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Relation between thermal diffusivity and positron annihilation lifetime in ceramic materials

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Abstract

Ceramics are candidate for divertor material in the future fusion reactor, where thermal diffusivity is one of the most important factors. Many studies were reported on properties of post irradiated specimen, but there have been few studies on properties during irradiation. Now we attempt to estimate a dynamic effect during ion-beam irradiation via positron annihilation spectroscopy. In this study, positron annihilation measurements were performed on neutron or electron irradiated specimens that showed severe degradation in thermal diffusivity to clarify the relation between positron-annihilation lifetime and thermal diffusivity. The positron annihilation lifetime of irradiated α-Al_2O_3 and AlN showed obvious increase, while β-SiC showed no change in lifetime, and β-Si_3N_4 rather showed decrease.

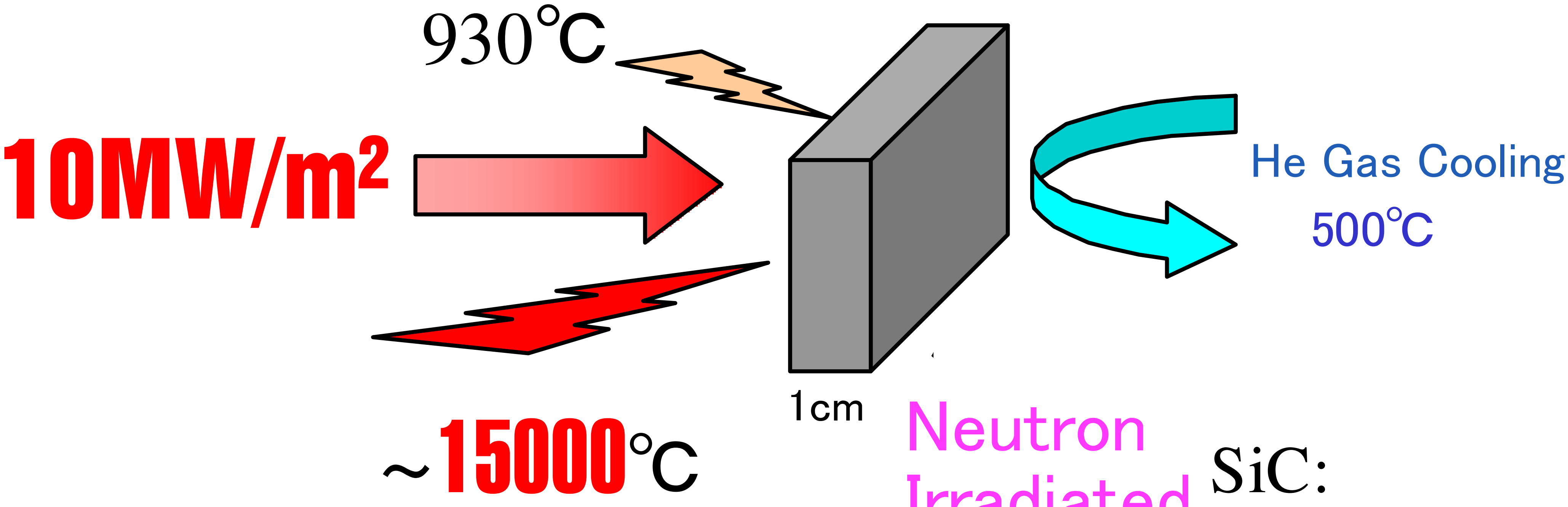
Keyword

ceramics, neutron irradiation, thermal diffusivity,
positron annihilation measurements

Degradation of Thermal Conductivity in Ceramics

Divertor of Fusion Reactor

Unirradiated SiC: 230 W/m・K



Neutron Irradiated SiC: **7** W/m・K

Thermal Diffusivity of Ceramics

- Heat is mainly carried by phonon unlike metals.
- In non-irradiated materials, thermal diffusivity is affected by microstructure of grain that was changed with sintering process.

In an irradiated specimen, mean free path of phonon is decreased to very small and the grain structure is too large to affect to the thermal diffusivity.

Thermal Diffusivity $\alpha = 1/3 \cdot \mu \cdot \lambda$

μ : Mean speed of phonon [m/s]

λ : Mean free path of phonon [m]

$$1/\lambda = 1/\lambda_a + 1/\lambda_b + 1/\lambda_c \cdots$$

Almost constant with each material

Phonon-Phonon Scattering

Changes with the temperature *T* as k/T^n (for non-irradiated single crystal, n=1)

Phonon-Lattice Scattering

Increased with density of irradiation defects

Temperature Change

During the Irradiation

After the Irradiation

Estimation of Thermal Diffusivity During the Irradiation

3 Assumption

1. Degradation of thermal diffusivity with neutron dose was saturated ～ 3×10^{26} n/m^2
2. Defects were stable until annealed above the irradiation temperature
3. Post irradiated specimens kept the same amount of defects as during the irradiation

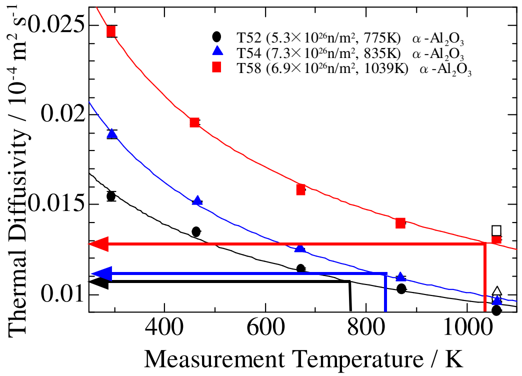
Thermal diffusivity at the irradiation temperature

$$\alpha_{irr}=k/T_{irr}^{n}$$

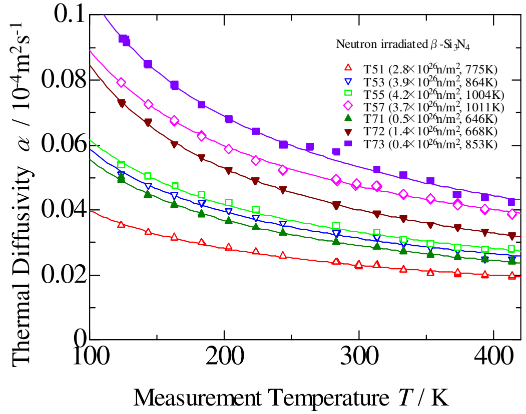
T_{irr}: the Irradiation Temperature

k, n: fitting parameters obtained from measurement of thermal diffusivity with various temperature

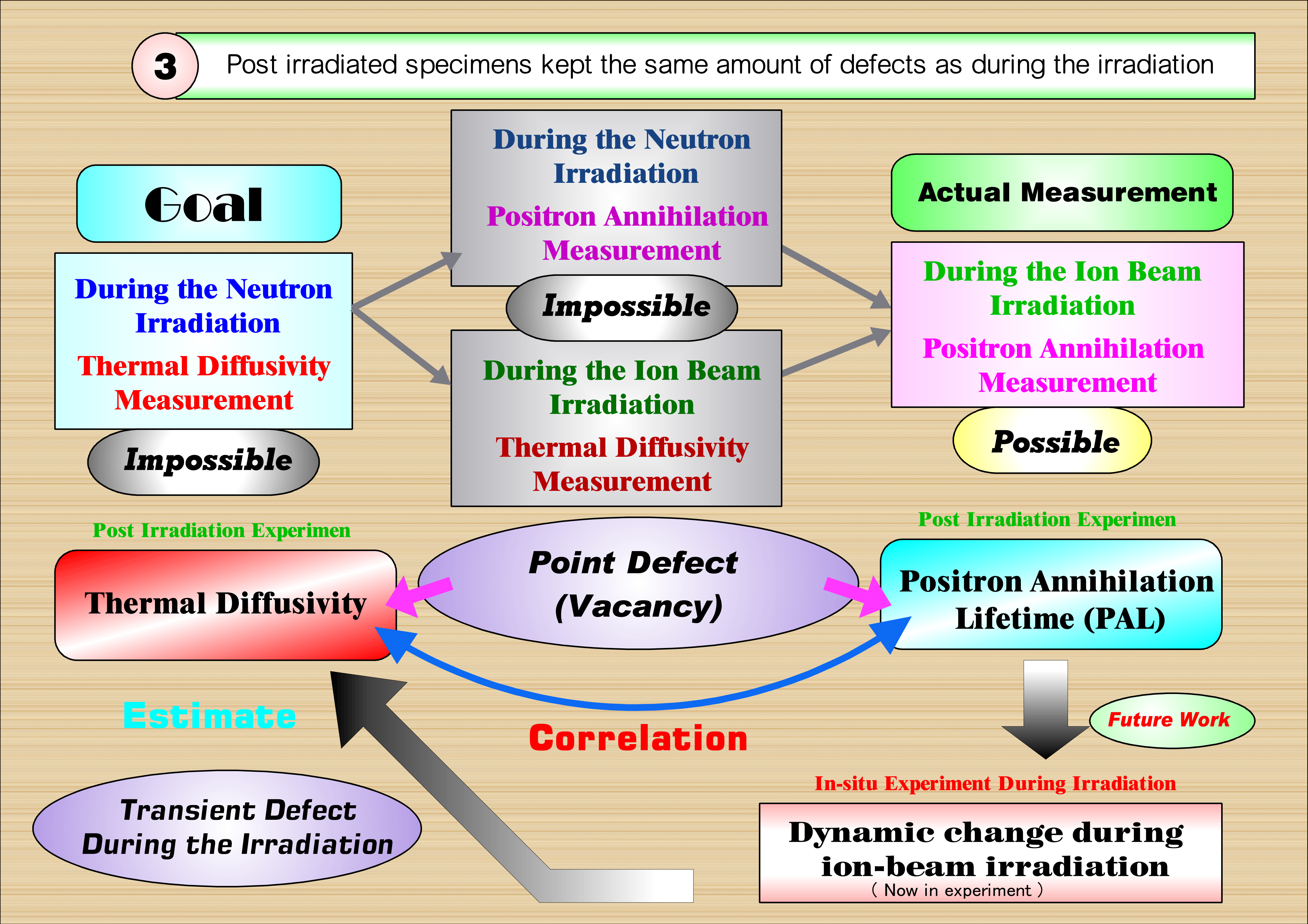
Post irradiation measurement at the irradiation temperature represent the thermal diffusivity during the irradiation.



Thermal Diffusivity Measured at Elevated Temperature



Thermal Diffusivity Measured below the irradiation temperature


3
Post irradiated specimens kept the same amount of defects as during the irradiation
Goal
During the Neutron Irradiation
Thermal Diffusivity Measurement
Impossible
During the Neutron Irradiation
Positron Annihilation Measurement
Impossible
During the Ion Beam Irradiation
Thermal Diffusivity Measurement
Actual Measurement
During the Ion Beam Irradiation
Positron Annihilation Measurement
Possible
Post Irradiation Experimen
Thermal Diffusivity
Point Defect (Vacancy)
Post Irradiation Experimen
Positron Annihilation Lifetime (PAL)
Estimate
Correlation
Future Work
Transient Defect During the Irradiation
In-situ Experiment During Irradiation
Dynamic change during ion-beam irradiation
(Now in experiment)

Experimental

Thermal Diffusivity Measurement

Thermal diffusivity was measured by **laser flash method** (ULVAC TC-7000L Special) and analyzed with $t_{1/2}$ method at room temperature.

Specimen Properties

	α-Al_2O_3	AlN	β-Si_3N_4	β-SiC
Manufacturer	Nippon Steel	Tokuyama	Nippon Steel	Nippon Steel
Raw material	α-Al_2O_3 100wt%	AlN (unknown)	β-Si_3N_4 > 88wt% Y_2O_3 < 10wt% $ZrSi_2$ < 2wt%	β-SiC > 99wt% Al_2O_3 < 1wt%
Sintering method	pressureless	pressureless	hot press	hot press
Relative density	99.5%	~99.9%	100%	99.7%
Thermal Diffusivity ($10^{-4}m^2/s$)	0.118	0.991	0.257	0.41

Positron Annihiration Measurement

Positron annihilation lifetime (PAL) measurement: conventional γ-γ fast coincidence method.

Neutron Irradiation

Experimental Fast Reactor

JOYO

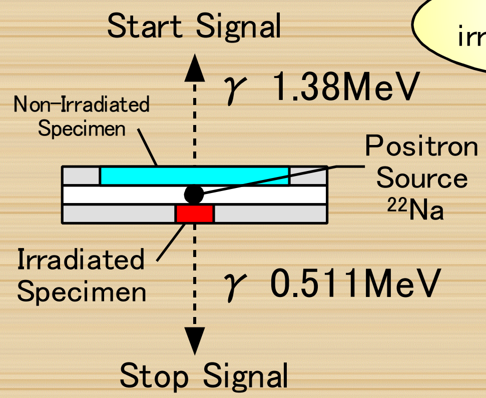
Mk II core, irradiation rig CMIR-4

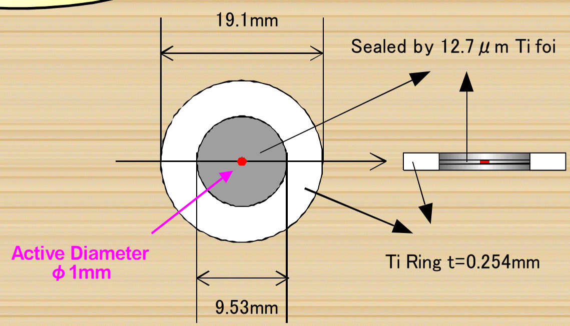
4 kind of specimens were irradiated in the same capsule

One disk each irradiation condition

Irradiation Condition

ID	Dose (n/m^2) (E >0.1MeV)	Irradiation Temperature (K)
3mmϕ × 0.5mm disk		
T51	2.8×10^{26}	775
T53	3.9×10^{26}	864
T55	4.2×10^{26}	1004
T57	3.7×10^{26}	1011



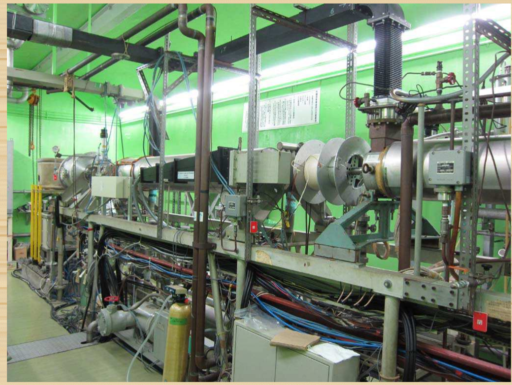


30MeV Electron Irradiation

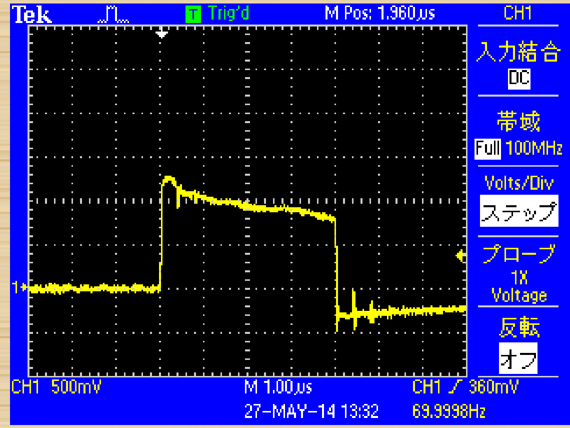
30MeV Electron Accelerator

KURRI-LINAC

Kyoto University
Research Reactor Institute, Kumatori



Accelerate Energy: 28-32MeV
The beam current: ～230 μ A
Peak current of pulse: ～600mA
Pulse width: 4 μ s
Pulse frequency: ～100Hz
Heat Load: ～7kW in 2cm^2 = 35MW/m^2
> the divertor in ITER, 10MW/m^2



The Range of 30MeV Electron → ～5cm (in ceramics)

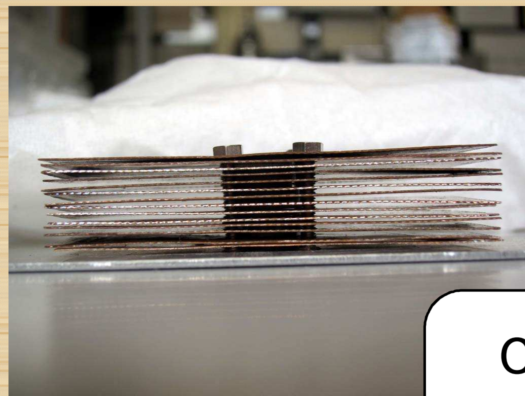
Energy of PKA (max): 125keV → Energy of PKA (average): 225eV
Number of Displacement Atoms per PKA: 3-4 atoms
→ Mainly Point Defects

Low Temperature Irradiation

KURL0604

Liquid Nitrogen Supply

KURL0604: −165℃ / 1.35kW
Liquid Nitrogen cooled system
30MeV, 3 μs x 500mA x 30Hz



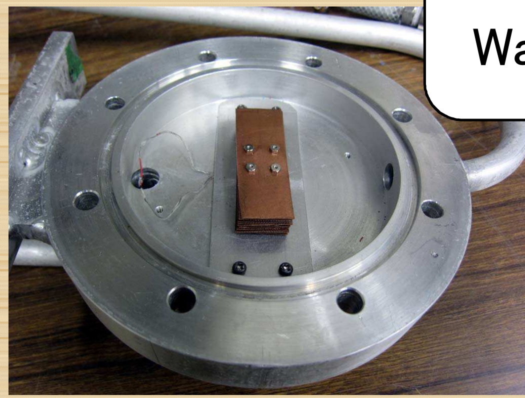
KURL1201
KURL1401

Cu heat sink in Water chamber

KURL1201: 80℃ / 6.0kW
32MeV, 4 μs x 590mA x 80Hz

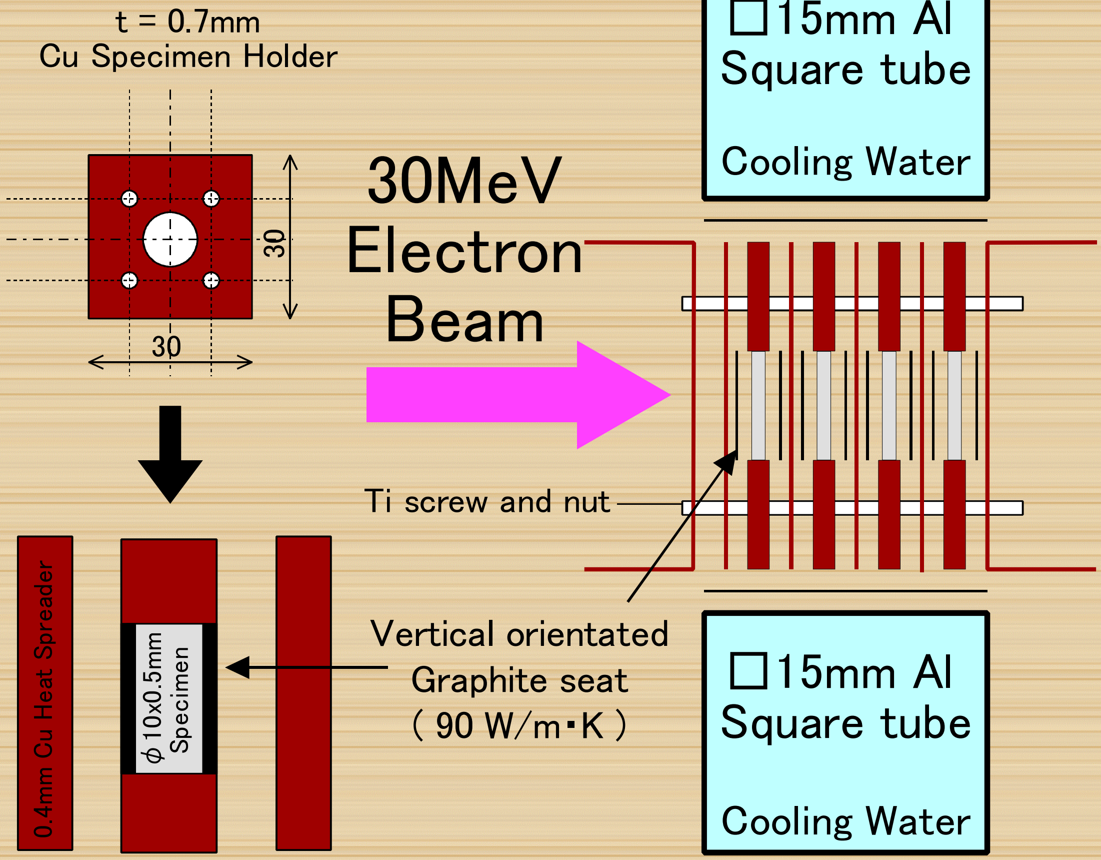
KURL1401: 90℃ / 4.7kW
28MeV, 4 μs x 600mA x 70Hz

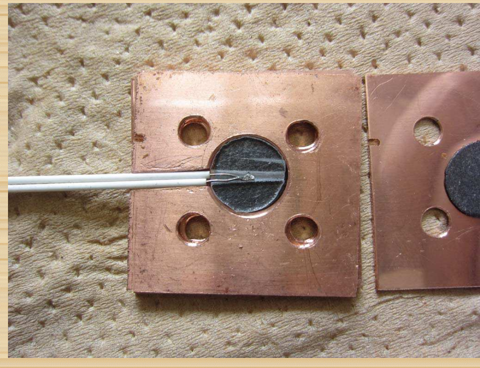
Cu Heat sink put in Water chamber

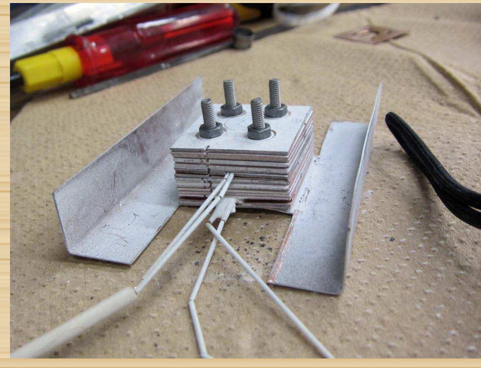


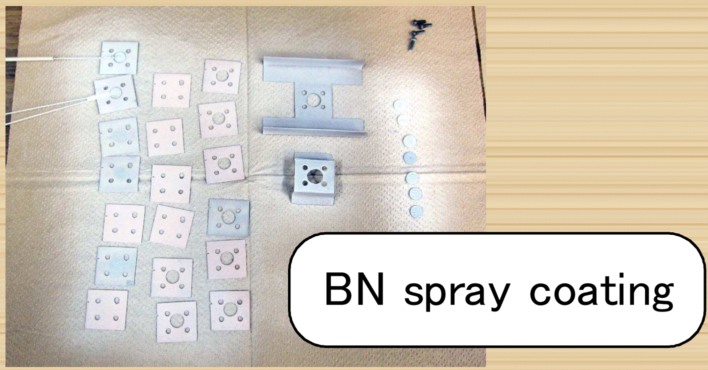
Very Stable and Established Irradiation System

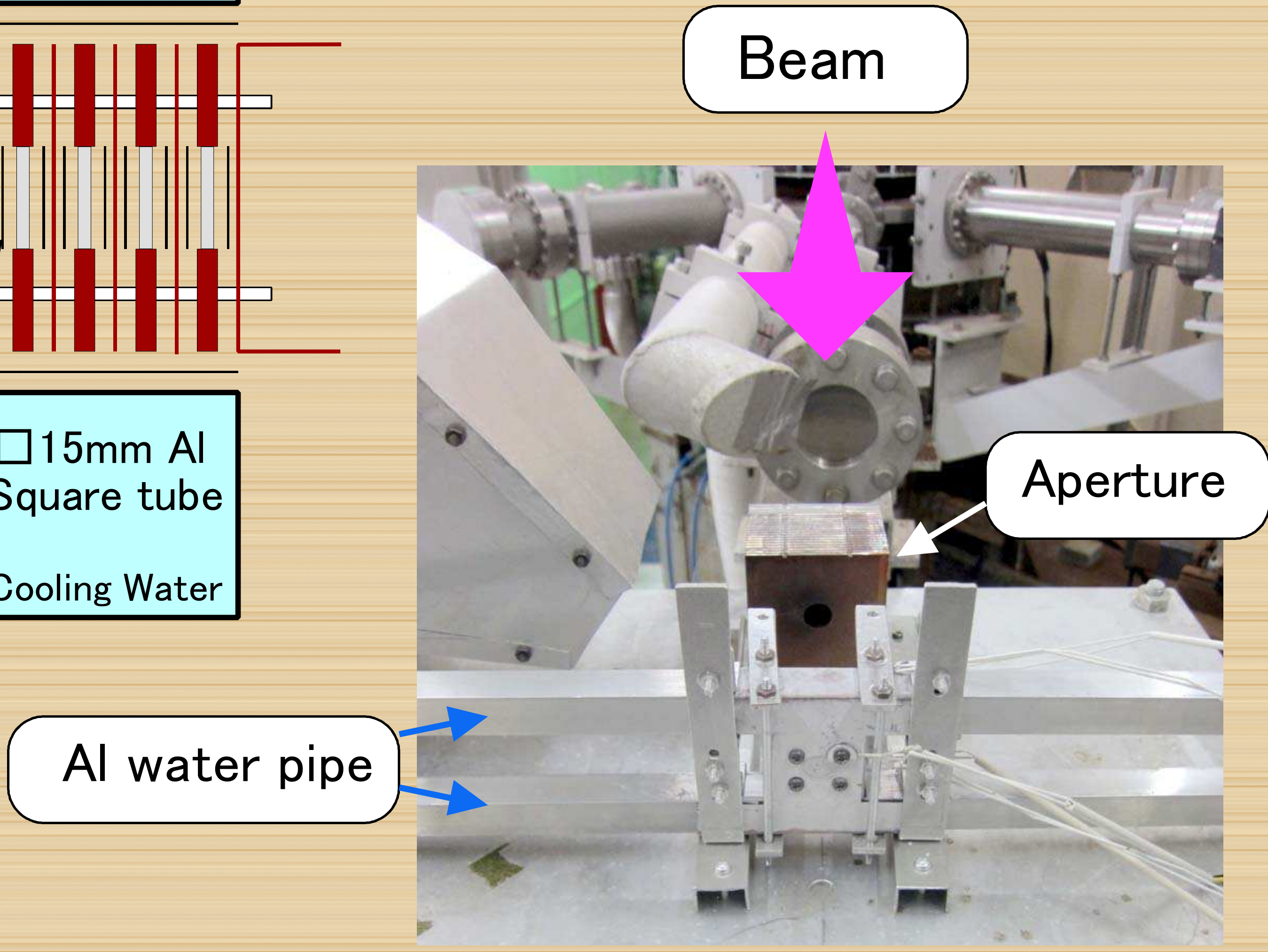
Medium Temperature Irradiation







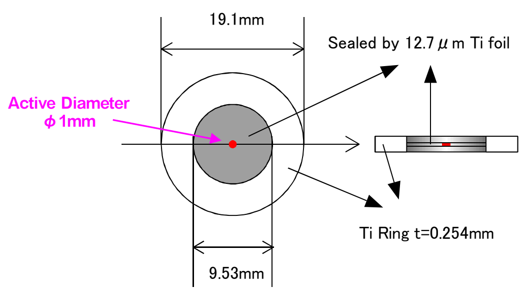


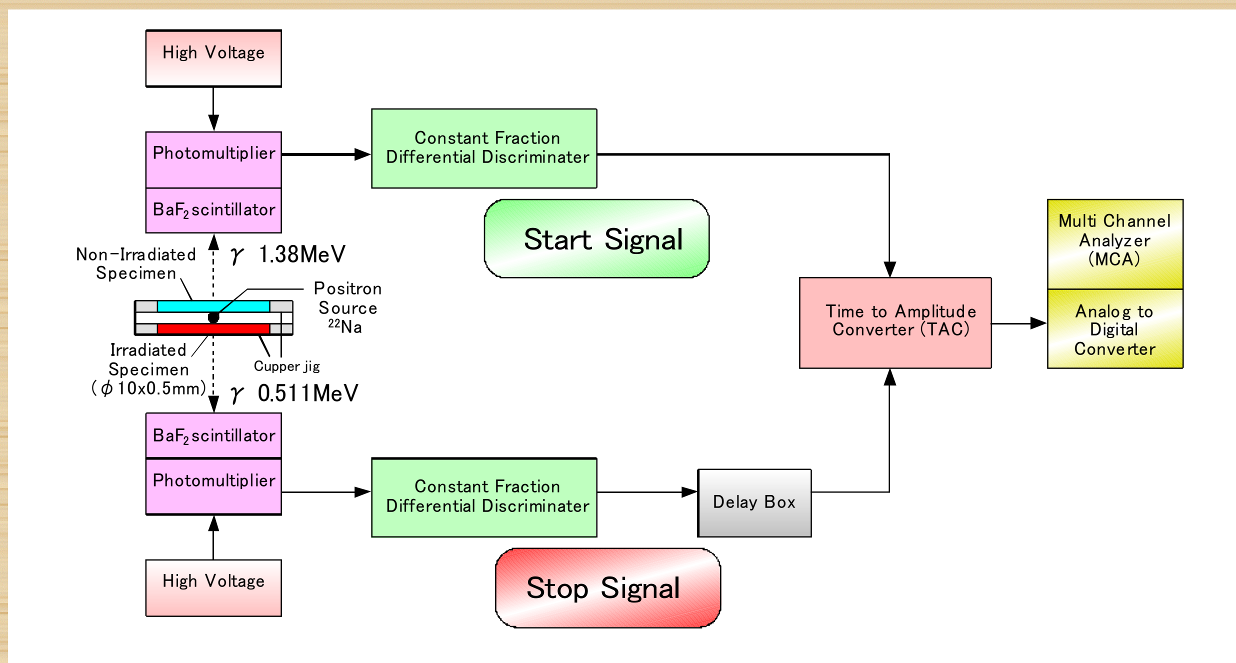


PAL and Thermal Diffusivity Measurement

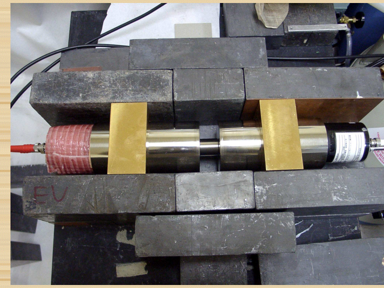
γ-γ Fast Coincidence Positron Annihilation Lifetime Measurement Circuit Diagram

Sealed Positron Source ^{22}Na < 1MBq

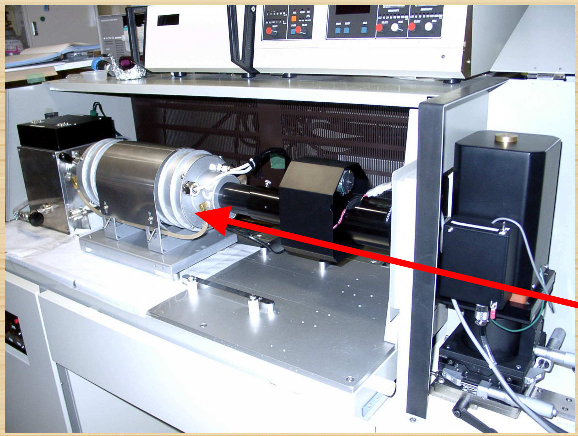




BaF2 Scintillator: OKEN φ30x40mm
Photomultiplier: H33778-51



Thermal Diffusivity Measurement System



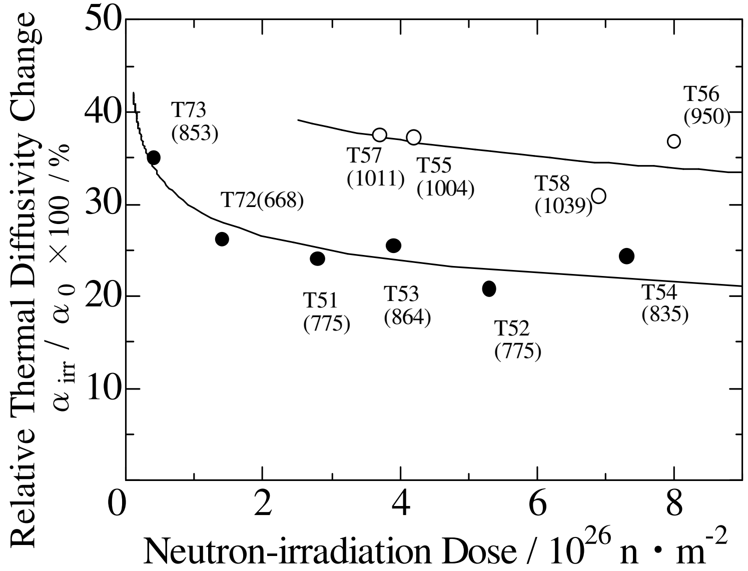
ULVAC Techno. TC-7000H/Special

Conventional laser-flash method / t1/2 analysis
Specially ordered system → φ3mm disk

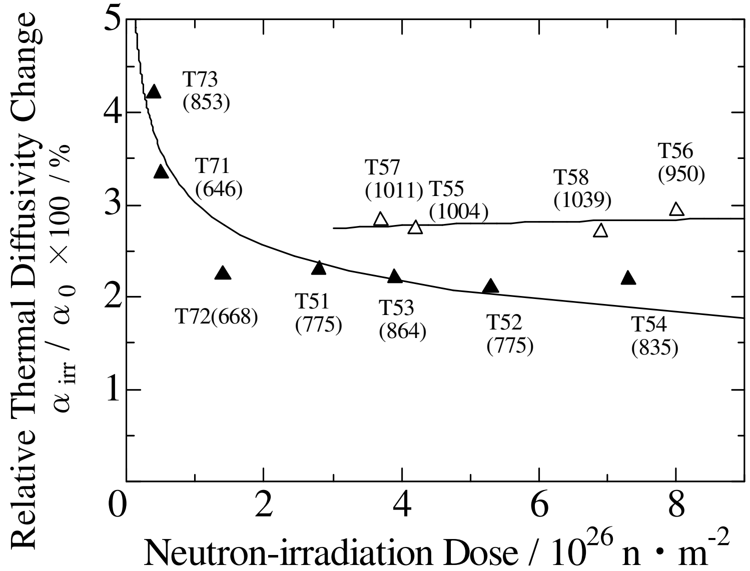
Isochronal annealings in a SiC furnace up to 1500℃ in vacuum

Degradation of thermal diffusivity after neutron irradiations

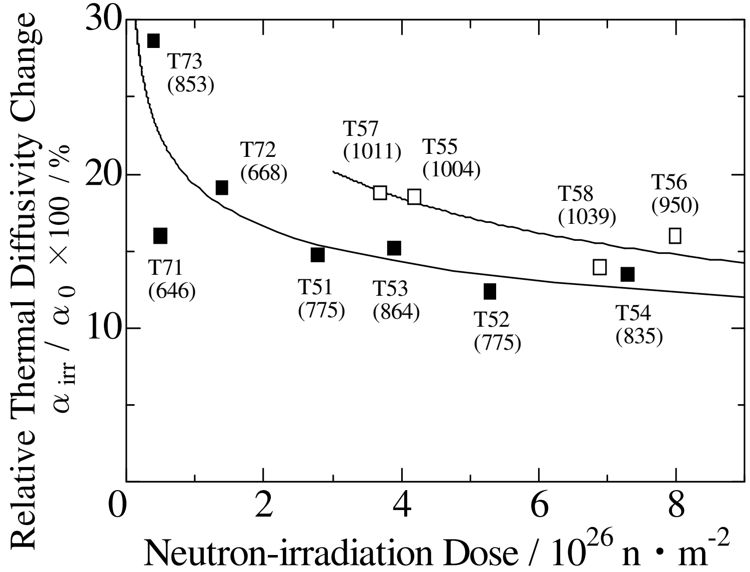
α-Al_2O_3



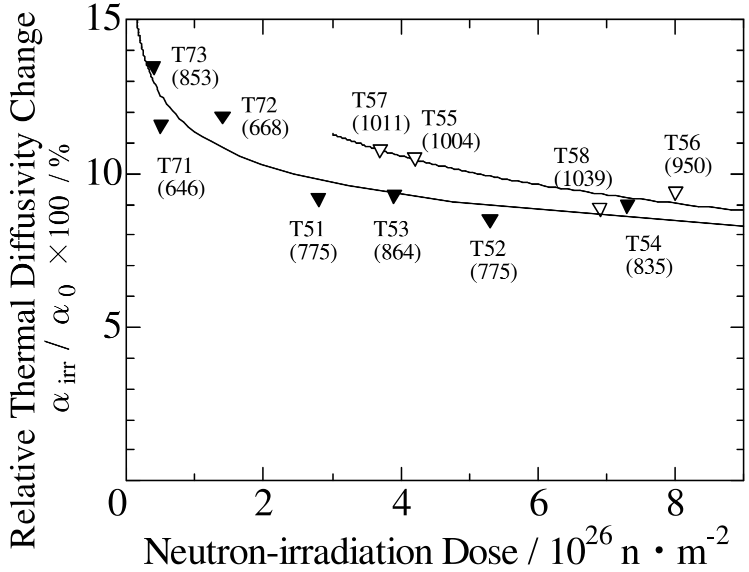
AlN



β-Si_3N_4

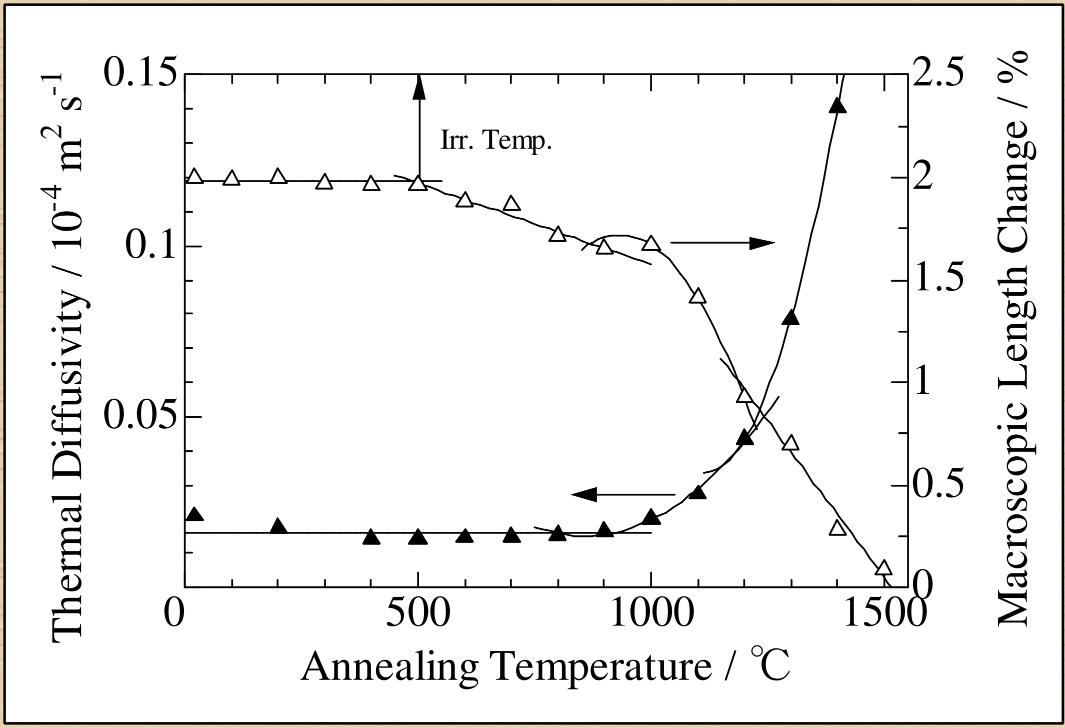


β-SiC

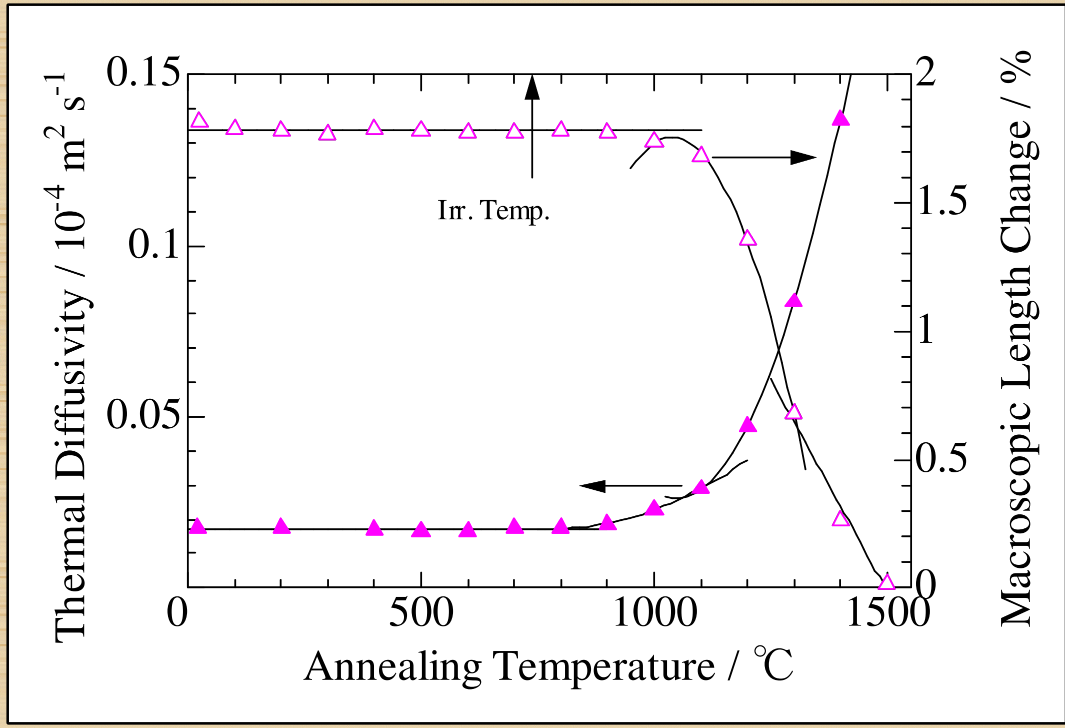


Annealing Behavior

AlN



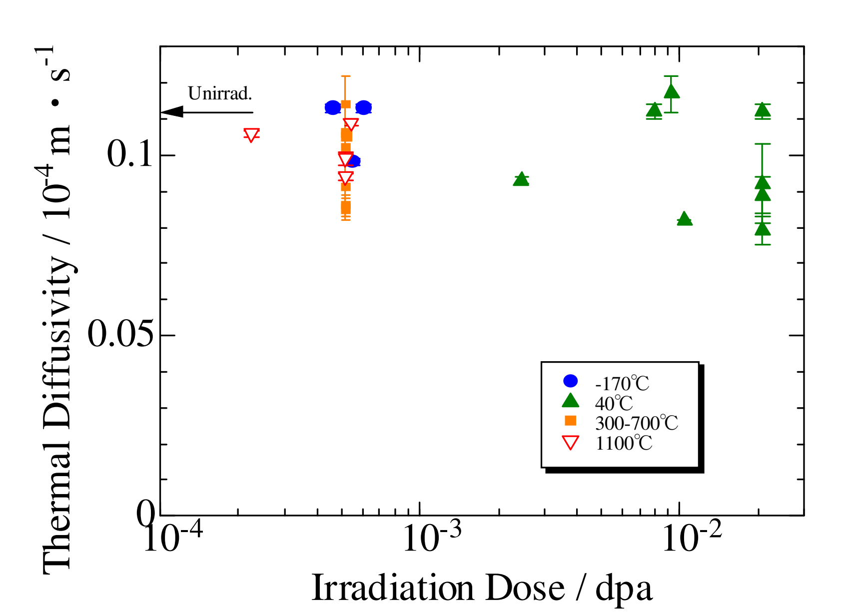
T51



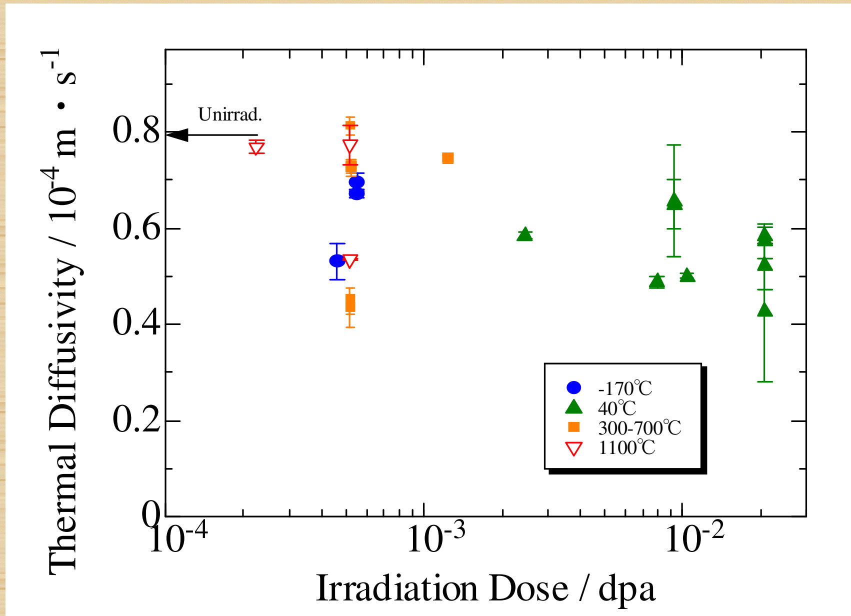
T57

Degradation of thermal diffusivity after electron irradiations

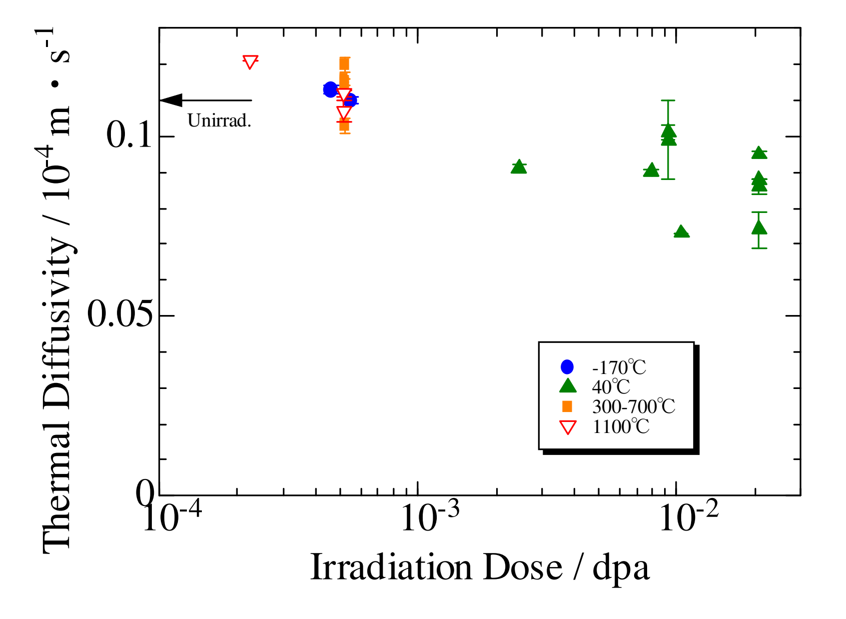
α-Al_2O_3



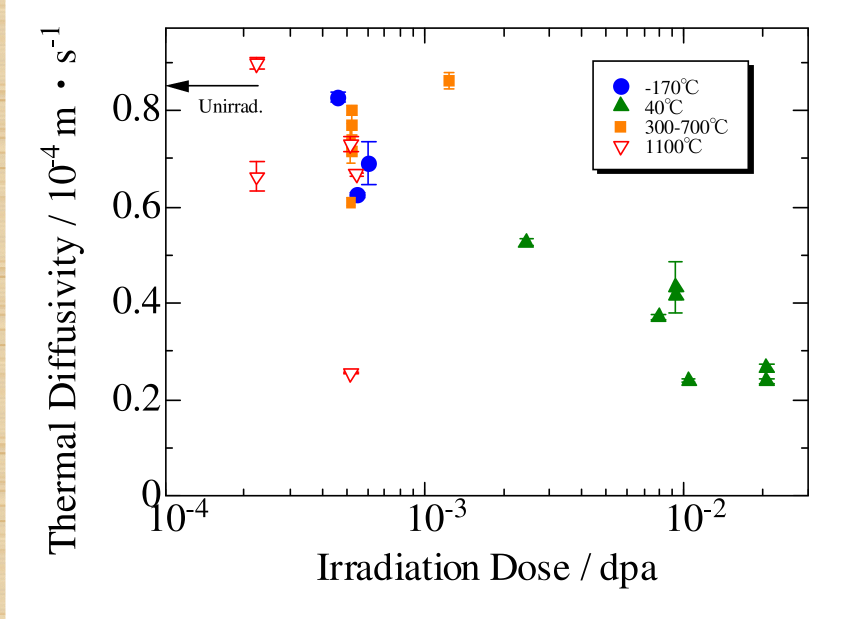
AlN



β-Si_3N_4



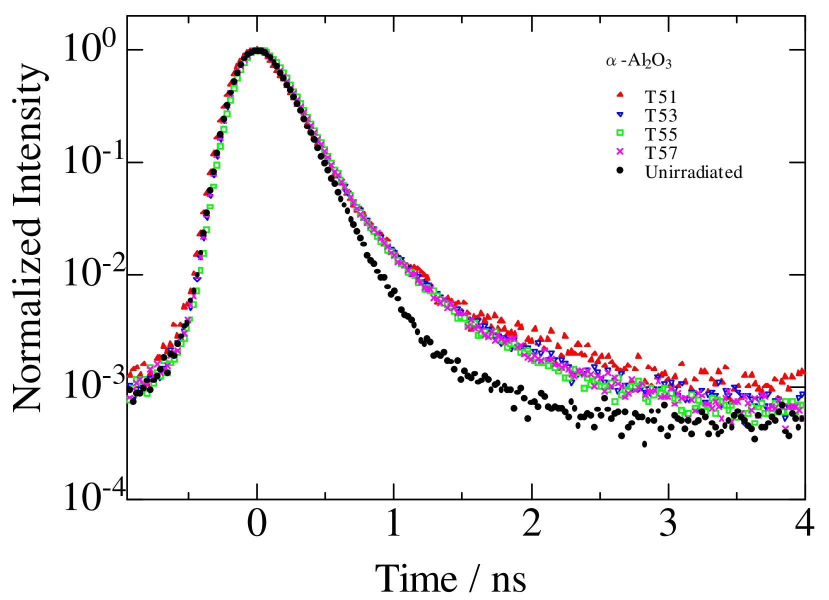
β-SiC



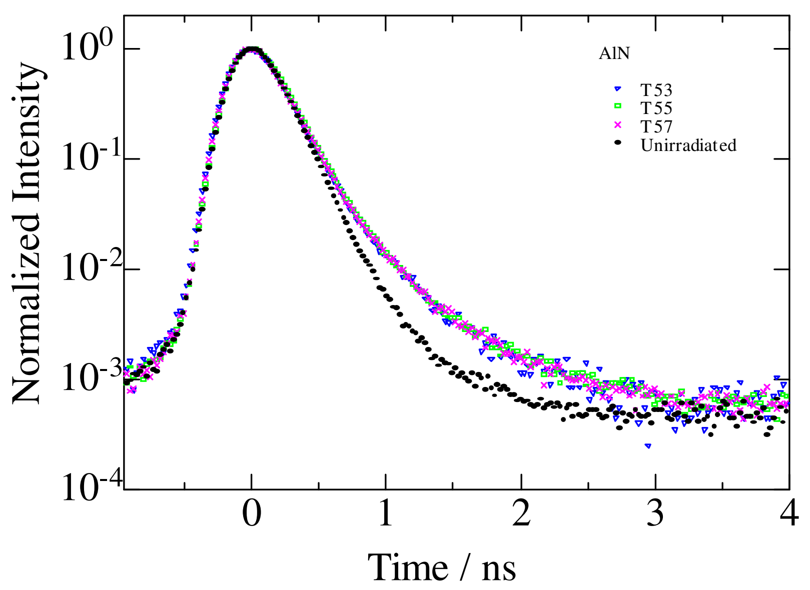
Result

Positron Annihilation Lifetime Spectrum

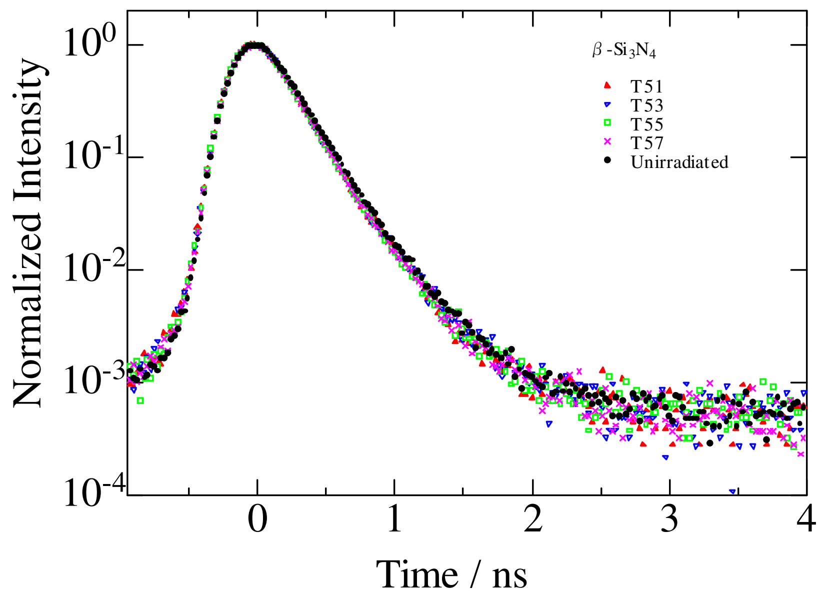
α-Al_2O_3



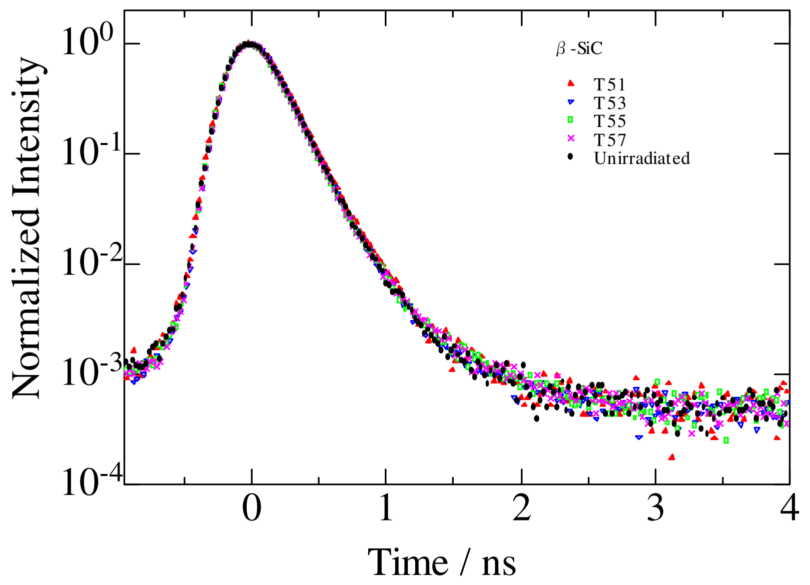
AlN



β-Si_3N_4



β-SiC

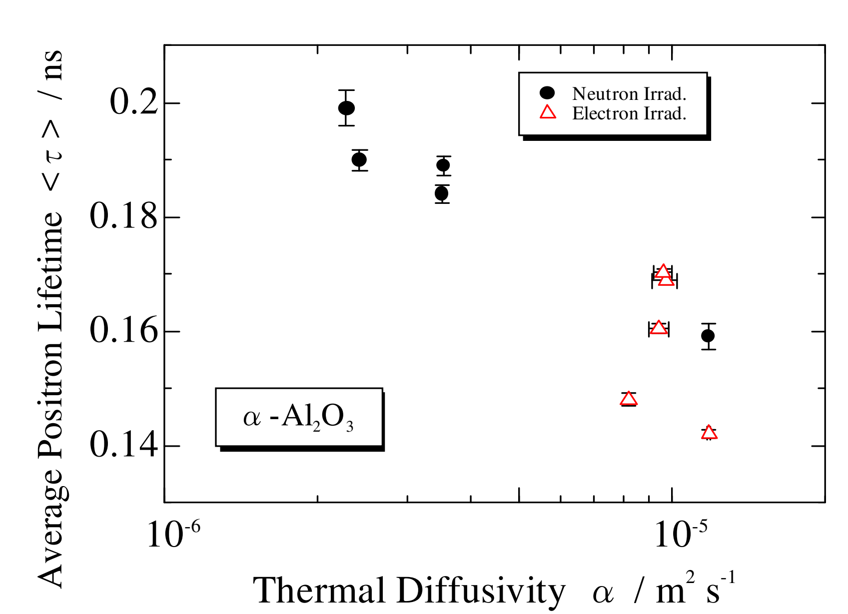


As-irradiated

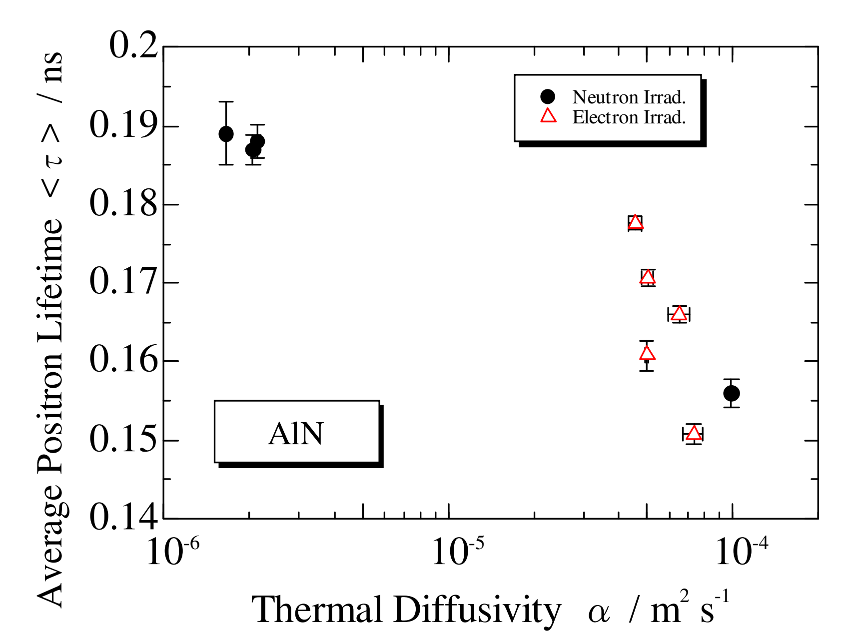
Averaged Positron Lifetime vs Thermal Diffusivity

Average lifetime $<\tau> = \tau_1 I_1 + \tau_2 I_2$
Here, τ_i are the analyzed positron lifetimes, and I_i are the intensities for the components.

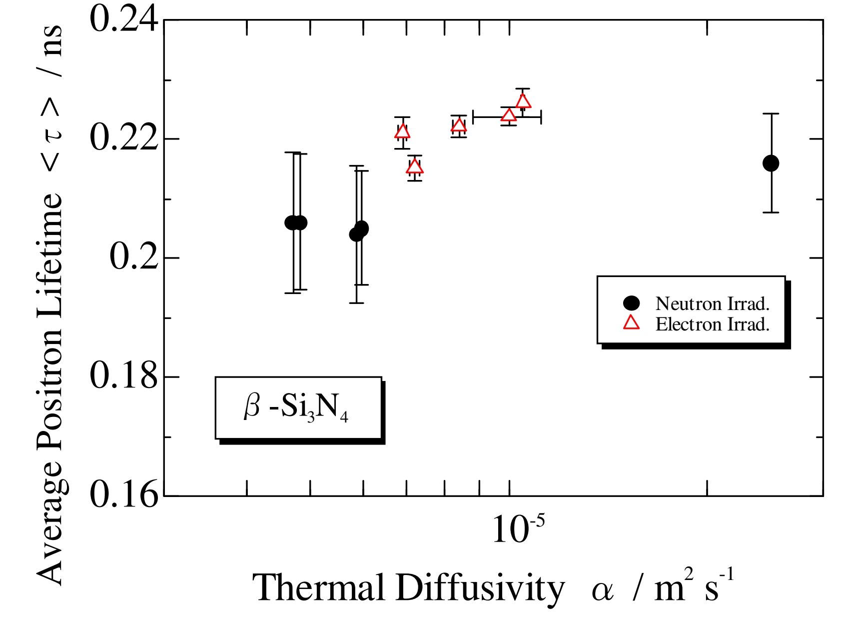
α-Al_2O_3



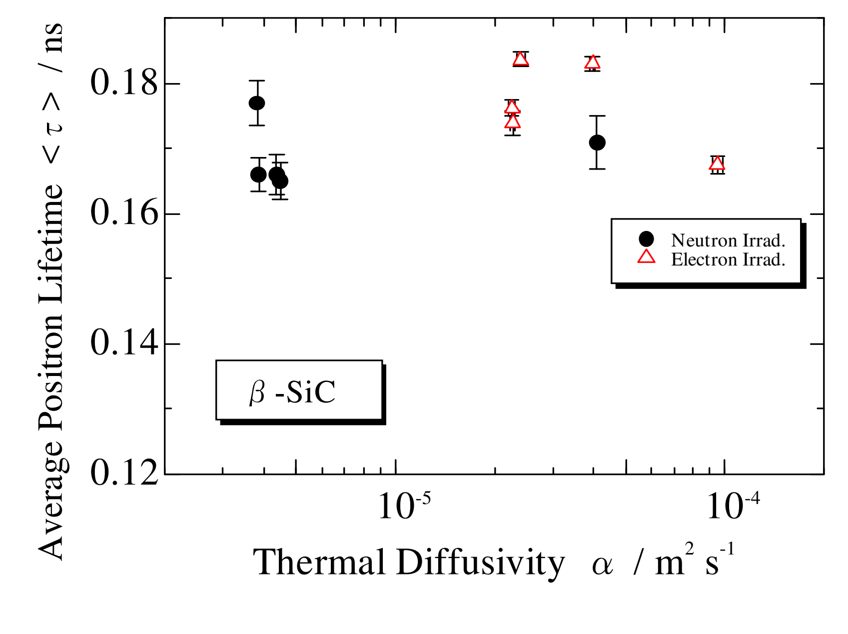
AlN



β-Si_3N_4

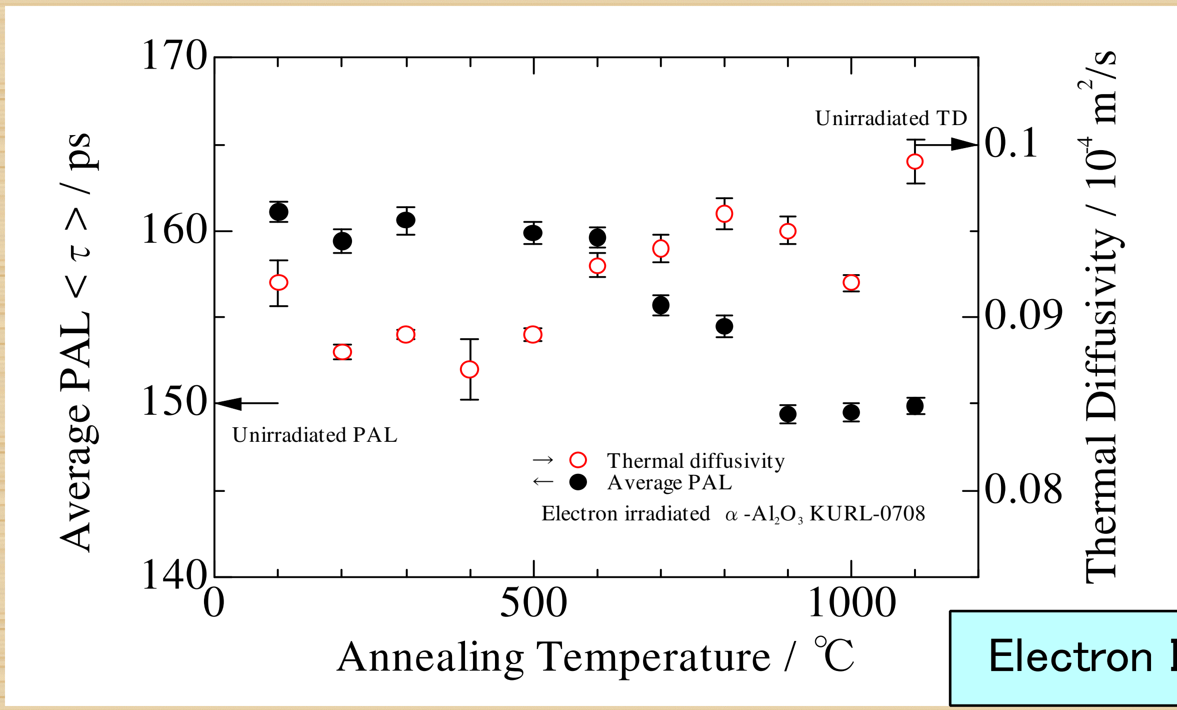


β-SiC



α-Al_2O_3

Annealing behavior

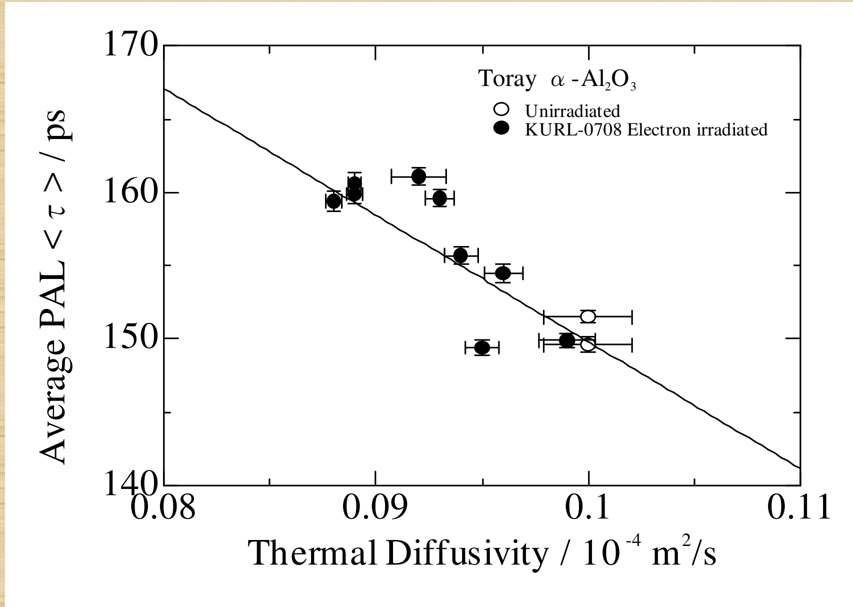


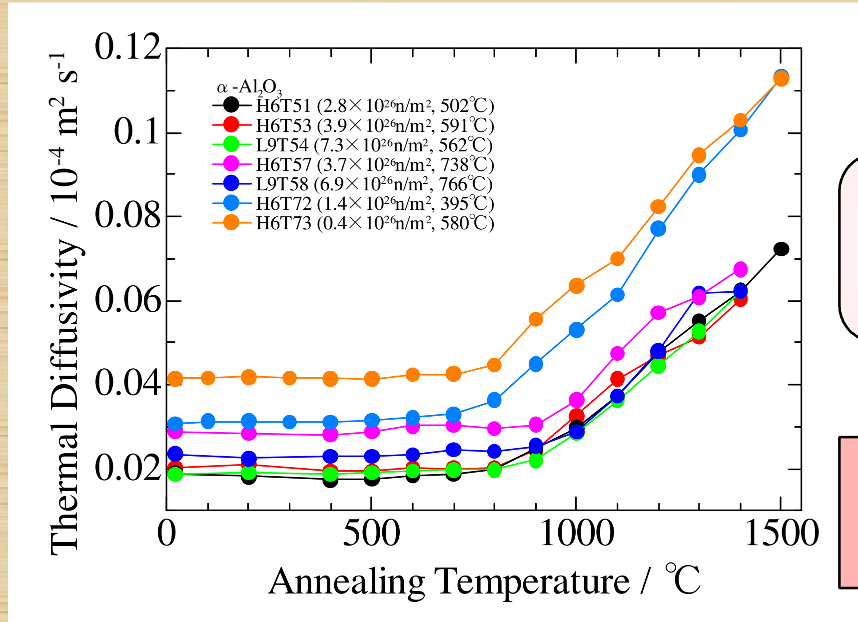
PAL: Recovered from 600℃ and completed at 900℃

Thermal diffusivity: Recovered from around 500℃ and almost completed at 1100℃

Correlation: Showed weak correlation arrose from small change of thermal diffusivity.

Electron Irradiated



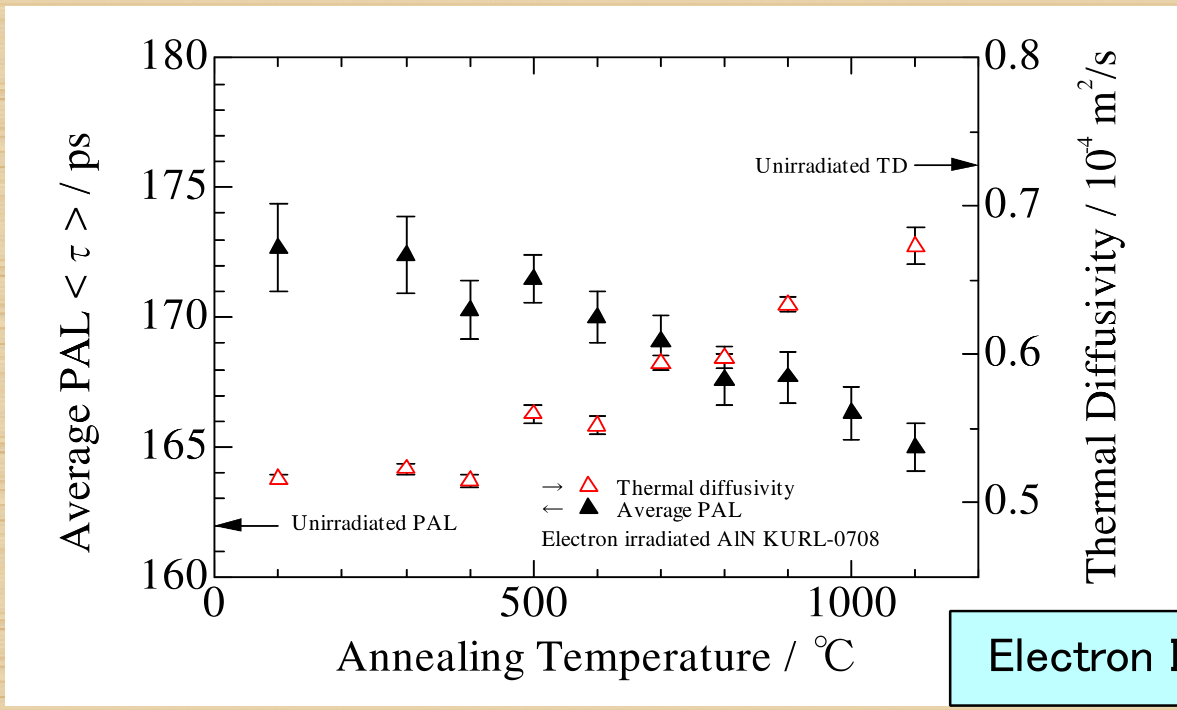


Recovered from 700–800℃ linearly

Neutron Irradiated

AlN

Annealing behavior

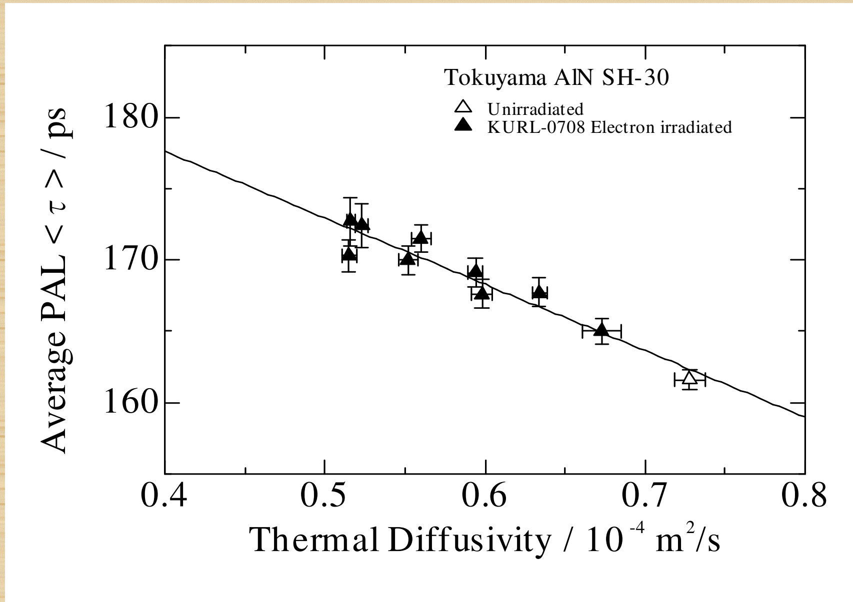


PAL: Recovered from 400℃ linearly and completed at 1100℃

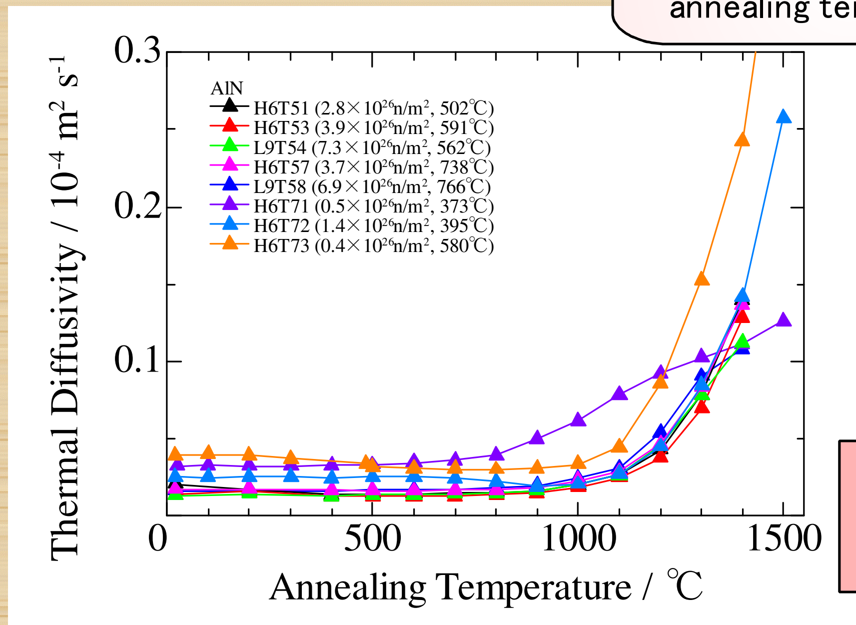
Thermal diffusivity: Same trend as the PAL result.

Correlation: Showed very good correlation.

Electron Irradiated



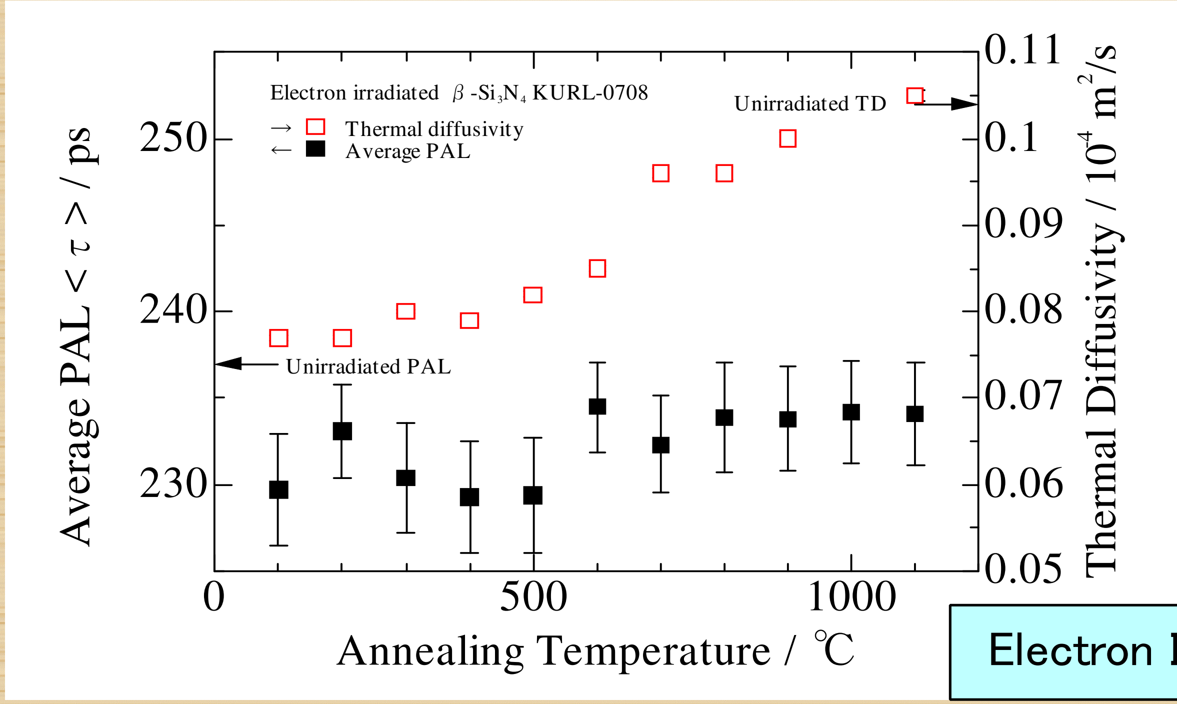
Recovered from 1100℃ as the square of the annealing temperature



Neutron Irradiated

β-Si_3N_4

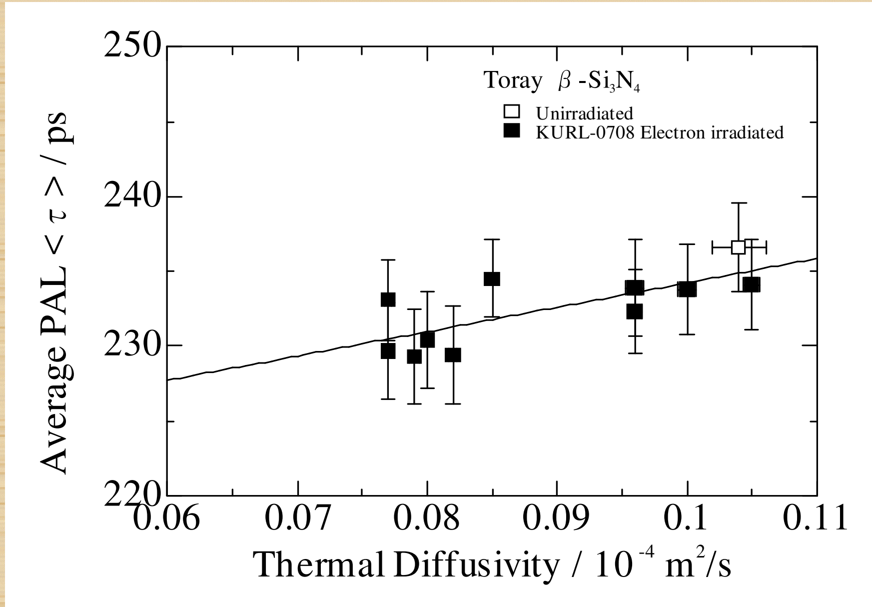
Annealing behavior

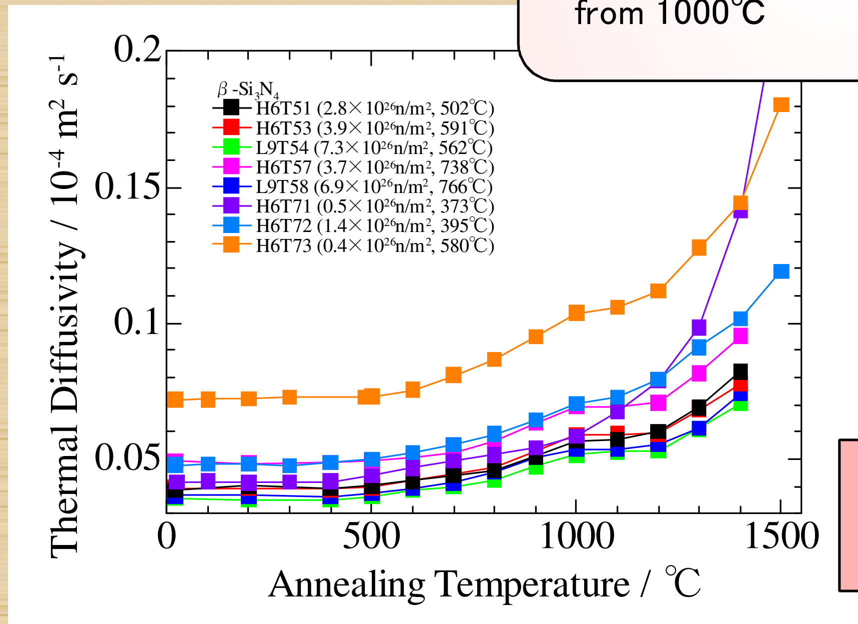


PAL: As-irradiated specimen showed slight decrease, and it recovered at 600℃ completely.
Thermal diffusivity: Recovered from 400℃ linearly and completed at 1100℃
Correlation: PAL showed inverse change as the other specimens, then showed a little positive correlation to the thermal diffusivity

Electron Irradiated

Recovered from 400℃ linearly, and then recovered as the square of the annealing temperature from 1000℃

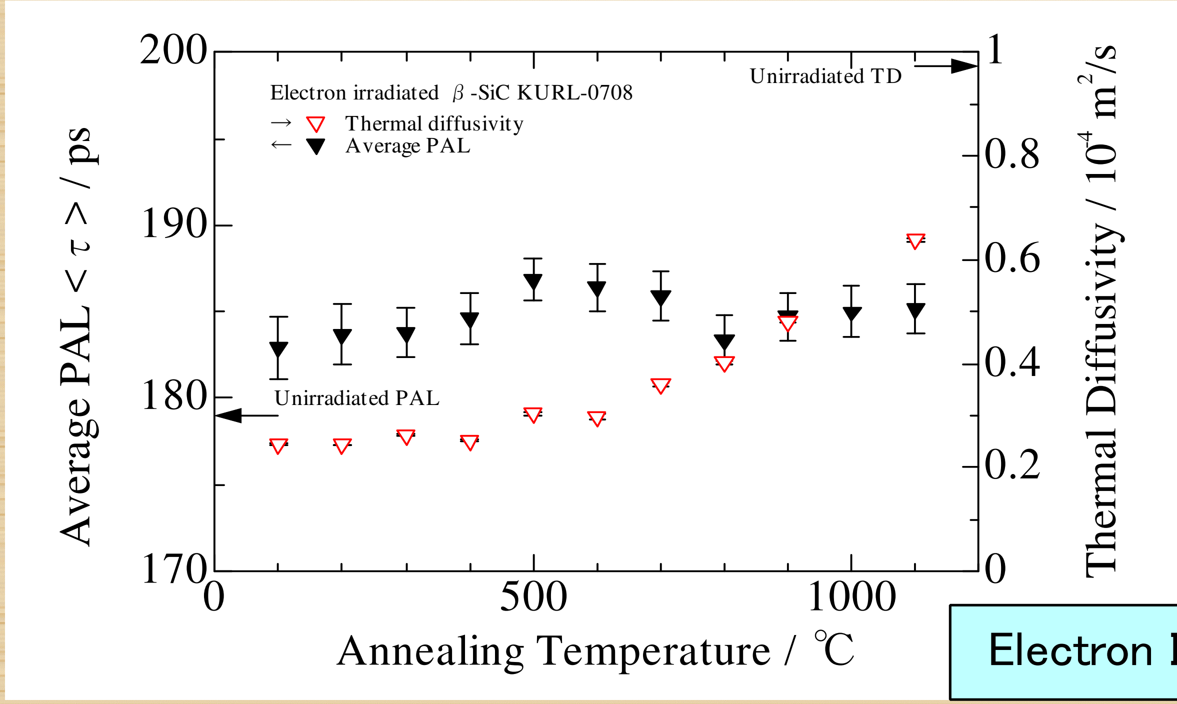




Neutron Irradiated

β-SiC

Annealing behavior



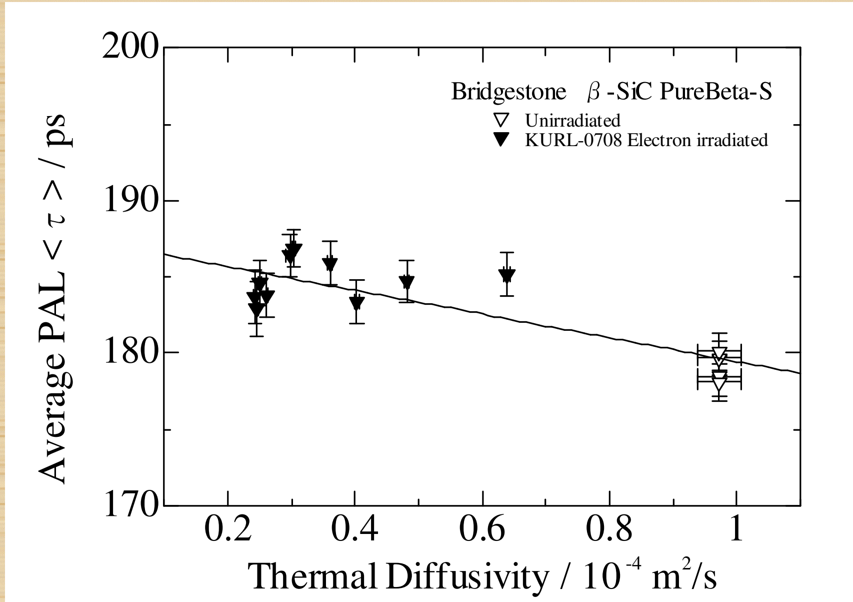
PAL: As-irradiated specimen showed a little increase, while it did not show recovery. Furthermore, a little increase was observed arround 500℃ to 800℃

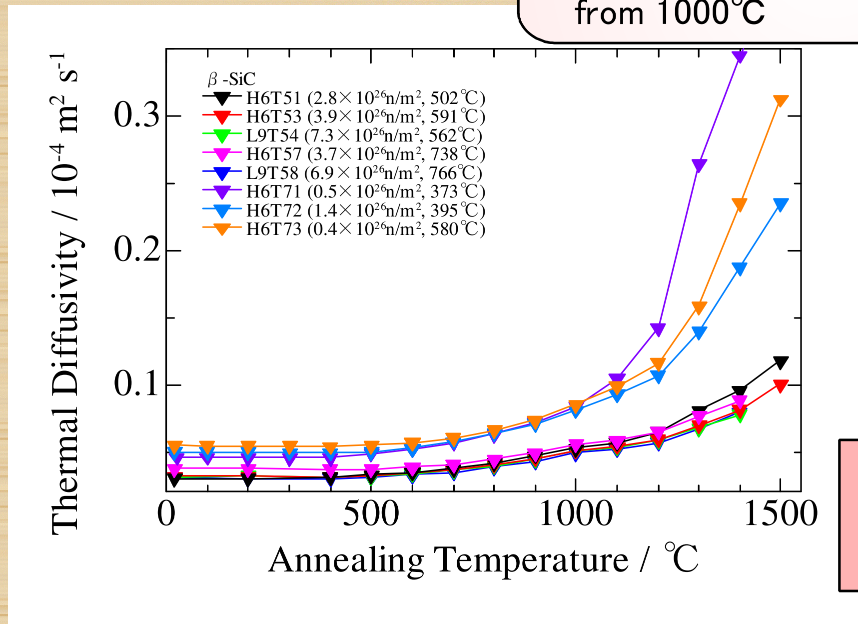
Thermal diffusivity: Recovered from 400℃ but did not completed at 1100℃.

Correlation: Obvious correlation was not observed

Electron Irradiated

Recovered from 400℃ linearly, and then recovered as the square of the annealing temperature from 1000℃

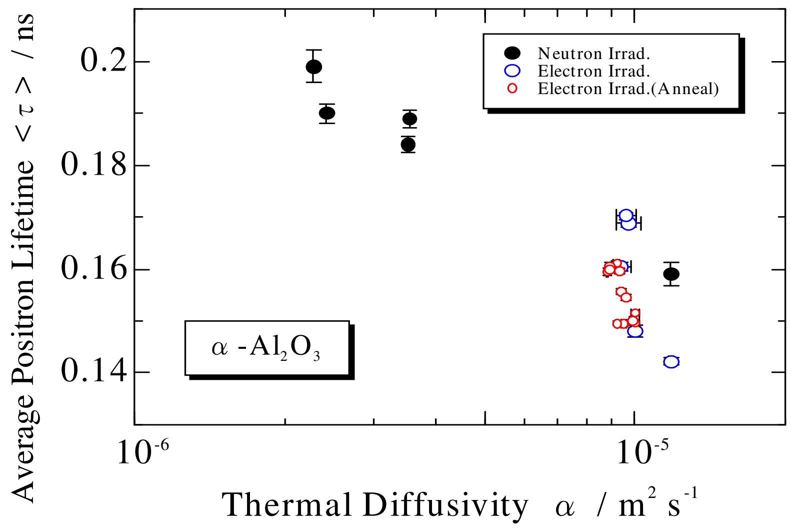




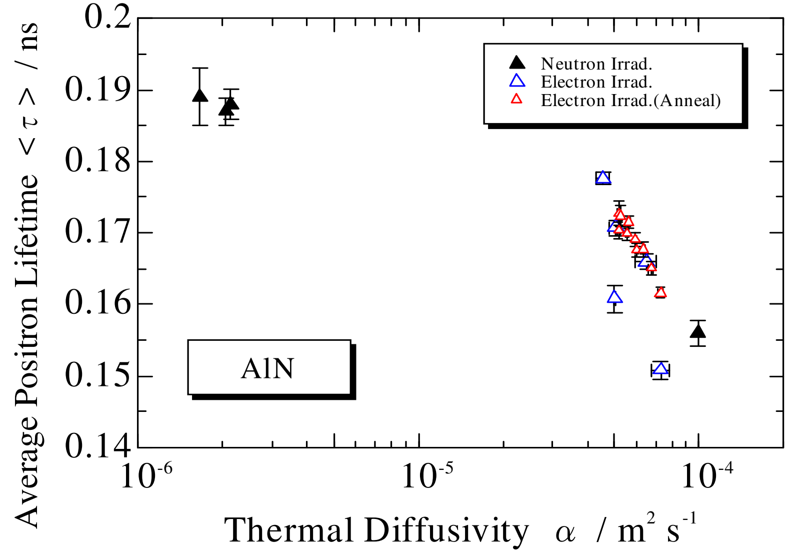
Neutron Irradiated

As-irradiated and annealed

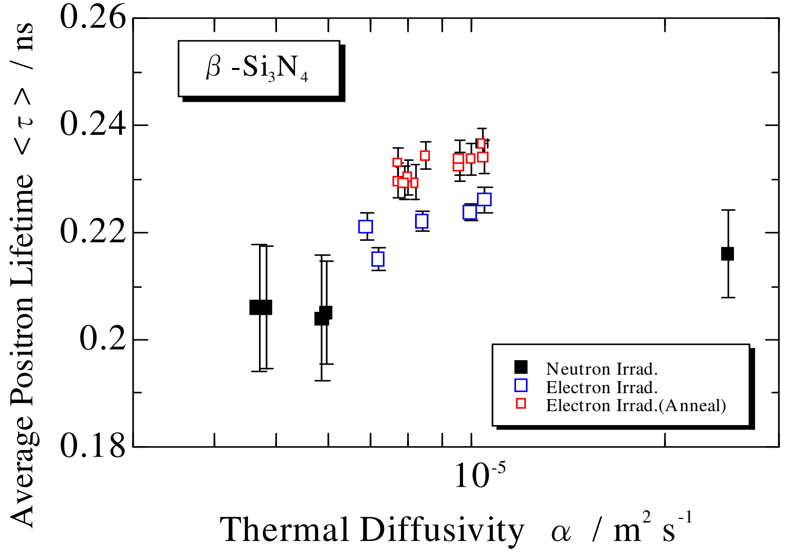
α-Al_2O_3



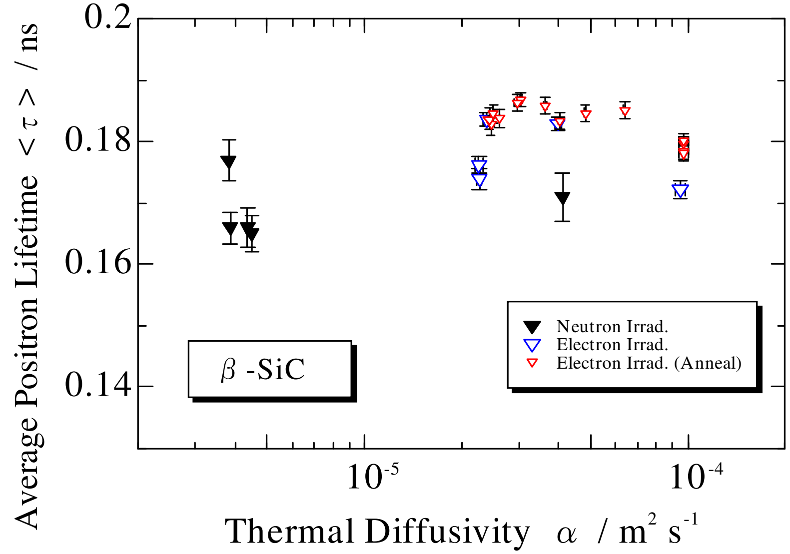
AlN



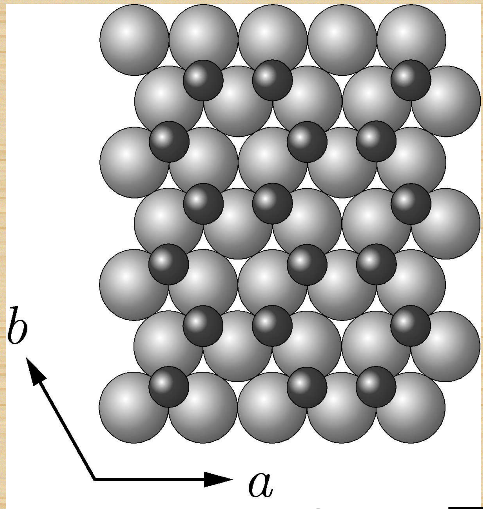
β-Si_3N_4



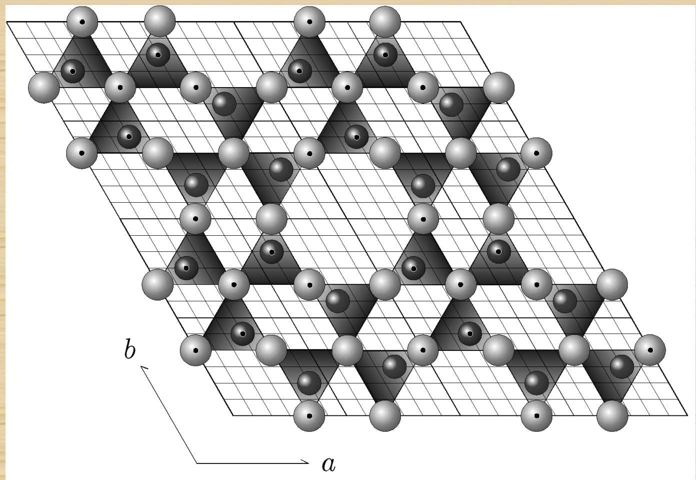
β-SiC



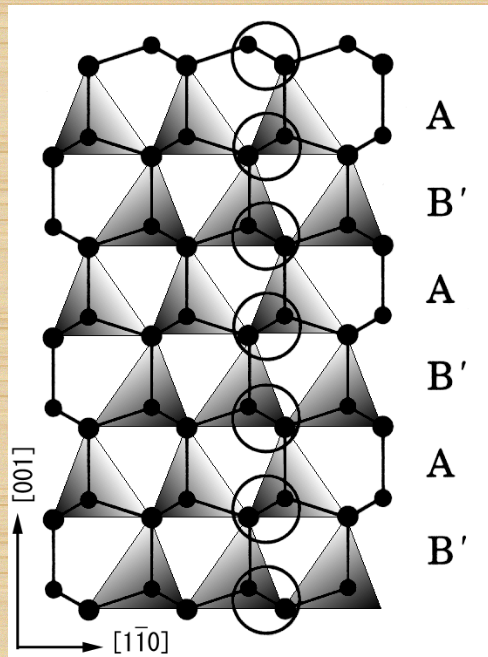
Structure of each material



α-Al_2O_3 の(001)面。
最密面 : (001)面

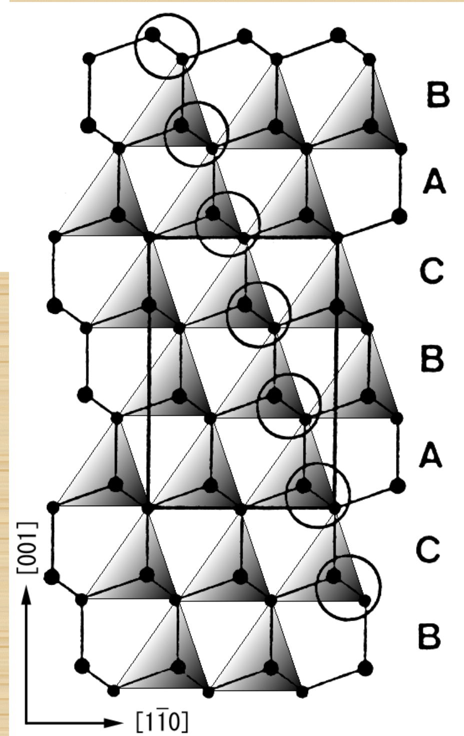


β-Si_3N_4 の(001)面。最密面 : {100}面



AlN の(110)面。
最密面 : (001)面

β-SiCの(110)面。
最密面 : {111}面



Conclusion

Positron annihilation lifetime and thermal diffusivity of electron and heavily neutron irradiated α-Al_2O_3, AlN, β-Si_3N_4 and β-SiC were measured and compared.

○ α-Al_2O_3 and AlN →(Irradiation)→ obvious change in PAL
(even in electron irradiated (0.01dpa) specimen)

× β-Si_3N_4 and β-SiC →(Irradiation)→ no change in PAL
(even in heavily neutron irradiated (28-42dpa) specimen)

Neutron Irradiation → too large degradation in thermal diffusivity (Saturated)
30MeV electron irradiation up to 0.01-0.02 dpa → thermal diffusivity in wide range

α-Al_2O_3 and AlN showed good correlation between thermal diffusivity and PAL.

△ α-Al_2O_3 : Not high thermal diffusivity (10mm^2/s)

◎ AlN: Very high thermal diffusivity (100mm^2/s)
→ easy to evaluate the difference after an irradiation

× β-Si_3N_4: Negative small PAL change arised from the crystal structure

× β-SiC: Very high thermal diffusivity (100mm^2/s)
× p-type: No PAL change → n-type: ?