OECD Nuclear Energy Agency
International Workshop on
Structural Materials for
Innovative Nuclear Systems
Idaho National Laboratory, Idaho Falls, USA
7-10 October, 2013

Based on the presentation at
"ETTORE MAJORANA" FOUNDATION and CENTRE FOR SCIENTIFIC CULTURE
INTERNATIONAL SCHOOL OF FUSION REACTOR TECHNOLOGY
11th Course: ENERGY CONVERSION SYSTEMS IN TOKAMAK REACTORS
ERICE-SICILY: 9-15 SEPTEMBER 2007

Current Status of NITE-SiC/SiC and Radiation Damage Studies Accomplished

Revised for BLIP Meeting

RaDIATE Meeting for BLIP Irradiation Test Preparation
November 23-25, 2015
Brookhaven National Laboratory, NY., USA

AKIRA KOHYAMA AND HIROTATSU KISHIMOTO
OASIS, MURORAN INSTITUTE OF TECHNOLOGY

INTRODUCTION

- SiC/SiC composites are considered as advanced structural materials in fission and fusion energy systems
  - Heat resistance
  - Intrinsic safety features
  - Radiation damage resistance

  Issues:
  - High cost
  - Flexibilities in Shape and Size
  - Weak industrial support
    (Availability of materials, and lots more)

- Continuing Efforts to establish SiC/SiC Industries in Japan
  - Many Japanese Government Funded Programs have been carried out to support these efforts

BACKGROUND: SILICON CARBIDE COMPOSITE

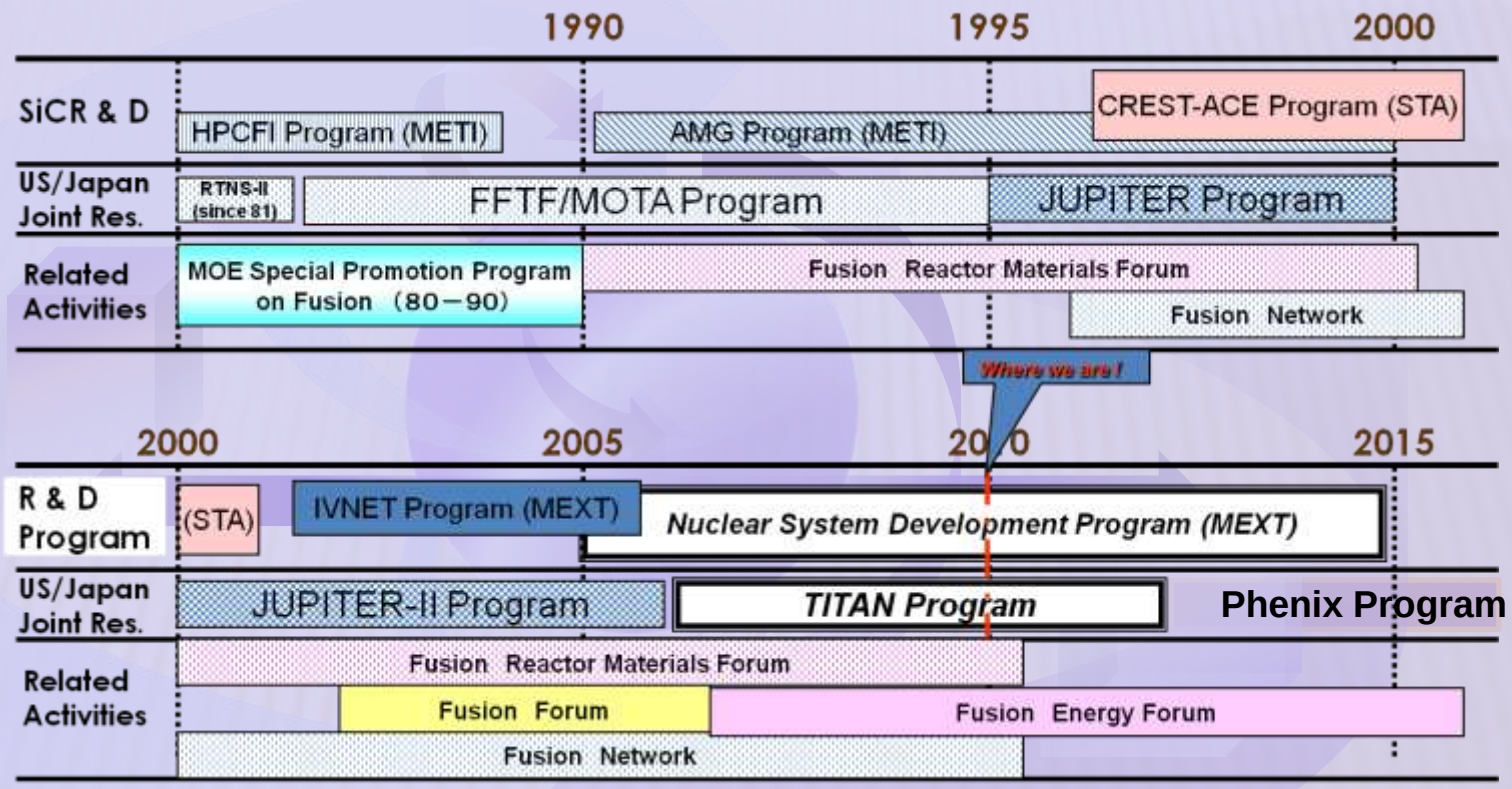
- From the Japanese efforts in these decades, starting from the invention of PCS-type SiC fibers, 70' NITE method to fabricate SiC/SiC composites was invented.
  - International Patents by A. Kohyama and Y. Katoh
- Efforts toward Industrialization:
  - IEST Co. Ltd., has started production of SiC fibers and SiC/SiC by NITE process since 2004.
  - Integrated industrial alliance toward early utilization of SiC/SiC is growing as *JESSiCA* since 2010 under OASIS, Muroran Institute of Technology.
  - Continuous production line prototype operation in OASIS is on-going

Two Major Directions

- *High Performance, Highly tailored Components.*
  - *Fuel cladding and Channel Box for LWR and Core Components for Gen IV Reactor*
  - *Blanket and HHF component for Fusion Reactor*
  - *Target for neutron/muon production*
  - *DHCE for geothermal electricity generation*
  - *Rocket/Gas Turbine components*
- *Moderate Performance, large (UD,2D, 3D) blocks*

The Past, Present and Future
- Fusion Reactor Materials R & D in Japanese Universities-

Muroran Institute of Technology



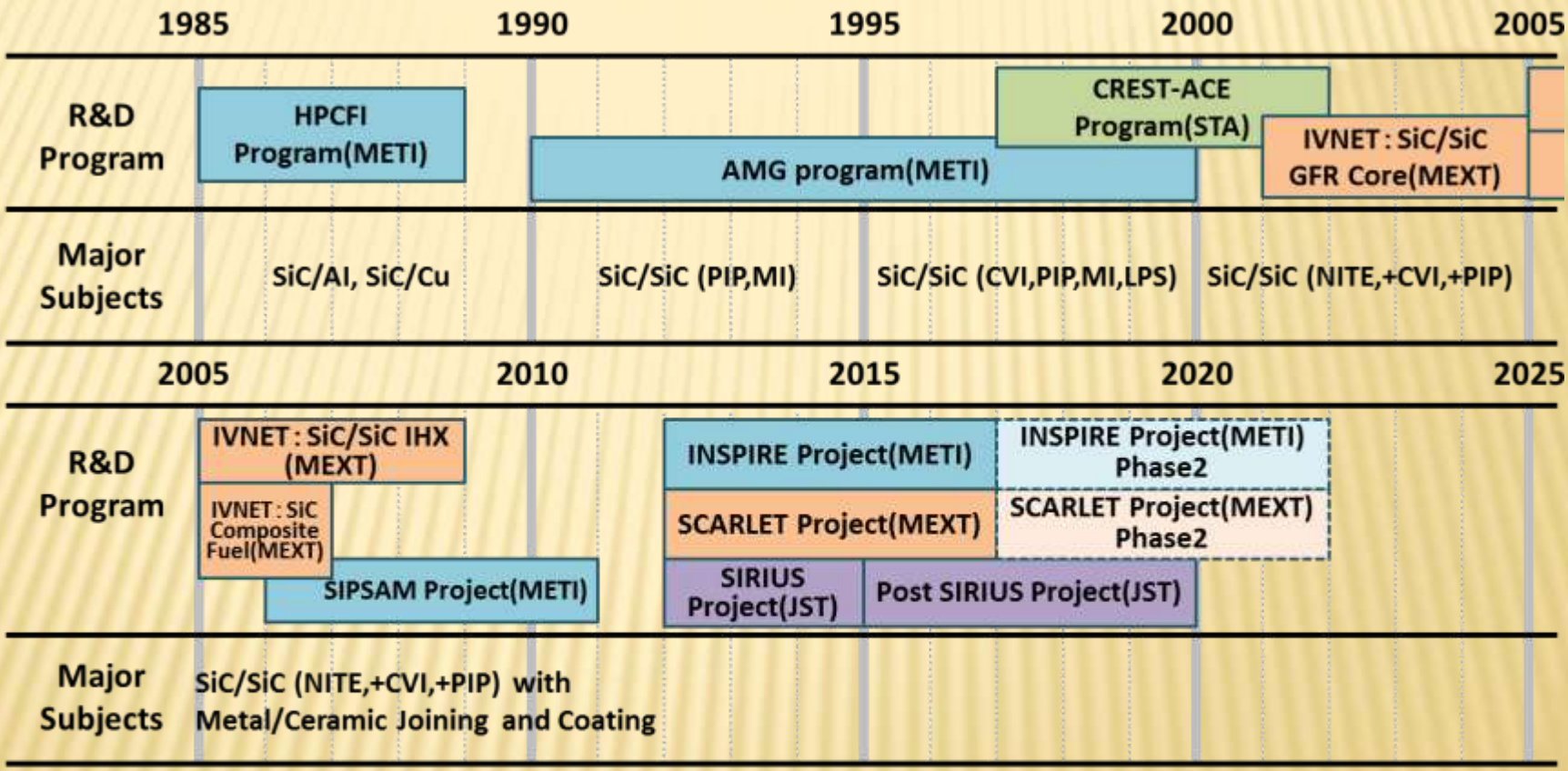
HPCFI : High Performance Composite Materials for Future Industries
AMG: Advanced Materials Gas-Generator
CREST-ACE: Core Research for Evolutional Science and Technology – Advanced materials for Conversion of Energy
IVNET: Innovative Nuclear Energy Technology development
FFTF/MOTA: Fast Flux Test Facility-Materials Open Test Assembly
JUPITER: Japan US Program of Irradiation Test for Energy Research

Is there any steady progress ? ⇒ Without learning the past No success anticipated !

No! Too simple model materials & without knowledge about process importance

THE HISTORY OF SiC COMPOSITE R & D

UNDER KOHYAMA GROUP



HPCFI : High Performance Composite Materials for Future Industries AMG : Advanced Materials Gas-Generator
CREST-ACE : Core Research for Evolutional Science and Technology –Advanced materials for Conversion of Energy
IVNET : Innovative Nuclear Energy Technology development SIRIUS : SiC Integration Research for Innovative Utilization of Geothermal Energy Source
INSPIRE : Innovative SiC Fuel-Pin Research SIPSAM : Support Industry Program/SiC/SiC for Al Die-Casting Machine by Hot-Chamber Method
SCARLET : SiC Fuel Cladding/Assembly Research Launching Extra-Safe Technology

MANY KINDS OF "SIC/SIC" PROCESSING

PIP (Polymer Infiltration and Pyrolysis

Polymer precursor + SiC fibers → (Absence of gas elements; Pyrolysis) → **SiC/SiC**

Ex. PCS $-(Si-C)_n-$

- Lower density
- Low cost but it becomes very high cost to increase density.
- Non stoichiometric SiC because of residual Si and C

RS (Reaction Sintering)

Carbon + SiC fibers → (molten Si) → **SiC/SiC**

- High density
- Non stoichiometric SiC because of residual Si and C
- Damage of fiber coating

CVI (Chemical vapor infiltration)

Gas including Si / **Gas including C** → **SiC fibers** → **SiC/SiC**

- Low density
- Stoichiometric and pure SiC
- Very high cost
- Limit of the thickness

NITE (Nano-Infiltration and Transient Eutectic Phase Process)

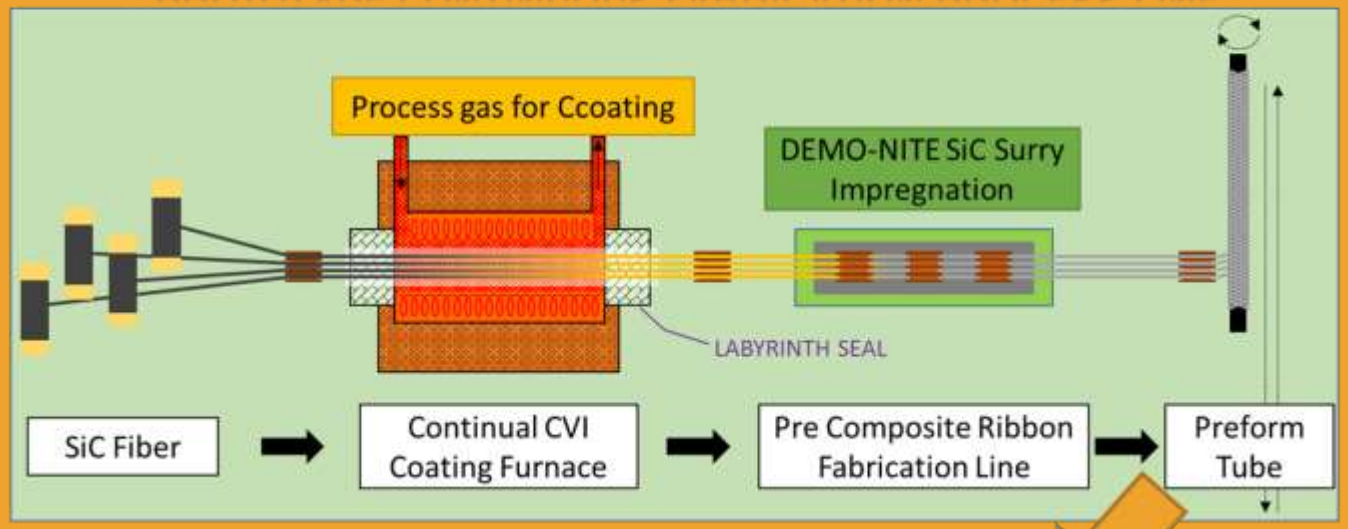
SiC nano powders + SiC fibers + Additives → (Sintering) → **SiC/SiC**

- High density and gas tightness
- Stoichiometric SiC
- Lower cost
- Residual additives

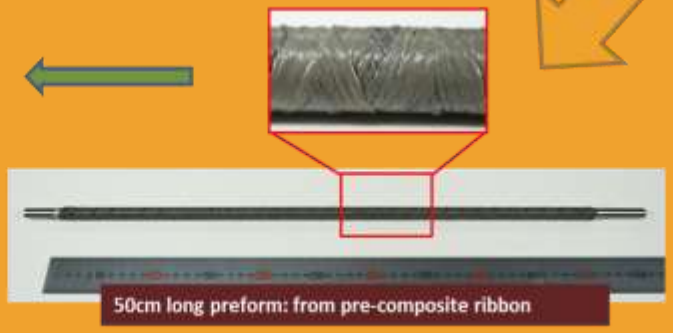
1: R & D HISTORY OF NITE-SIC/SIC

DEMO-NITE PROCESS GEN.II

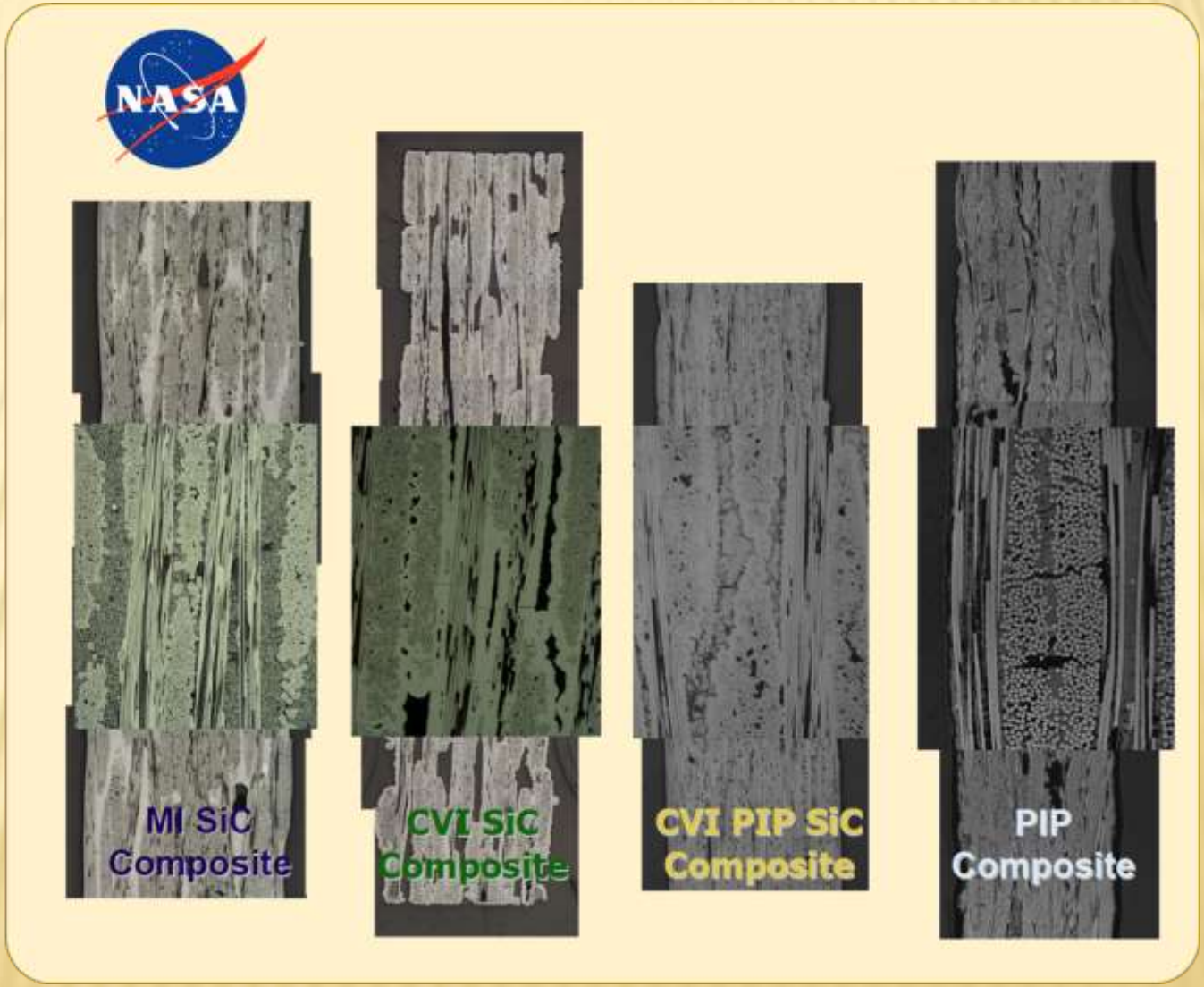
- PROTOTYPE CONTINUOUS FABRICATION PROCESS LINE -



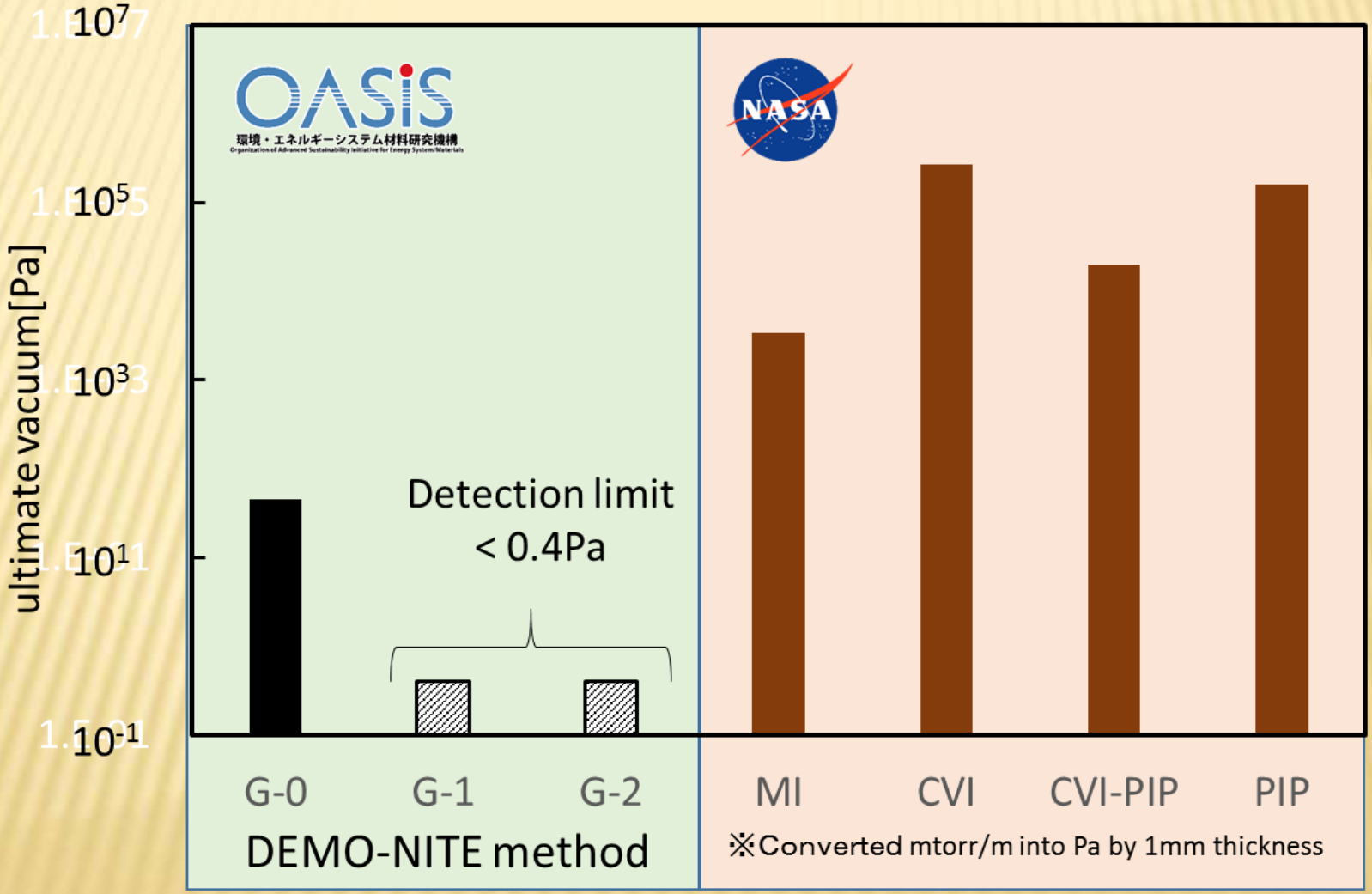
Surface of preform prepared by PCR winding method



EXCELLENCE ON GAS LEAK TIGHTNESS OF NITE-SIC/SIC

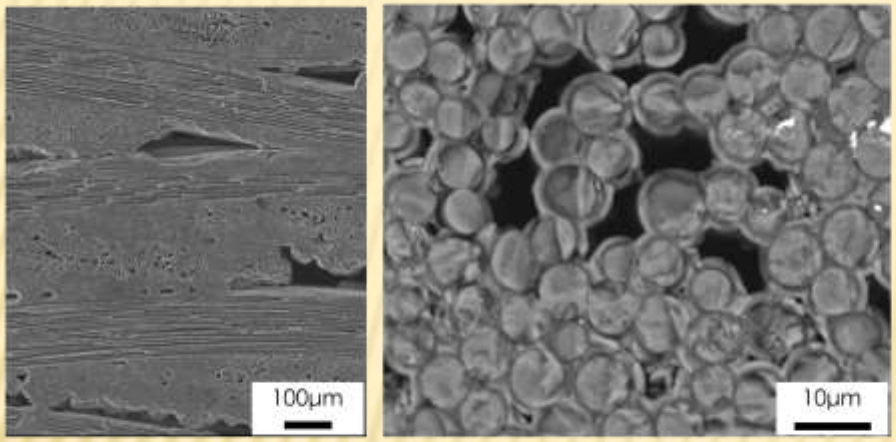


EXCELLENCE ON AIR LEAK TIGHTNESS OF NITE-SIC/SIC

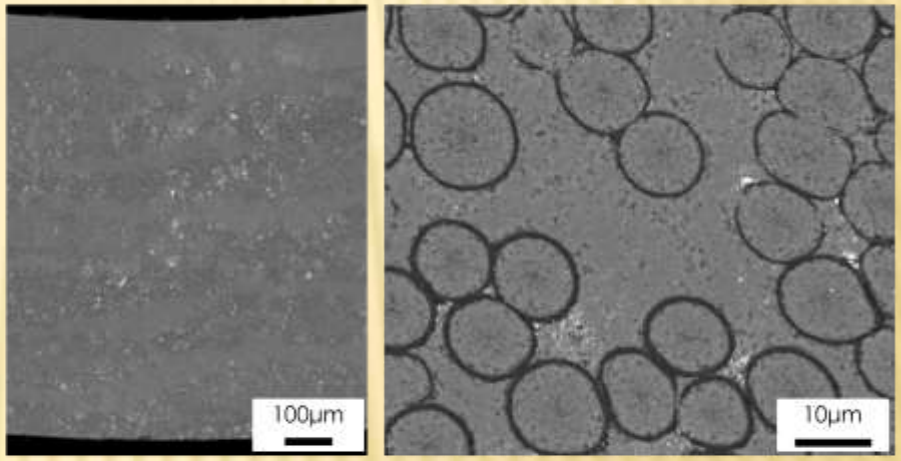


GAS LEAK TIGHTNESS OF SIC/SIC

CVI SiC/SiC



DEMO-NITE SiC/SiC



Macro / micro structure of SiC/SiC prepared by CVI and DEMO-NITE

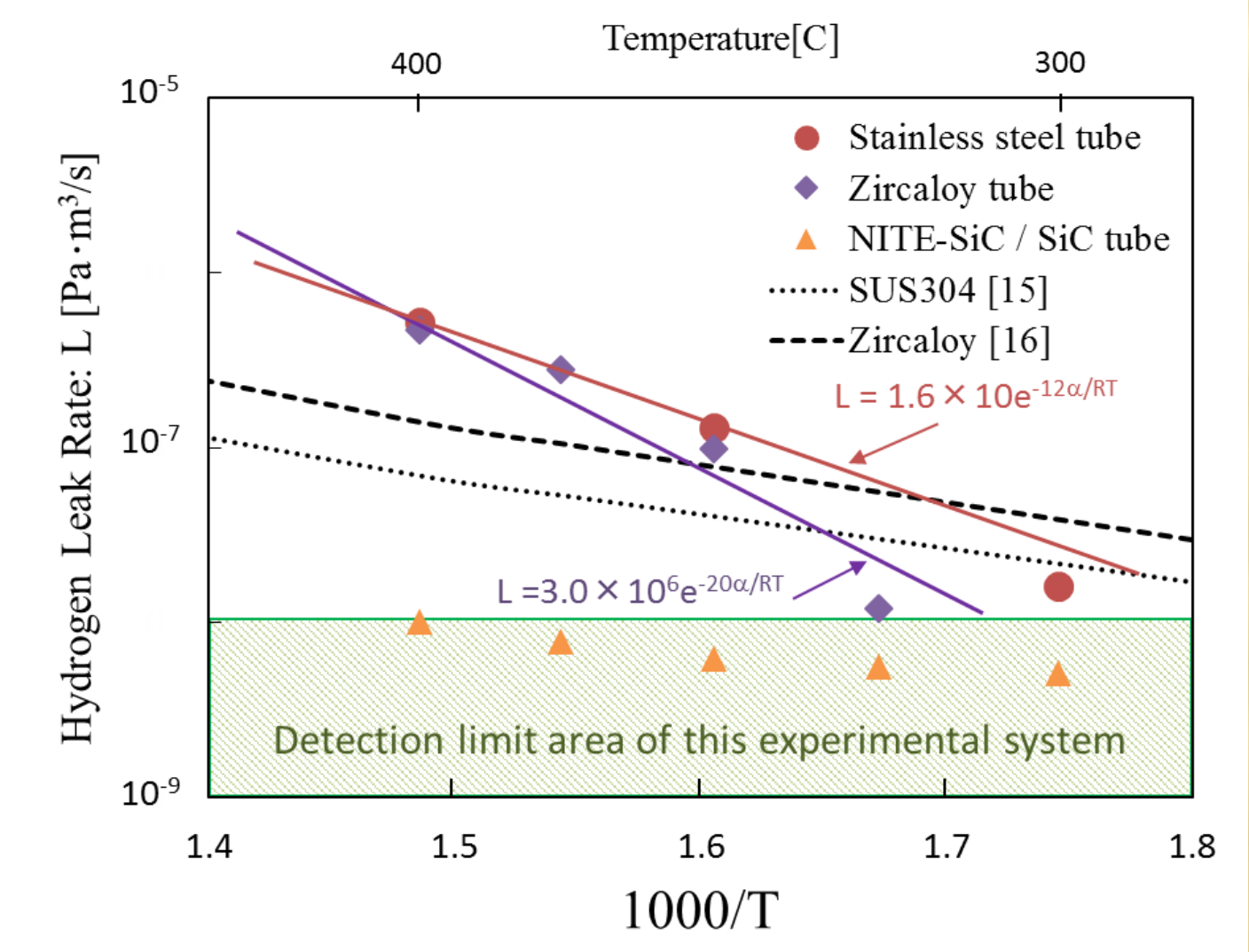
- CVI SiC/SiC
  - Porosity > 15%
    (in case of advanced CVI < 10%)
  - About 100μm pores in inter- fiber bundles
  - About 10μm pores in intra- fiber bundles

- DEMO-NITE SiC/SiC
  - Porosity: < 1%
  - No clear Pore

The similar excellent gas leak tightness with CVD monolithic SiC may indicate suppression of potential gas leak passes, such as cracks or misfit along SiC fibers and matrix micro-cracks.

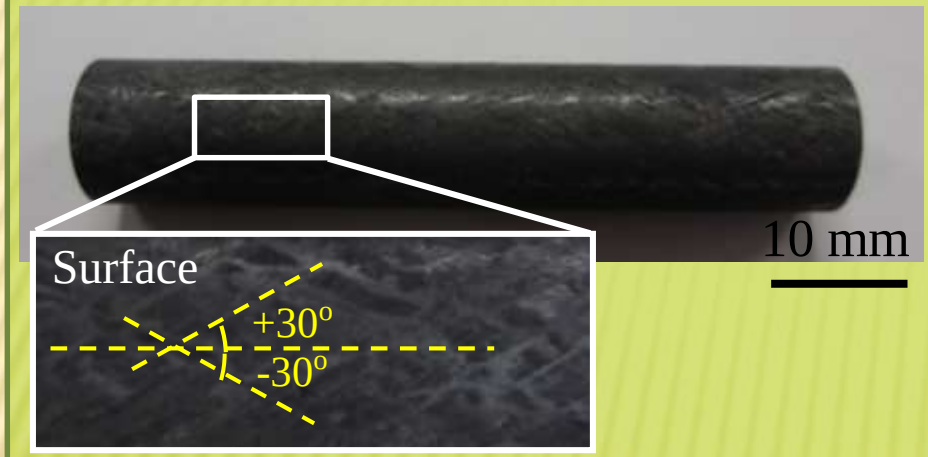
HYDROGEN LEAK TIGHTNESS OF NITE-SIC/SIC

- TEMPERATURE DEPENDENCE -



SiC/SiC Fuel Cladding

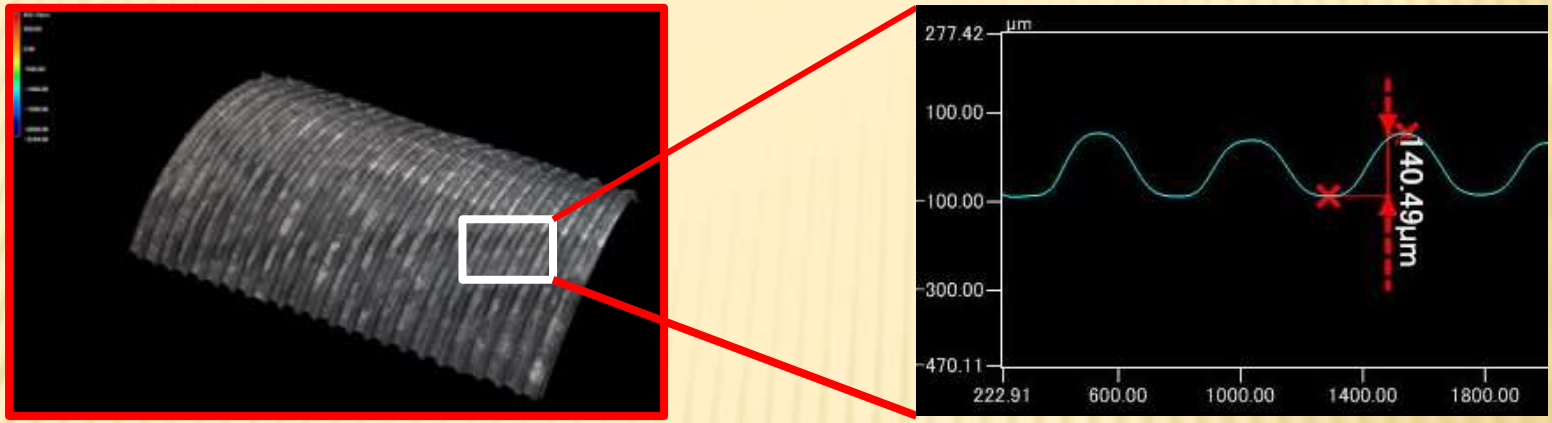
Un-coated SiC/SiC tube



Monolithic SiC coated SiC/SiC tube

Cross-section

ZIRCALOY + SIC/SIC JOINT TEST SAMPLE

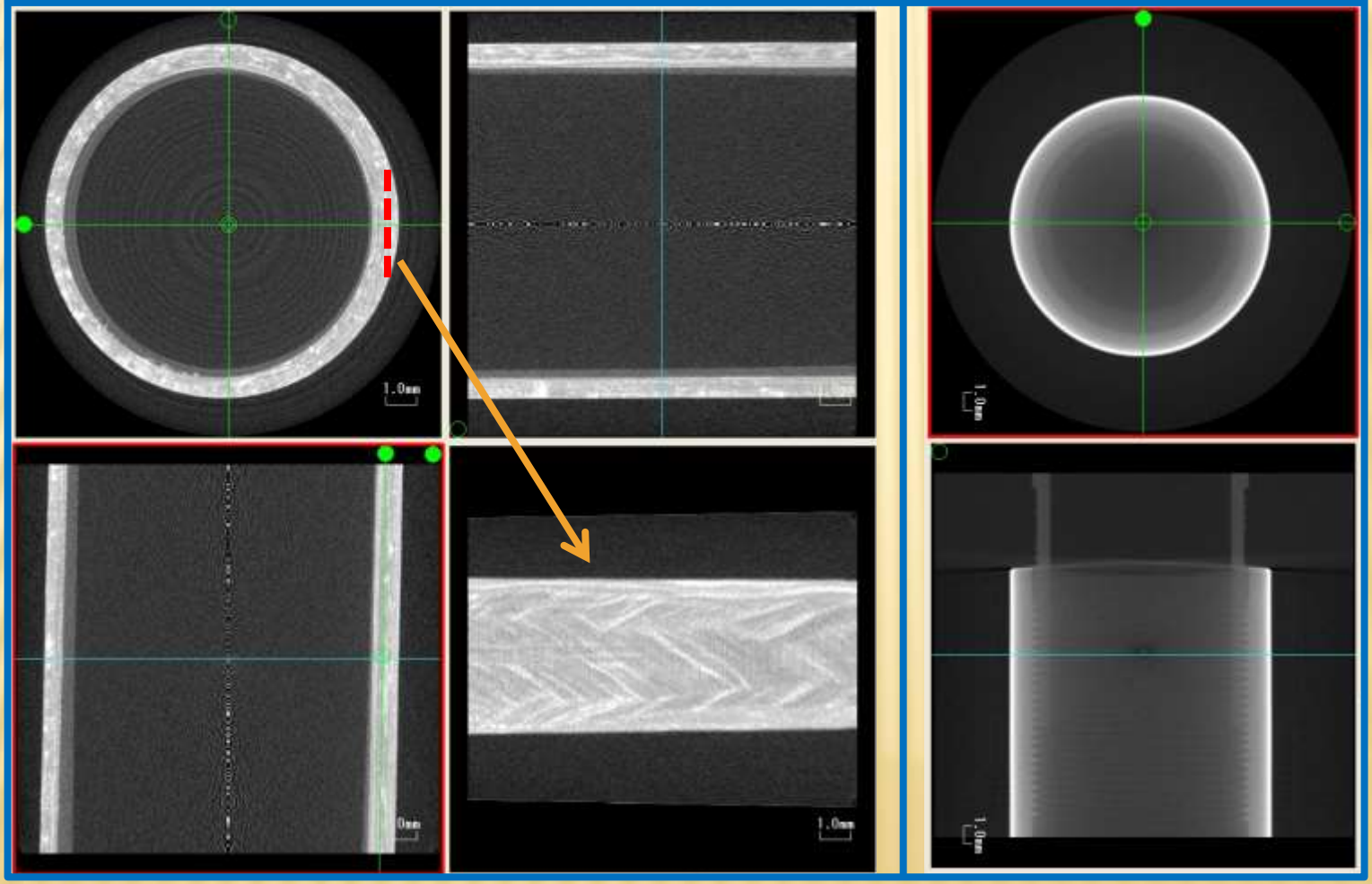


10mm^{id}, 0.7mm^{t} NITE-SiC/SiC Tube

Zircaloy End-cap SiC/SiC Cladding

X-RAY CT IMAGES OF SIC/SIC AND ZIRCALOY JOINT

SiC/SiC	SiC/SiC+Zircaloy



SIC/SIC FUEL PIN SEGMENT FOR HBWR IRRADIATION TEST

- PROCEDURE DEMONSTRATION FOR FUELED PIN -

http://josephmiller.typepad.com/b
ttle-to-stabilize-the-f/2011/03/

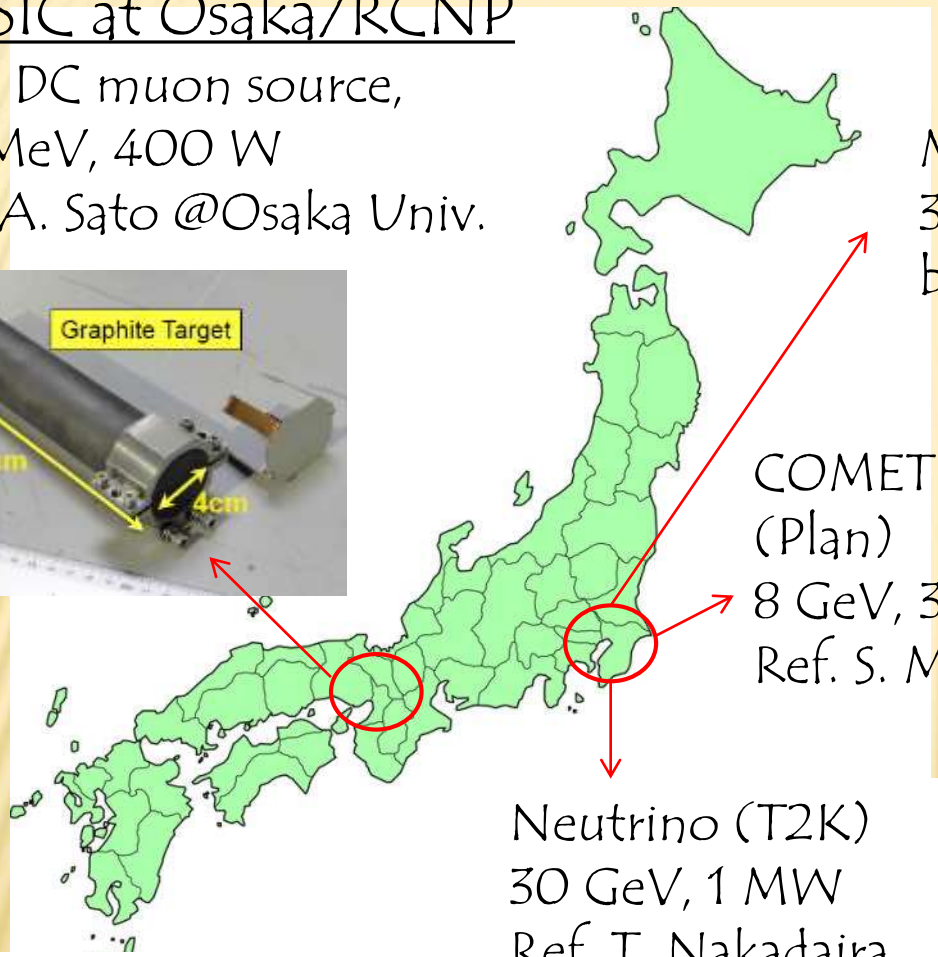
- Demonstration for 2015 INSPIRE irradiation project 2015 with simulation fuel pellets has been successfully done.
- Large delay and basic policy change of nuclear research project under METI funding results in temporally cancellation of fueled pin irradiation.

Stainless dummy fuel pellets have been inserted.

Muon/Pion Production Target for proton accelerator in Japan

MuSIC at Osaka/RCNP

New DC muon source,
392MeV, 400 W
Ref. A. Sato @Osaka Univ.

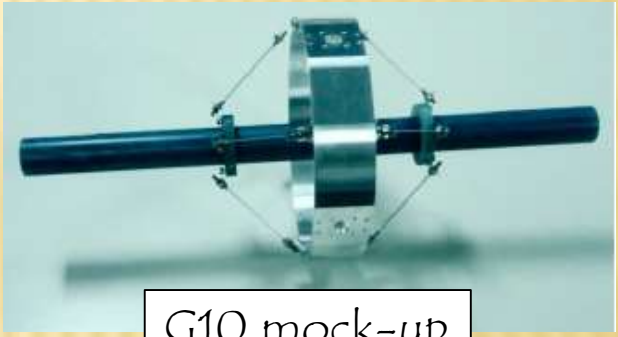


J-PARC

MLF/Muon
3 GeV, 1MW
by S. Makimura

COMET Phase 1
(Plan)
8 GeV, 3.2 kW
Ref. S. Mihara

Neutrino (T2K)
30 GeV, 1 MW
Ref. T. Nakadaira



G10 mock-up

From t2k experiment.org

Graphite Target in general

Polycrystalline Graphite

Conventional material for High Power Proton Target

- Not only Japan but also, PSI, ISIS,,,,
- Low density~ Low Beam loss
  Large disperse of muon/pion production
- High-temperature resistance
- Low Young's modulus~ Low thermal stress
- High thermal-shock resistance
- Loss by Oxidation
- Tritium Production

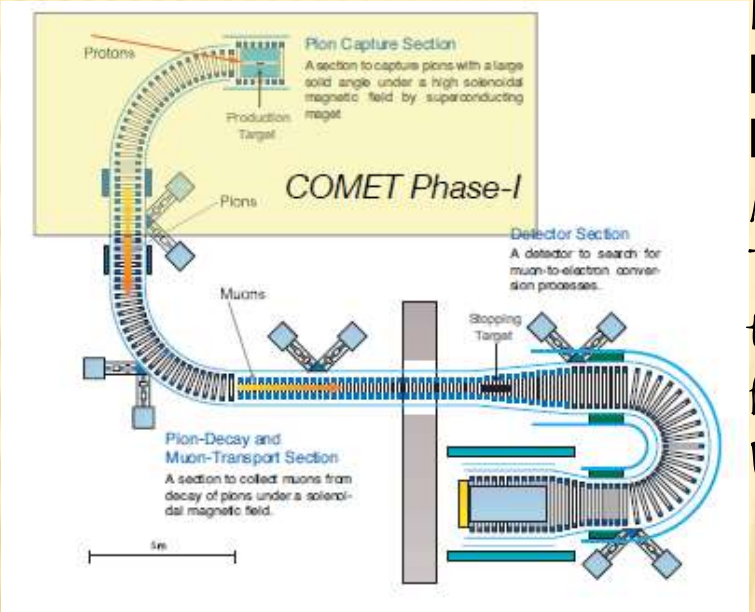
SiC/SiC composite material

Candidate of new target material with High Performance

Advantage of SiC with regard to Physics requirements

COMET Experiment;

These information were supplied by Dr. Mihara.



Muon-Electron Conversion

Hadron Experimental Facility at J-PARC

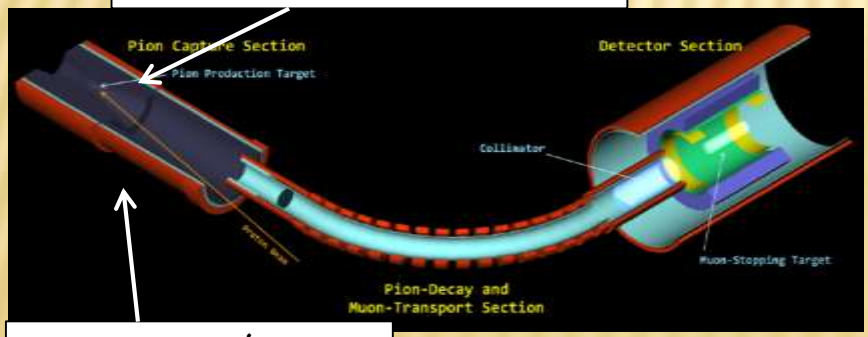
- Phase-I; 8 GeV-MR, 3.2 kW, Graphite target
- Phase-II; 8 GeV-MR, 56 kW, High-Z target

Muon Transport Section is designed for Phase-II.

To reduce the disperse of muon production in the solenoid, SiC target is preferable to graphite for Phase-I.

Under discussion, with Dr. Mihara.

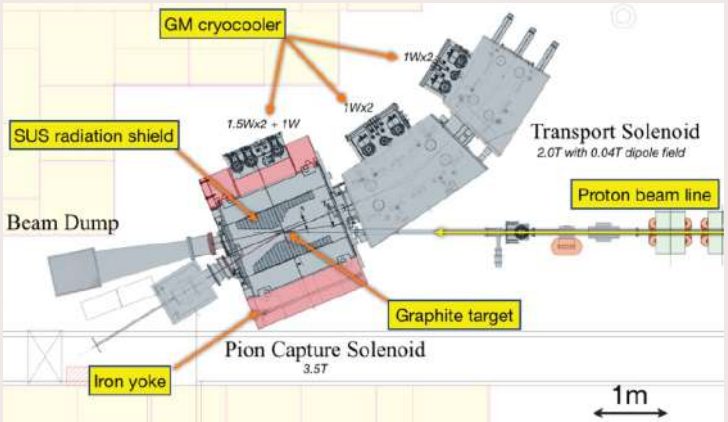
Pion Production Target



Capture Solenoid

MuSIC/RCNP at Osaka Univ. by Dr. Sato

Target is also located in capture solenoid.



Advantage of SiC with regard to Physics requirements

DeeMe Experiment;

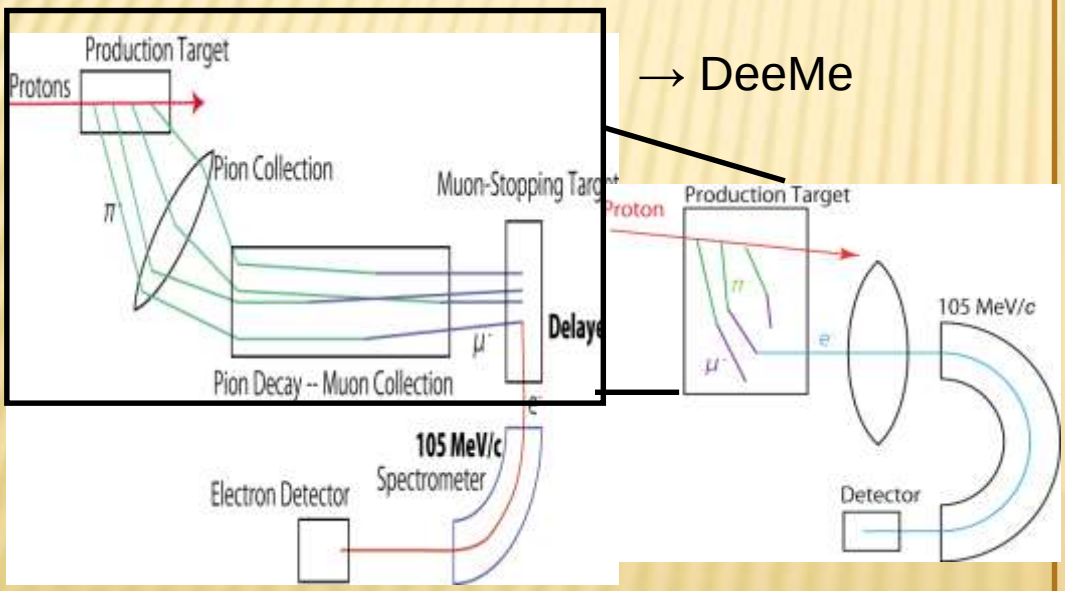
These information were supplied by Dr. Aoki in Osaka Univ..

Muon-Electron Conversion

Materials and Life Science Facility at J-PARC

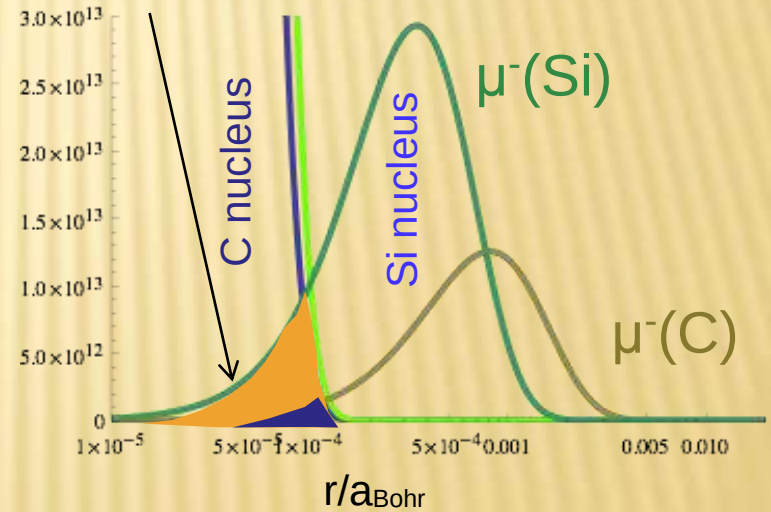
- Sensitivity of DeeMe; 10^{-14}
- (Sensitivity of COMET
  Phase-I; 10^{-15}, Phase-II; 10^{-17})

Better to replace graphite with SiC.



Concept of DeeMe, simple, quick and low-cost.

Graphite (C) → Si: 11-times larger overlap
μ^- reaction ε: 8%(C)→67%(Si)



Silicon Carbide

Eff. of Muon Reaction: 6 times larger than graphite.

SiC/SiC Target for DeeMe

Merits of SiC target

- Higher efficiency of the production of muon than graphite targets.
- Higher density (3.21g/cm^3) than graphite (1.8-1.9 g/cm^3)
- Expected better tritium confinement in material.
- Safety and flame retardant in a loss of vacuum accident.

Issues of SiC/SiC target related to operation

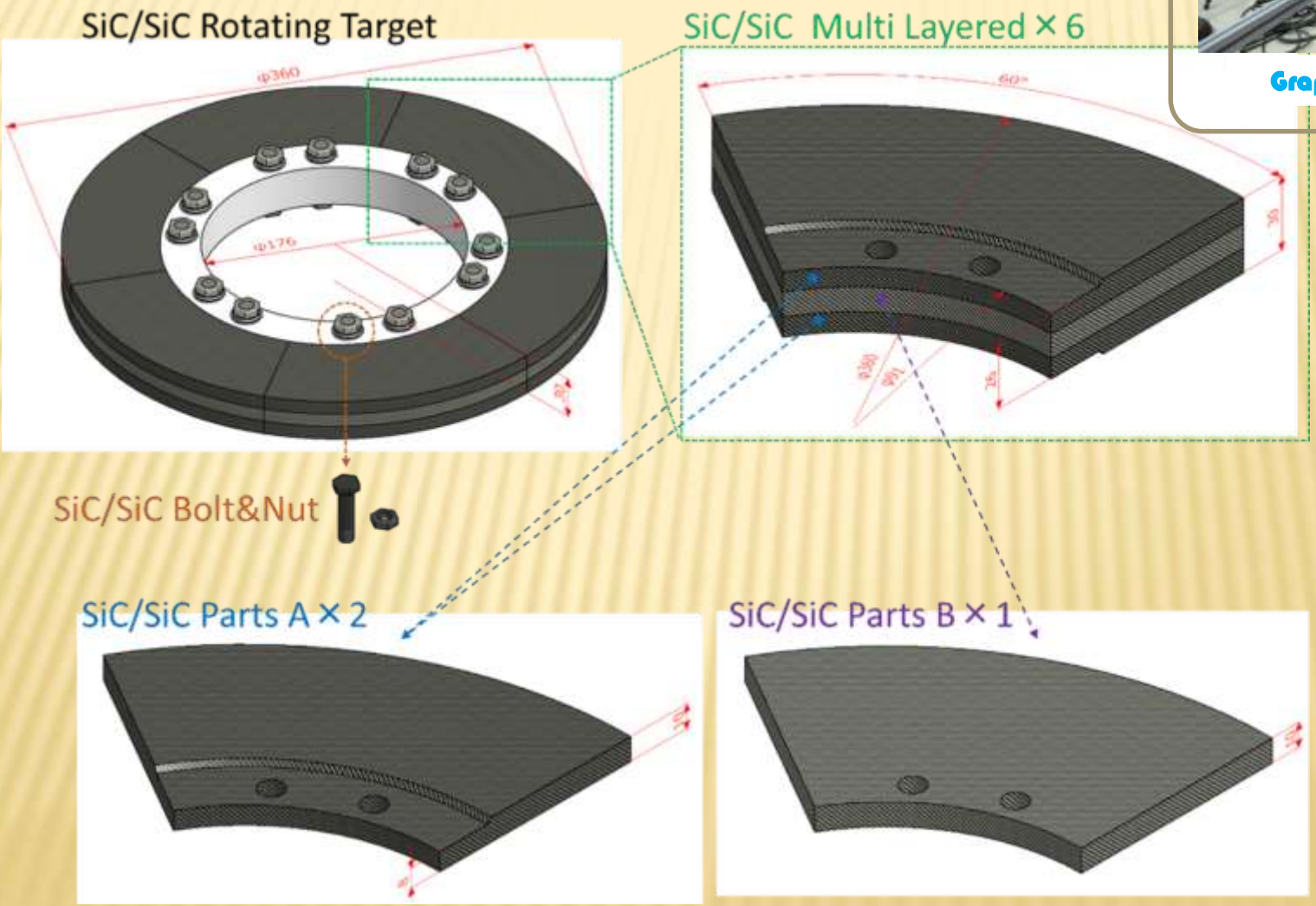
- Radio activation caused by impurities (additives) such as Al and Y
- Thermal stress and thermal shock during the operation.
- Tritium production and release rate during the operation.
- Life time.

Issues of SiC/SiC target fabrication

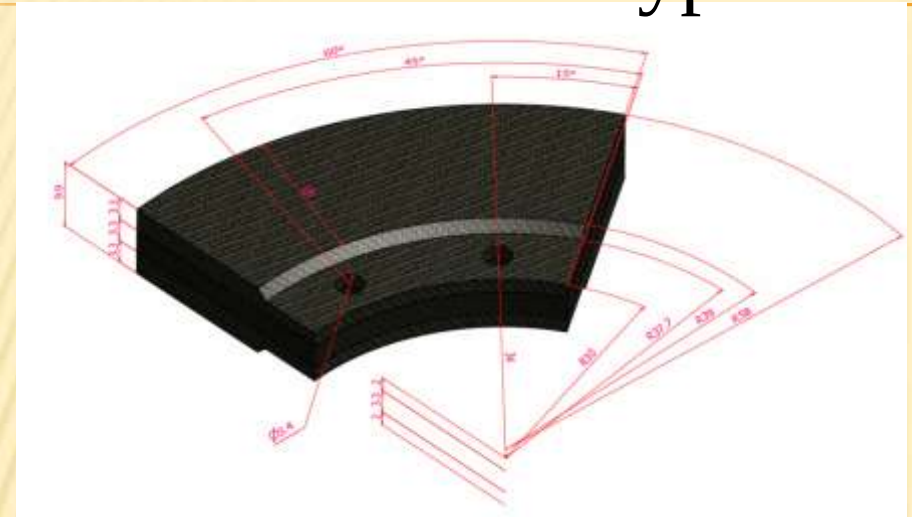
- Shape of targets and architecture of reinforcements.
- Processing method and device, especially, for large scale targets.
- Cost and process time, especially for larger targets.

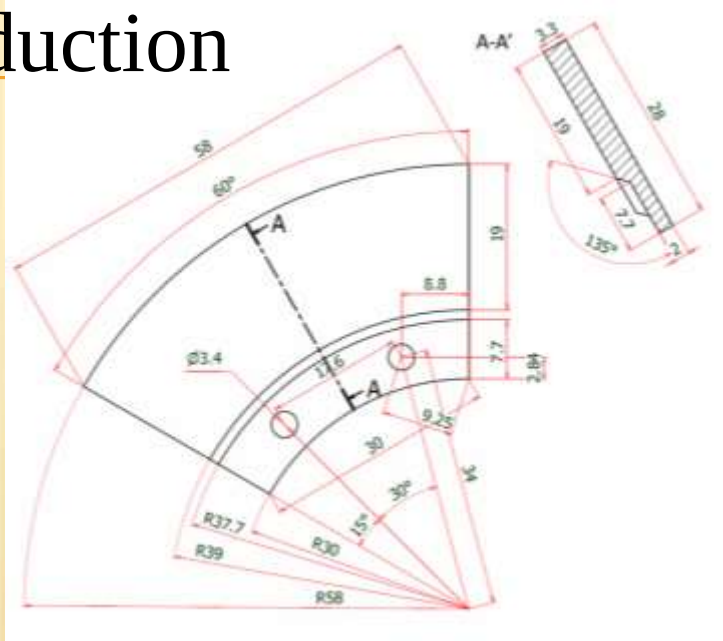
The Current Design of SiC/SiC Target for DeeMe

Graphite Rotating Target

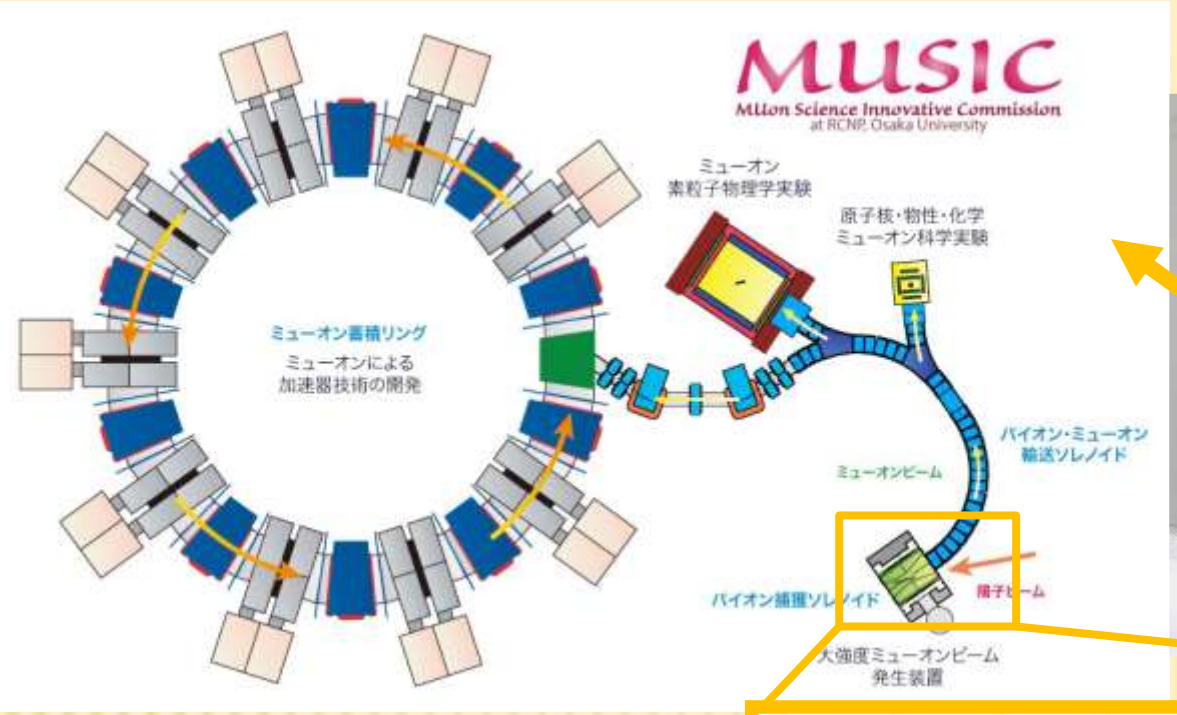


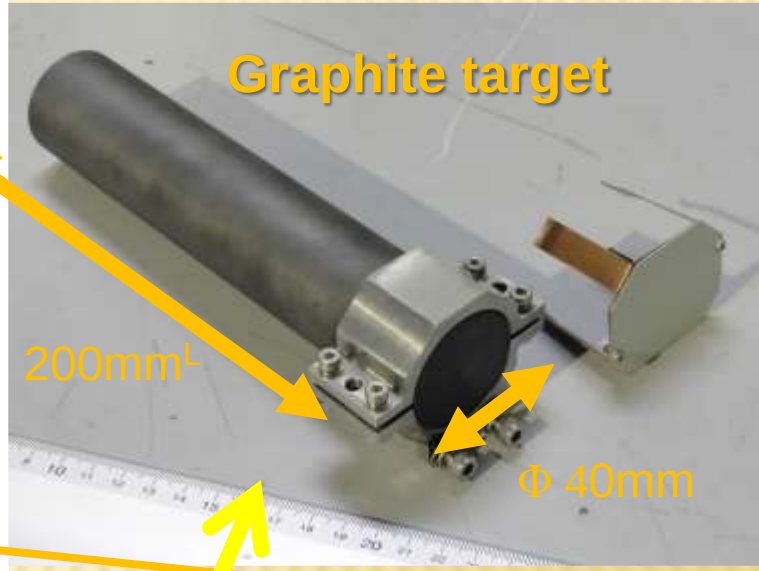
Drawing for 1/3 Scale Target for DeeMe and Prototype Production

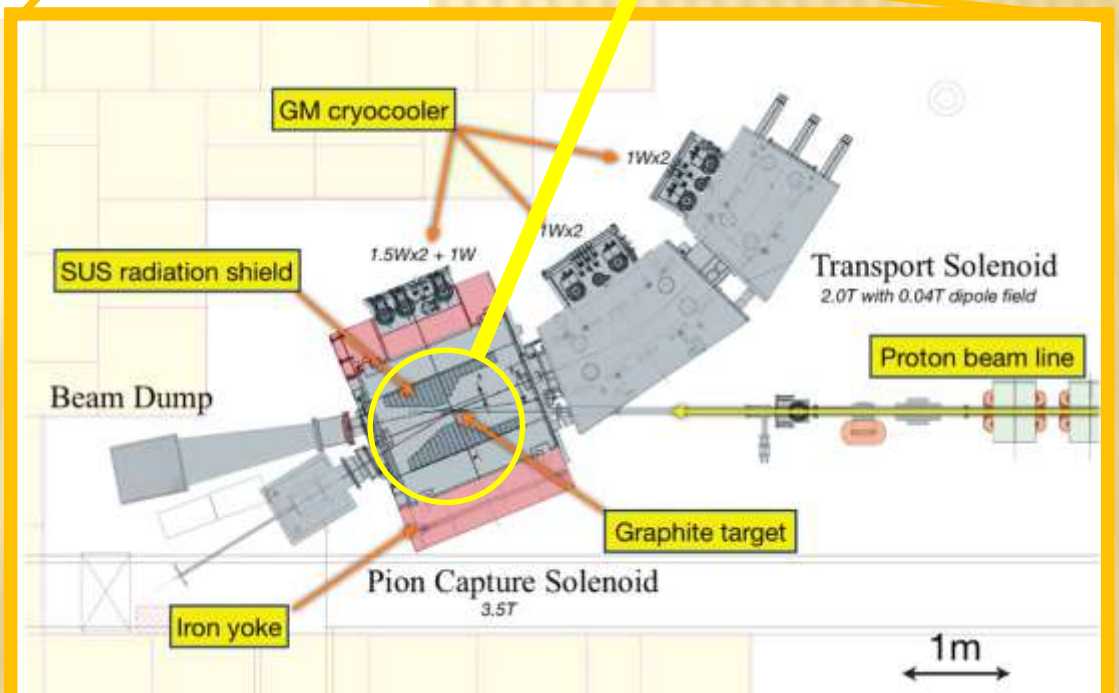




- Simulation of temperature distribution and thermal stress distribution by FEM analysis will be also done.
- Depending on the simulation results and performance evaluation test results of NITE-SiC/SiC target, fiber architecture design modification, matrix design modification will be done together with the target design modification.

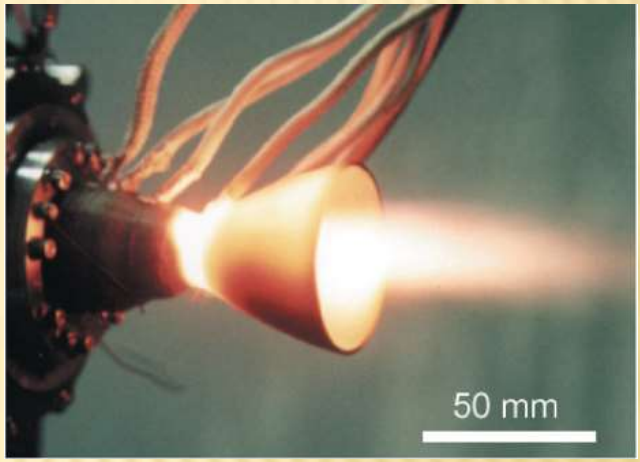




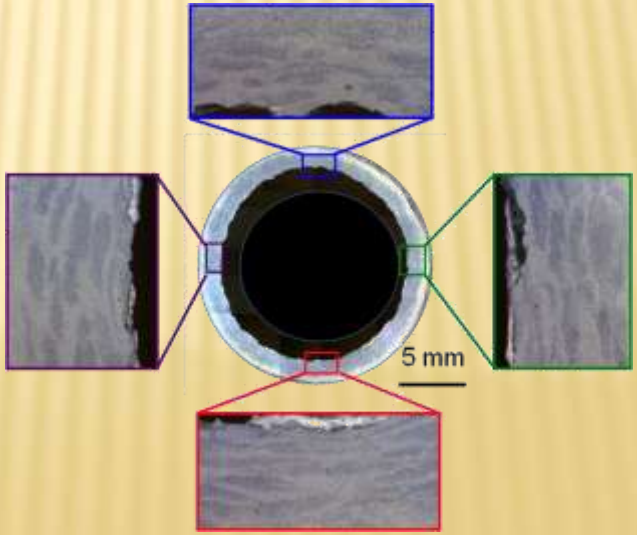


Presented by A Sato, Osaka univ., Sep 28 in 2012

ALL SIC/SIC THRUSTER NOZZLE



Propulsion Test



Cross-section of 20N class thruster (throat position)

SiC/SiC Material Database on "Radiation Effects" Required

- Radiation damage tolerance (influencing parameters)
  - Bombarding particles
  - Bombarding scenario
  - Energy/Flux/Fluence
  - Environment (temperature, gas, pressure)
- Primary Effects on
  - Displacement damage
  - Transmutation damage (mainly, H, D, T, He)
- Secondary Effects
  - Mechanical Properties (Static/Dynamic/Cyclic)
  - Physical/Chemical Properties (Thermal/Electrical)
  - Microstructure/Dimensional Evolution (swelling)

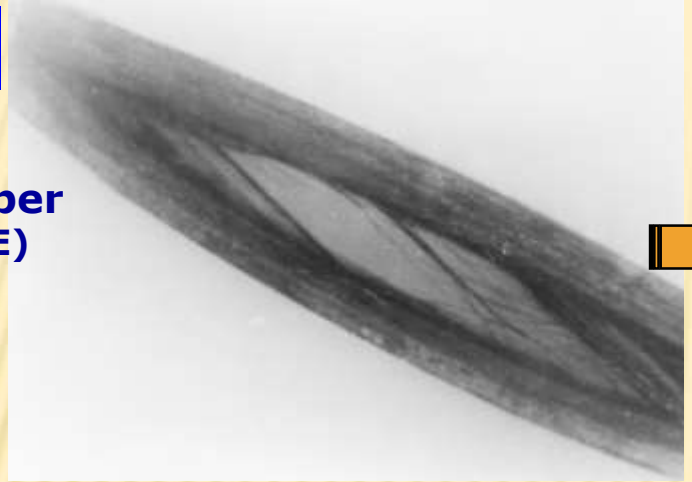
To prioritize radiation effect importance in this work is essential

Neutron Irradiation Effect on Dimensional Stability

- CVI SiC/SiC with different SiC fibers -

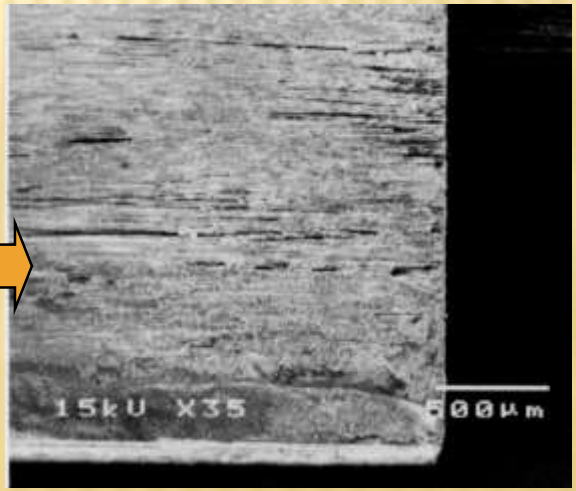
Old Fiber

Amorphous Si-Ti-C-O Fiber (Tyranno™-TE)



New Fiber

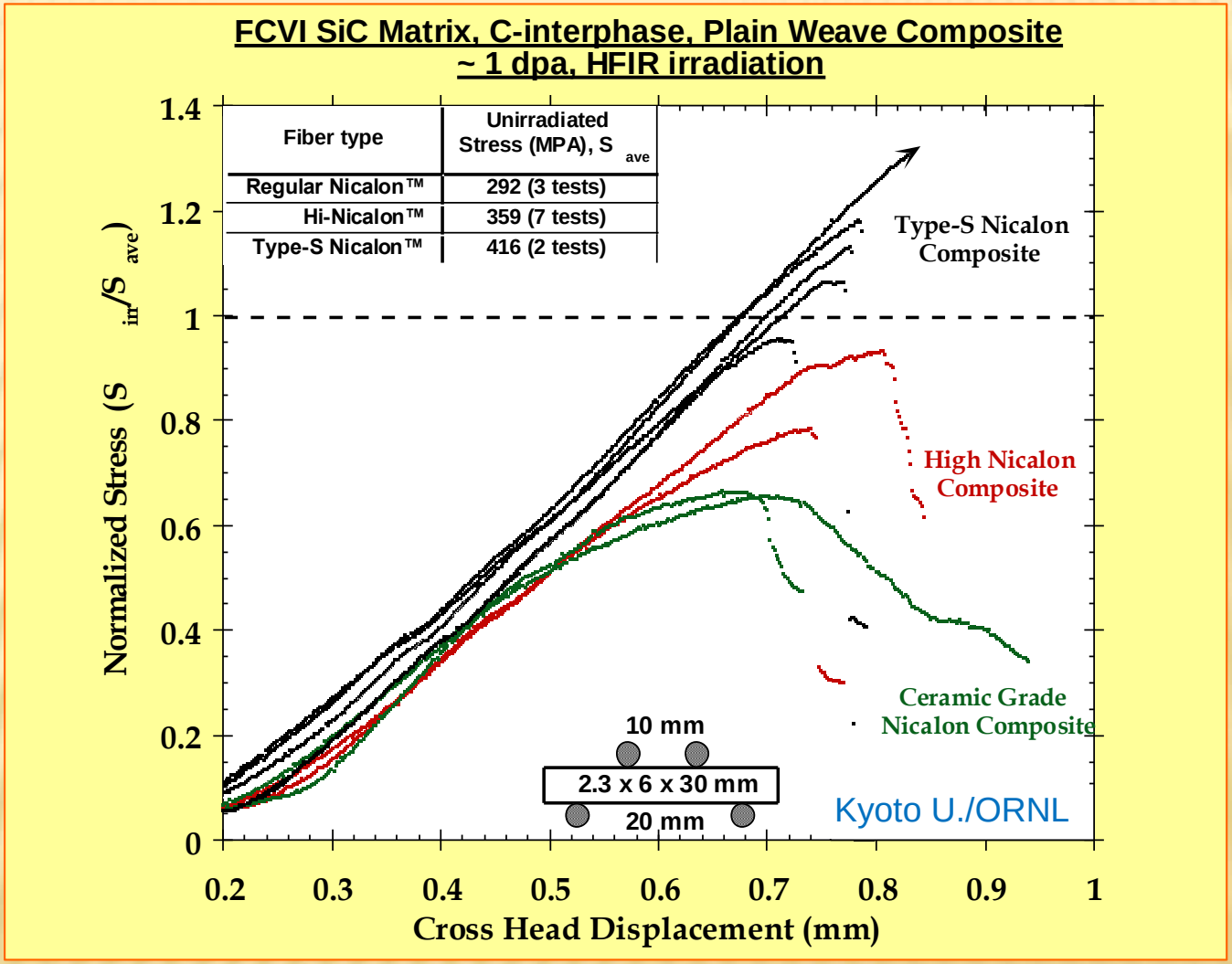
Stoichiometry Si-C Fiber (Hi-NicalonTM -Type-S)



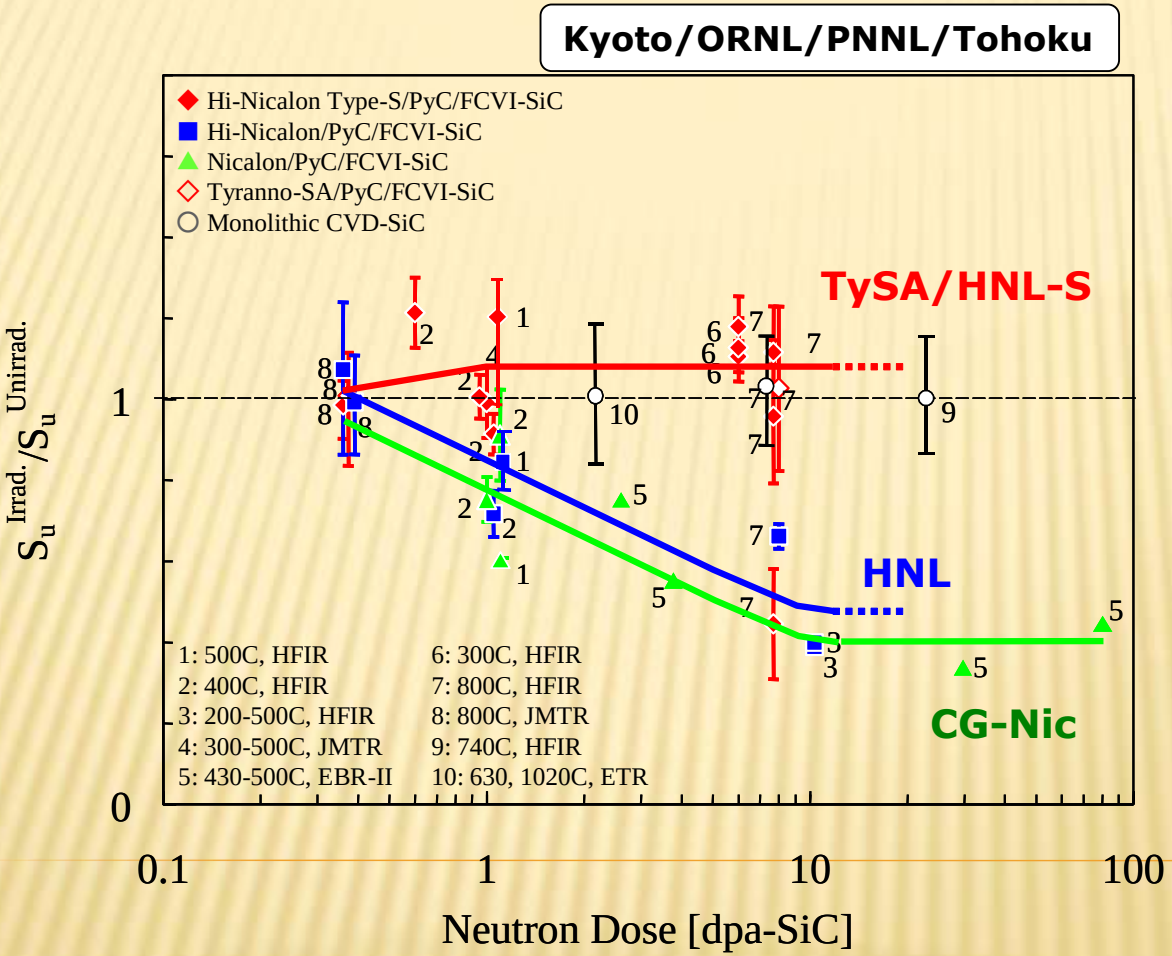
Excellent Stabilities in; Dimension, Strength

JMTR Irradiation: at (573K +) 773 K, - 1 dpa-SiC

Four Point Bending Strength after Neutron Irradiation



EFFECT OF NEUTRON IRRADIATION; STRENGTH VS. FLUENCE



- Advanced SiC-fiber composites with a thin pyrolytic carbon interphase maintain the original strength up to at least 10 dpa of neutron irradiation at 300~800C.
- Absence of progressive strength degradation in the conventional composite ('CG-Nic') suggests tolerance of advanced composites to higher fluence levels provided that the interphase maintain its functionality.

[1, 2] L.L. Snead, et al., JNM 283-287 (2000) 551-555.
[3, 4] T. Hinoki, et al., PRICM4 (2001), to be presented.
[5] R.H. Jones, et al., 1st IEA-SiC/SiC (1996)
[6, 7] L.L. Snead, et al., JMR, submitted.
[8] T. Nozawa, et al., ICFRM-10 (2001), to be presented.
[9] R.J. Price, et al., JNM 108-109 (1982) 732-738.
[10] R.J. Price, et al., JNM 33 (1969) 17-22.

Radiation Damage Study on SiC

- Objective and Procedure -

To get clear insights about microstructural development under irradiation
- Need to clarify electronic excitation effect -

Microstructure Analysis by TEM:
"In-situ" observation under HVEM.

"Post-ion-irradiation" observation by TEM

Modeling :
The kinetic rate theory calculation of defect accumulation by a conventional computer

Research Facilities for Radiation Damage Study for Fission and Fusion Materials

- **Fission Reactor**

(HFIR, *FFTF/MOTA, EBR-II, HFBR,* BR-II, HBWR, JMTR, JOYO)

- **Ion and Neutron Sources**

(Simulated Irradiation with Ion Beam such as DuET, HIT, TIARA, *RTNS-II*)

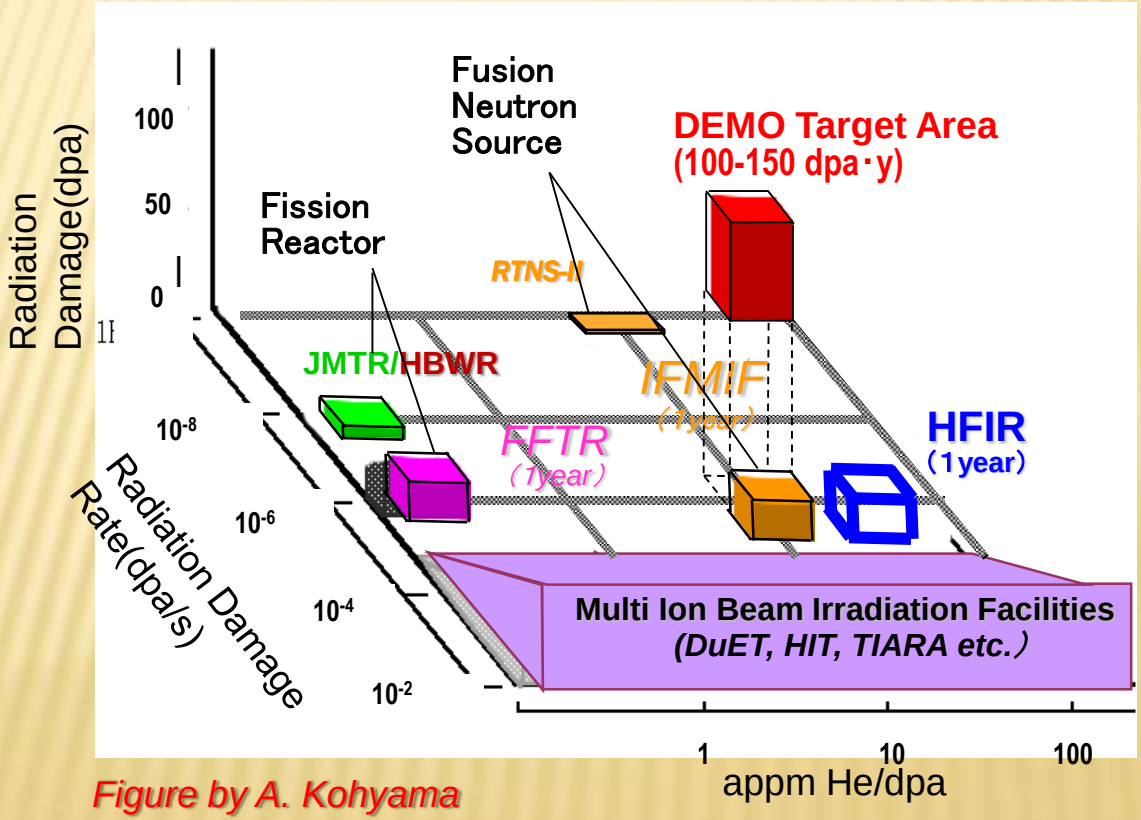
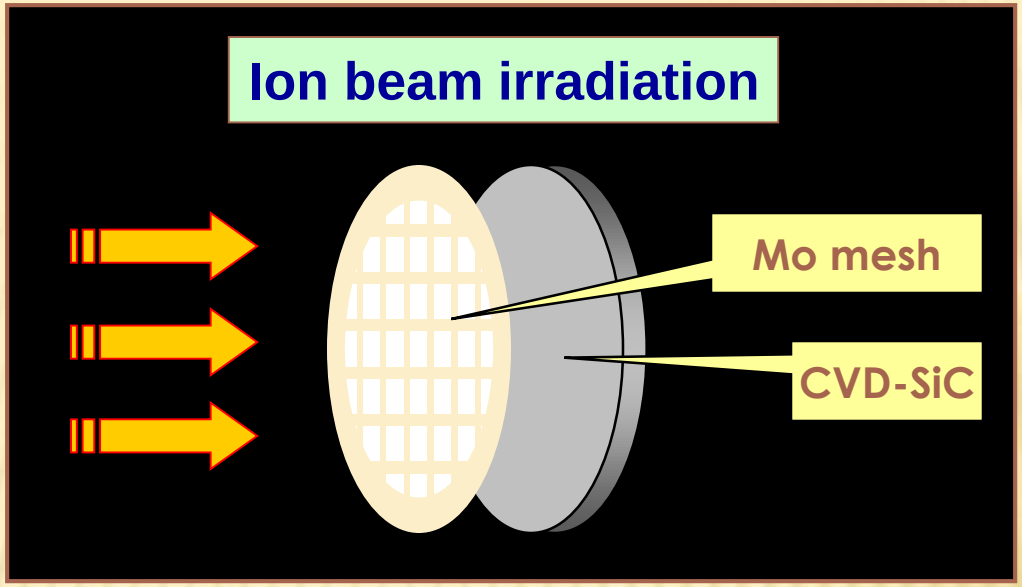
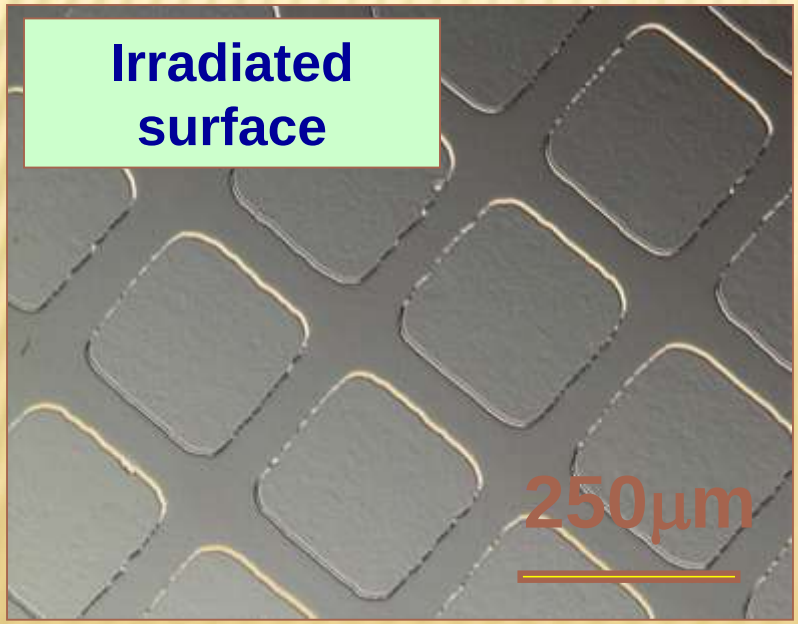
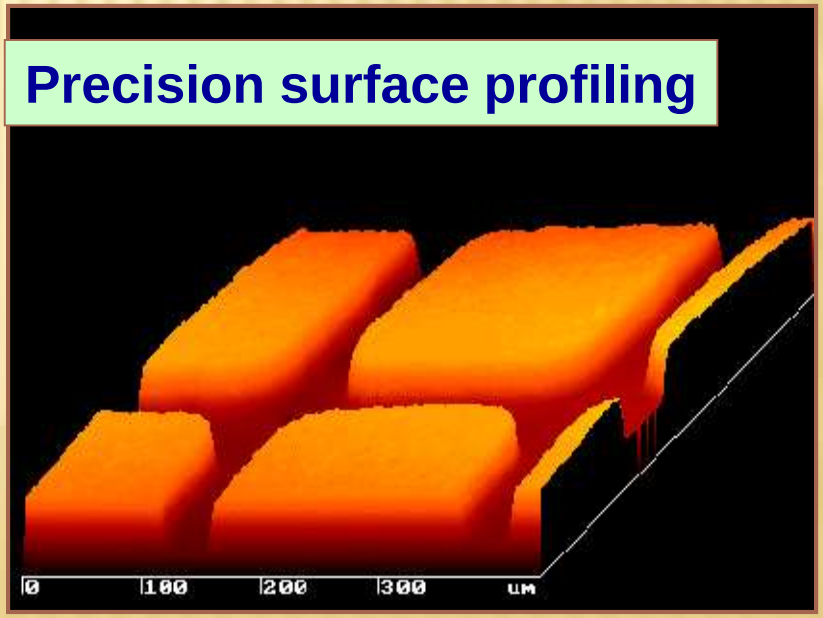


Figure by A. Kohyama

Ion Irradiation and Surface Profile Characterization

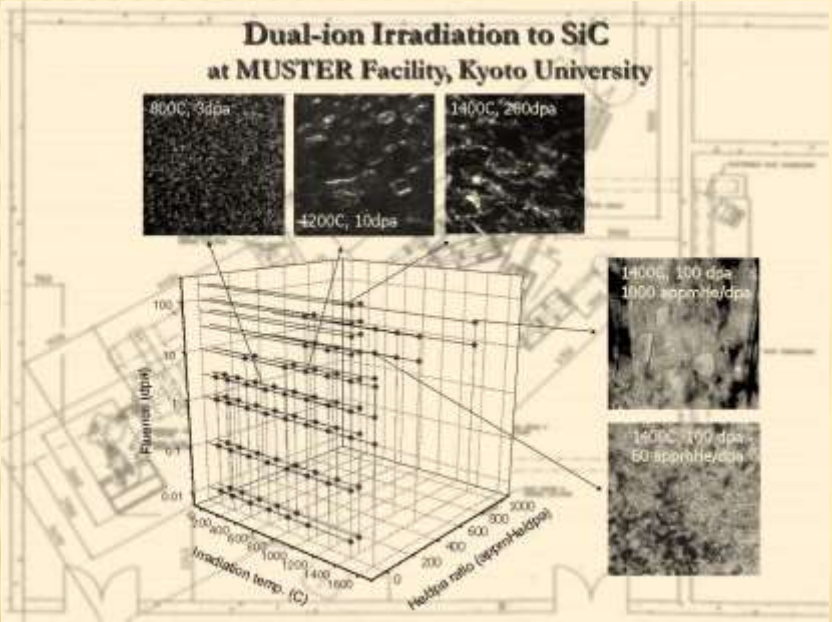




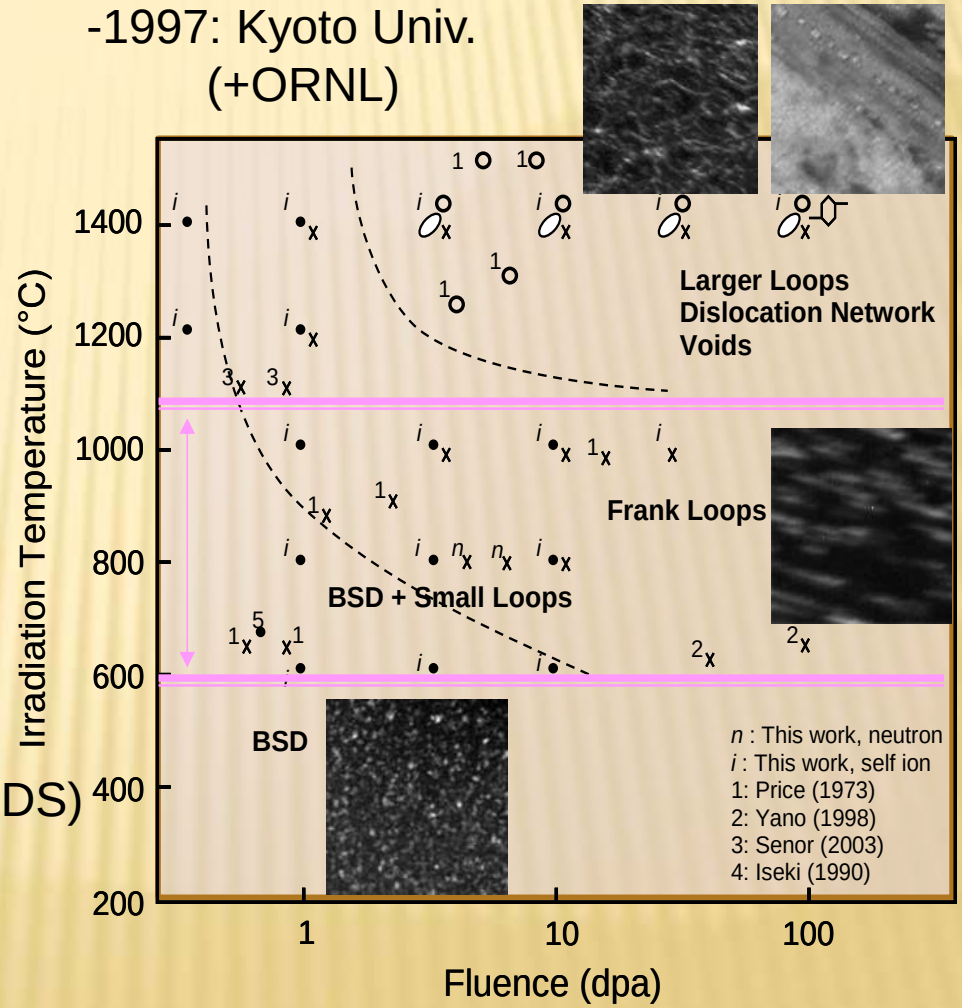


Significant Advancement in Mapping

- Intrinsic Microstructural Stability under Radiation Damage -



-1997: Kyoto Univ.
(+ORNL)

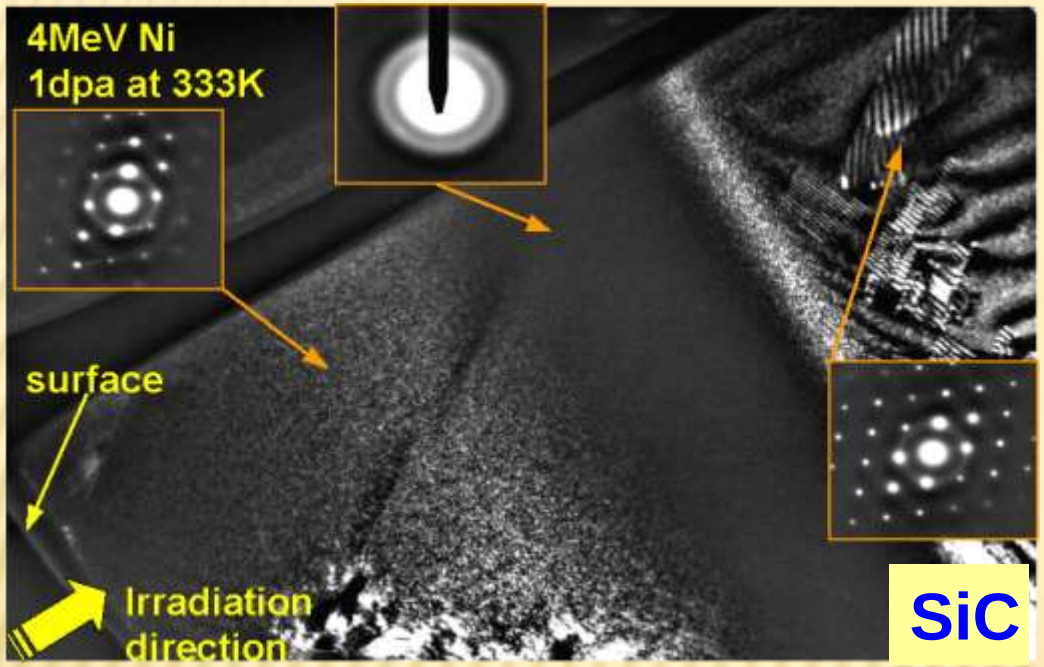


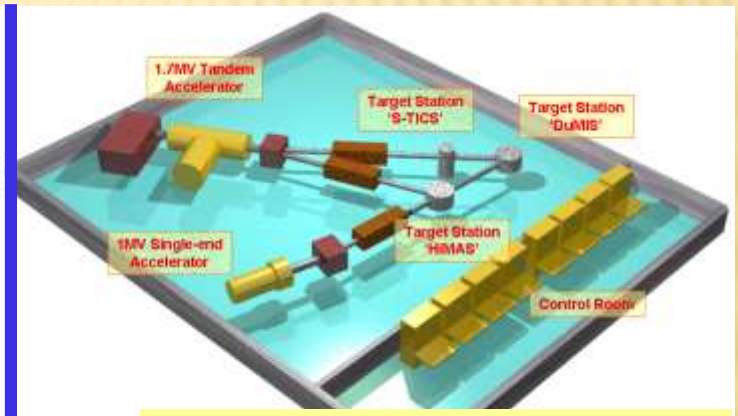
1976: ANL Dual-ion exp.; SUS316
1977-: in-situ HVEM (Univ. Tokyo); 316
1985-:HIT Facility (Univ. Tokyo); JLF-1(ODS)
1997-: DuET Facility (Kyoto Univ.); +SiC
Later I named as MUSTER Facility

Mid-temperature range may suggest little concern about swelling/degradation

Advanced Multiple Beam Irradiation Facility-DuET

a powerful tool for analysis and basic study of microstructural evolution under neutron damage





The world most advanced Multiple-Beam Facility: DuET(Kyoto Univ.) Provides
Varieties of data
With high accuracy
Under excellent control

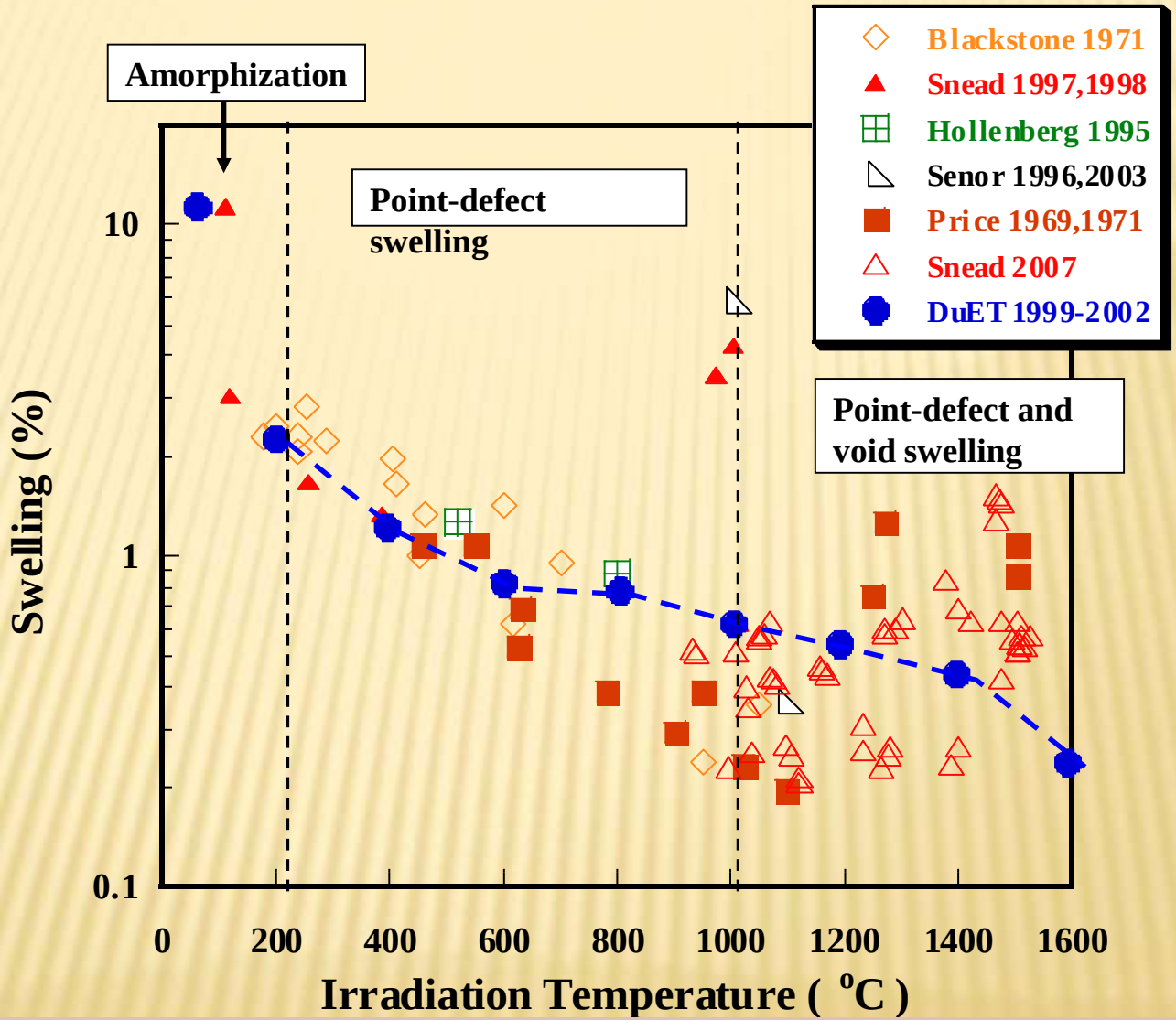
Point defect accumulation

Deformation by swelling

Amorphization behavior (critical condition for amorphization)

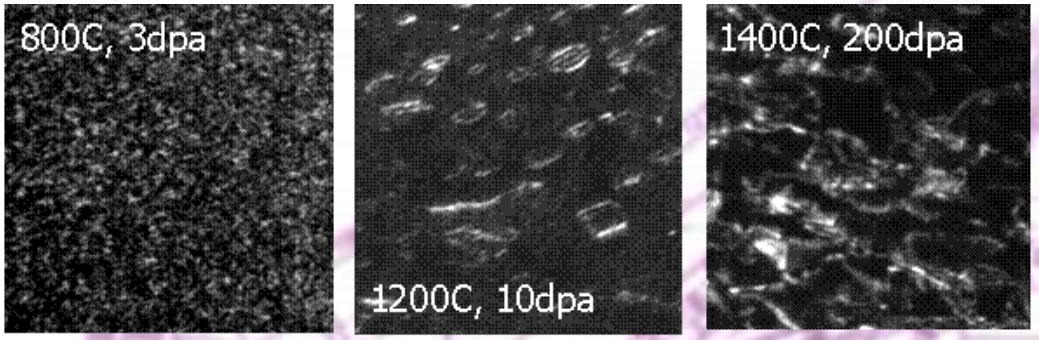
IAE, Kyoto University

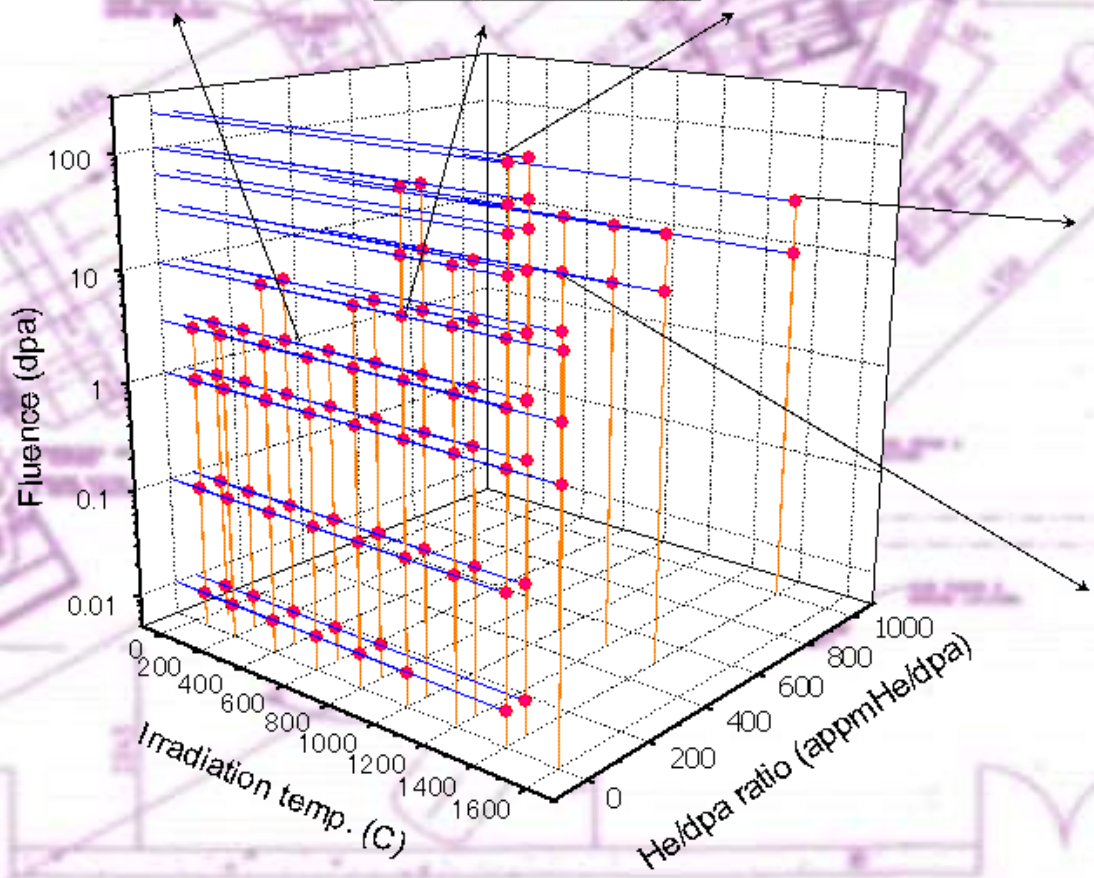
Swelling of SiC by Neutron and Ion Irradiation

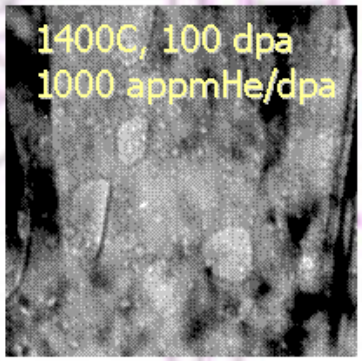


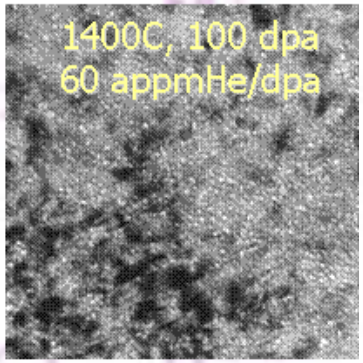
The credibility of SiC as a fusion material had been presented by the ion irradiation research using accelerators

Dual-ion Irradiation to SiC
at MUSTER Facility, Kyoto University

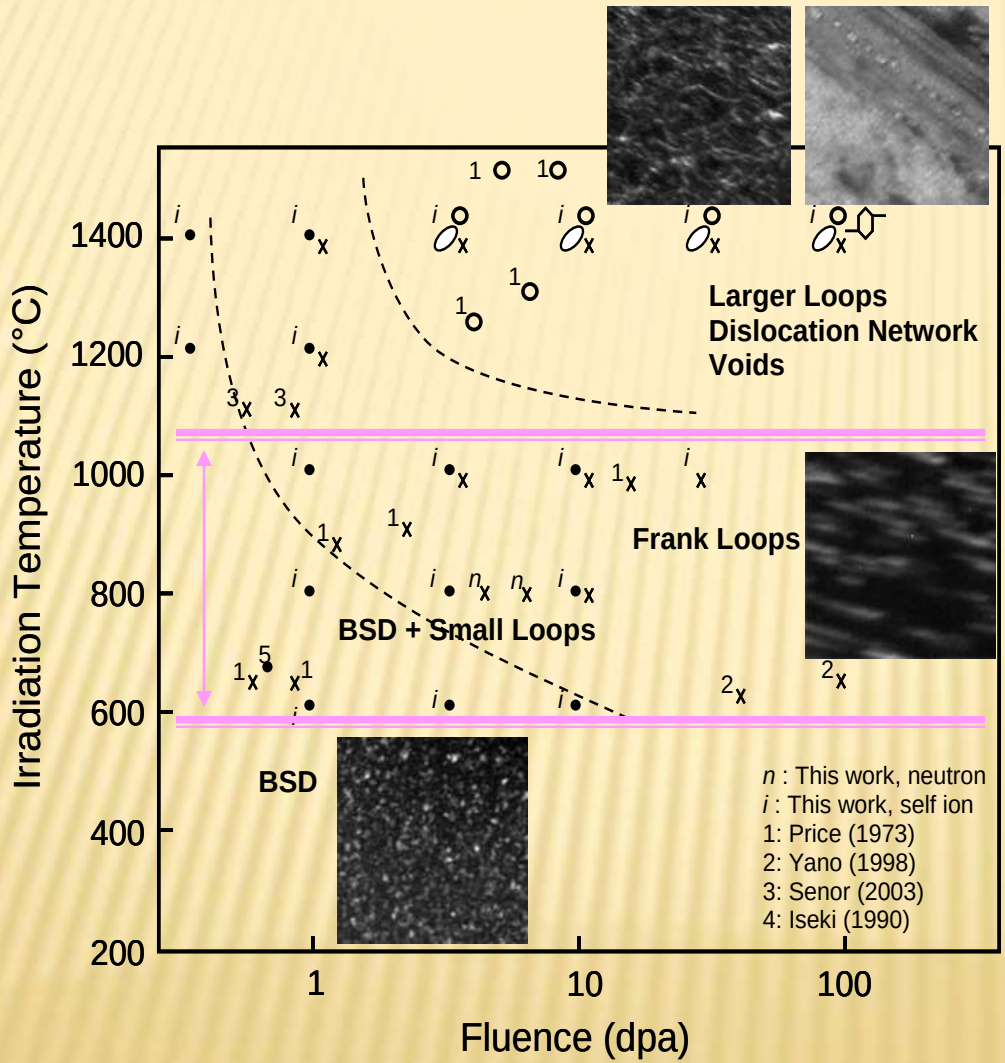








Significant Advancement in Mapping
- Microstructural Development -

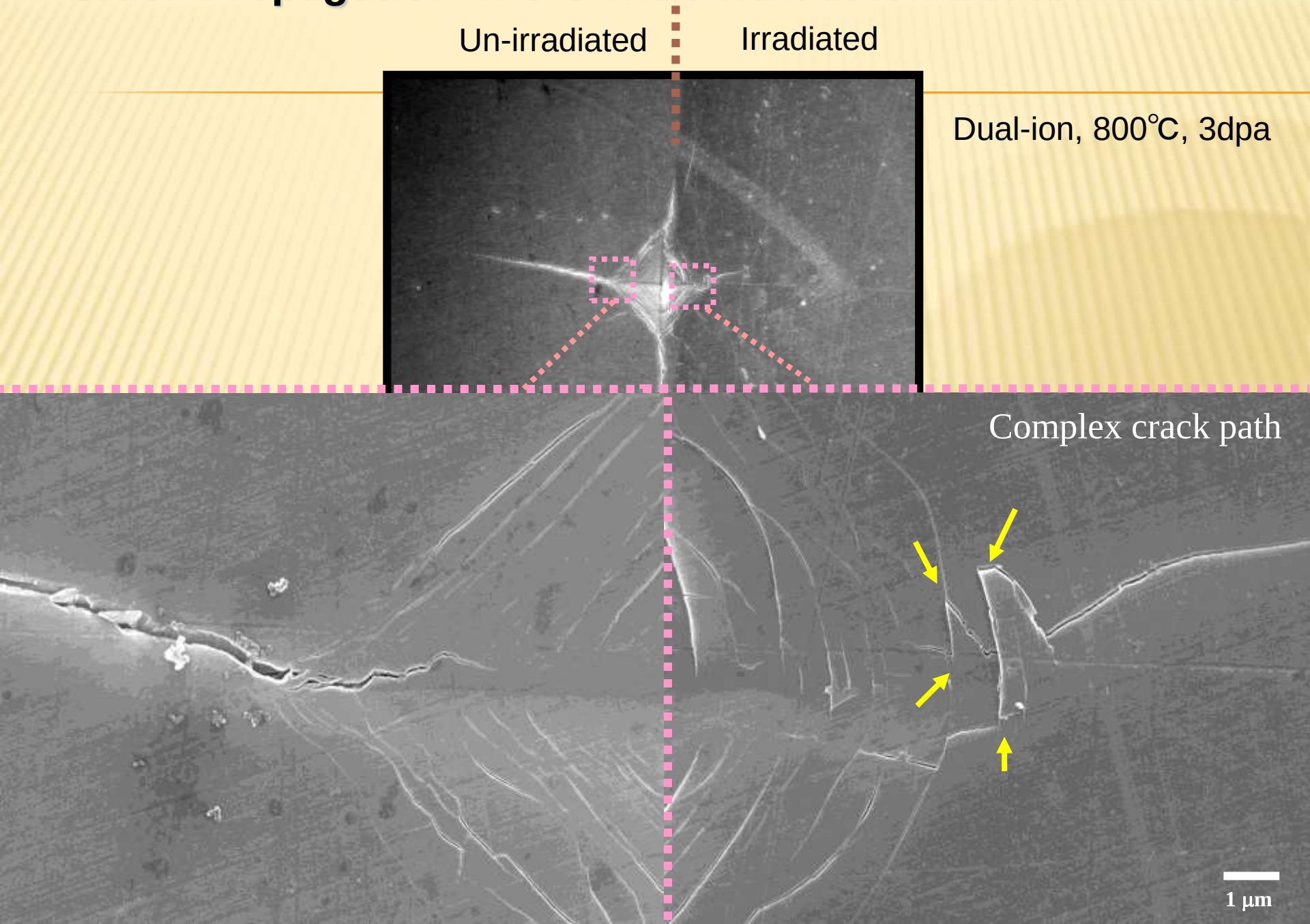


- Black Spot Defects (BSD)
- Small / Frank Loops
- Large Loops
- Dislocation Network
- Voids

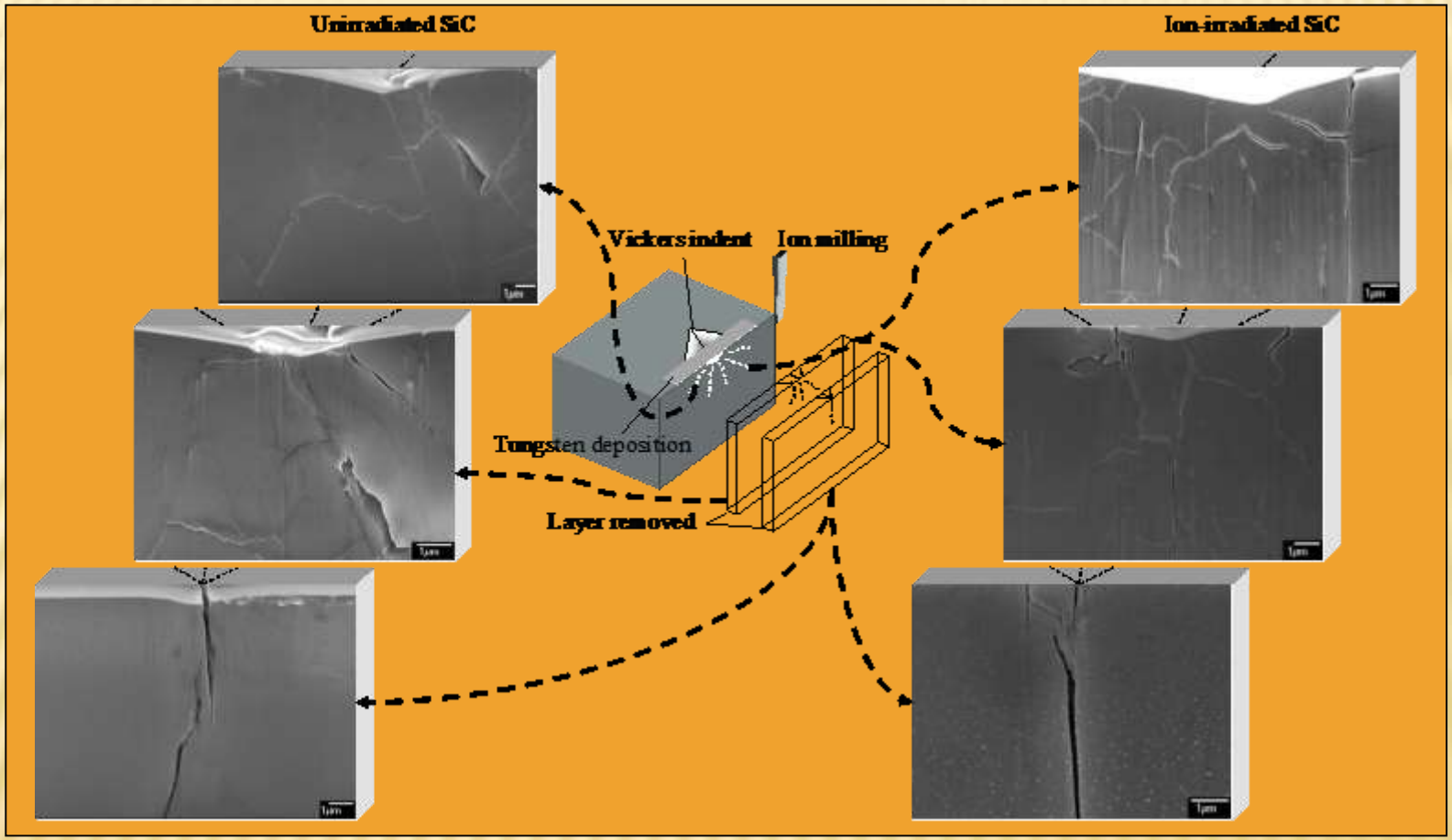
- **'Black spots' are probably small loops or SIA cluster as loop embryo.**
- **Perfect loops develop into dislocation network at high T and high doses.**
- **Microstructural development in the high-T & high-dose regime is similar to some of fcc metals.**

Mid-temperature range may suggest little concern about swelling/degradation

Crack Propagation in SiC with/without Ion Bombardment



3D Analysis of Crack Propagation in SiC
（1st: by FE-SEM）

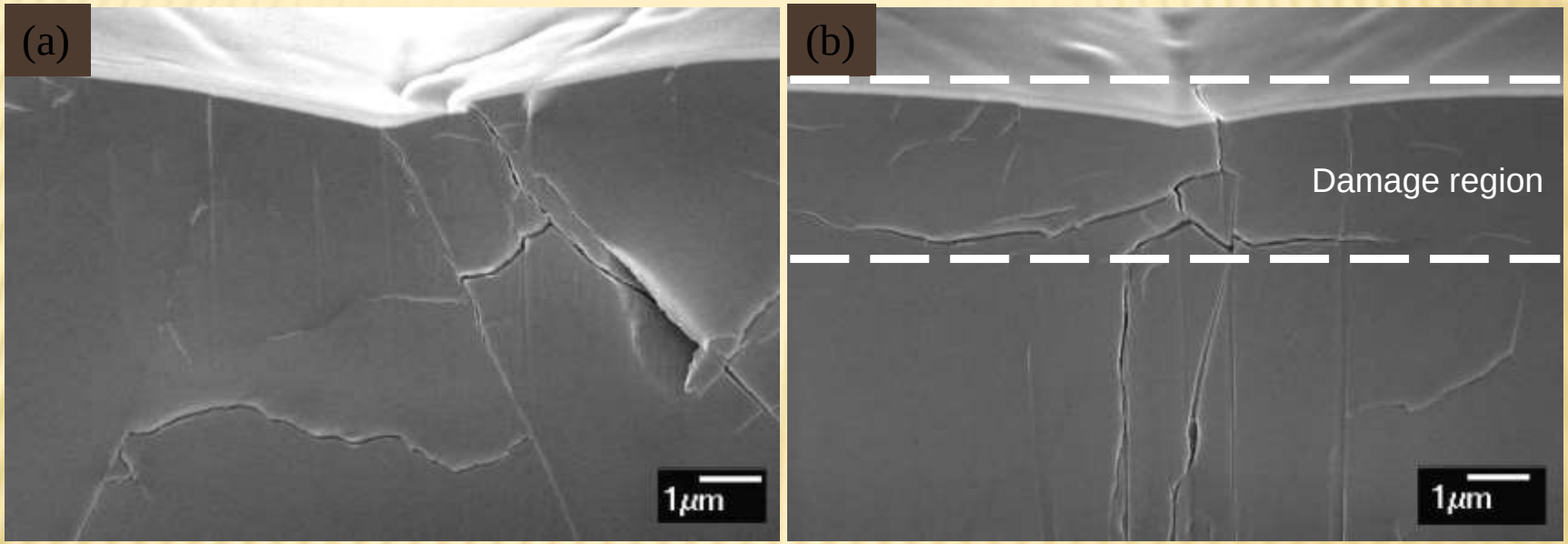


Thin-foil slicing by Ga ions (FIB)

Crack Propagation in SiC with/without Ion Bombardment

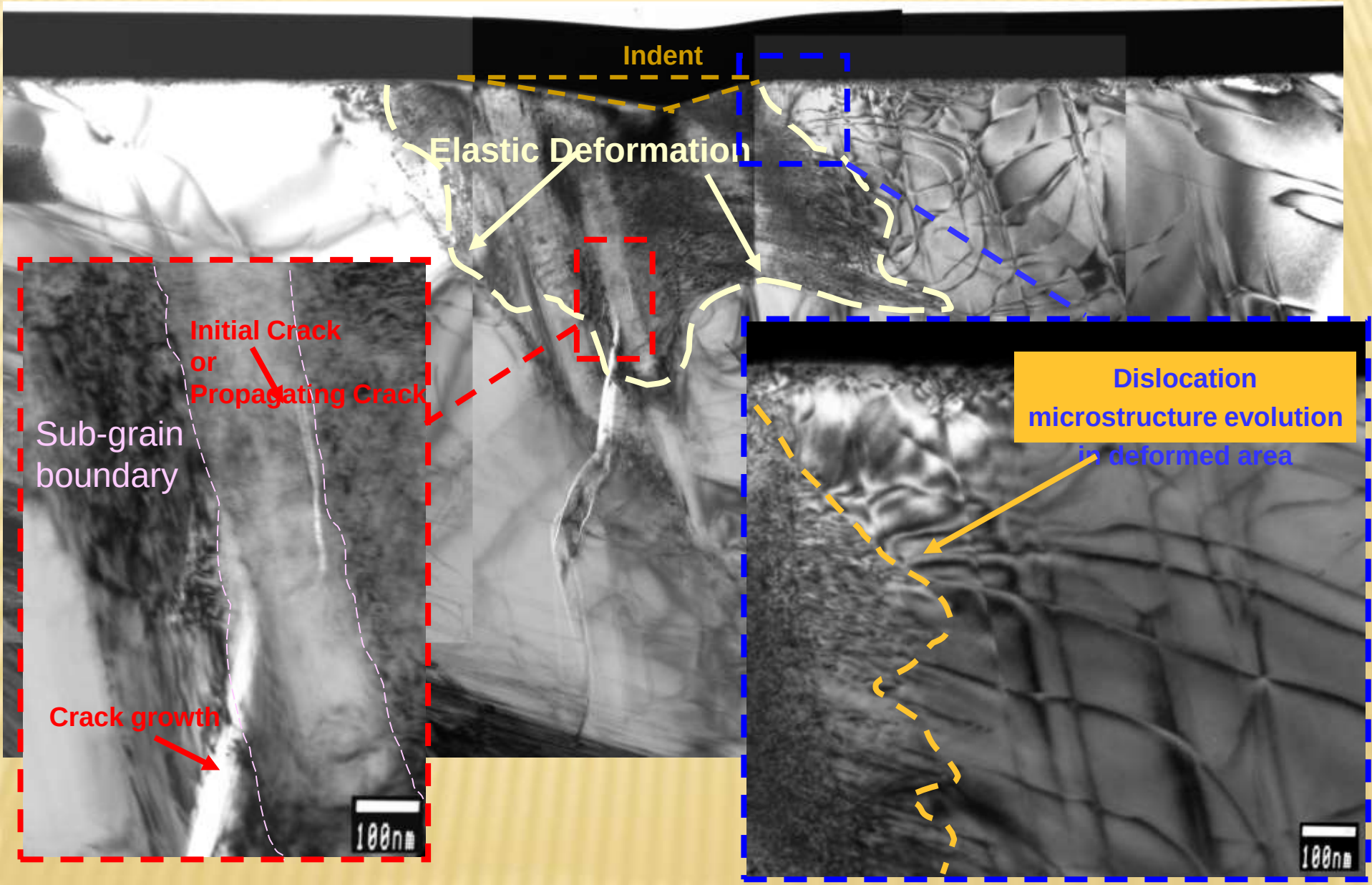
Un-irradiated SiC

Duai-ion Irradiated SiC
600ºC, 10dpa



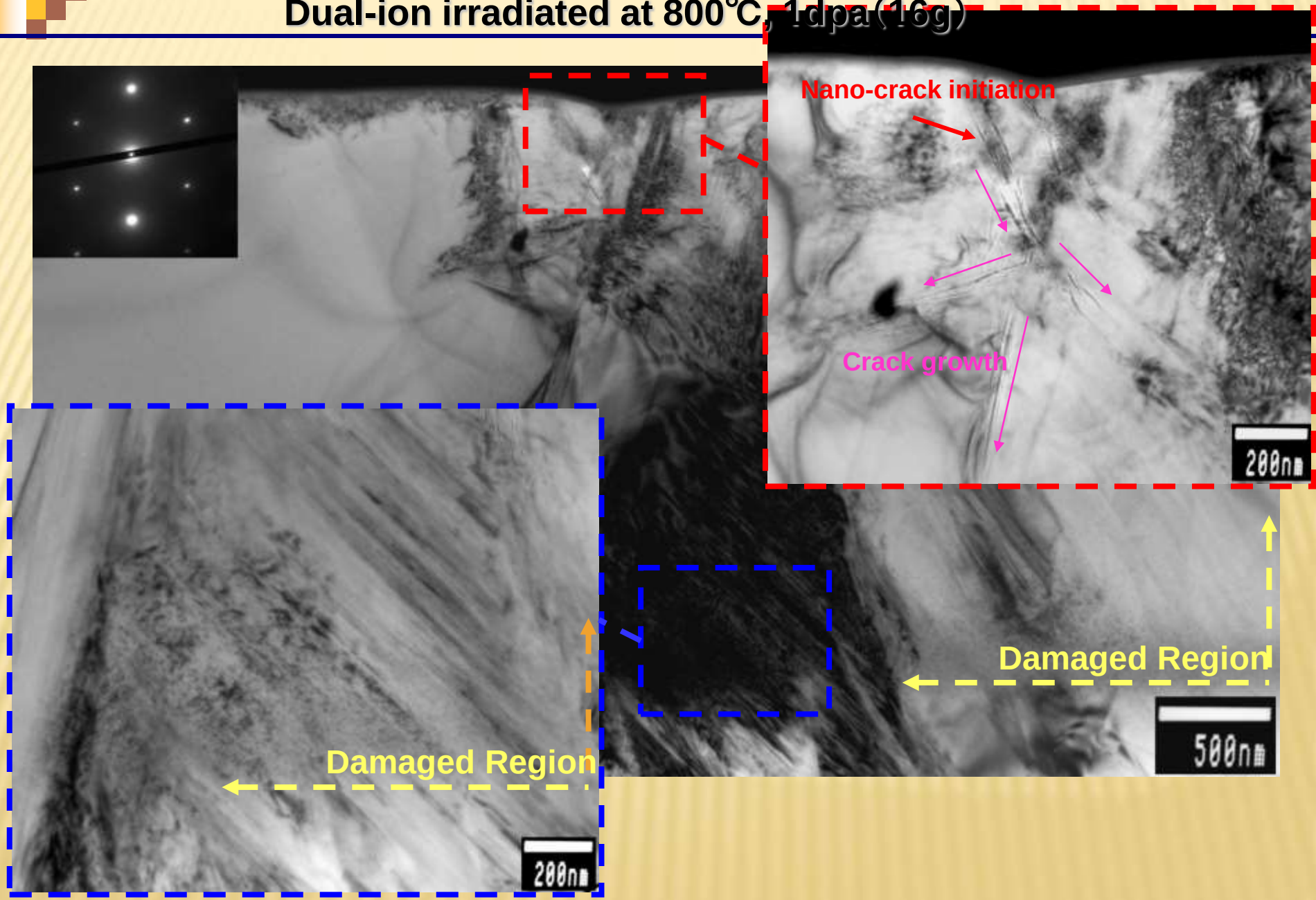
- **Lateral crack propagation near damage peak**
- **Vertical crack propagation beneath the damage region**

Microstructure in un-irradiated SiC
（after 16g indentation）



Microstructure after indentation test

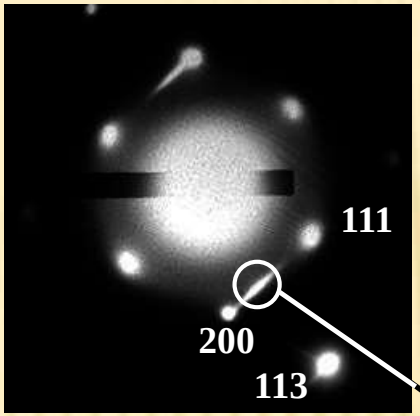
Dual-ion irradiated at 800℃, 1dpa (16g)

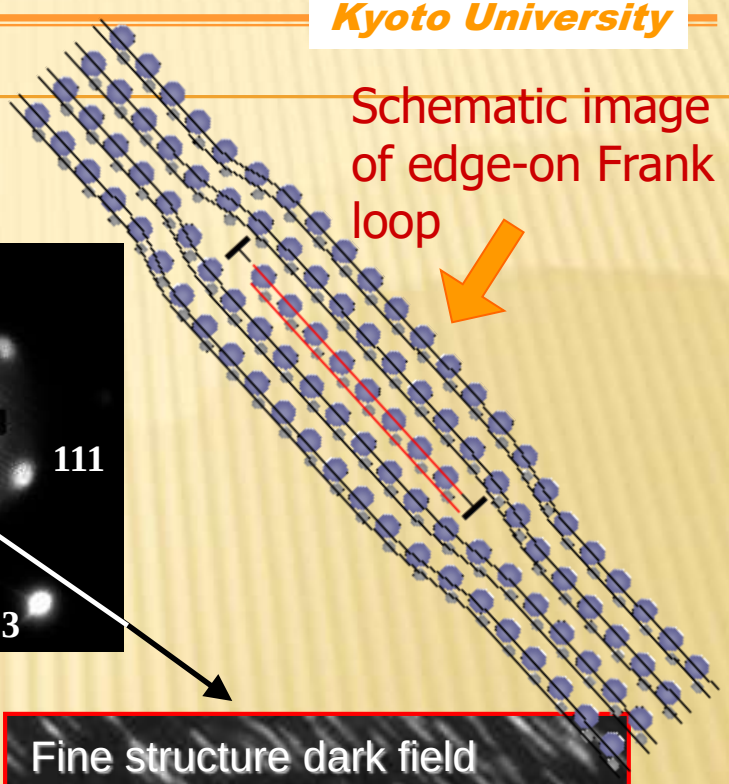


TEM Imaging Method for Frank Loops

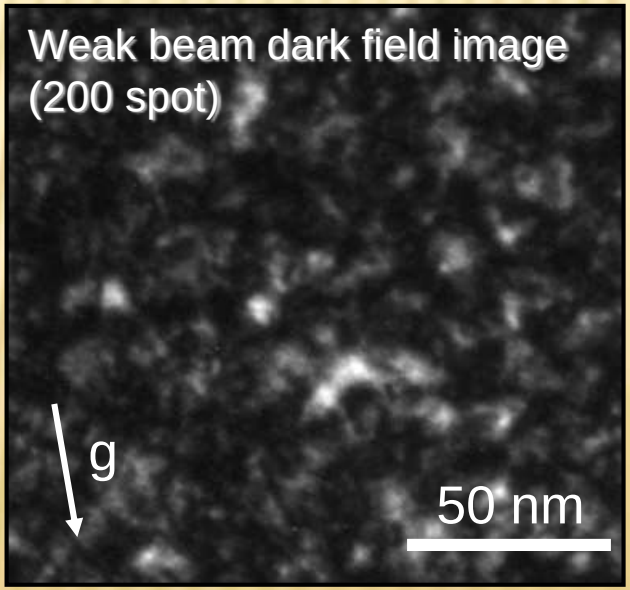
Kyoto University

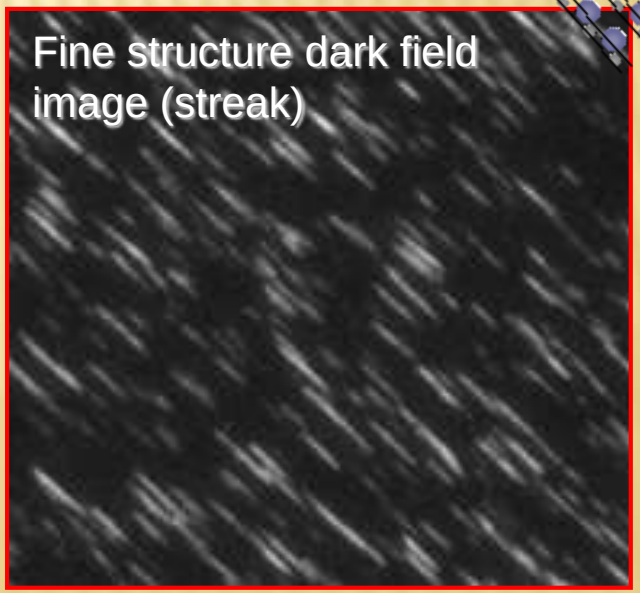
- Frank loops are faulted dislocation loops lying on {111} family planes.
- The streak (encircled) are due to presence of the {111} stacking faults in the loops and can be used to image the Frank loops independent of the matrix.
- Using the streak allows the Frank loops to be measured to sizes less than 1 nm.



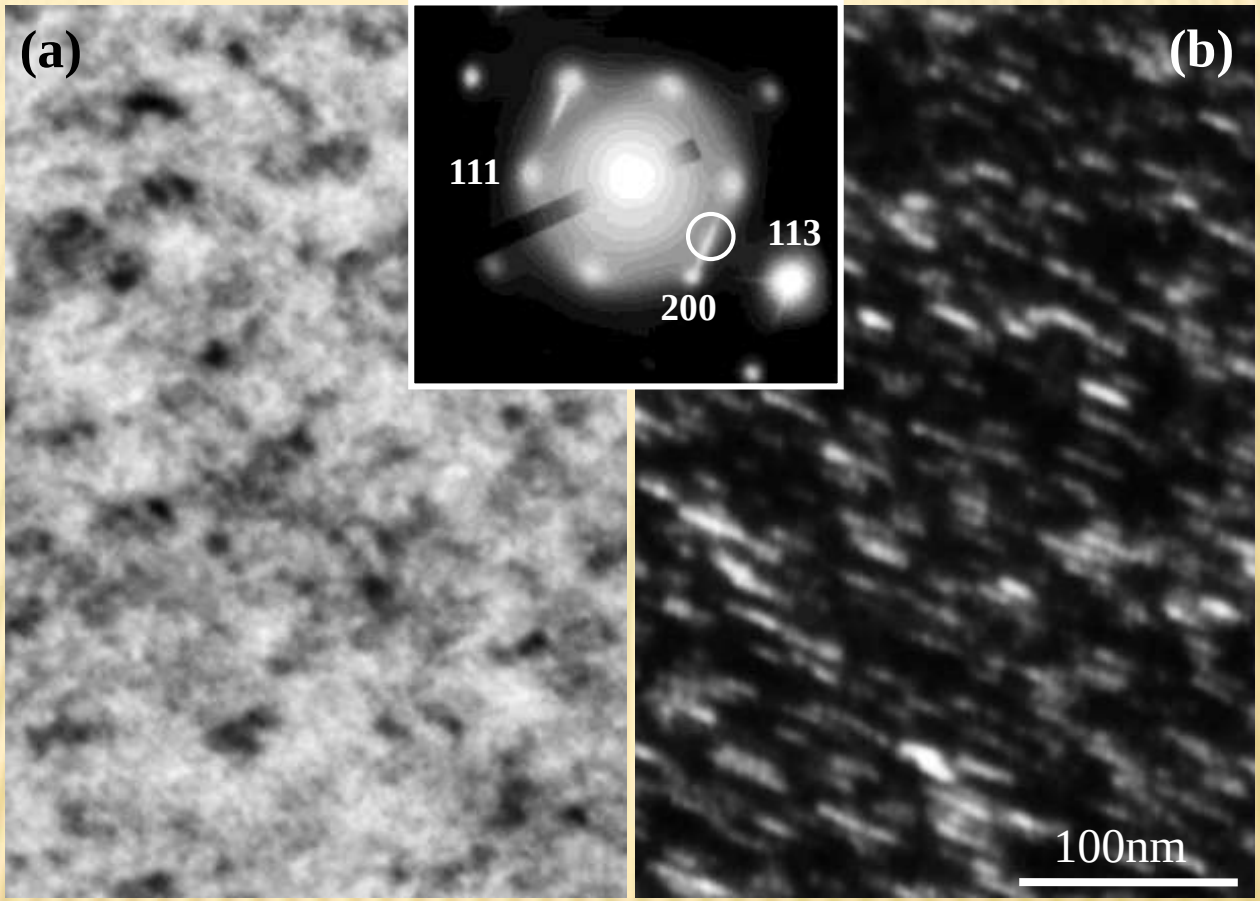


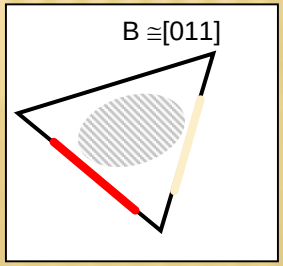
Schematic image of edge-on Frank loop





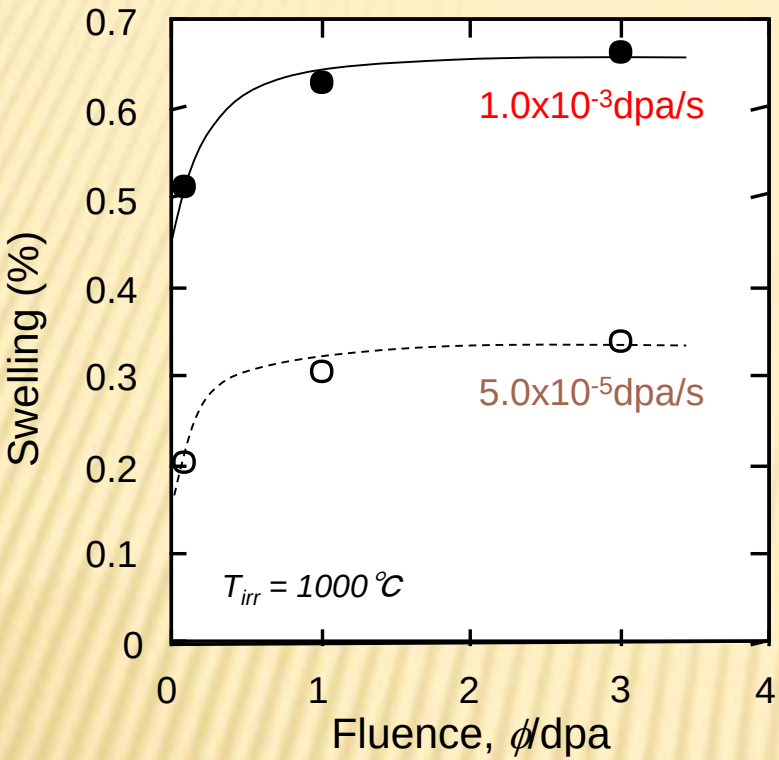
Typical Frank Loops



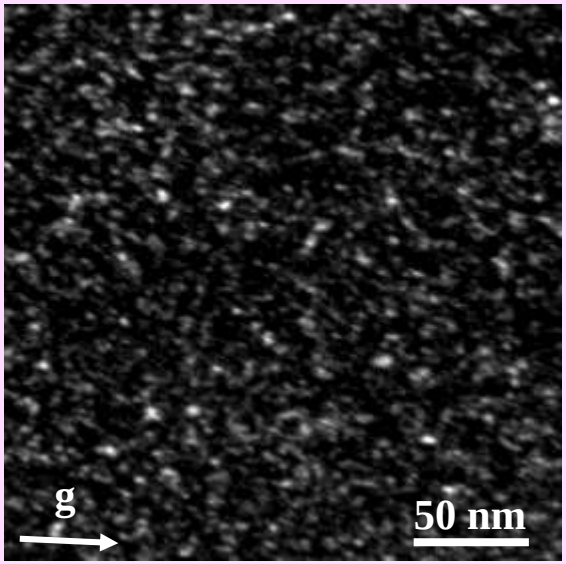


Damage rate dependence on swelling & microstructure

Kyoto University



- Swelling in lower dpa/s conditions show approximately half the values in higher dpa/s conditions.
- Higher dpa/s condition delay the growth of loops.



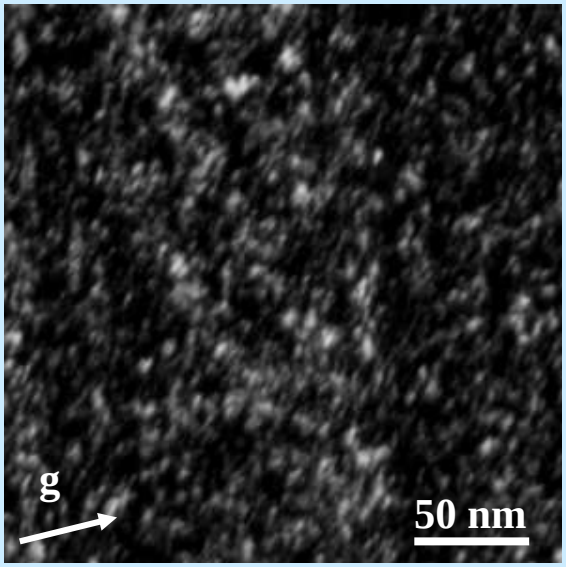
1.0×10^{-3} dpa/s

3 dpa

ΔV = 0.66 (%)

$N_L = 2.6 \times 10^{23}$ (m^{-3})

d_L = 3.1 (nm)



5.0×10^{-5} dpa/s

3 dpa

ΔV = 0.32 (%)

$N_L = 6.9 \times 10^{22}$ (m^{-3})

d_L = 5.1 (nm)

EXPERIMENTAL FACILITIES OF OARAI CENTER, TOHOKU UNIV.

Facility	Specification
Instrumented Charpy Impact Testing Machine	Sample size : (1-10) mm^2
FE-SEM	JEOL JSM-6701F
Touch-screen Remote-controlled SEM	JEOL JSM-6010
Superconducting Magnet	JASTEC Magnetic field : 15.5 T (Max.)
Ion Slicer	JEOL EM-09100IS Accelerating voltage : 1-8 kV
Fatigue Testing Machine	Intesco 200kgf, 7 x 10^{-5} Pa, Temp. : 700°C (Max.)
Electric Discharger	Brother, HS-300
Material Testing Machines	Intesco Temp. : 700°C (Max.), Load : 200 kg
Supercritical Water Generator	TOSHIN KOGYO CO., LTD. Pressure : 25 MPa, Temp. : 600°C (Max.)

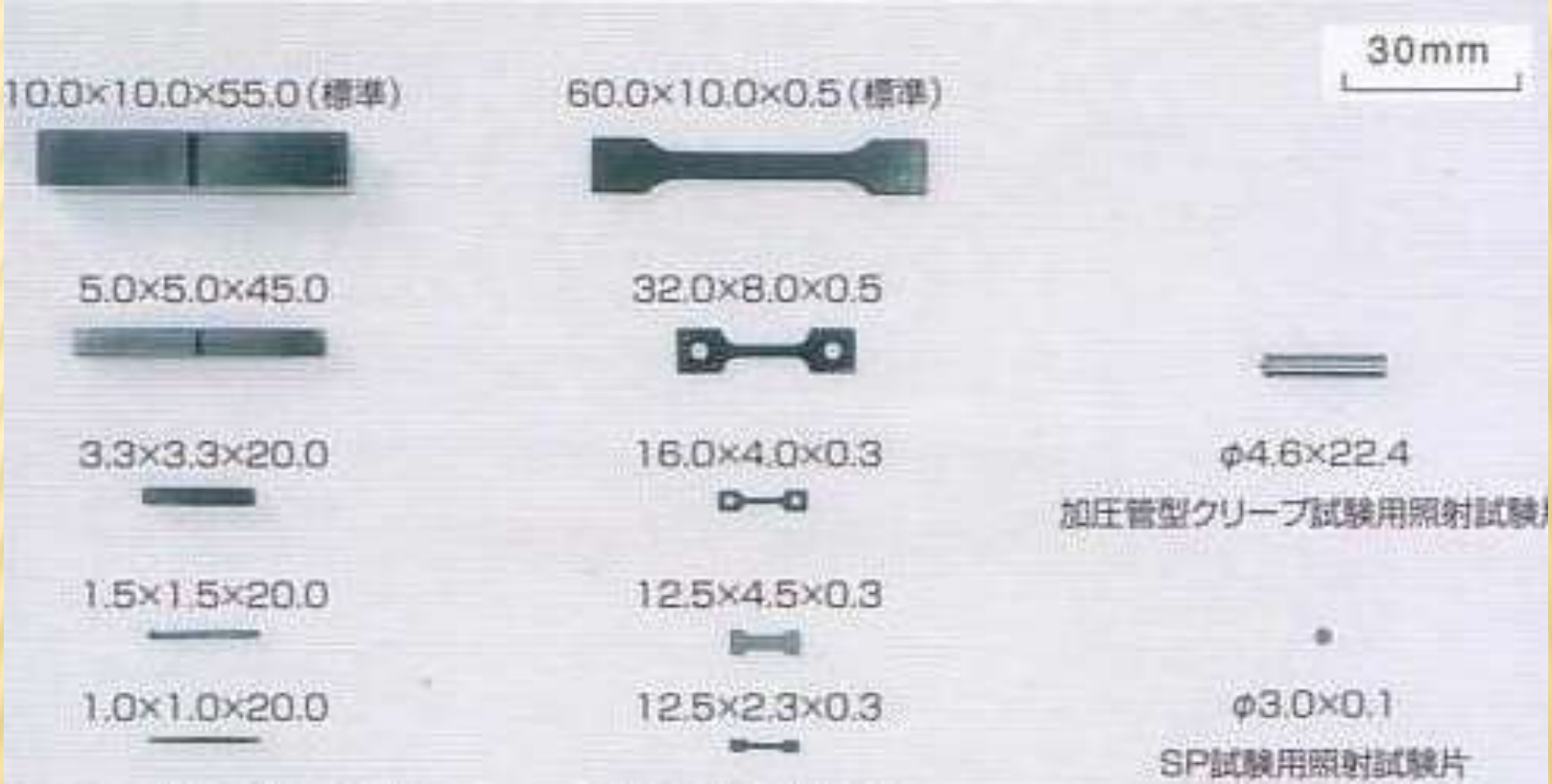
EXPERIMENTAL FACILITIES OF OARAI CENTER, TOHOKU UNIV.

Facility	Specification
FE-TEM	JEM-ARM200F
Positron Annihilation Coincidence Doppler Broadening and Positron Lifetime Apparatus	
Energy Variable Positron Beam Generator	
Three-Dimensional Atom Probe Tomography (3D-AP)	CAMECA LEAP-4000XHR
Focused Ion Beam (FIB) Apparatus	FEI Quanta 200 3D
Thermal Desorption Spectrometry (TDS) Apparatus	Qmass : MKS Microvision2 1-6 Temp. : R.T.-1000°C
Nano-indenter	ELIONIX ENT-1100a
Two-Dimensional Angular Correlation of Positron Annihilation Radiation (2D-ACAR) Apparatus	

EXPERIMENTAL FACILITIES OF OARAI CENTER, TOHOKU UNIV.

Facility	Specification
Hot Cell for Transuranium Elements	Hitachi Zosen
Glove Box for Transuranium Elements	
Tetra-arc Furnace	7×10^{-4} Pa, <10 g
Powder X-ray Diffractometer	Regaku RINT2500V
Dilution Refrigerator	Magnetic field : 15 T (Max.) Temp. : 30 mK
SQUID Magnetization Measuring Equipment	Magnetic field : 5.5 T (Max.) Temp. : 1.8-350 K
NMR Spectroscopes for low and high temperatures	Magnetic field : 12 T (Max.) Temp. : 1.4-873 K
Mossbauer Spectroscopes	Magnetic field : 1.2 T (Max.) Temp. : 3.5-300 K
Ultraviolet Visible Near-infrared Spectroscope	Perkin Elmer lamda750 Wavelength : 190-3300 nm

TYPICAL SPECIMENS FOR SSTM



This has been a standard for international fusion material community.
With sufficient data on size effects including irradiated metallic samples.

PROJECT PROPOSAL TO OARAI CENTER

- 2016 Proposal can be made: by December 9, 2015
- I will meet Director General, Prof. Nagai of Oarai Center: Afternoon of December 1st.
- Brief explanation to him about this BLIP and CERN activity has been done.
- Oarai center collaborative research project can cover PIE cost and travel cost for researchers.

My group is the largest user group and the center needs our collaboration.

- I will try to find another budget for international collaboration.
- H. Kishimoto will be the representative of this proposal.

Summary

- SiC and SiC/SiC composite have been studied as nuclear materials since late 70's using reactors and accelerators. SiC is attractive material especially for severe condition usage such as high temperature, radiation and corrosive environments.
- NITE method for SiC/SiC composite is useful technology to fabricate dense, crystalized SiC/SiC composites. The NITE SiC/SiC composite has excellent thermal stress and thermal shock resistance, experimentally verified.
- Muroran Institute of Technology is developing SiC/SiC targets under the collaboration among KEK and Osaka University for both MuSIC and DeeMe project.
- Fundamental property investigation of SiC/SiC target is being planned to perform at Osaka University/ RCNP /MuSIC.
- SiC/SiC target for DeeMe is a challenge. Muroran and KEK are collaborating to progress the target production. Currently, 1/3-scale mock-up SiC/SiC target has been produced, and a full-size target production is on-going.