Microstructural Analysis of Nuclear-Grade Graphite Materials after Neutron Irradiation

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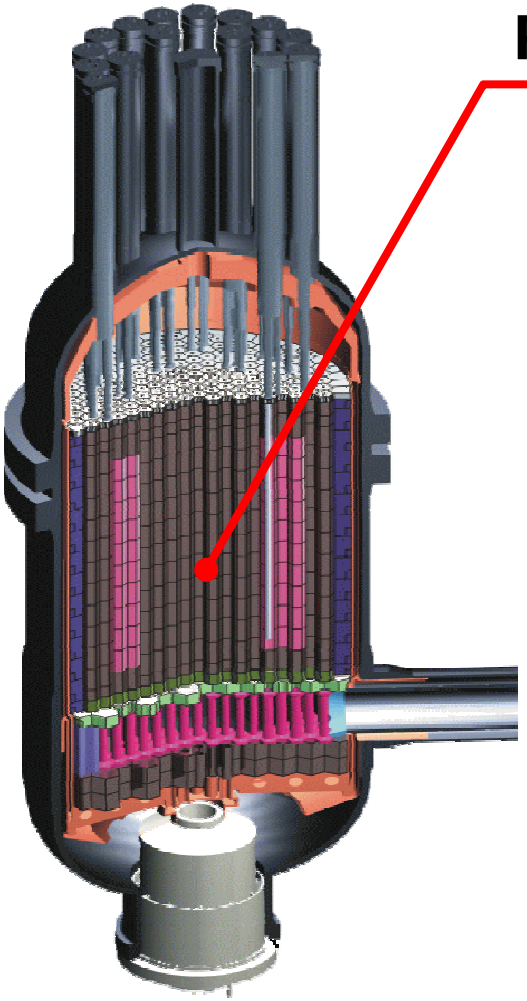
Graphite for VHTR



VHTR

Fine grained isotropic graphite

Used as the core structural material of a Very High Temperature gas-cooled Reactor which is one of the candidate next generation reactors

■ **VHTR has inherent safety features**

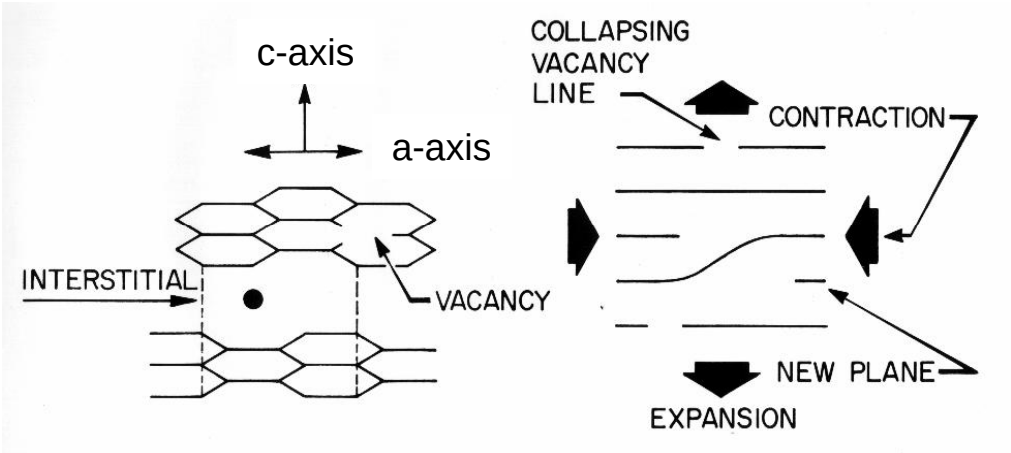
- ✓ No meltdown
- ✓ Slow temperature transient
- ✓ No chemical reaction

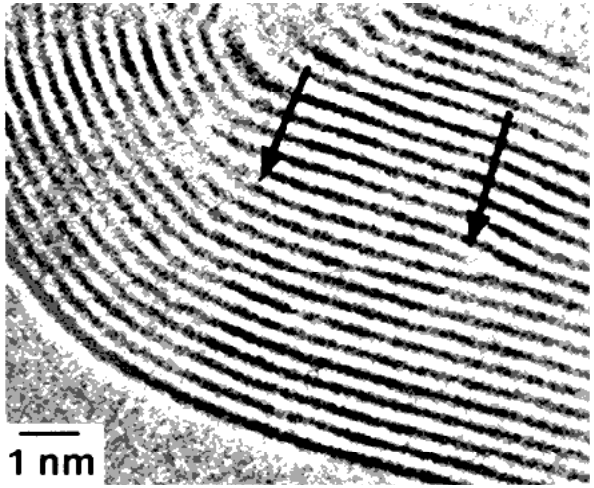
■ **Outlet gas temperature : 750-1000℃**

High process heat can be utilized as a heat source for variety of application

Neutron Irradiation Damage on Graphite

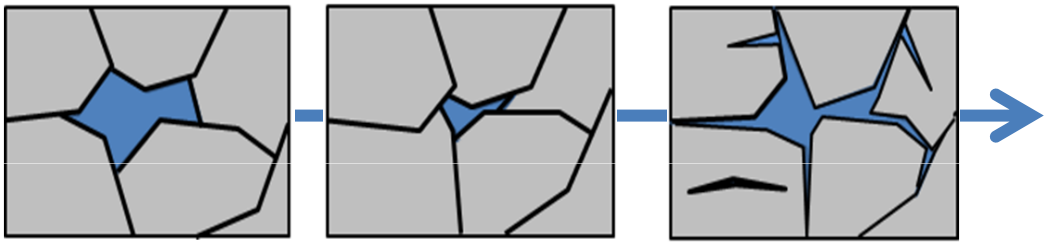
Microscopic changes

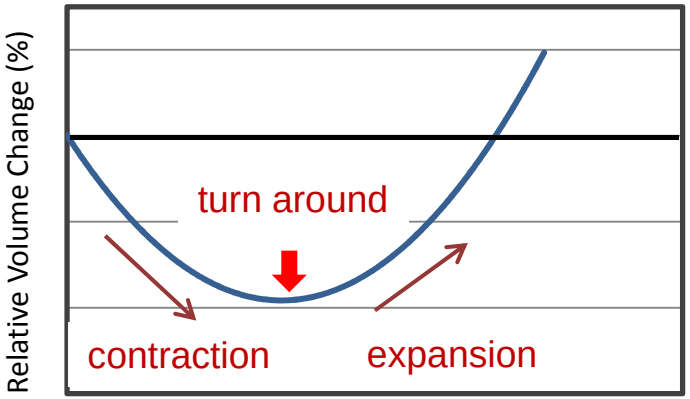




Banhart, F. *Rep. Prog. Phys.* **1999**, 62, 1181–1221.

Macroscopic changes





Fast Neutron Fluence (x 10^{26} n/m^2, E > 0.1 MeV)

Contract at first stage then expand
- pores accommodate expansion in c-axis

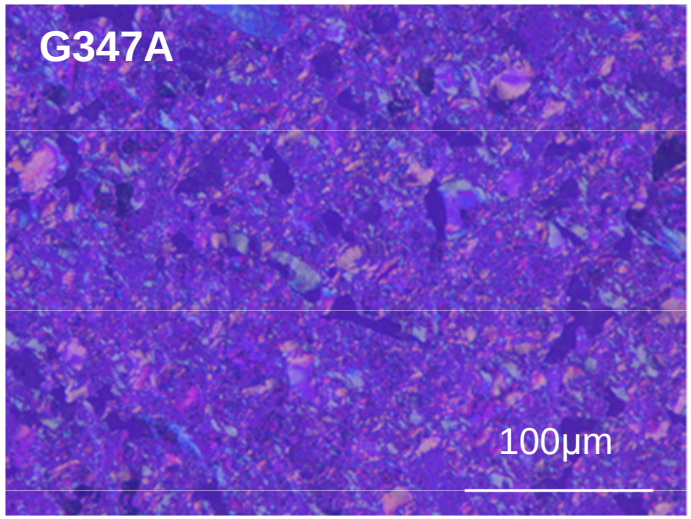
Objective

Graphite materials in nuclear services undergo very significant modifications in various thermal and mechanical properties resulting from the irradiation-induced microstructural changes.

It is essential to characterize the microstructures before and after irradiation and correlate the structural changes with the evolving macroscopic properties

To characterize the microstructural changes of isotropic fine-grained graphite following the neutron irradiation to help develop graphite for improved radiation service life

Material Information



Nuclear grade graphite **G347A** manufactured by Tokai Carbon is used for microstructural analysis

G347A is fine-grained, high strength, isotropic graphite

Typical properties of G347A and ASTM requirements

Grade	Bulk density	Dynamic Elastic Modulus	Strength			Thermal Conductivity	CTE	Purity	
			Tensile	Flexural	Compressive			Ash Content	Boron Equivalent
	(g/cm^3)	(Gpa)	(MPa)	(MPa)	(MPa)	(W/m･K)	(10^{-6}/K)	(ppm)	(ppm)
G347A	1.85	10.8	**31.4**	**49.0**	**98.0**	116	4.9	<5	<0.5
ASTM Requirement	>1.7	8-15	>22	>35	>65	>90	3.5-5.5	<300	<2

Satisfy all ASTM requirements

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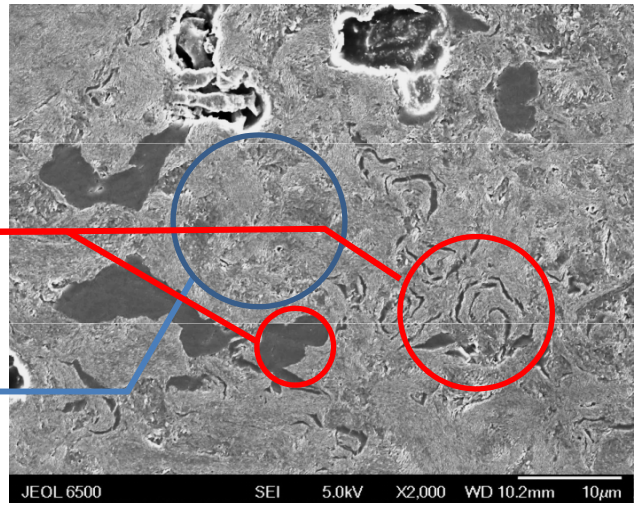
Microstructural Analysis ~ Matrix

Microstructural constituents

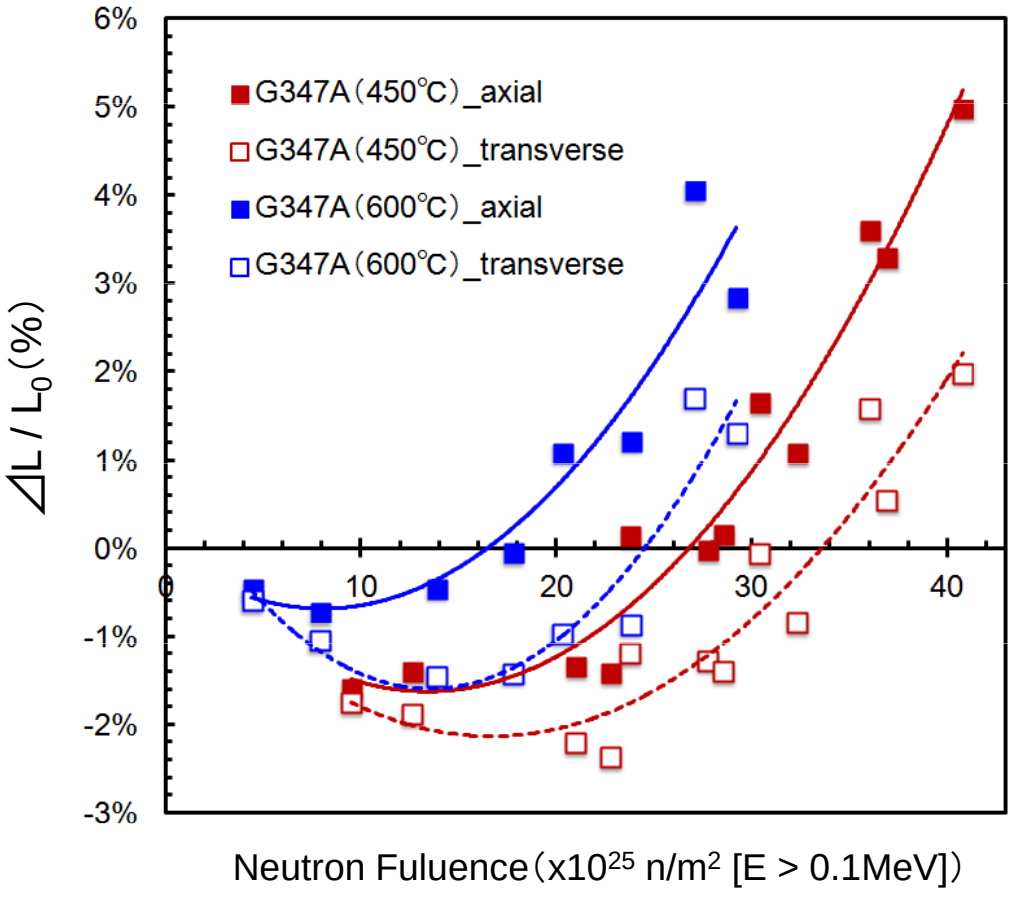
Pores (including cracks)

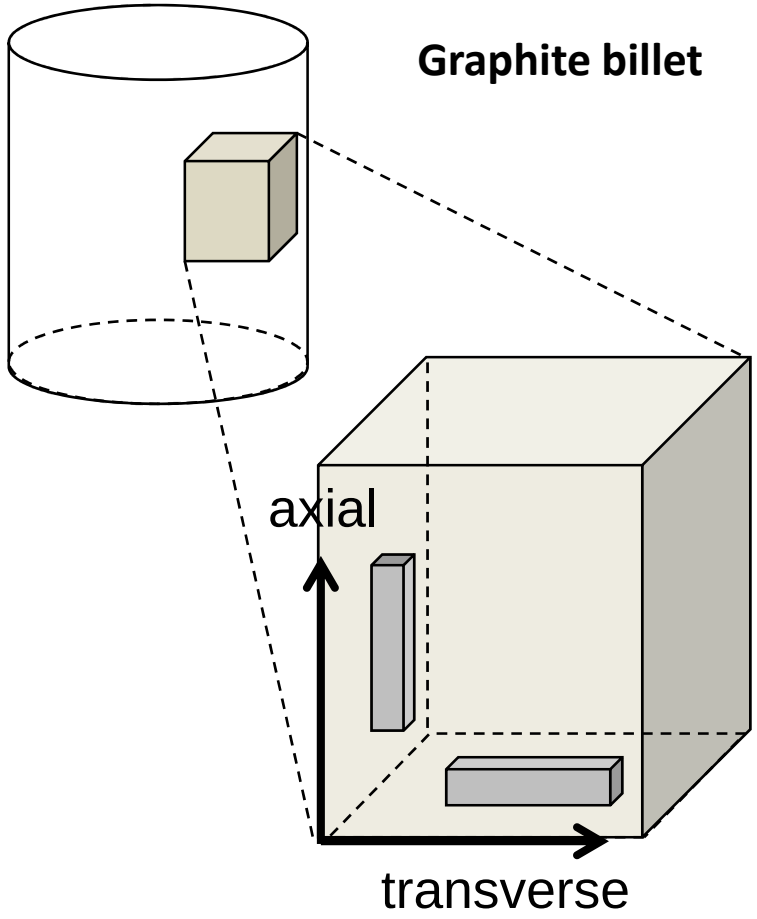
Cokes (or binder)



Constituents	Shape	Size	Anisotropy	Crystallinity
Cokes	Raman microscopy	Raman microscopy	2-MGEM（ellipsometry）	Raman microscopy
Pores	Image Analysis X-ray CT Hg porosimetry N_2 adsorption		Image Analysis X-ray CT	–
whole	SEM, TEM		XRD, CTE Electric resistivity	XRD

Dimensional changes after irradiation





G347A showed
- quadratic-like changes
- temperature dependency
- anisotropic changes

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Microstructural Analysis ~ focal point

Place the focus on 2 properties

- **Pores (distribution changes)**

  Considered to be the most important factor directly related to the dimensional changes (i.e. life time)

- **Cokes (orientation changes)**

  Even isotropic graphite shows definite anisotropic changes

	Unirr.	Irrad.
CTE (RT-500C)	4%	4%
Young's modulus	2%	2%
Dimension	-	**Significant**

- shown **only** in dimensional changes
- more pronounced at higher fluence

To evaluate these properties in detail help understand complicated changes occurred during irradiation

Microstructural Analysis ~ Matrix

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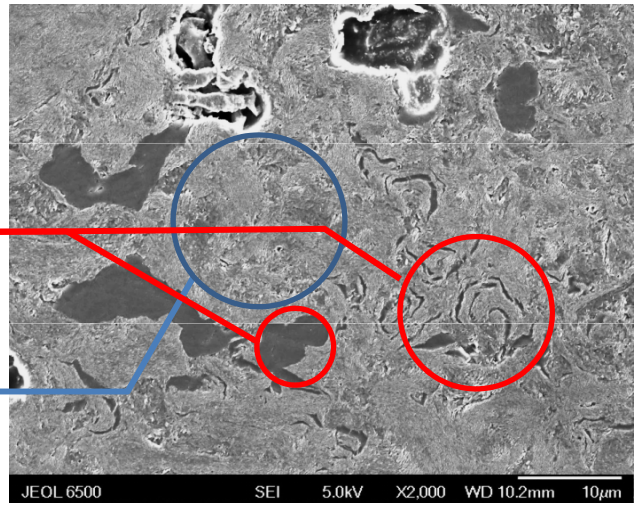

Constituents	Shape	Size	Anisotropy	Crystallinity
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Pores	**Image Analysis** X-ray CT Hg porosimetry N_2 adsorption		Image Analysis X-ray CT	–
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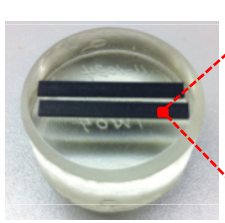
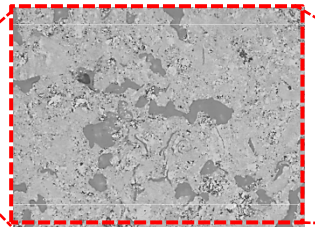
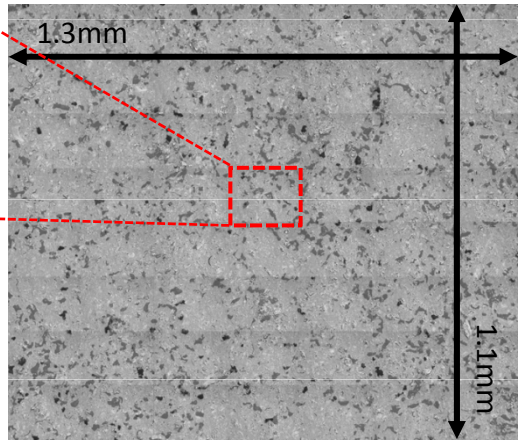
Pore distribution measurement

■ **The changes of pore distribution are evaluated by optical image analysis**

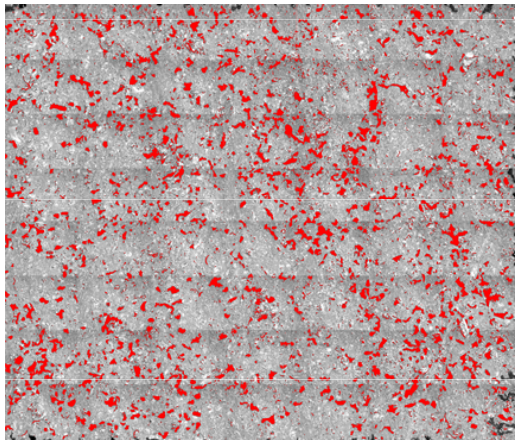




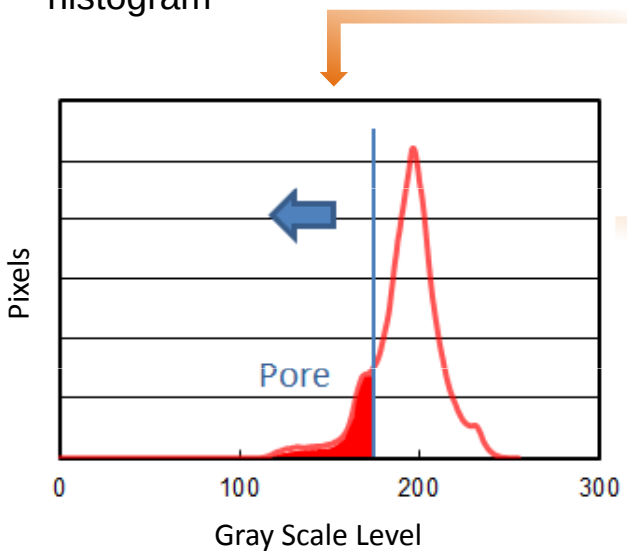
8 bit gray scale composite image is changed into a set of 256 colored histogram



Composite microstructural image(56 images)



Minimum evaluation area is 1.6μm^2

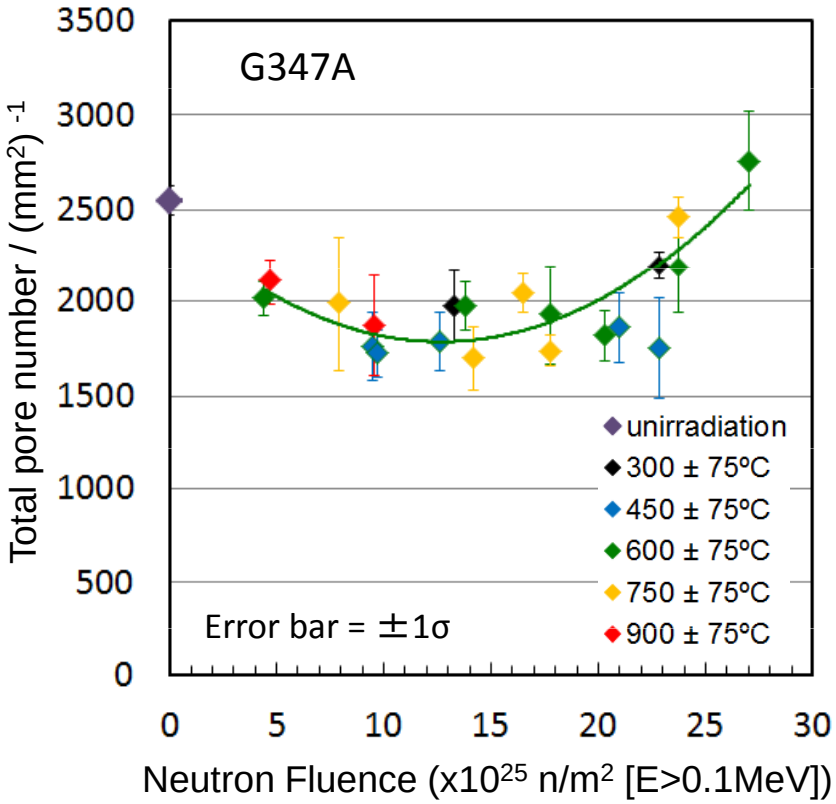


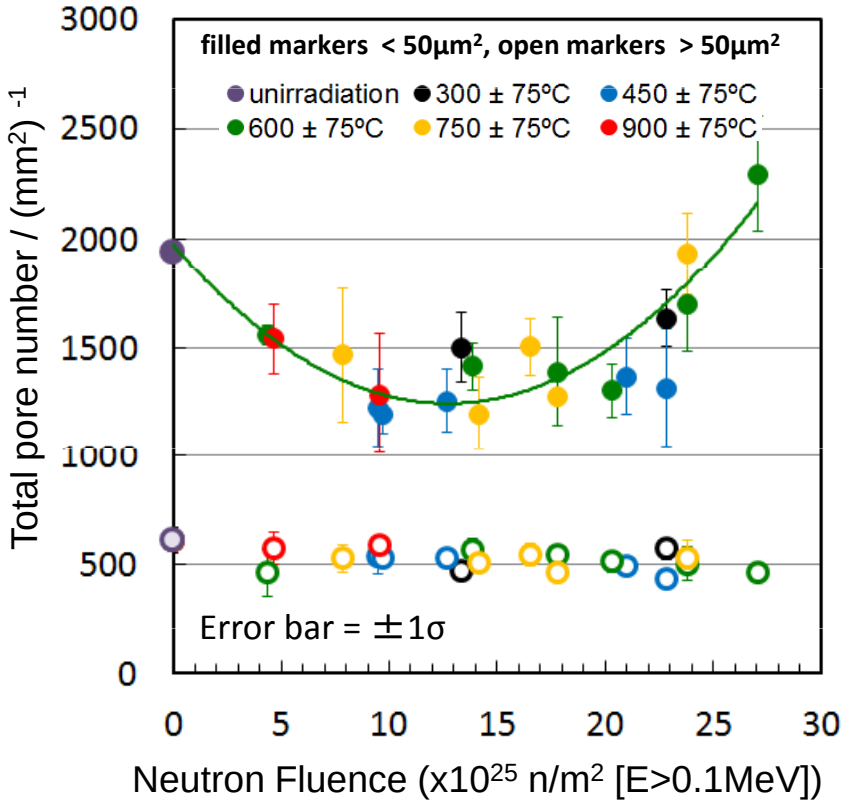
As existing pores do not reflect visible light, the dark region is recognized as pores

⇒ Pores are automatically identified and calculated

In this study pores less than 5μm^2 were disregarded to remove any errors by dot noises in the microscope

Result : pore distribution





Change of the total number showed quadratic-like curve, similar to the dimensional changes

- Changes of the small pores (<50µm²) are the dominant behavior
- Extinction and generation of pores reaches equilibrium around 15x10²⁵ n/m²

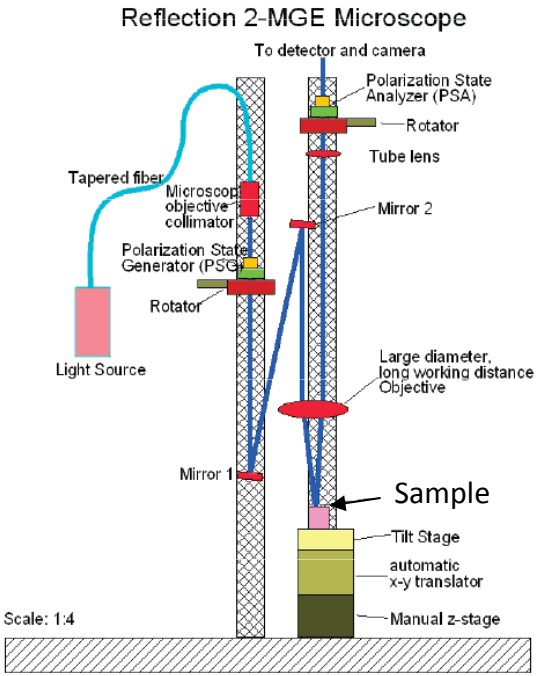
Conclusion : pore distribution

- Characteristic changes during irradiation mostly depends on the changes of smaller pores
- The curve had its vertex around $15x10^{25}$ n/m^2 which was close to the value of average turn around of G347A (300~900ºC, $10~20x10^{25}$ n/m^2)
- Each result **DID NOT** show the temperature dependence which dimensional changes clearly showed

Undetectable submicron sized pores have a temperature dependence and a great influence on dimensional changes?

Orientation measurement

2-MGEM



(**2**- **M**odulator **G**eneralized **E**llipsometry **M**icroscope)

2-MGEM is configured as a reflection microscope at near-normal incidence and measures eight different parameters*

Time-dependent intensity can be expressed as

$$\text{Intensity}(t) = I_{dc} + I_{X0}X0 + I_{Y0}Y0 + I_{X1}X1 + I_{Y1}Y1 + I_{X0X1}X0X1 + I_{X0Y1}X0Y1 + I_{Y0X1}Y0X1 + I_{Y0Y1}Y0Y1$$

Two of eight coefficients of the basis functions are used to evaluate graphite orientation

i.e. $I_{Y0} = \sin(2\gamma)N$ ∵ $\tan(2\gamma) = -(I_{Y0}/I_{Y1})$

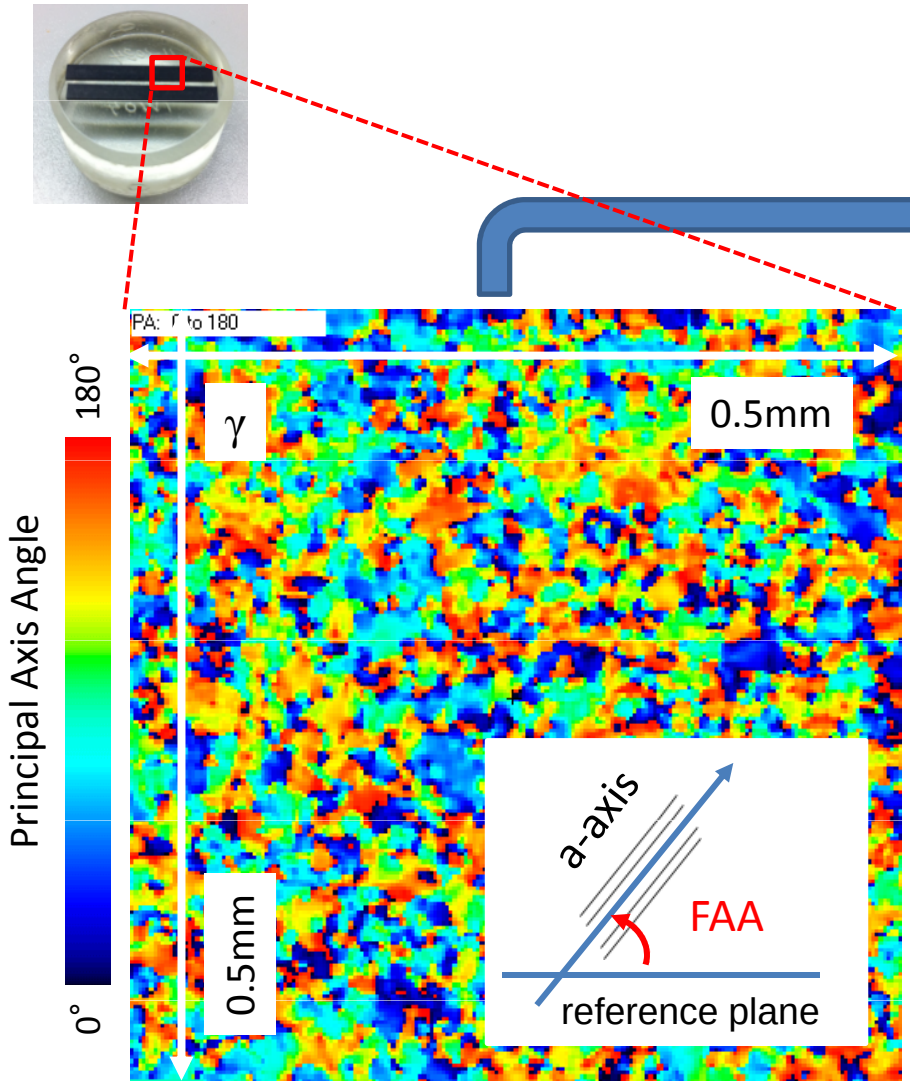
$I_{Y1} = -\cos(2\gamma)N$ $N = \sqrt{I_{Y0}^2 + I_{Y1}^2}$

I_{Y0}, I_{Y1} = coefficients of basis function,
γ **= principal axis**, N = diattenuation

Preferential orientation of the principal axis in graphite is parallel to the a-axis (perpendicular to the c-axis)

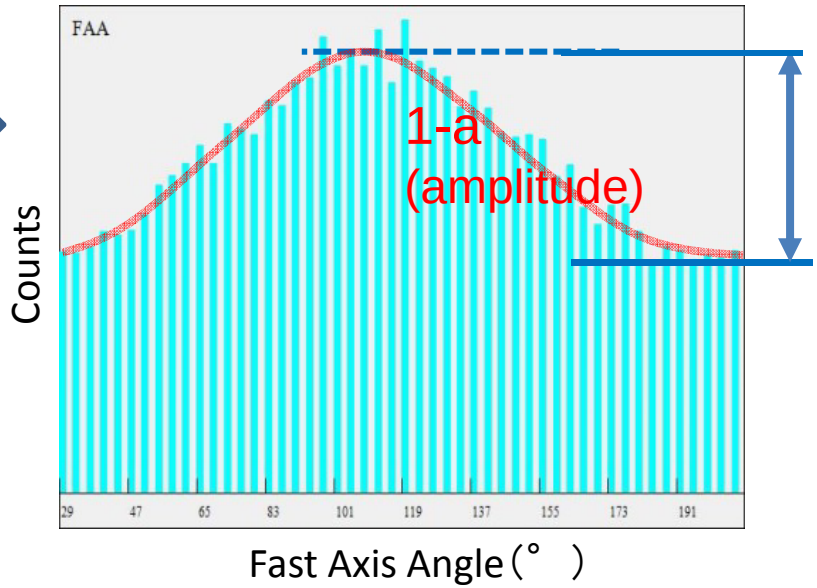
*G. E. Jellison, Jr. and J. D. Hunn, "Optical anisotropy measurements of TRISO nuclear fuel particle cross-sections: The method" J. Nuclear Mat. **372**, 36-44, (2008)

Orientation measurement



Mapping results of G347A（pixel size：□5μm）

Principal axis angle collections are converted into histogram and normalized for calculation



Each histogram was fitted with sign curve;

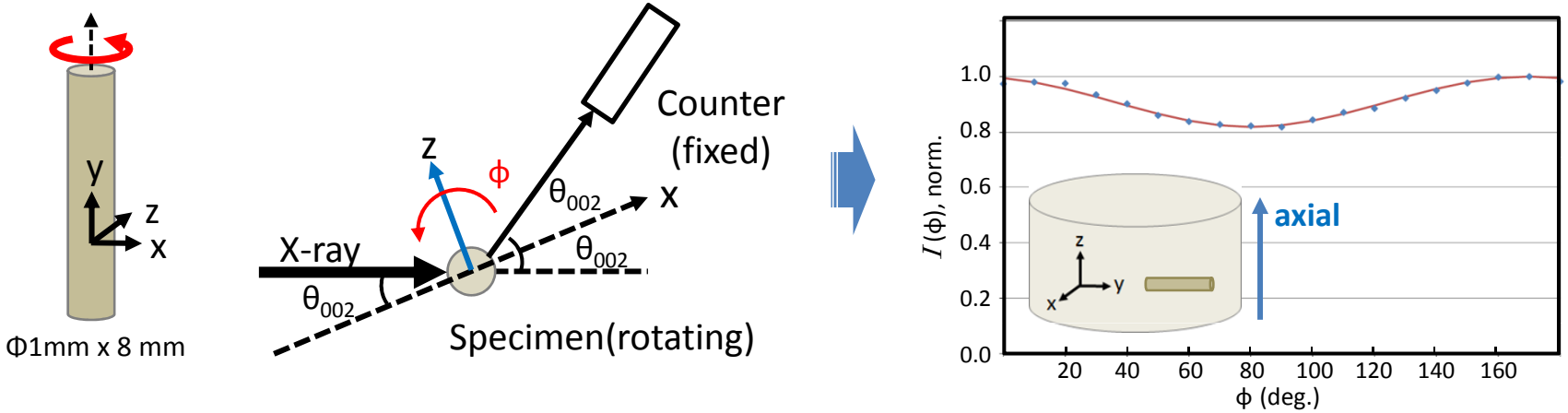
Count（nor.）= (1-a)sin^n (2(x-b))+a

Graphite orientation was estimated by the value of amplitude for simple evaluation

Result : Comparison with XRD method

- Orientation function *I* (φ) based on the diffraction curves of (002) were calculated by using X-ray diffractometer to compare with the results obtained by 2-MGEM

The diffraction pattern of (002) for two characteristic graphite (G347A, Graphite A) were obtained as a function of rotating angle



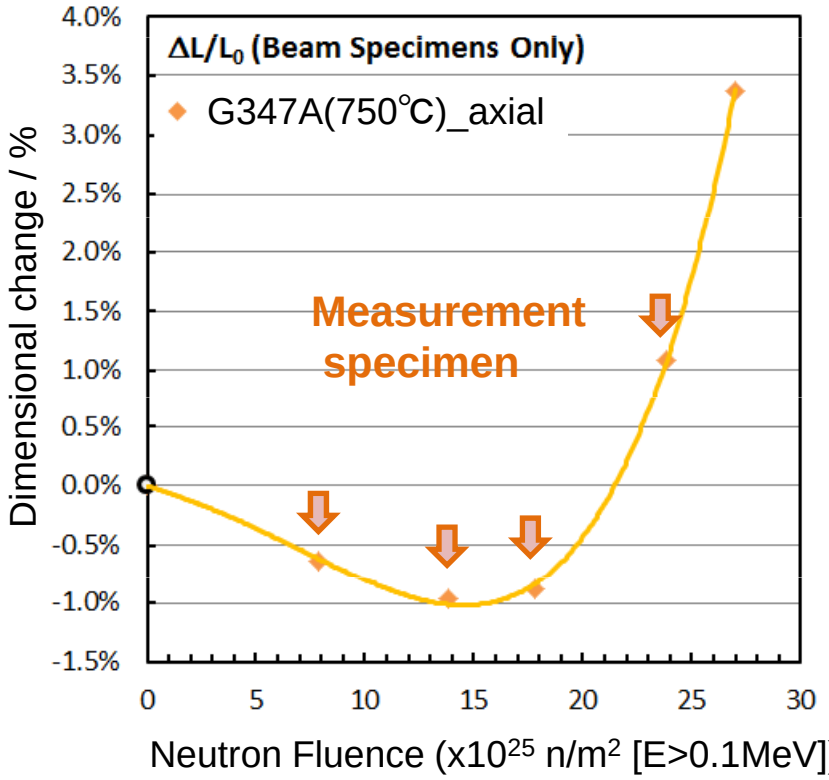
Grade	Amplitude	
	XRD	2-MGEM*
G347A	0.18	0.33
Graphite A	0.24	0.54

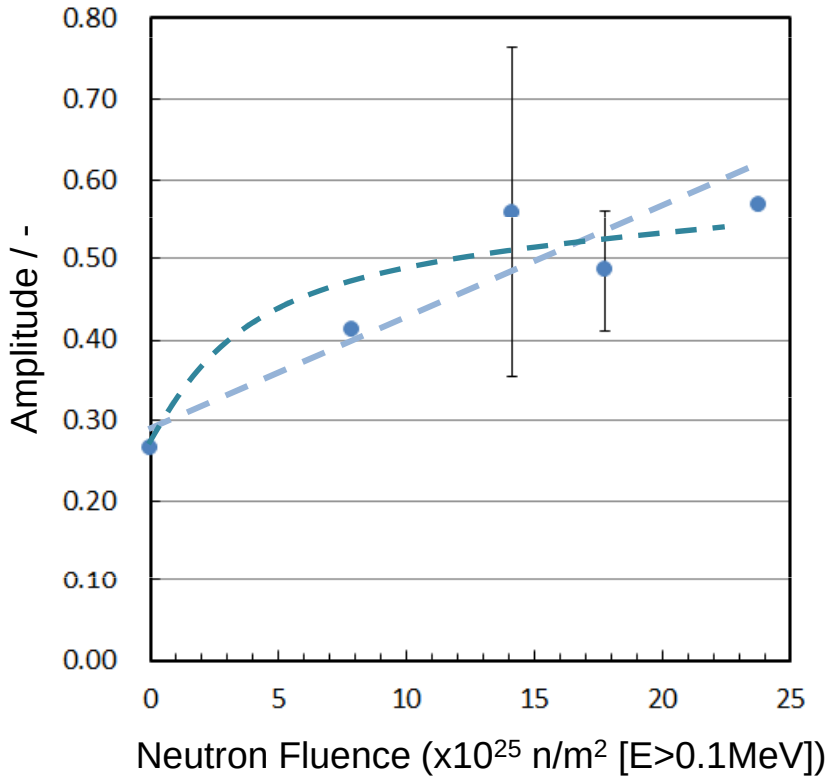
2-MGEM : y-z plane was measured

2-MGEM method showed the same trend as XRD method indicating its availability for orientation evaluation

Result : orientation

■ Irradiated graphite (G347A, 750ºC) were evaluated by using 2-MGEM

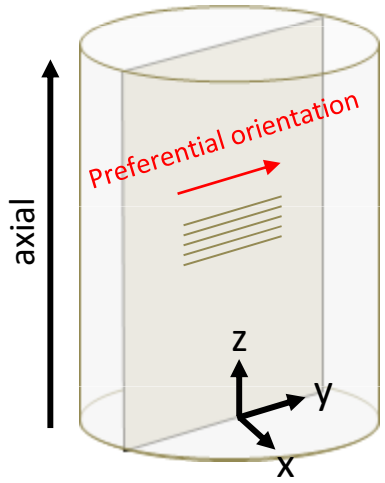




Increment of amplitude was observed with increasing neutron fluence

- Internal orientation became more anisotropic after irradiation
- This tendency well agree with the anisotropic dimensional changes

Conclusion : orientation



- 2-MGEM is of use in evaluating graphite orientation having the advantage in terms of sensitivity to preferential orientation compared with the XRD method
- G347A showed more anisotropic properties after irradiation agreeing with anisotropic dimensional changes
- 2-MGEM measurement indicated the possibility that rearrangement of cokes particles occurred by shrinkage

Summary

Focused on pore distribution/orientation changes during irradiation

- Pore changes observed as a function of neutron fluence had the same tendency as what dimensional changes had shown

  We have to come up with another method to investigate the thermal sensitive element with statistical volume

- Shrinkage during irradiation lead to rearrangement of internal crystal strengthening the originally existing preferential orientation

  Still unclear why mechanical /thermal measurements showed isotropic properties even after irradiation

  – Crystallographic orientation is not vital for mechanical/thermal anisotropic properties?

    In our recent study with unirradiated graphite, BAFs obtained by XRD showed a different trend than that of mechanical/thermal properties

Thank you for your attention!

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