e + \gamma$ → 1154 keV

  e⁺ diffusion = f (material)

  e⁺ trapping = f (defects)

  Time [ps] = F (material, defects)

- $e^{-} + e^{+}$ annihilation → 511 keV ±ΔE   ΔE = F (material, defects)



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External vs. Internal positron source

Benefits of using internal $^{44}Ti/^{44}Sc$ source:

- The $^{44}Ti/^{44}Sc$ source has a much longer half-life (59.6 y) which ensures a long-term stable production of positrons.
- Homogeneous probing of the whole bulk
- No surface treatment required
- There are practically no limitations as regards the sample size and shape.
- Methodology is suitable for very active samples (^{60}Co activity is usually equal or lower than ^{44}Ti activity).

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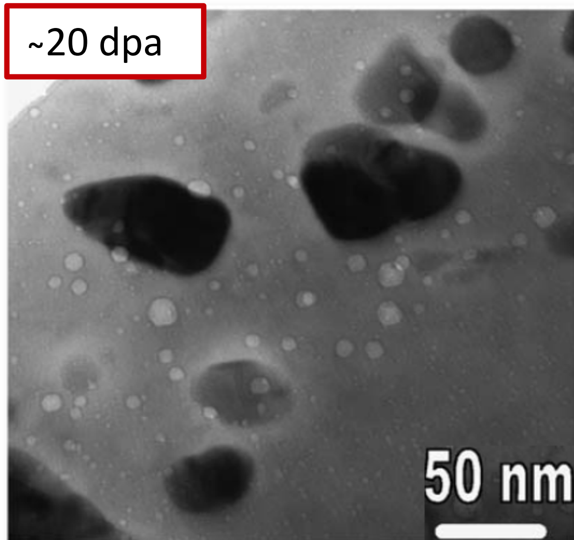
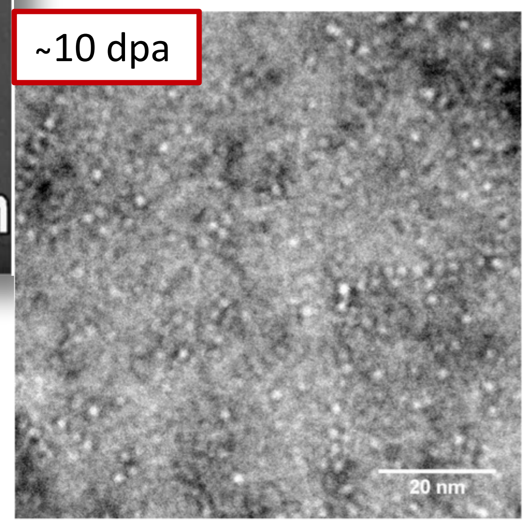
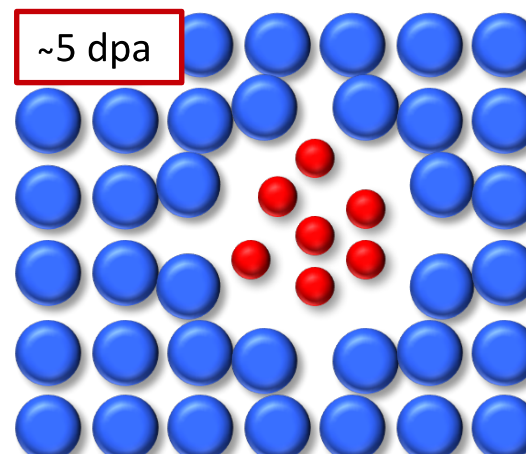
Imaging techniques limitations



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

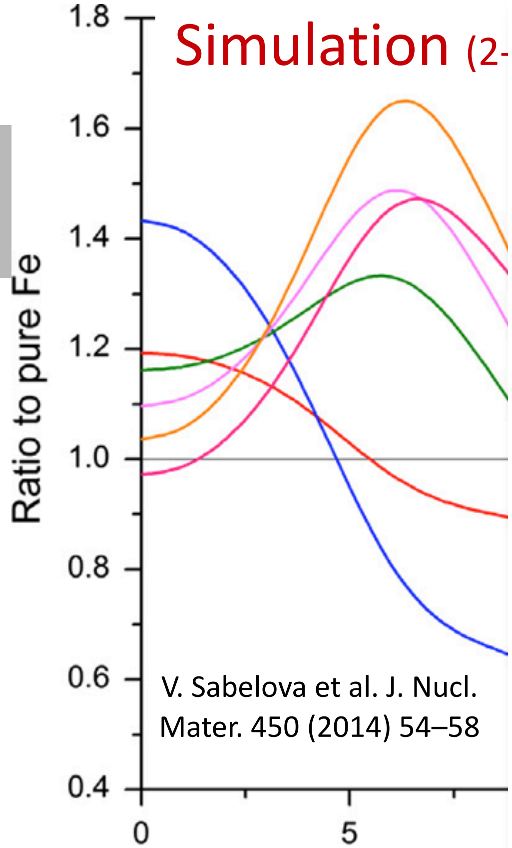


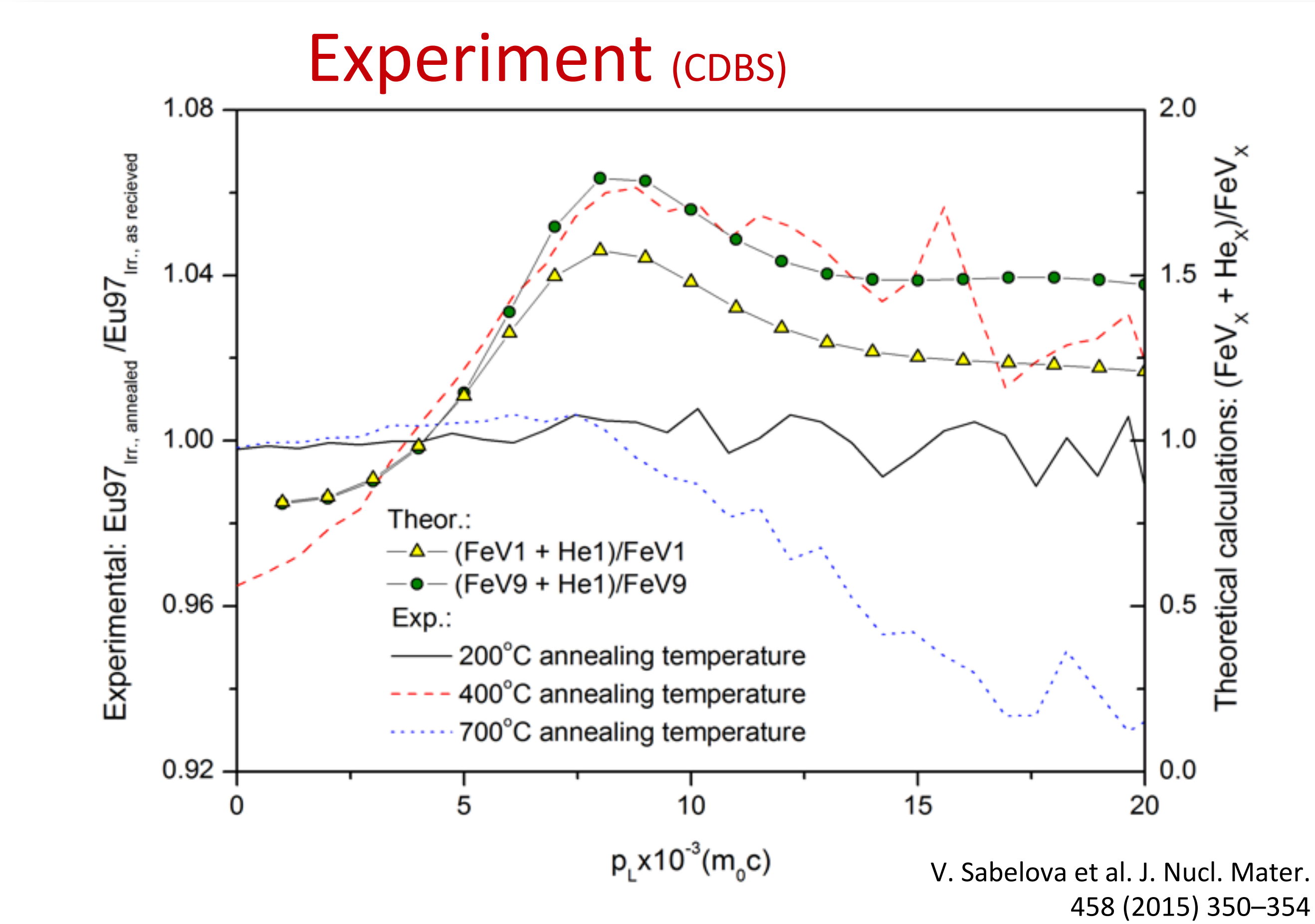
Early stage displacement damage, He-V clusters; almost no experimental data from electron microscopy techniques



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

Do we actually see Helium? CDBS Results





The distinct peak appears in the experimental spectra at the stage when we expect a maximum concentration of He in vacancy clusters (He/V = max.)

Helium introduces a distinct peak in the momentum spectra around 7×10^{-3} m_0c. In the conventional approach with the W parameter set to 15-25 m_0c, He effect is practically invisible.

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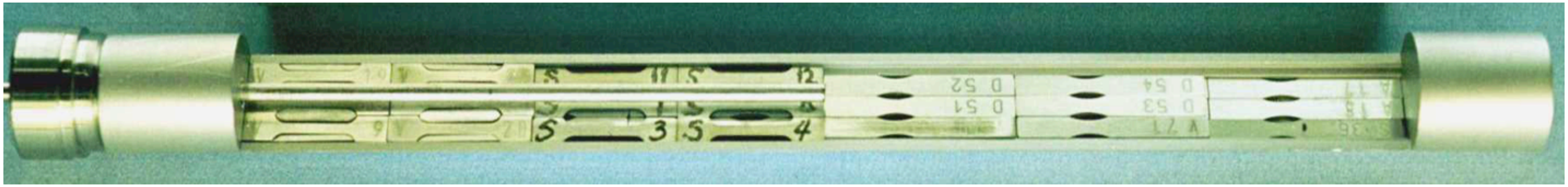
Experiment on the STIP sample

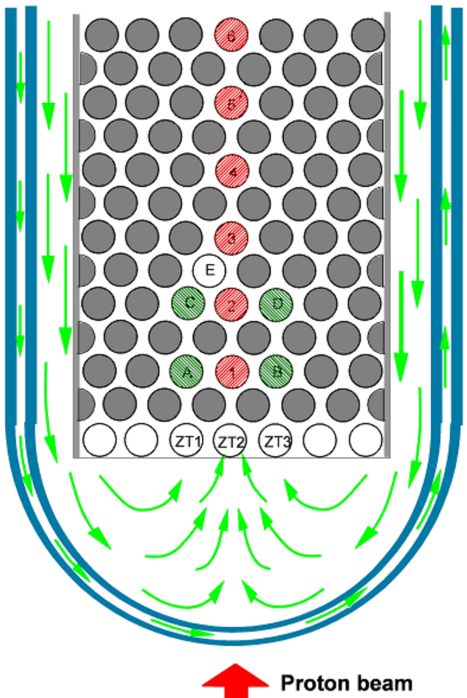
Material: EM10 (9Cr-1Mo) - Cr – 8.97%, Mo – 1.06%, Mn – 0.49%, Si – 0.46, C – 0.099%



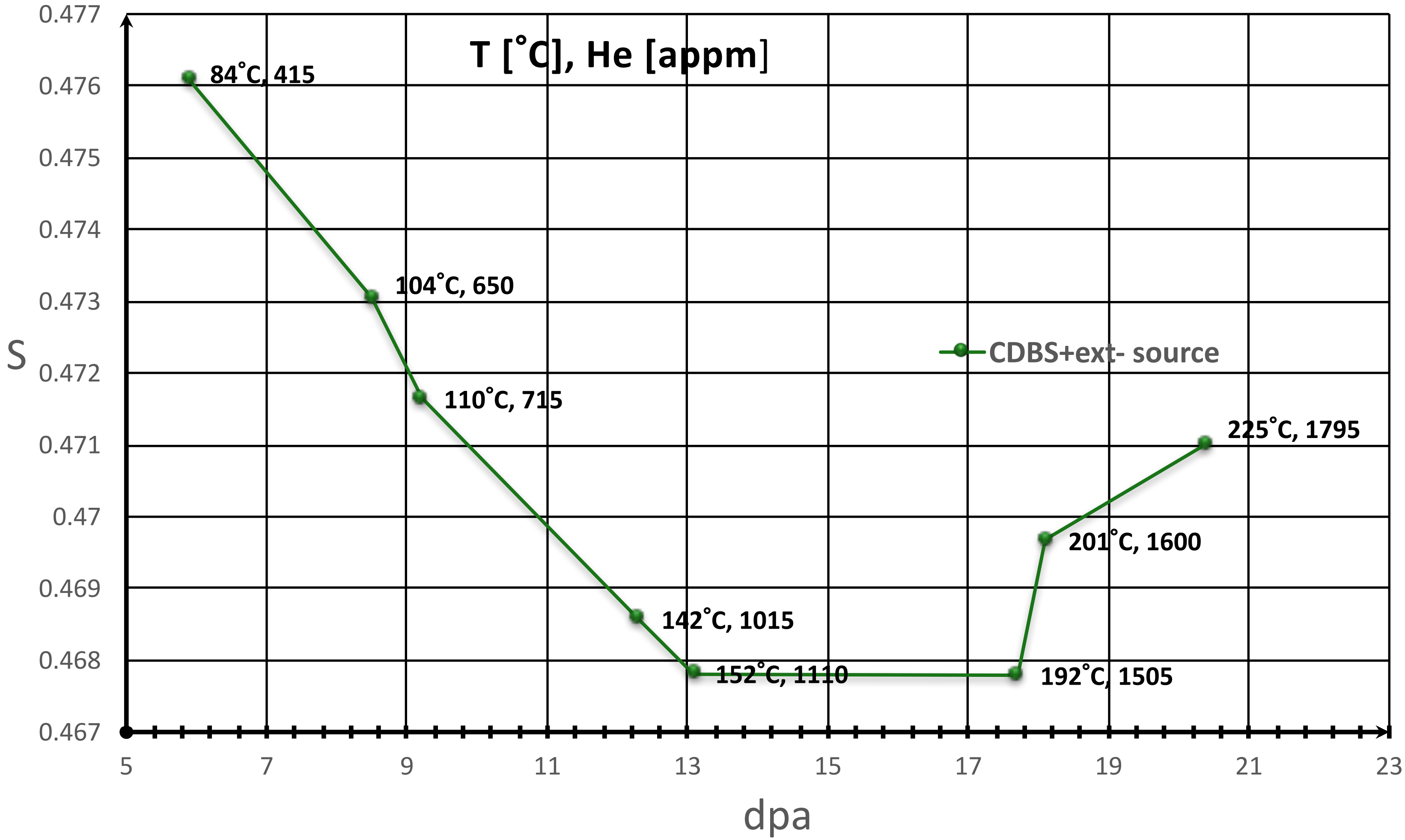
Irradiation doses: (5.9 – 20.4 dpa)

Irradiation temperatures: (84 – 225˚C)

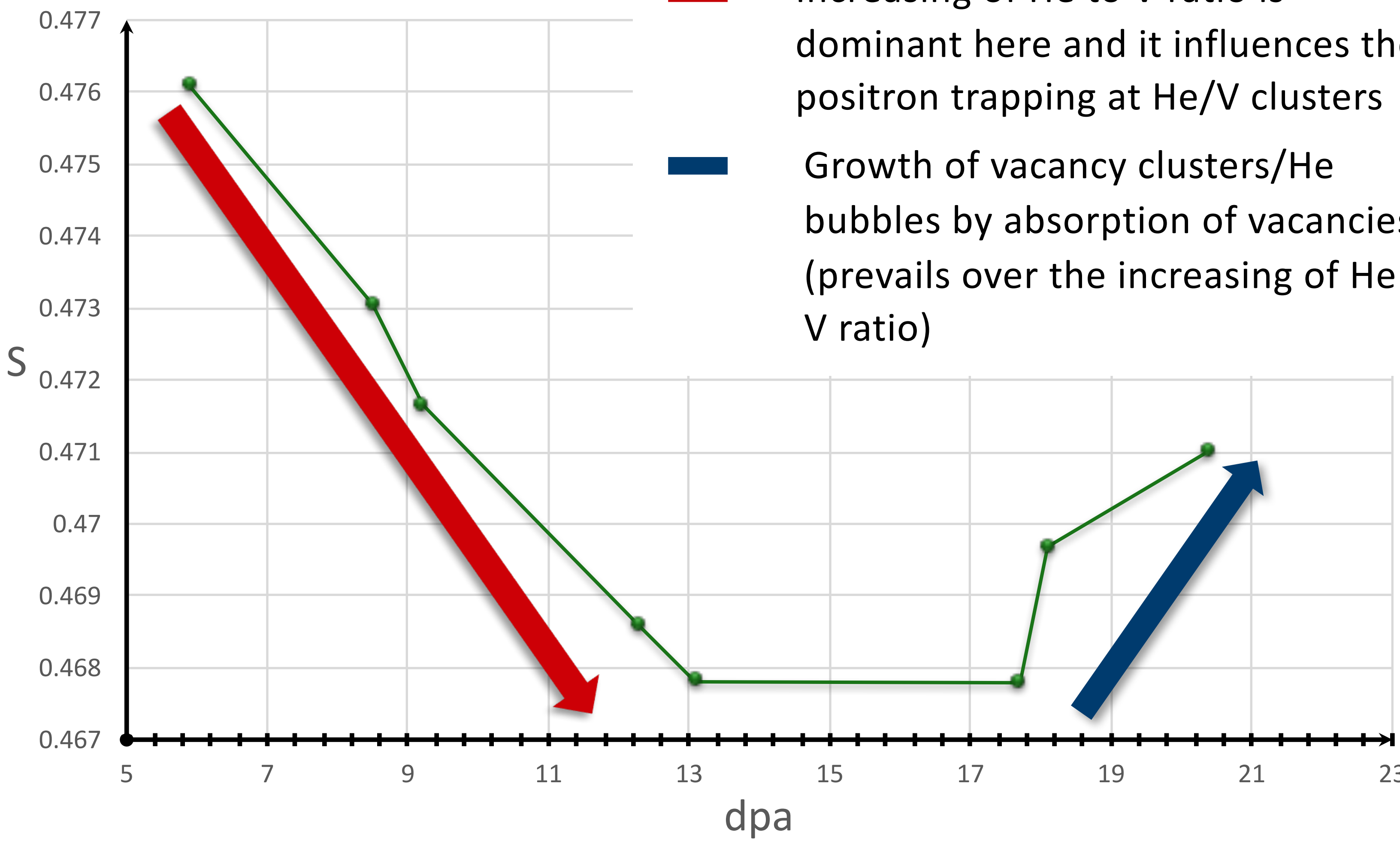
Helium density: (415 – 1795 appm)



EM10 samples measured by CDBS with external positron source – S parameter



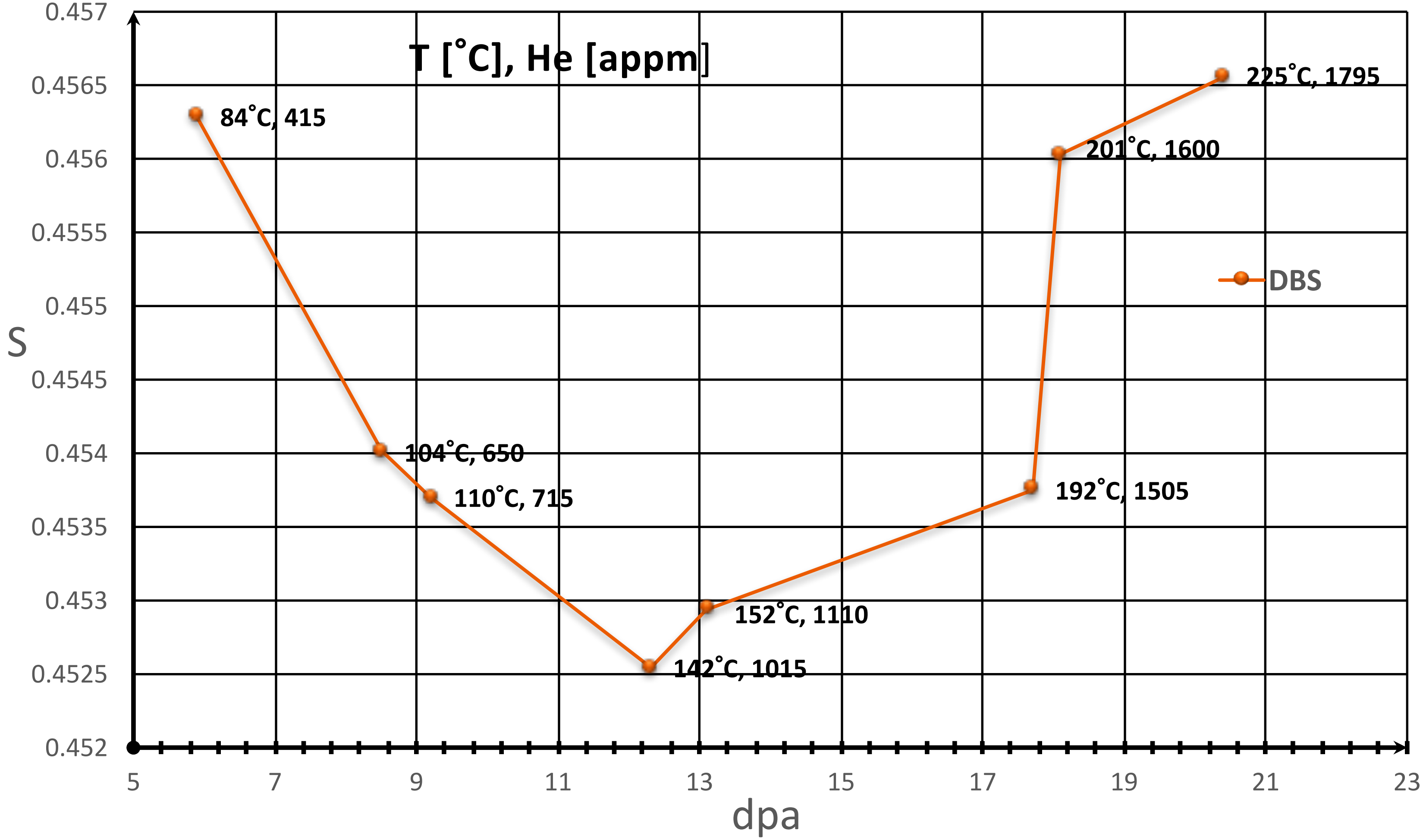
EM10 samples measured by CDBS with external positron source – S parameter



Increasing of He to V ratio is dominant here and it influences the positron trapping at He/V clusters

Growth of vacancy clusters/He bubbles by absorption of vacancies (prevails over the increasing of He to V ratio)

EM10 samples measured by DBS with only internal positron source – S parameter



EM10 samples measured by DBS with only internal positron source – S parameter

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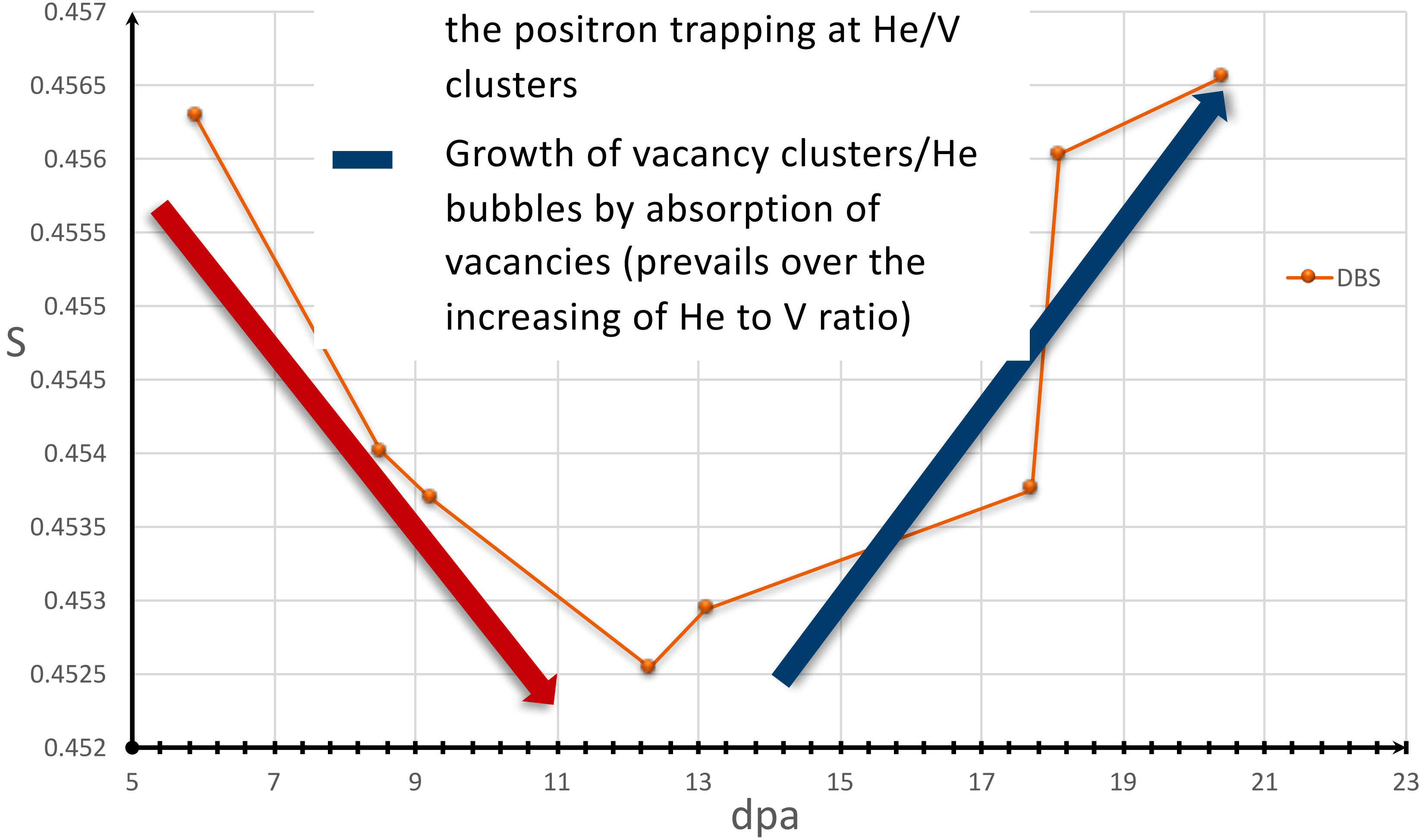
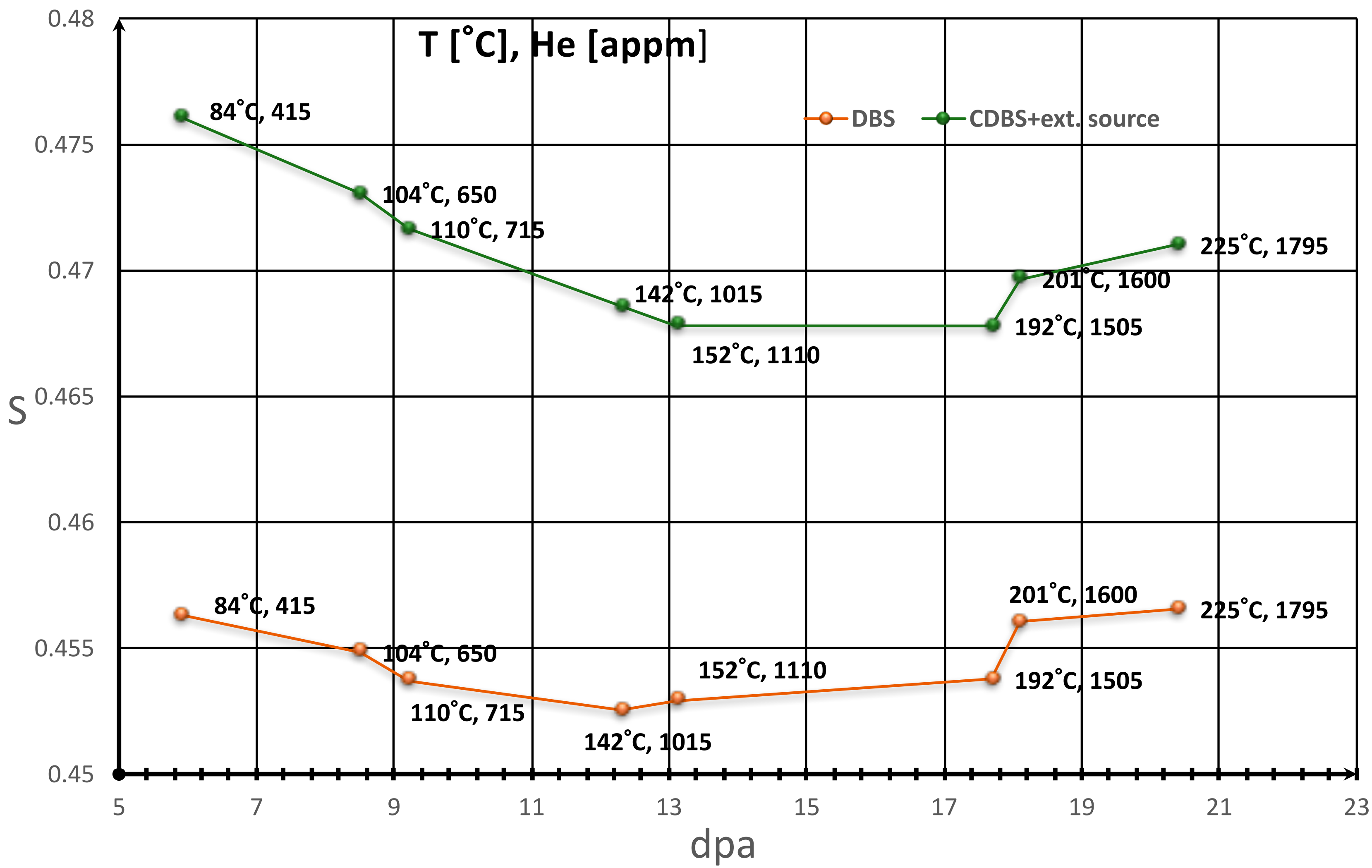
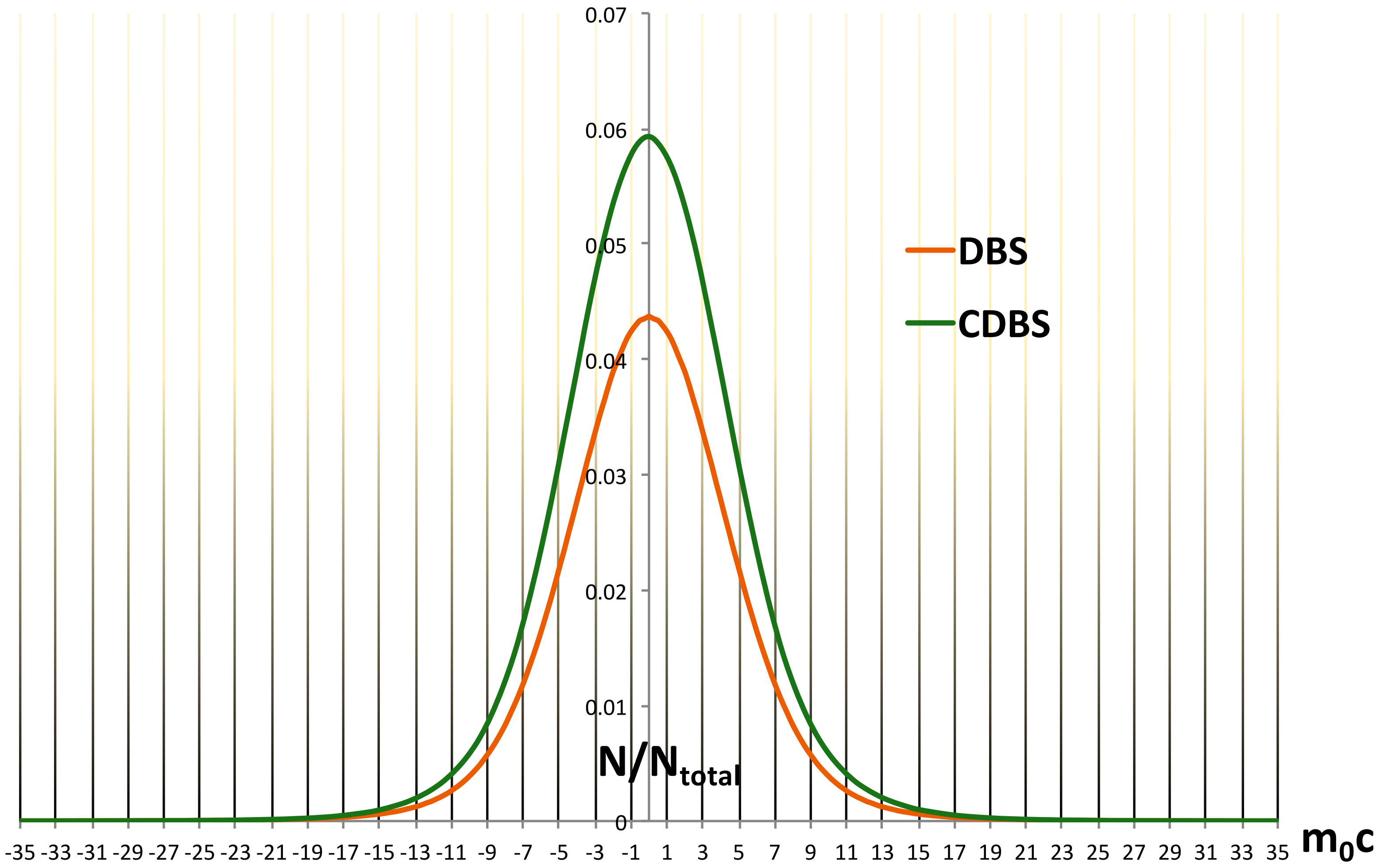


Comparison of DBS and CDBS S curves of EM10 in absolute values



Graph of DBS and CDBS – N/N_{total} (20.4 dpa)



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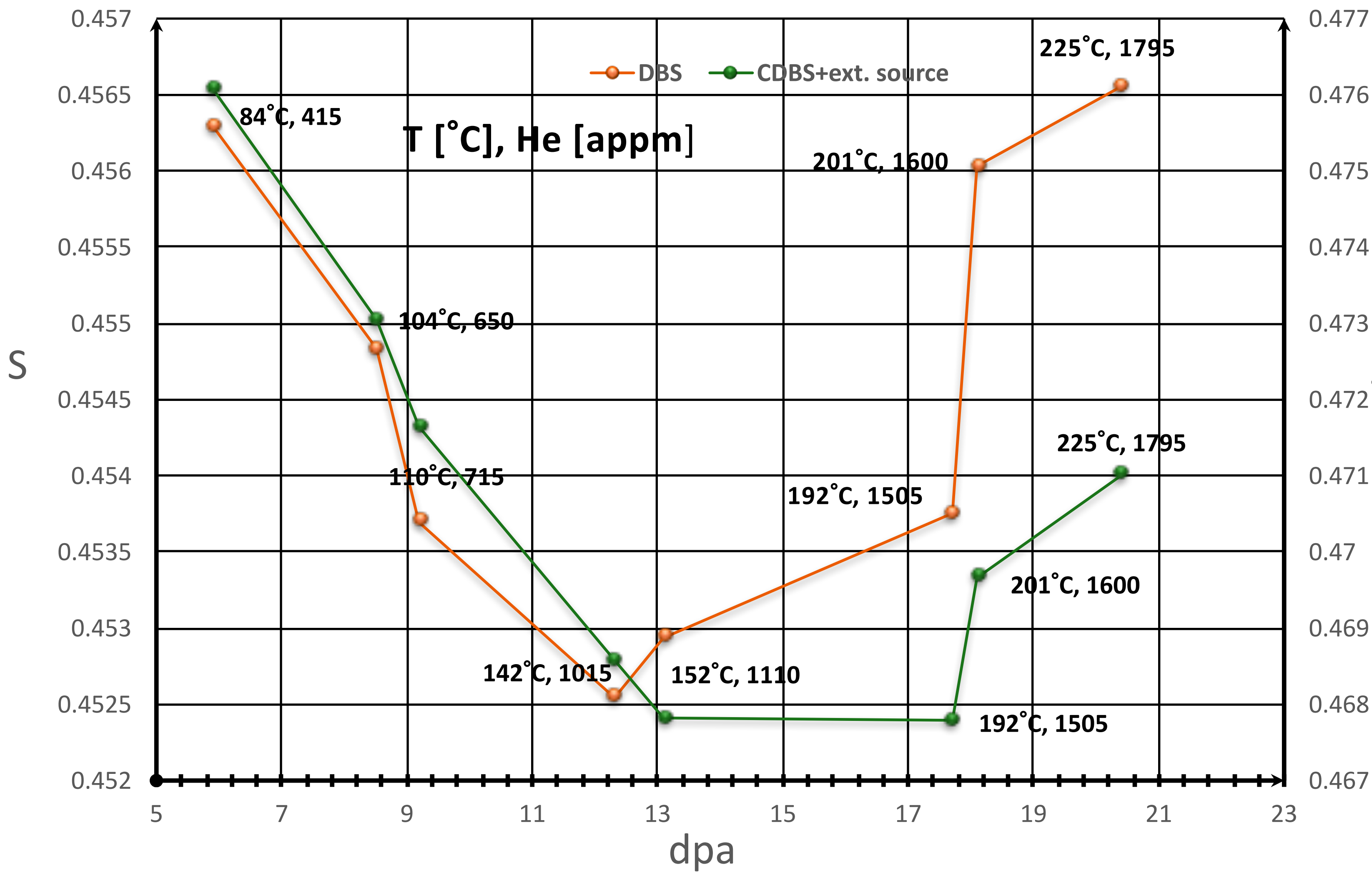
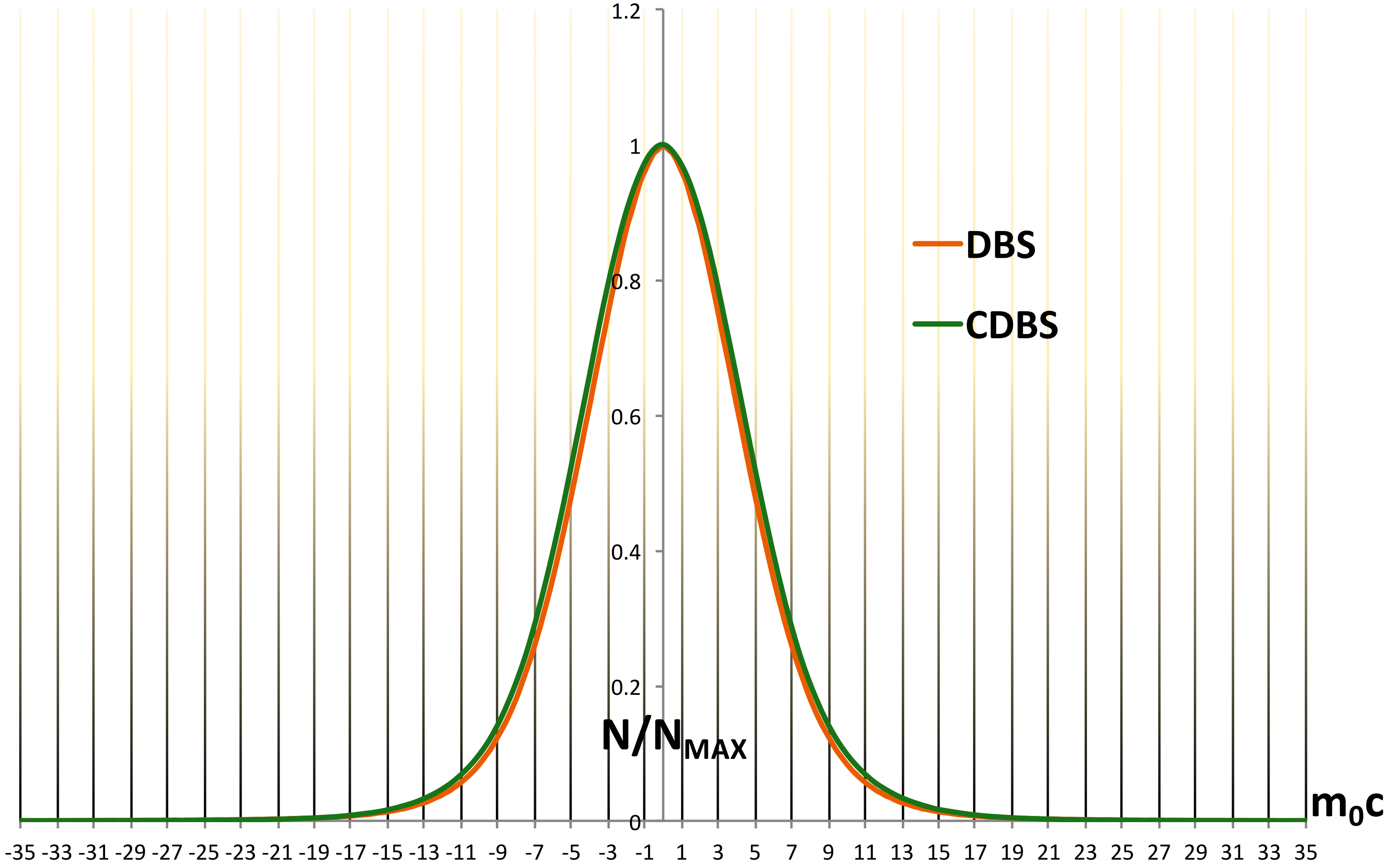
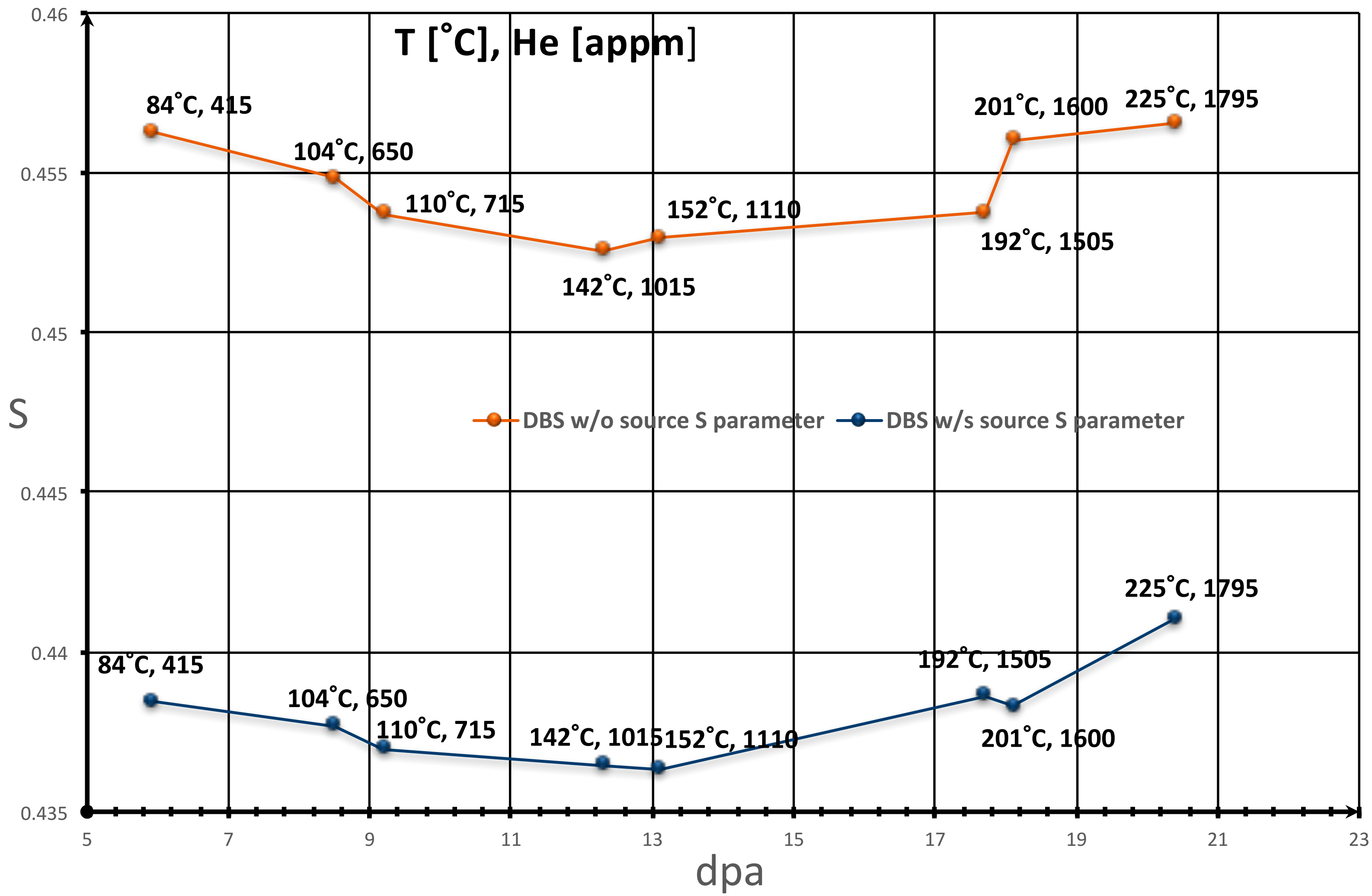
Comparison of CDBS and DBS measurement curves of EM10



Graph of DBS and CDBS – N/N_{MAX} (20.4 dpa)



Comparison of DBS S curves of EM10 in absolute values WITH or WITHOUT ext. source



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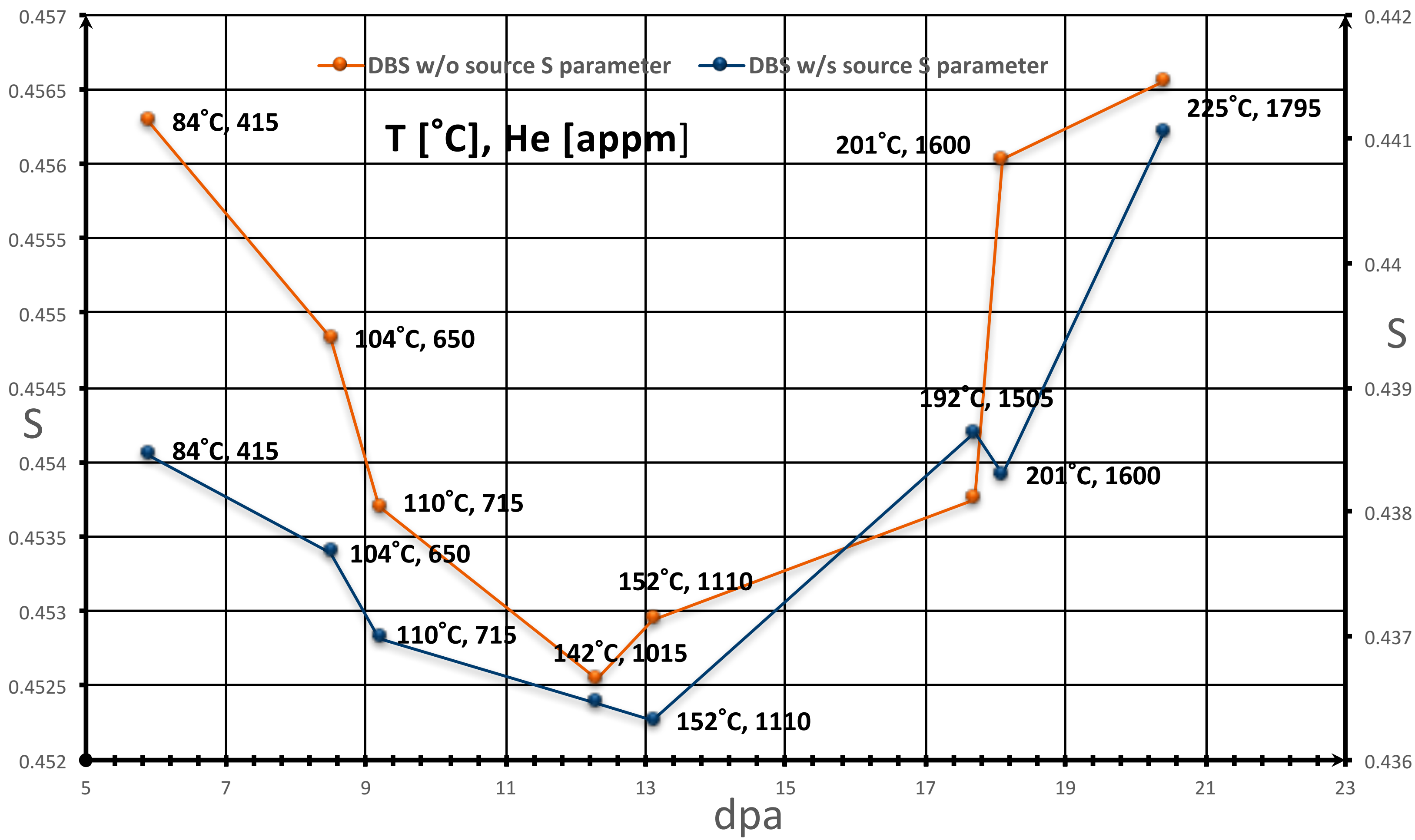
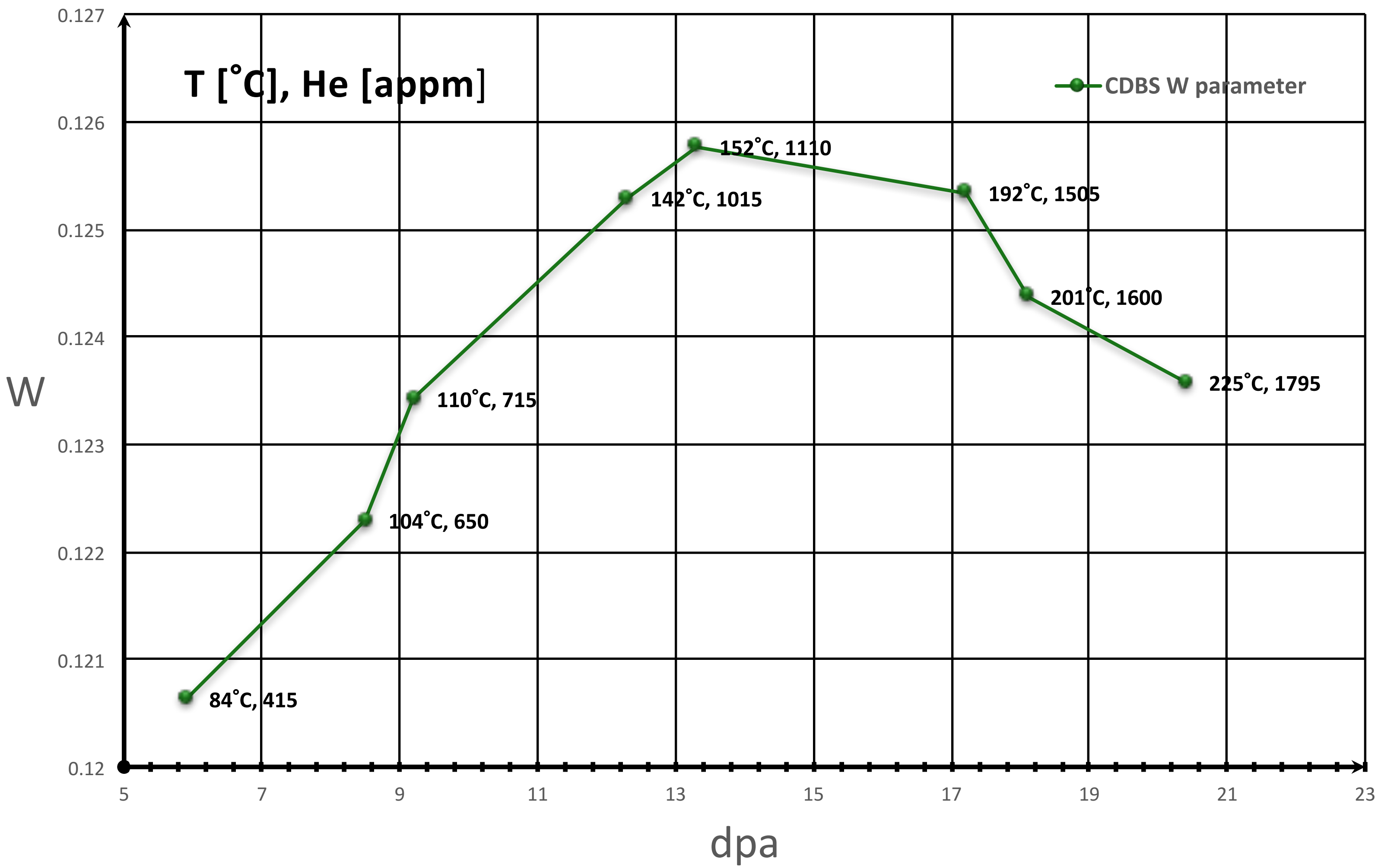
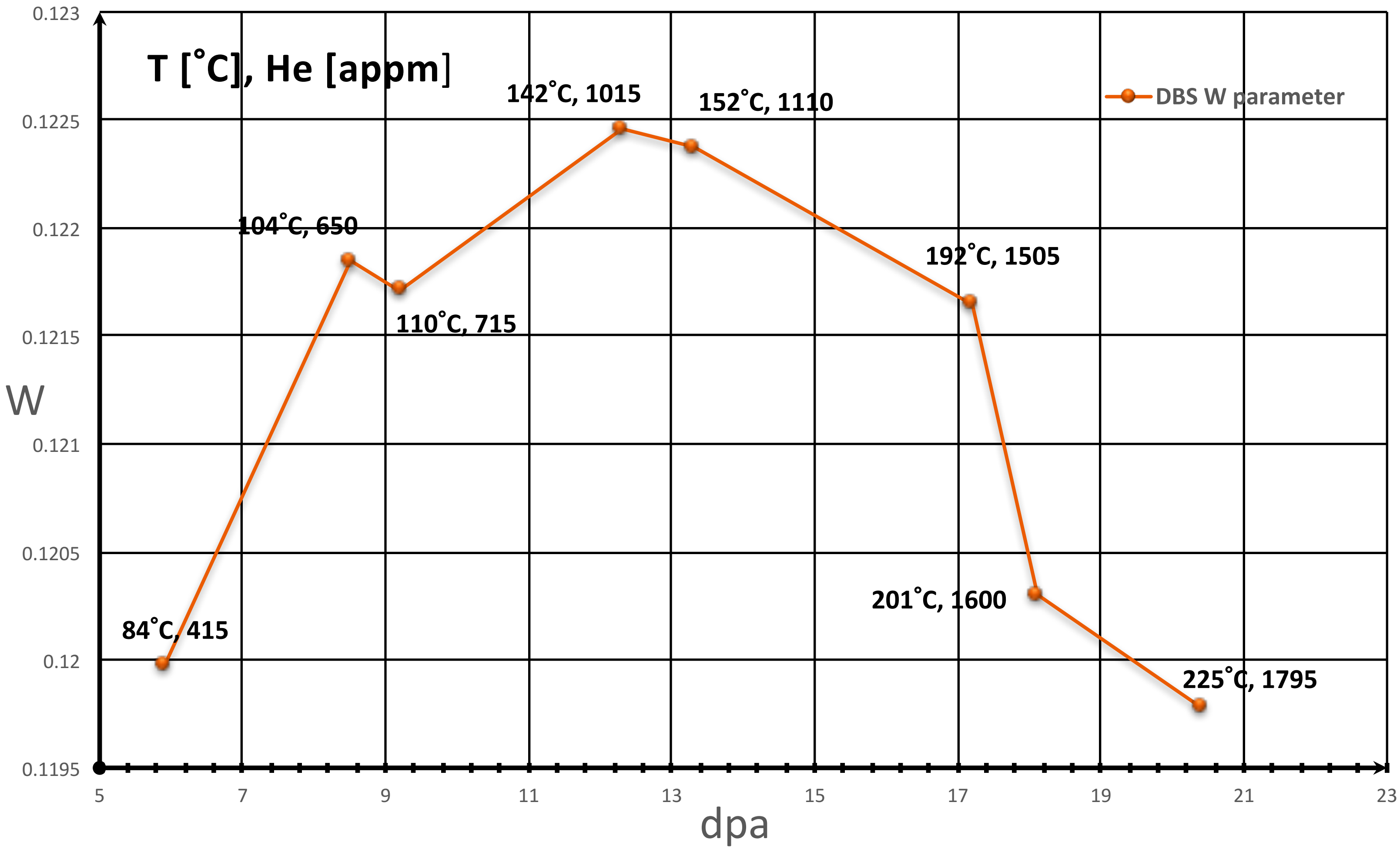
Comparison of DBS S curves of EM10 WITH or WITHOUT ext. source



EM10 samples measured by CDBS with external positron source – W parameter



EM10 samples measured by DBS with only internal positron source – W parameter



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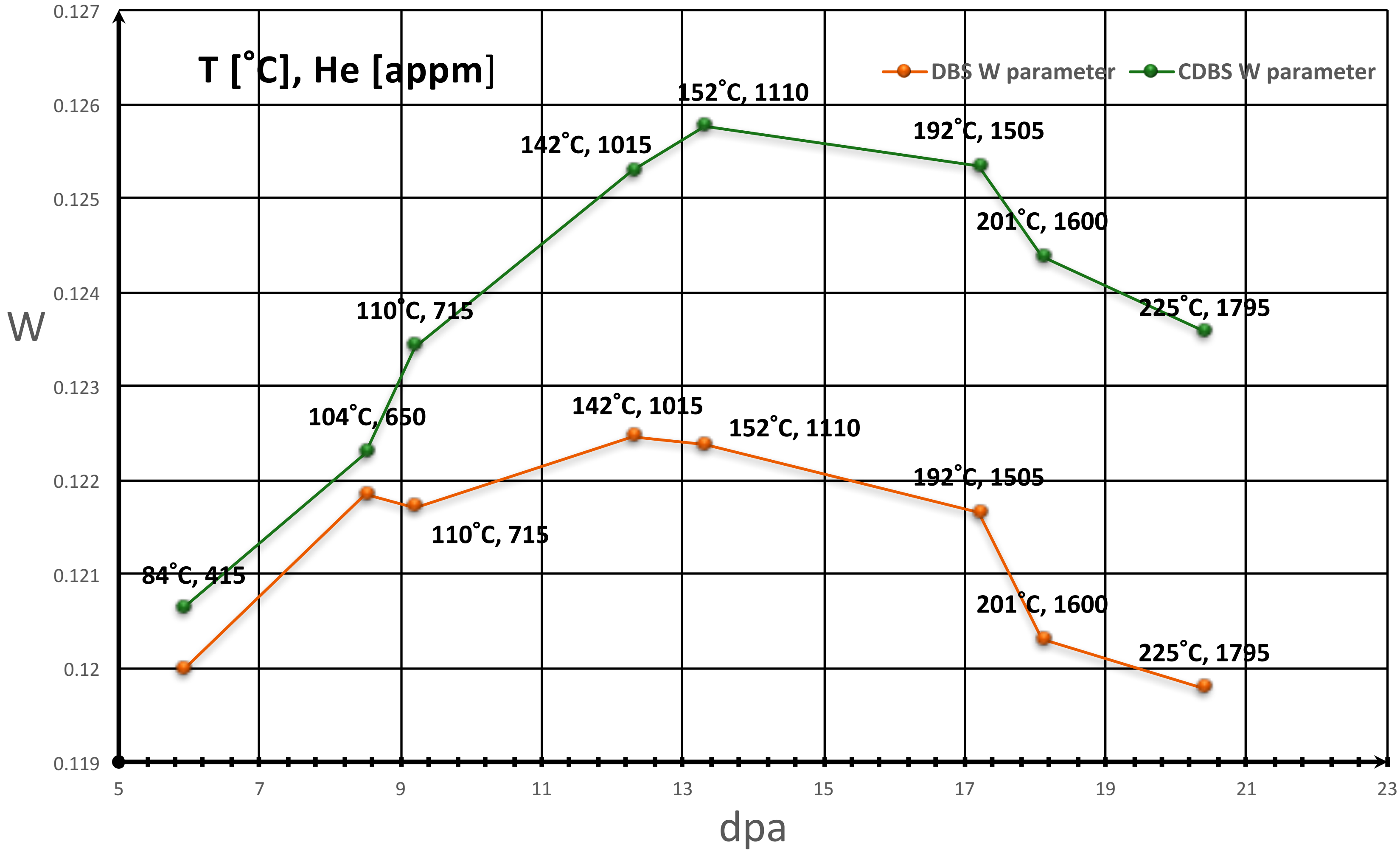
Comparison of DBS and CDBS W curves of EM10 in absolute values



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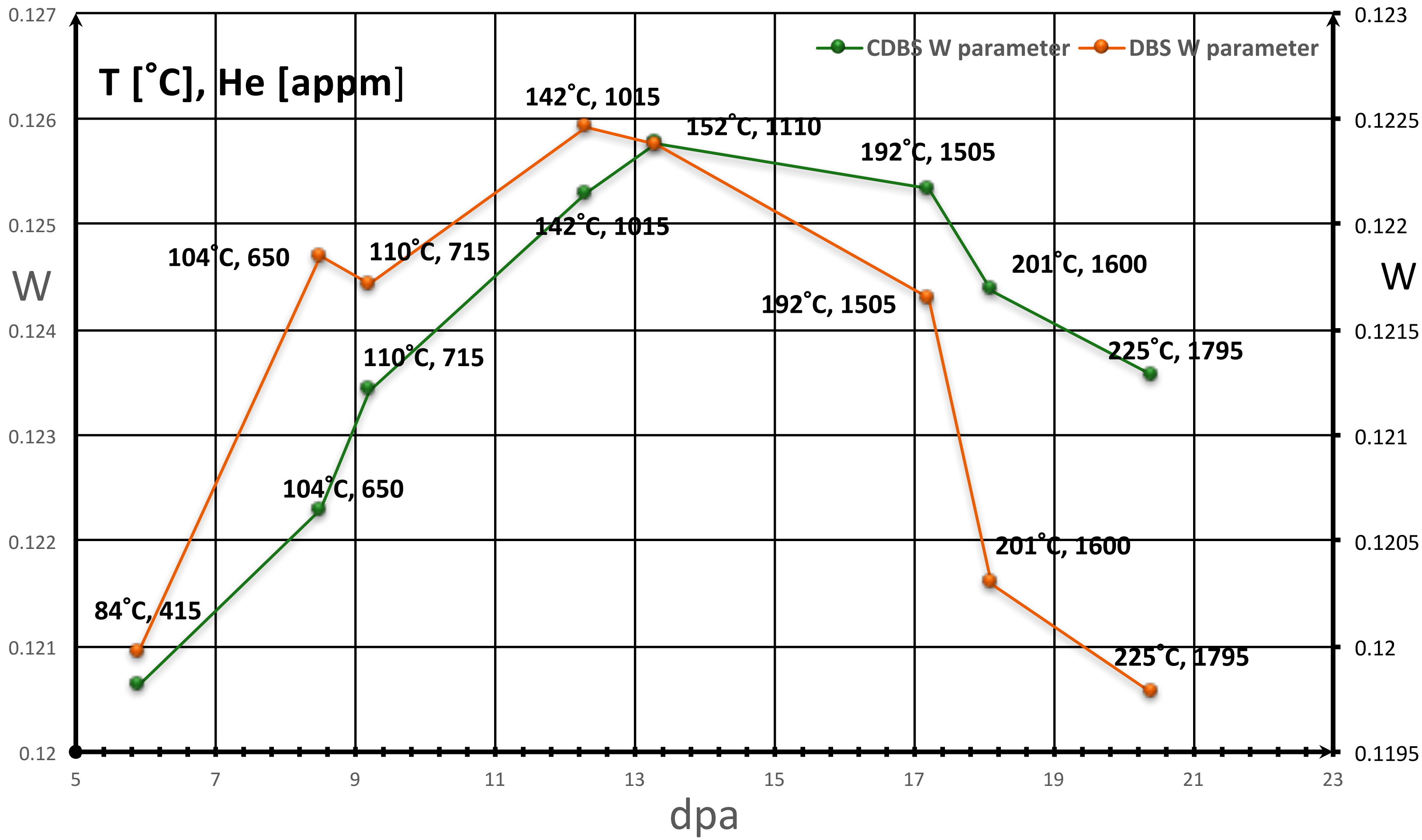
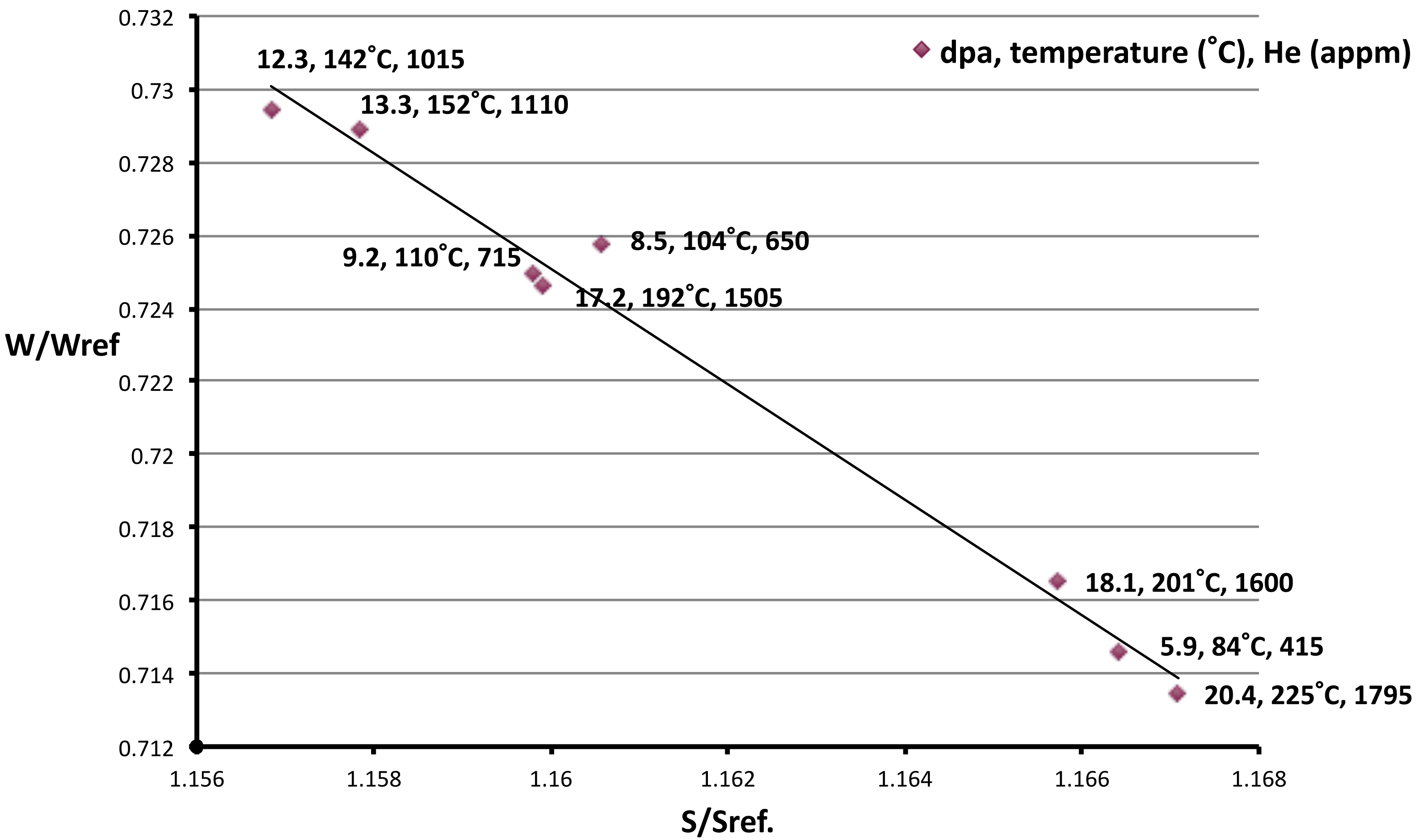
Comparison of DBS and CDBS W curves of EM10



S-W Graph of EM10, DBS, internal source



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Summary and Conclusion

- Internal transmutation-based positron source produced in steel samples irradiated in spallation targets offer a new potential for experimental research using PALS and DBS
- Conventional gamma spectrometer can provide interesting complementary data on spallation samples in very short time and for reasonable cost
- Experimental DBS data of He to V ratios are in a good agreement with theoretical modeling data
- Bellow ~12 dpa, a competition between new defects production and absorption of helium by existing ones takes place
- Growth of helium bubbles was identified for samples irradiated to more than 12 dpa

My thanks go to

- Dr. Vladimír Kršjak
- Dr. Yong Dai

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Thank you for your kind attention