Positron annihilation lifetime measurements of stainless steels irradiated in the SINQ target irradiation program

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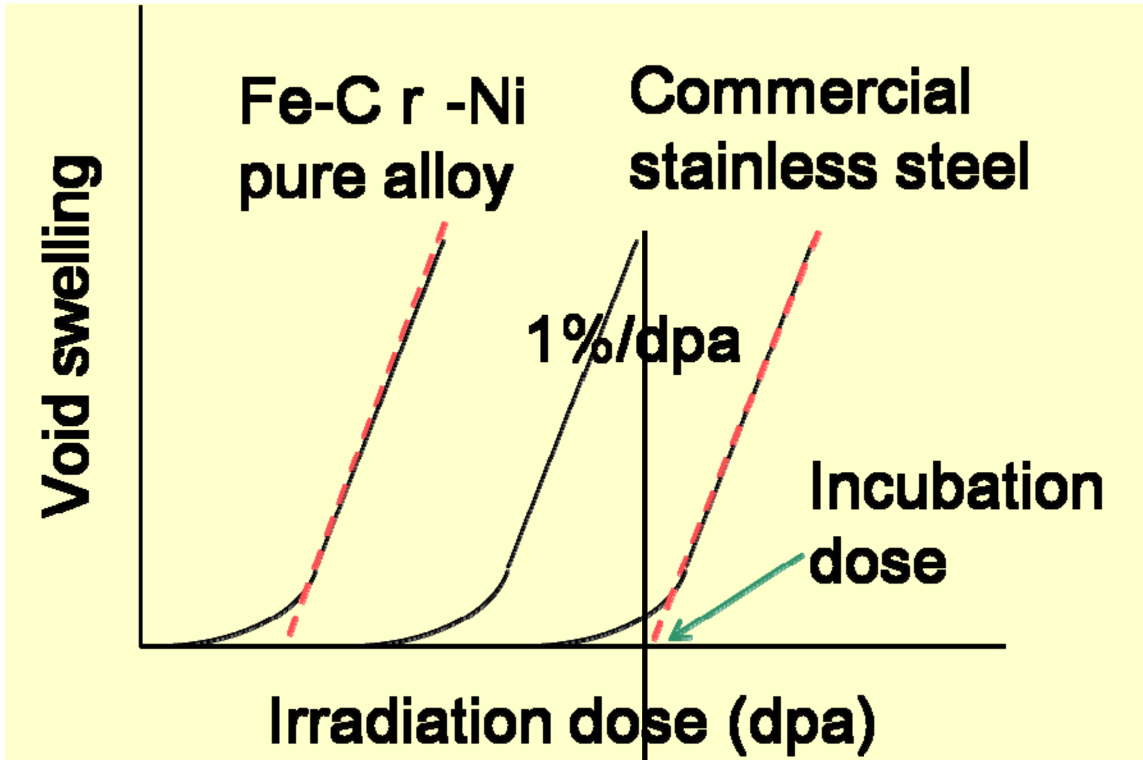
Background

- Facilities with high irradiation dose are required.
  - Fast breeder reactor
  - Spallation neutron source / Accelerator driven system
  - Fusion reactor etc.
- Reduced activated ferritic/martensitic steel and austenitic stainless steels are candidates for the structural materials of the facilities.

- For development of irradiation-resistant materials, point defect processes in the incubation period of void formation is very important.

Void swelling of stainless steel



Voids cannot be observed by TEM | Voids can be observed by TEM

Positron annihilation spectroscopy is very powerful tool to detect vacancy type defects.

[F. A. Garner, Nuclear Materials, vol. 10A, 1994, p.419.]

Purpose of this study

- Defect structures in reduced activated ferritic/martensitic steel F82H and austenitic stainless steel JPCA (Ti-added modified 316SS) during the incubation period were investigated after proton irradiation using positron annihilation lifetime measurement.

Experimental procedure

- Sample: F82H, JPCA (Ti-added modified 316SS)
- Irradiation condition (STIP-1)

F82H Tensile test sample

ID	Temp (℃)	dpa	He (appm)	H (appm)
2-ST-L11-L 2-ST-L12-L	98	5.9	413	1680
2-ST-L11-H 2-ST-L12-H	124	8.5	650	2530
2-ST-L13-L 2-ST-L14-L	133	9.2	715	2850
2-ST-L13-H 2-ST-L14-H	175	12.3	1015	4200
2-ST-L15-H 2-ST-L16-H	240	17.2	1505	6200
2-ST-L17-H 2-ST-L18-H	287	20.4	1795	7720

Red: isochronal annealing (200, 300, 400, 600℃)

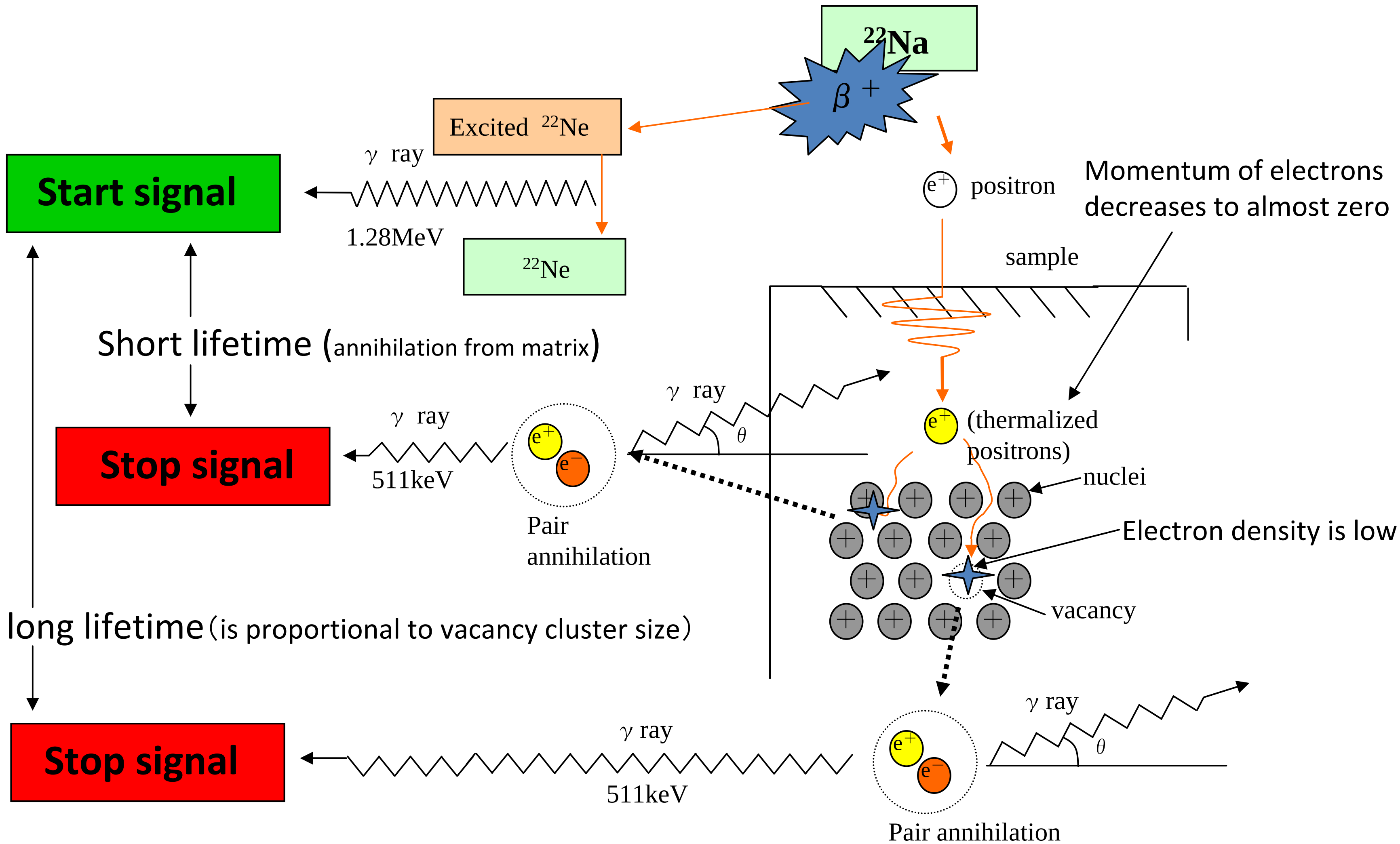
JPCA Fatigue test sample

ID	Temp (℃)	dpa	He (appm)	H (appm)
F21	140	6.3		

isochronal annealing (every 100℃ from 150℃ to 1050℃)

- Positron annihilation lifetime measurement

Positron annihilation lifetime measurement



Calculated positron annihilation lifetime

Table 1
The calculated positron lifetimes and binding energies for vacancy clusters in Ni, Cu, and Fe as a function of the cluster size

Ni			Cu			Fe		
Defect	τ (ps)	E_b (eV)	Defect	τ (ps)	E_b (eV)	Defect	τ (ps)	E_b (eV)
Bulk	100	0.00	Bulk	110	0.00	Bulk	104	0.00
V_1	169	3.34	V_1	173	2.35	V_1	180	3.56
V_2	188	3.82	V_2	196	2.74	V_2[a]	187/202	3.86/4.11
V_4	246	4.66	V_4	255	3.36	V_5	246	4.89
V_7	265	4.92	V_7	274	3.57	V_9	280	5.32
V_{13}	341	5.54	V_{13}	348	4.07	V_{15}	368	6.01
V_{19}	371	5.77	V_{19}	377	4.28	V_{27}	396	6.27
V_{43}	410	6.15	V_{43}	413	4.62	V_{51}	419	6.55
V_{55}	420	6.28	V_{55}	421	4.74	V_{59}	426	6.69
V_{79}	427	6.42	V_{79}	428	4.86	V_{65}	427	6.72
V_{177}	435	6.60	V_{177}	436	5.02	V_{137}	435	6.91

[a] The values are listed for two distinct divacancy geometries, i.e. V_2 along [111] and [100] directions.

[H. Ohkubo et al., Mater. Sci. Eng. A350 (2003) 95.]

- Positron lifetime is proportional to the size of vacancy clusters.

- In metallic system, Positron lifetime is less than 500ps.
  500ps is saturation value of positron lifetime.
  Even if void grows and is observed by TEM, positron lifetime of the void is less than 500ps.

Positron lifetime of vacancy clusters-He complexes in Fe

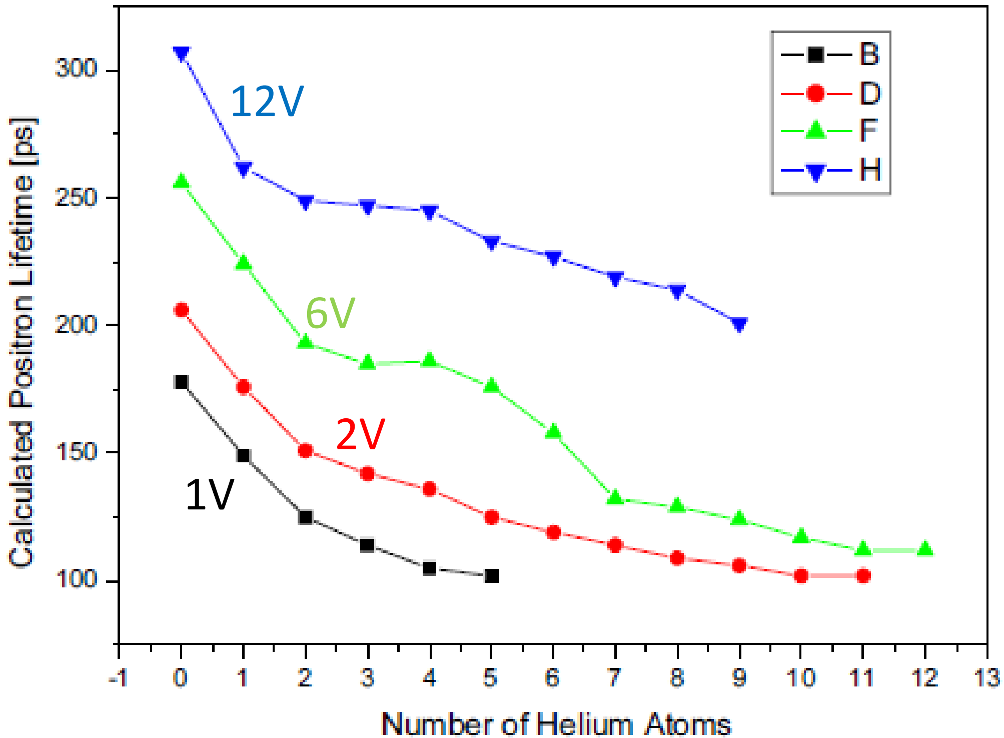
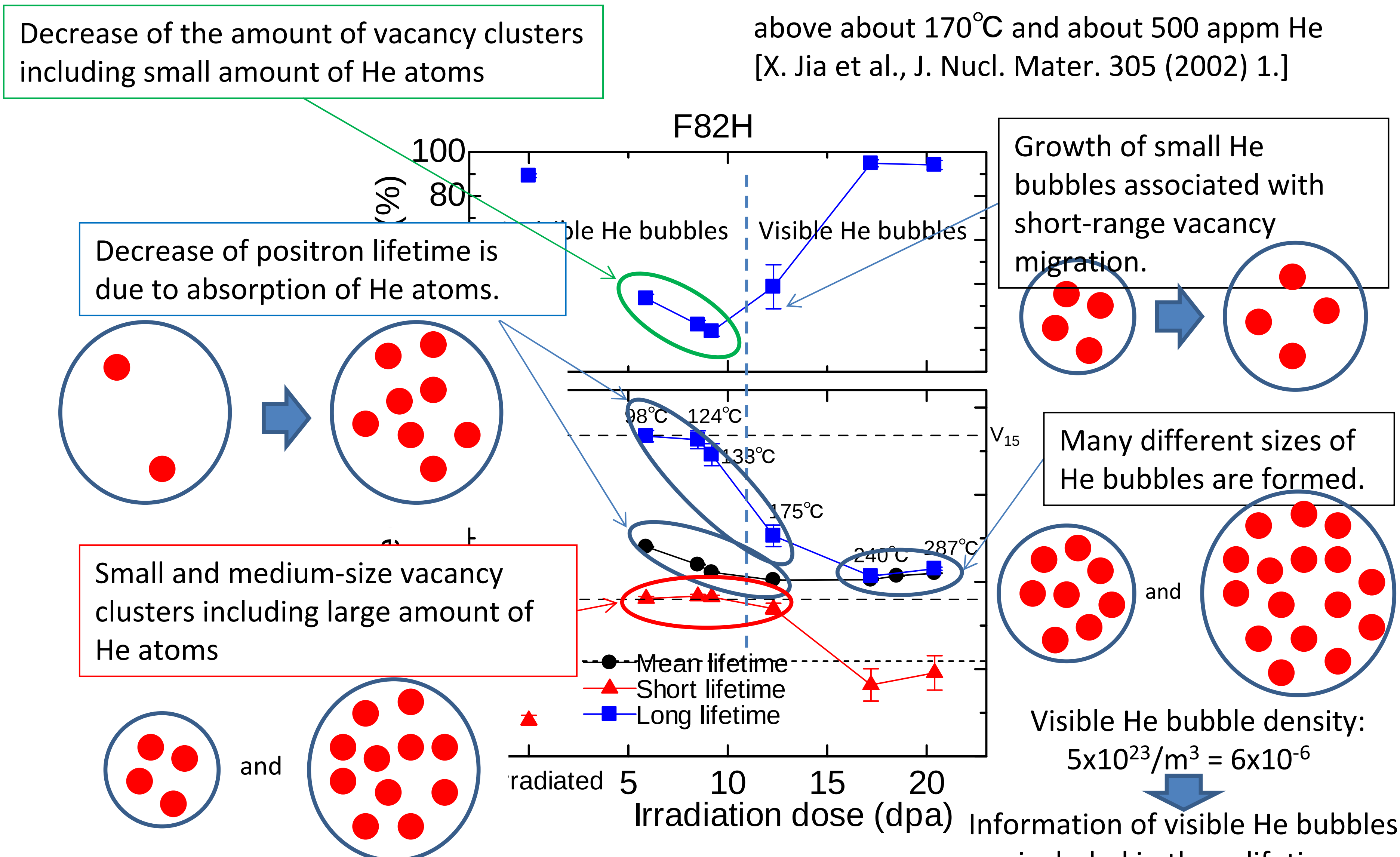


Figure 4 Correlation between positron lifetime and the number of helium atoms in nano-void (B) 1V+nHe, (D) 2V+nHe, (F) 6V+nHe, (H) 12V+nHe.

[T. Troev et al., Phys. Status Solidi C 6 (2009) 2373]

Dose dependence of positron lifetime in F82H

Formation of visible He bubbles in F82H: above about 170℃ and about 500 appm He [X. Jia et al., J. Nucl. Mater. 305 (2002) 1.]

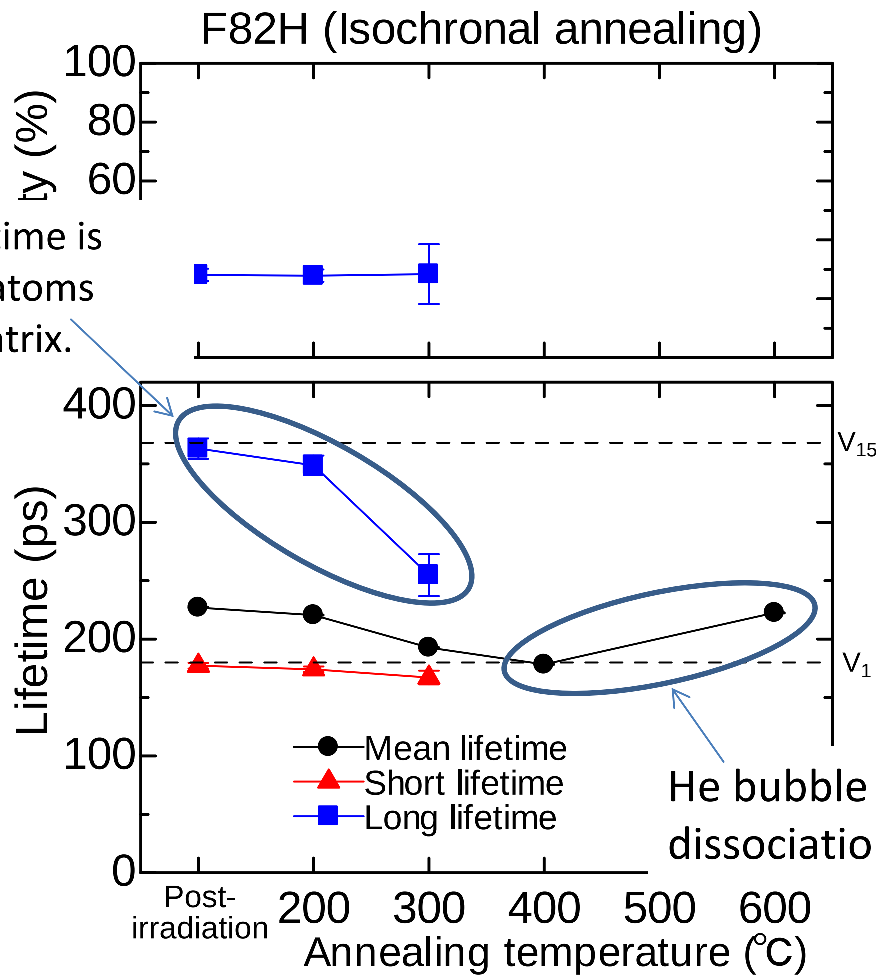


Visible He bubble density: $5x10^{23}/m^3 = 6x10^{-6}$

Information of visible He bubbles are included in these lifetimes

Annealing behavior of F82H

Decrease of positron lifetime is due to absorption of He atoms weakly trapped in the matrix.



He bubble size increase because of dissociation of V-He$_n$ complexes.

500°C: V-He$_n$ complexes dissociate

700°C: V$_m$-He$_n$ complexes dissociate

1100°C: Large He bubbles dissociate

TDS measurements of Fe-Cr alloys

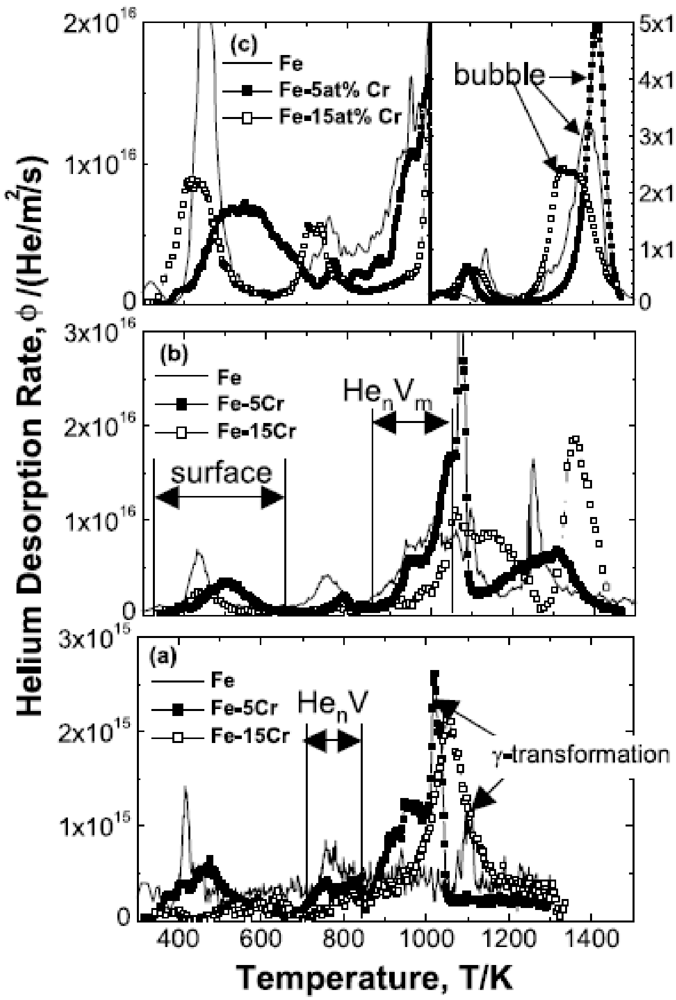
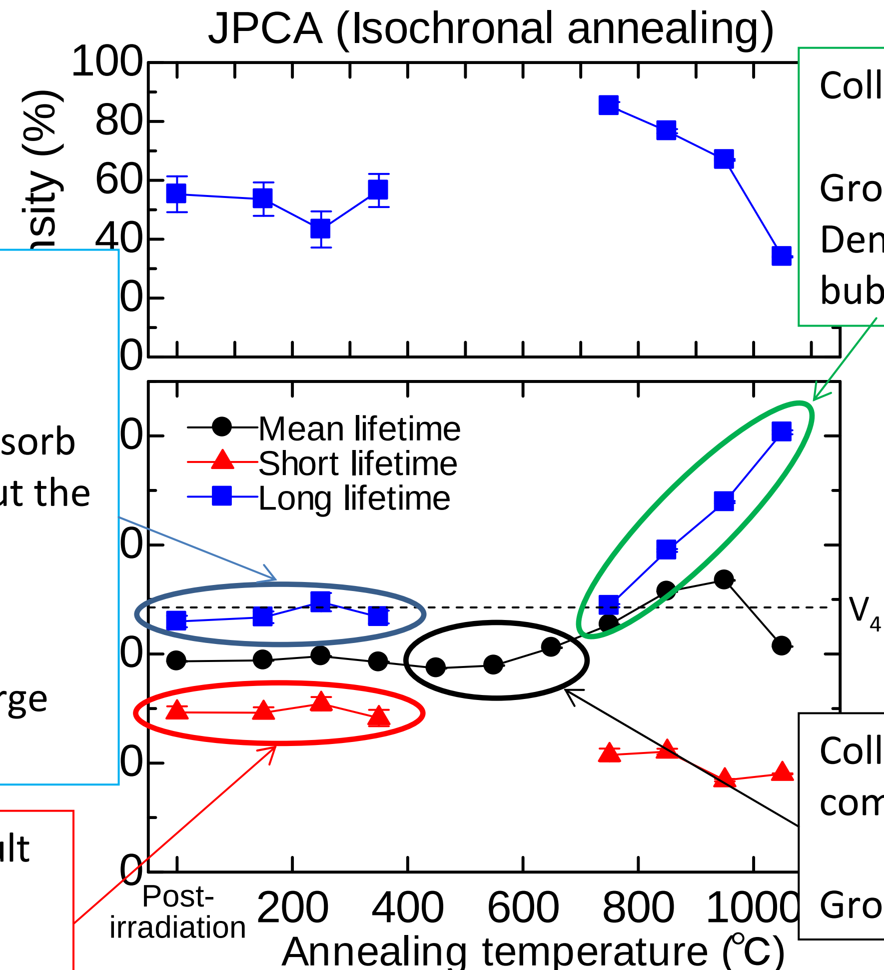


Fig. 1. He desorption spectra of Fe, Fe–5Cr and Fe–15Cr irradiated by 8 keV He+ ions at room temperature. The irradiation doses are (a) 1017, (b) 1018 and (c) 10^{19} He^{+}/m^{2}.

500℃: V-He_n complexes dissociate
700℃: V_m-He_n complexes dissociate
1100℃: Large He bubbles dissociate

[R. Sugano et al., J.Nucl. Mater. 329–333 (2004) 942]

Annealing behavior of JPCA



Formation of V_m-He_n complexes

It is expected that they absorb He atoms by annealing, but the lifetime do not change.

After irradiation, V_m-He_n complexes may include large amount of He atoms.

Formation of stacking fault tetrahedra (SFTs), dislocation loops, V-He_n complexes.

Collapse of V_m-He_n complexes

Growth of He bubbles
Density of He atoms in He bubbles decreases.

Collapse of SFTs and V-He_n complexes

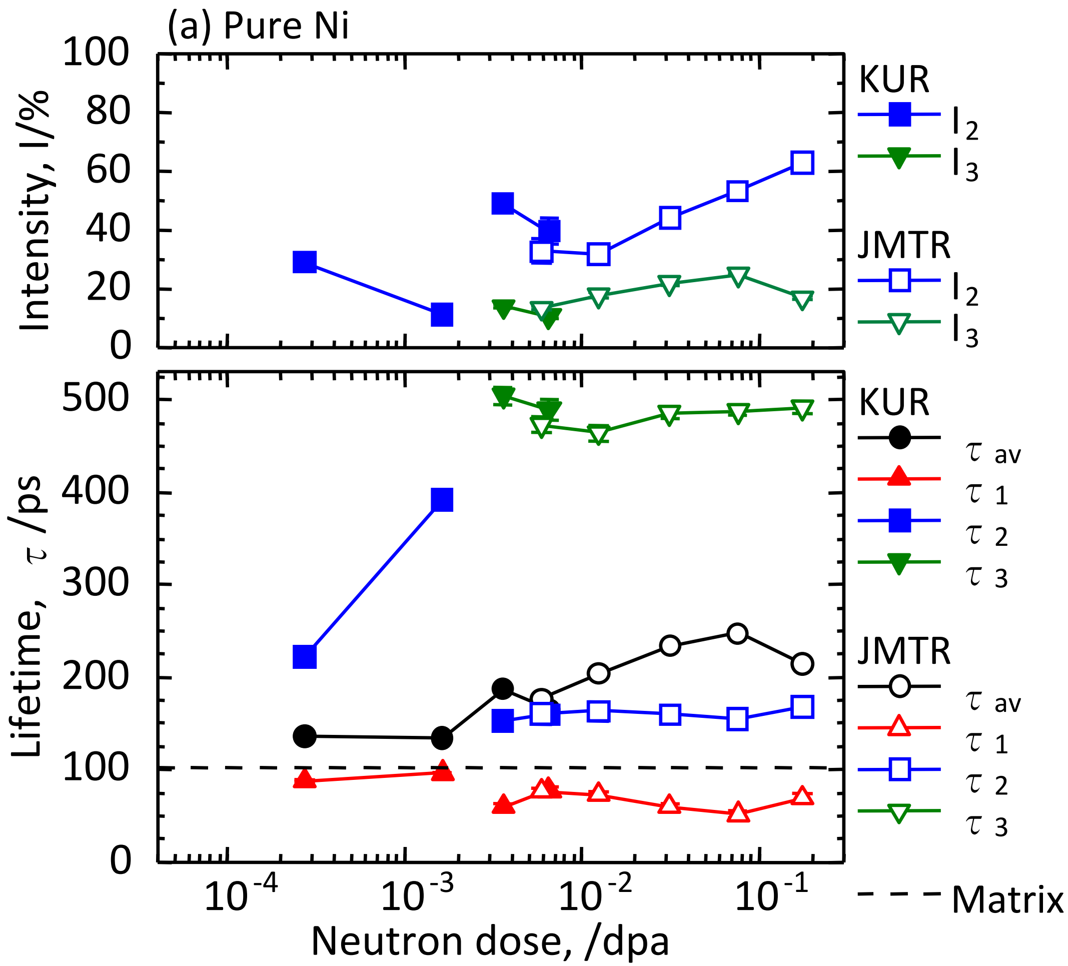
Growth of He bubbles

500℃: V-He_n complexes dissociate
700℃: V_m-He_n complexes dissociate
1100℃: Large He bubbles dissociate

Conclusion

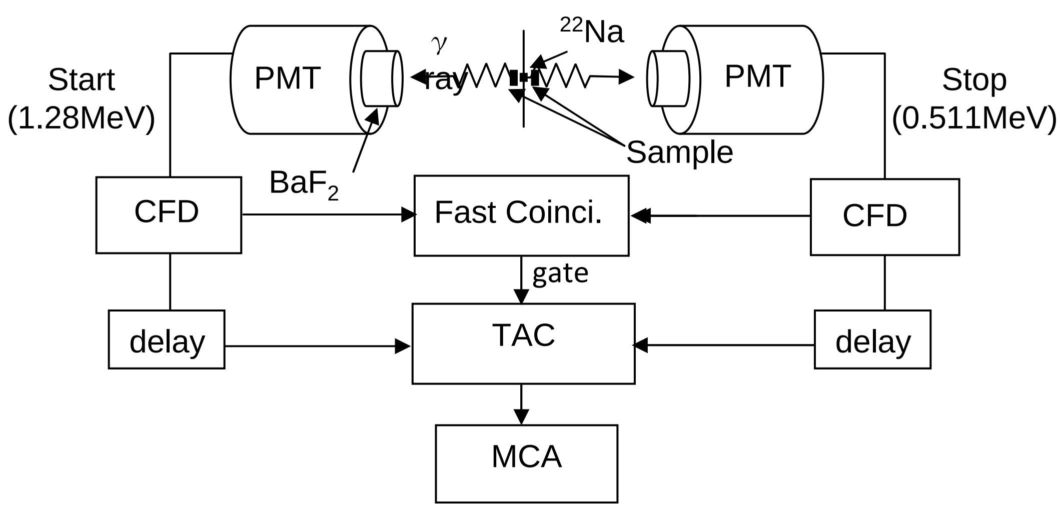
- Positron annihilation lifetime (PAL) measurements of F82H and JPCA irradiated with protons at SINQ were performed.
- We could detect the small vacancy clusters by PAL, which cannot be observed by TEM.
- In F82H, positron annihilation lifetime of He bubbles decreases with increasing the irradiation dose. Because the He bubbles absorb more He atoms.
- In isochronal annealing, we can detect the growth process of He bubbles.
- This is the first detection of small vacancy clusters and He bubbles in F82H and JPCA irradiated in the STIP.

Positron annihilation lifetimes in fission neutron-irradiated Ni

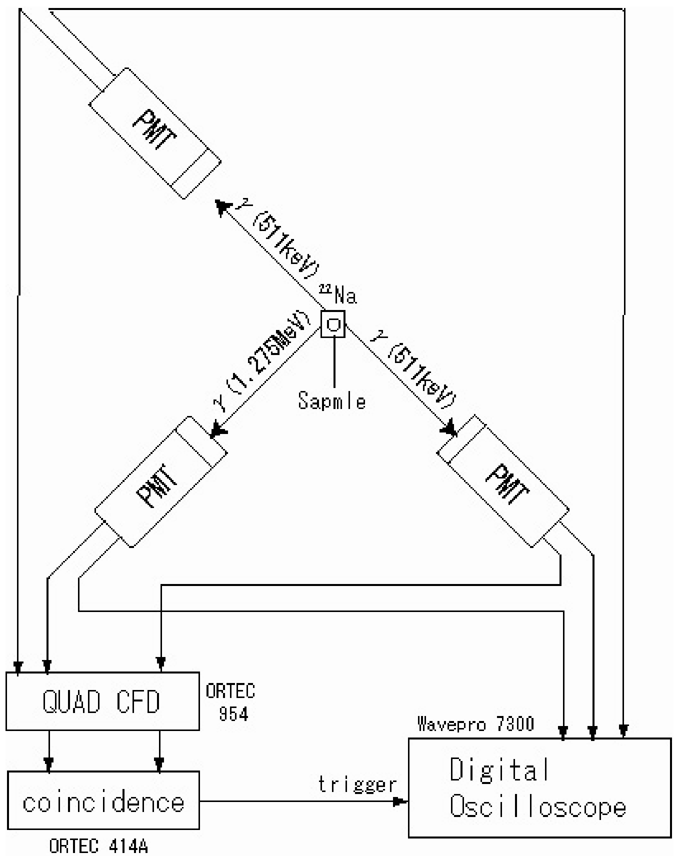


Void growth is observed by TEM in more than 0.01dpa, but positron lifetime is saturated.

Positron annihilation lifetime measurement system



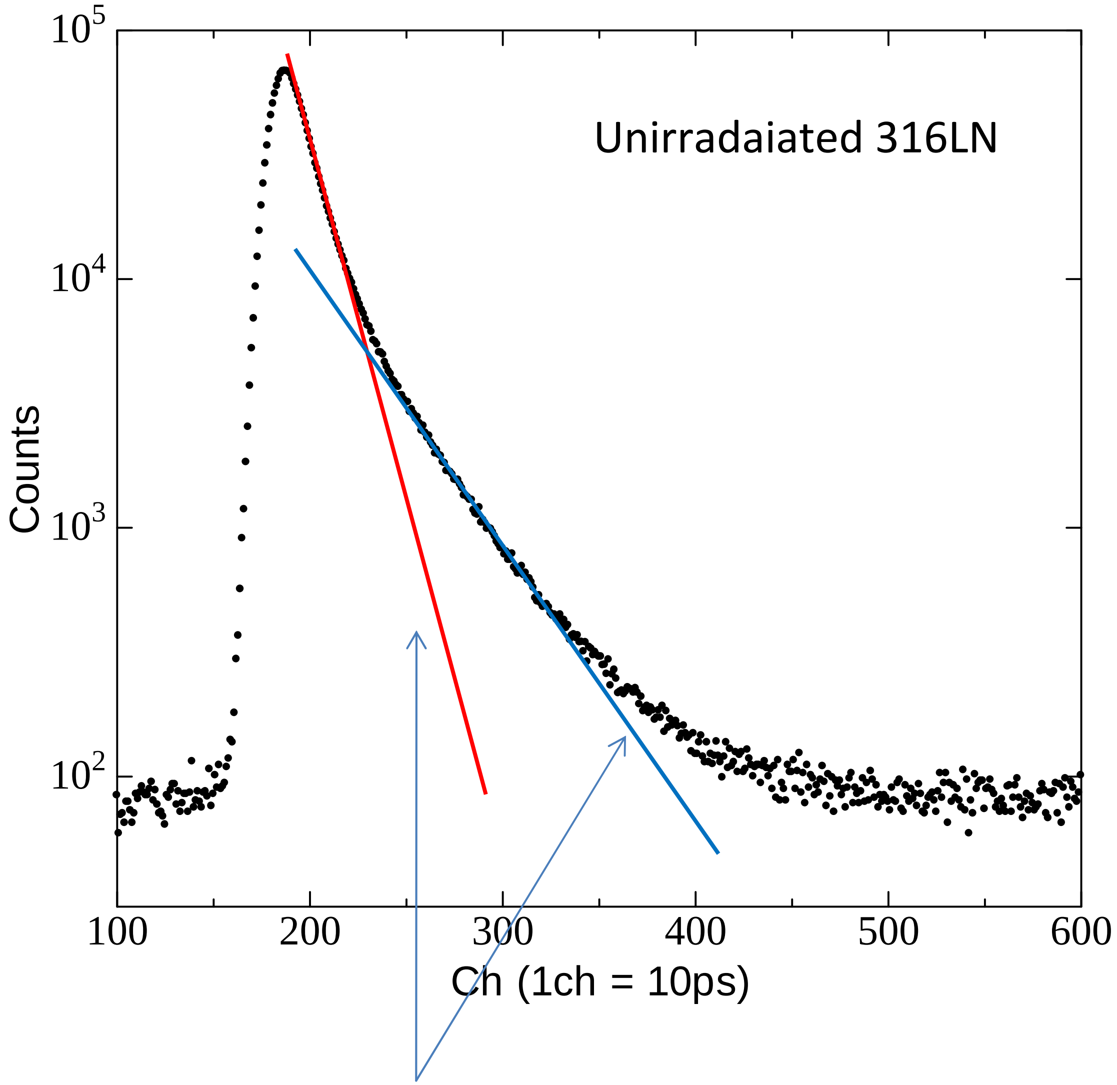
Conventional measurement system
(two-detector system)



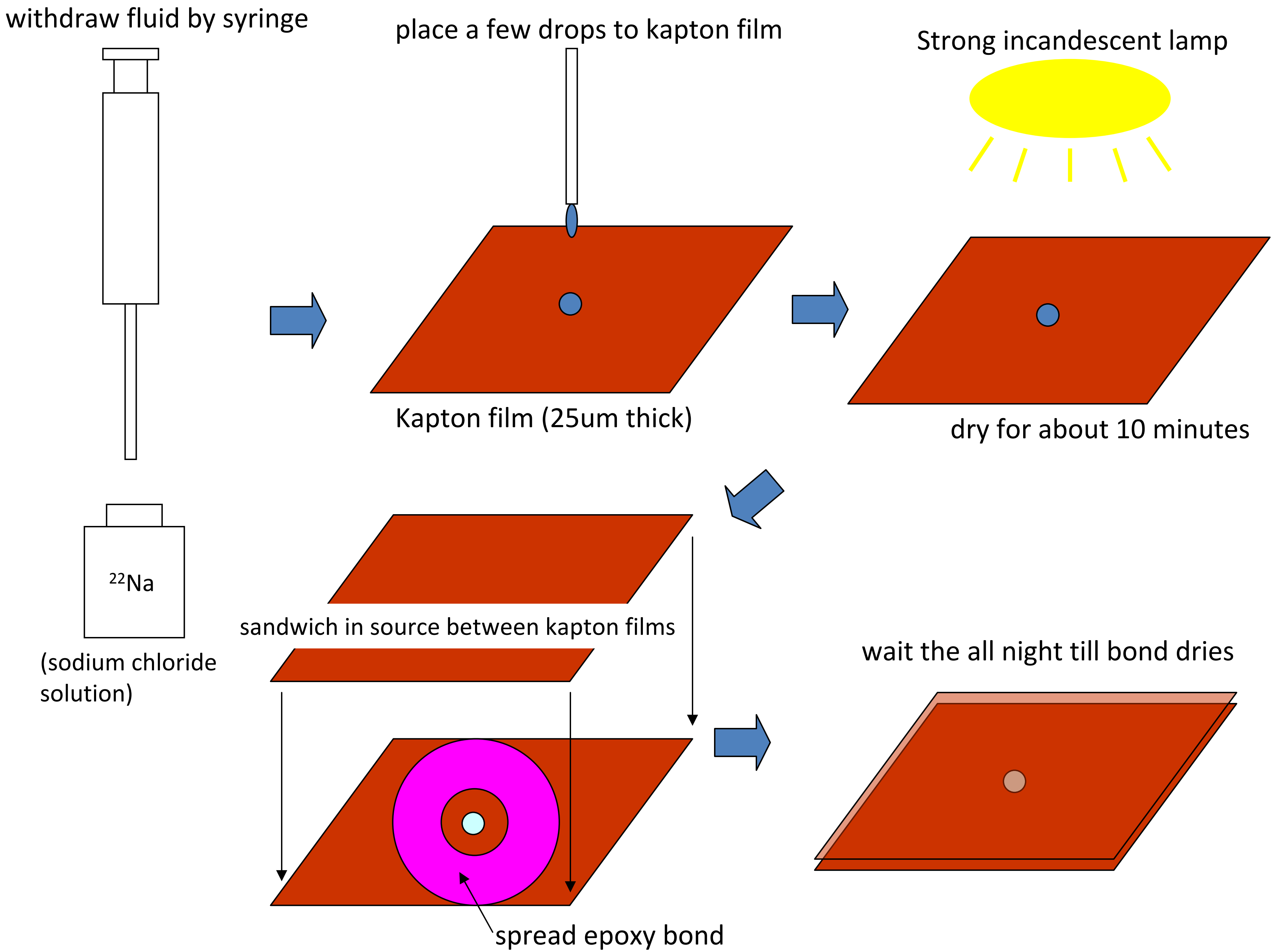
Improved measurement system
using a digital oscilloscope
(three-detector system)

Merit: Reduction of background
Demerit: Decrease of count rate

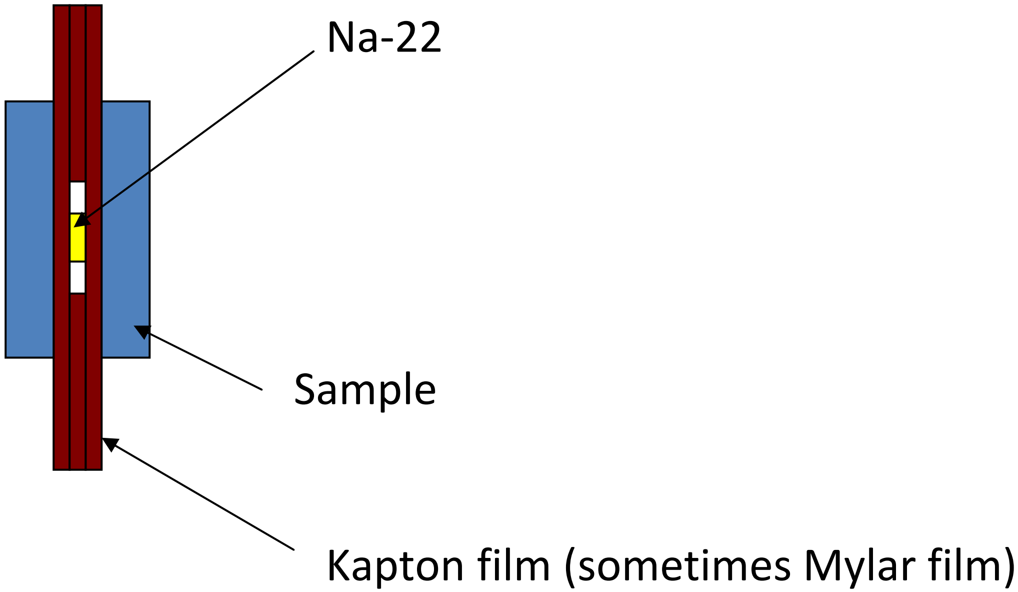
Positron annihilation lifetime spectrum



This spectrum is composed of these two curves.


withdraw fluid by syringe
place a few drops to kapton film
Strong incandescent lamp
Kapton film (25um thick)
dry for about 10 minutes
²²Na
(sodium chloride solution)
sandwich in source between kapton films
wait the all night till bond dries
spread epoxy bond

Set of samples

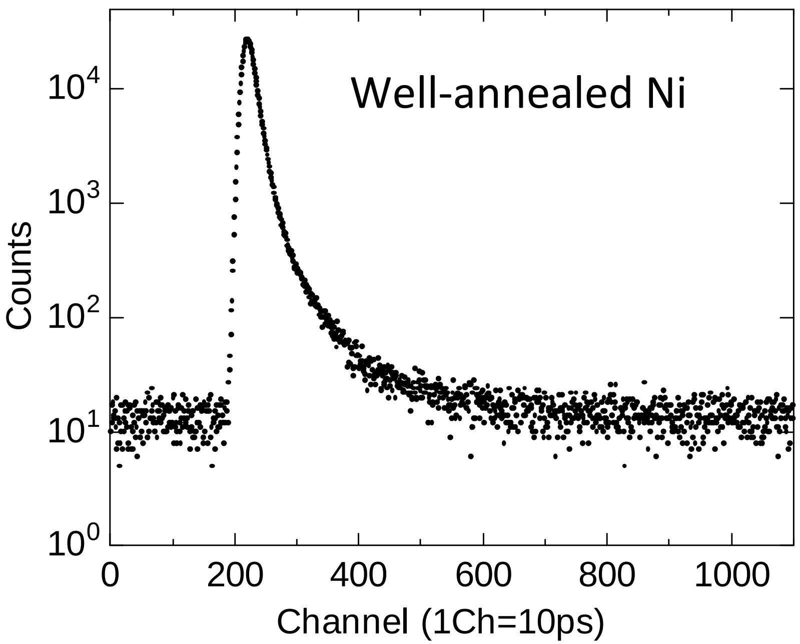


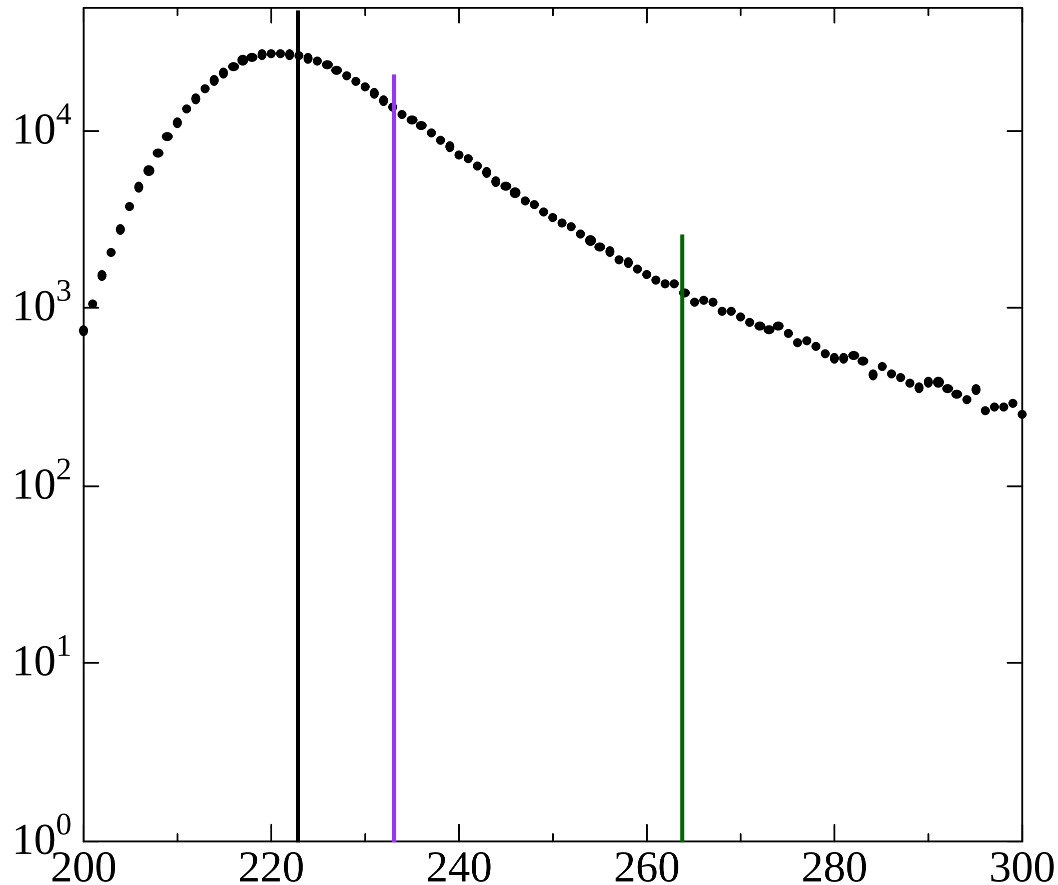
A part of positrons annihilate in the Kapton film.

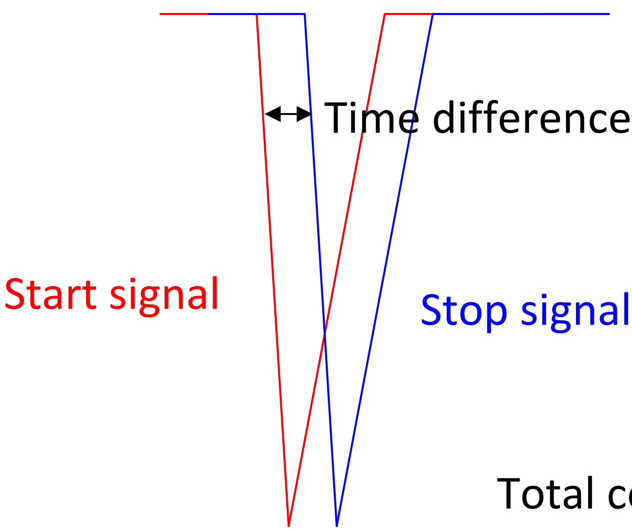
Ratio of positrons, which annihilate at Kapton film, depends on the thickness.

5um: ~13%, 10um: ~20%, 25um: ~33%

How to make lifetime spectrum





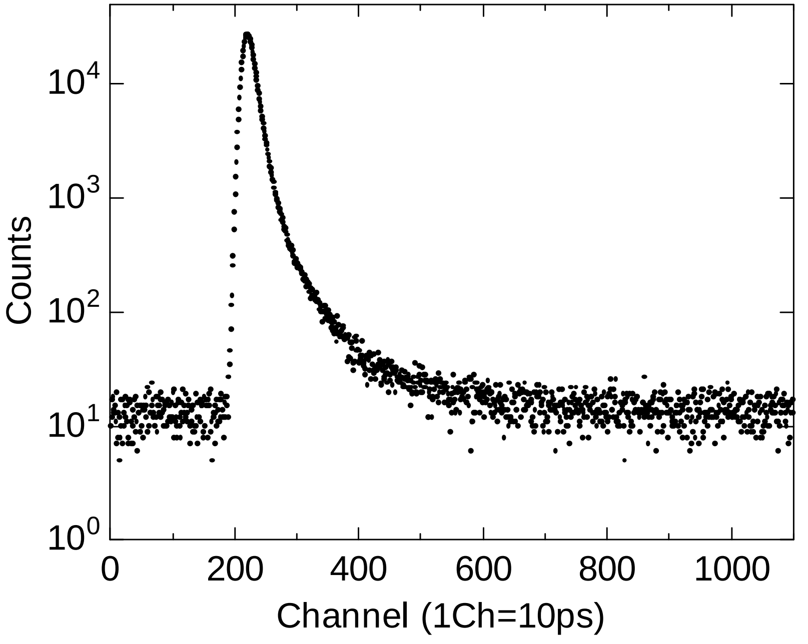


Time difference: 100ps

Time difference: 400ps

Total count of more than 1M is needed for good statistics.

Analysis of lifetime spectrum



We usually use PALSfit program,
which is developed by one group of Riso DTU.

$$T'(t) = \int_{-\infty}^{\infty} T(x) G(t-x) dx + B$$

$$\int_{-\infty}^{\infty} G(t) dt = 1$$

T': Lifetime spectrum (left figure)
T: Decay function
G: Time-resolution function
B: Background

G is given by a sum of two or three Gaussians

One component

$$T(t) = \frac{I_1}{\tau_1} \exp\left(-\frac{t}{\tau_1}\right)$$

Two components

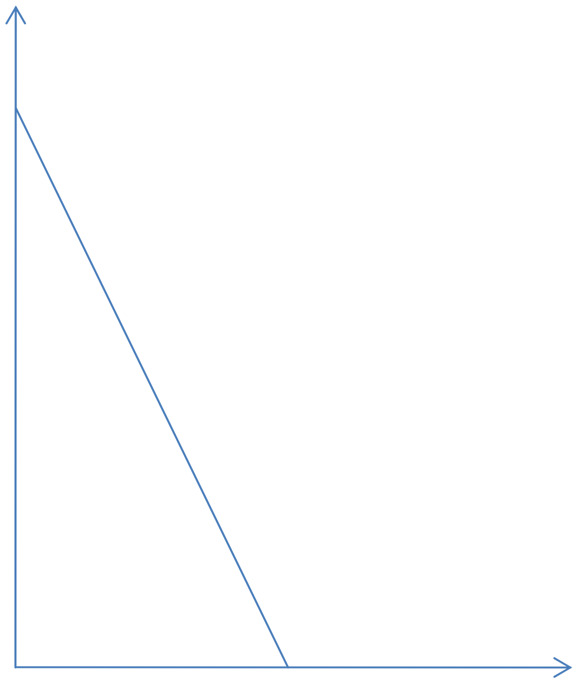
$$T(t) = \frac{I_1}{\tau_1} \exp\left(-\frac{t}{\tau_1}\right) + \frac{I_2}{\tau_2} \exp\left(-\frac{t}{\tau_2}\right)$$

τ : lifetime
I : lifetime intensity

Three components

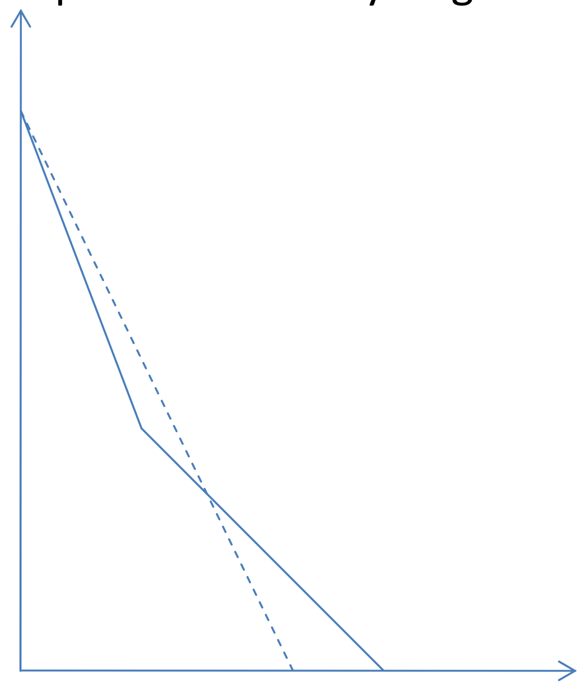
$$T(t) = \frac{I_1}{\tau_1} \exp\left(-\frac{t}{\tau_1}\right) + \frac{I_2}{\tau_2} \exp\left(-\frac{t}{\tau_2}\right) + \frac{I_3}{\tau_3} \exp\left(-\frac{t}{\tau_3}\right)$$

Without any defects



$$\tau = \frac{1}{\lambda_m} \approx 100\,ps$$

With single vacancies
(sample contains only single vacancies)



$$\tau_1 = \frac{1}{\lambda_m + \kappa} < 100\,ps$$

$$\tau_2 = \frac{1}{\lambda_d} \approx 170 - 500\,ps$$

λ_m : positron annihilation rate in the matrix

λ_d : positron annihilation rate at the defect site

κ : positron transition rate from the matrix to the defect site