# Advanced Materials Studies for High Intensity Proton Production Targets and Windows


**Kavin Ammigan (FNAL),** on behalf of the scientific team and **Taku Ishida (KEK)**

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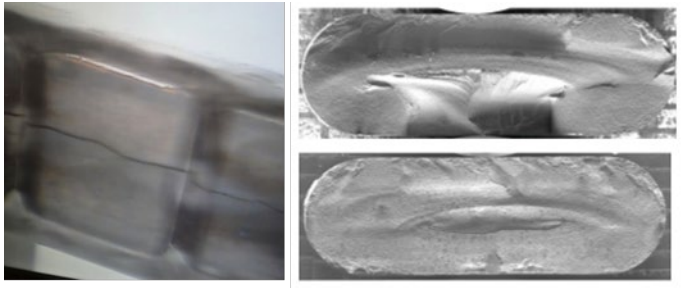
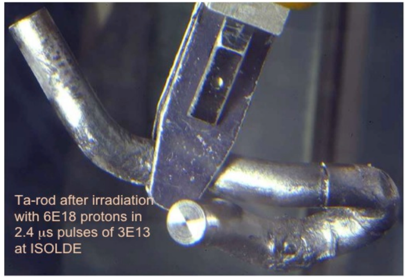
*Lead Principal Investigators

# Motivation from Past Experience

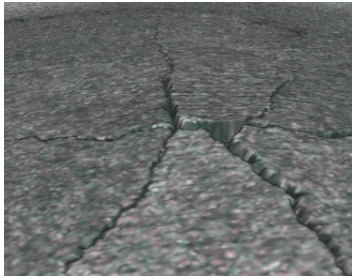


MINOS NT-02 target failure: radiation-induced swelling (FNAL)

MINOS NT-01 target water leak (FNAL)




ISOLDE target (CERN)



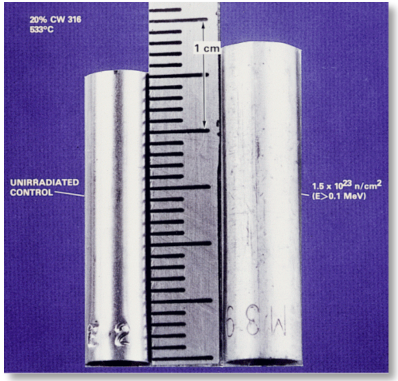
Be window embrittlement (FNAL)

- In the recent past, several major accelerator facilities have had to limit their beam powers due to **target and beam window survivability concerns**
  - Timely research into robust high-power targets and beam windows is essential to fully reap the physics benefit of future accelerator facilities (1.2 – 4 MW)
- **Imperative to research radiation damage effects to enable:**
  - **Accurate component lifetime prediction,**
  - **Design of robust multi-MW targetry components, and**
  - **Development of new materials to extend lifetimes**

# Primary Challenges to Targetry Materials

**Radiation Damage:** Displacements in crystal lattice expressed as Displacements Per Atom (DPA)

- Hardening and embrittlement
- Creep and swelling
- Fracture toughness reduction
- Thermal/electrical conductivity reduction
- Coefficient of thermal expansion
- Modulus of elasticity
- Transmutation products (H, He gas production (appm/DPA) can cause void formation and embrittlement)




D.L. Porter and F. A. Garner, J. Nuclear Materials, **159**, p. 114 (1988)

**Thermal Shock:** Sudden energy deposition from pulsed beam

- Localized area of compressive stress generated due to fast expansion of the material surrounded by cooler material
  - 1 MW target: ~250 K in 10 μs pulse (2.5 x $10^7$ K/s)
- Stress waves move through the material at sonic velocities
- Plastic deformation, cracking and fatigue failure can occur

Iridium target tested at CERN's HiRadMat facility



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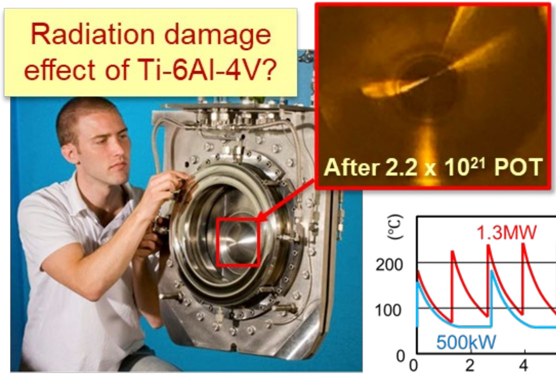
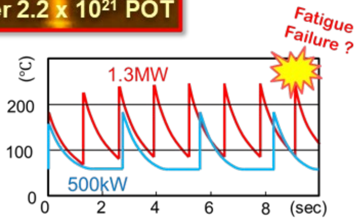
# J-PARC & FNAL Neutrino Target Facilities

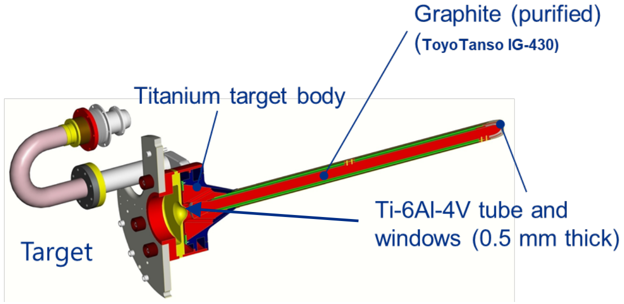
## T2HK 1.3 MW upgrade




| Beam Power | PPP | Cycle | #pulse /100 days | POT / 100 days |
|---|---|---|---|---|
| 500kW (now) | 2.6 x $10^{14}$ | 2.48 s | 3.5 x $10^{6}$ | 0.9 x $10^{21}$ |
| **1.3 MW** | **3.2 x $10^{14}$** | **1.16 s** | **7.4 x $10^{6}$** | **2.4 x $10^{21}$** |
| | **Thermal Shock × 1.2** | | **High Cycle Fatigue × 2.1** | **Radiation Damage × 2.6** |

## LBNF 1.2 MW target design




Both facilities (shielding) built for upgrades to beam-line components to **multi-MW**

- LBNF to 2.4 MW
- T2HK to 1.3+ MW

Targets and windows will experience:

- **Greater radiation damage (~2 DPA/yr for Ti window)**
- **Higher thermal shock and cyclic stresses**
- **Higher temperatures**

# Project Objective and Method

- Study proton **radiation damage effects in key beam-intercepting materials** for future high-power (+1.2 MW) accelerator facilities at J-PARC and Fermilab
  - **Titanium alloys:** primary window (T2HK) & target containment window (T2HK/LBNF)
  - **Beryllium:** primary window & decay pipe window (LBNF)
  - **New materials:** NITE SiC/SiC composite, ductile tungsten (TFGR), …


- 1st BLIP irradiation campaign completed in 2017-2018, 2nd irradiation planned in 2023-2024
- HiRadMat experiments completed in 2015 & 2018, next experiment planned in 2022

# RaDIATE Collaboration

## Radiation Damage In Accelerator Target Environments

https://radiate.fnal.gov


### Objective:

- Harness existing expertise in nuclear materials and accelerator targets
- Generate new and useful materials data for application within the accelerator and fission/fusion communities

### Activities include:

- Analysis of materials taken from existing beamline as well as new irradiations of candidate target materials at low and high energy beam facilities
- In-beam thermal shock experiments

Program manager: Dr. Frederique Pellemoine (Fermilab)

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Pacific Northwest NATIONAL LABORATORY
Los Alamos NATIONAL LABORATORY EST.1943
Ciemat Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas
Science & Technology Facilities Council

# RaDIATE BLIP 2017-2018 Irradiation Summary

- BNL BLIP irradiation executed in 3 phases and target box configurations
  - Over 200 specimens in 9 capsules from 7 RaDIATE institutions
    - Graphite, Beryllium, Aluminum, Titanium, Silicon, SiC-coated graphite, TZM, CuCrZr, Iridium, TaW, MoGr
  - **3 capsules containing Titanium specimens**

| | 2017 | | 2018 | Total |
|---|---|---|---|---|
| | Phase 1 | Phase 2 | Phase 3 | |
| Total µA-hr | 32464.49 | 45614.58 | 124979.89 | 203058.96 |
| Total hr | 226.27 | 302.94 | 789.09 | 1318.30 |
| Total days | 9.43 | 12.62 | 32.88 | 54.93 |
| Total weeks | 1.35 | 1.80 | 4.70 | 7.85 |
| Avg. current (µA) | 143.48 | 150.57 | 158.38 | 154.03 |
| **POT** | 7.30E+20 | 1.03E+21 | 2.81E+21 | **4.57E+21** |

181 MeV rastered beam
3 cm diameter footprint with
154 µA avg. current x 55 days
$4.6 \times 10^{21}$ POT

Comparable to MW facility operation

| Capsule | Capsule position | Material | Irrad. Phases | Peak DPA/p (NRT) | POT | Total DPA |
|---|---|---|---|---|---|---|
| DS Ti1 (KEK) | 6 | Titanium | 1 | 3.38E-22 | 7.30E+20 | **0.25** |
| DS Ti2 (KEK) | 6 | Titanium | 3 | 3.38E-22 | 2.81E+21 | **0.95** |
| US Ti (KEK) | 5 | Titanium | 1, 2, 3 | 3.34E-22 | 4.57E+21 | ~~**1.53**~~ Damaged |
| Si (CERN) | 3 | SiC | 1, 2 | 7.03E-23 | 1.76E+21 | **0.12** |
| | | Graphite | 1, 2 | 2.52E-23 | 1.76E+21 | **0.04** |

Past tensile data on titanium alloys only up to 0.3 DPA


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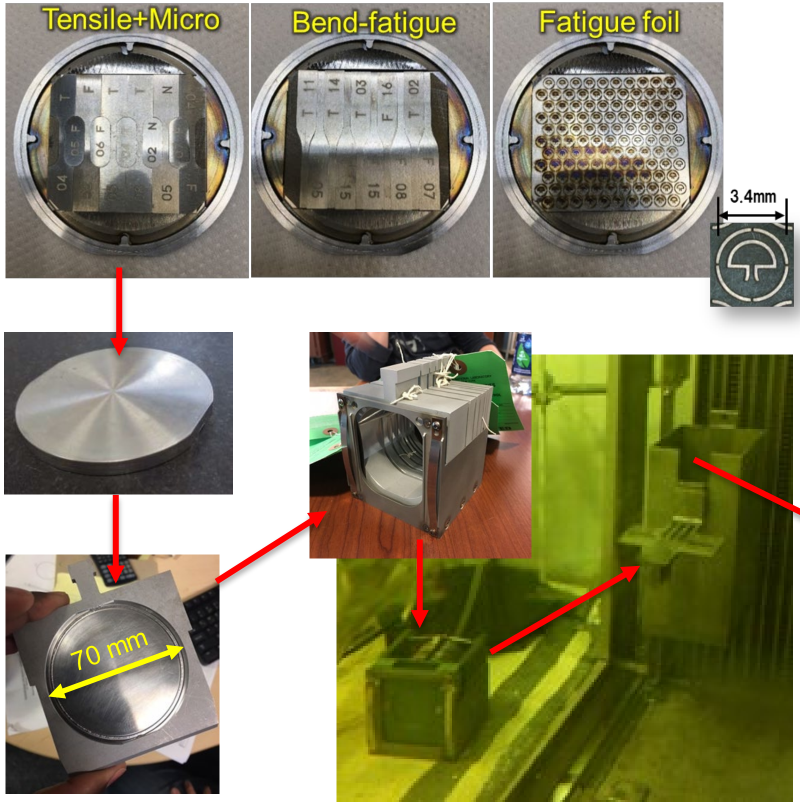
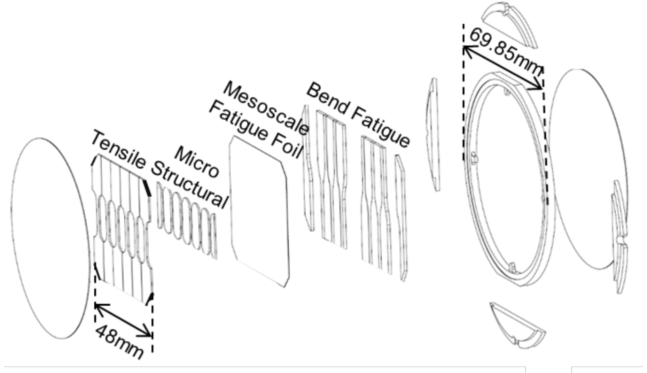
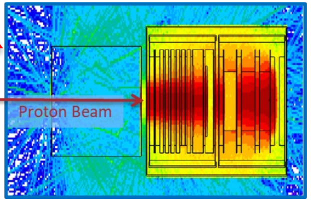
# BLIP 2017-2018 Irradiation of Titanium Alloys




No high-cycle fatigue strength data for irradiated titanium




## Titanium alloys irradiated

| Phase | Grade | Heat Treatment |
|---|---|---|
| α + β | Ti-6Al-4V Gr-5 | Annealed |
| α + β | Ti-6Al-4V Gr-5 | Ultrafine grain |
| α | Ti-3Al-2.5Sn Gr-6 | Annealed |
| β | Ti-15V-3Al-3Cr-3Sn | Solution Treated Aged |
| α | Pure Ti – Gr-2 | Annealed |
| α + β | Ti-3Al-2.5V Gr-9 | Annealed |
| α + β | Ti-6Al-4V Gr-23 ELI | Solution Treated Aged |
| α + β | Ti-6Al-4V Gr-23 ELI | Annealed |



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# U.S.-Japan Target Materials Project Achievements

## I. DS-Ti1 Post-Irradiation Examination (PIE) → publication & press release

- ✓ First observation of **radiation-induced ω-phase production**, that causes **hardening and loss of ductility in Ti-6Al-4V** (current T2HK beam window material)
- ✓ High-cycle fatigue testing shows evidence of **reduced fatigue strength in Ti-6Al-4V**

## II. DS-Ti2 Post-Irradiation Examination (PIE)

- ✓ First investigation of radiation damage behavior of various titanium alloys with **tensile tests and advanced microscopy up to 1 DPA**

## III. Low energy $Fe^{2+}$ ion irradiation at HIT Facility

- ✓ Radiation damage studies of Ti-6Al-4V and candidate titanium alloys **up to 10 DPA**

## IV. Thermal shock tests at CERN's HiRadMat facility → publication

- ✓ First and unique test to compare **thermal shock response of irradiated materials**

## V. Atomic Displacement (DPA) Cross Section Measurements → publication and press release

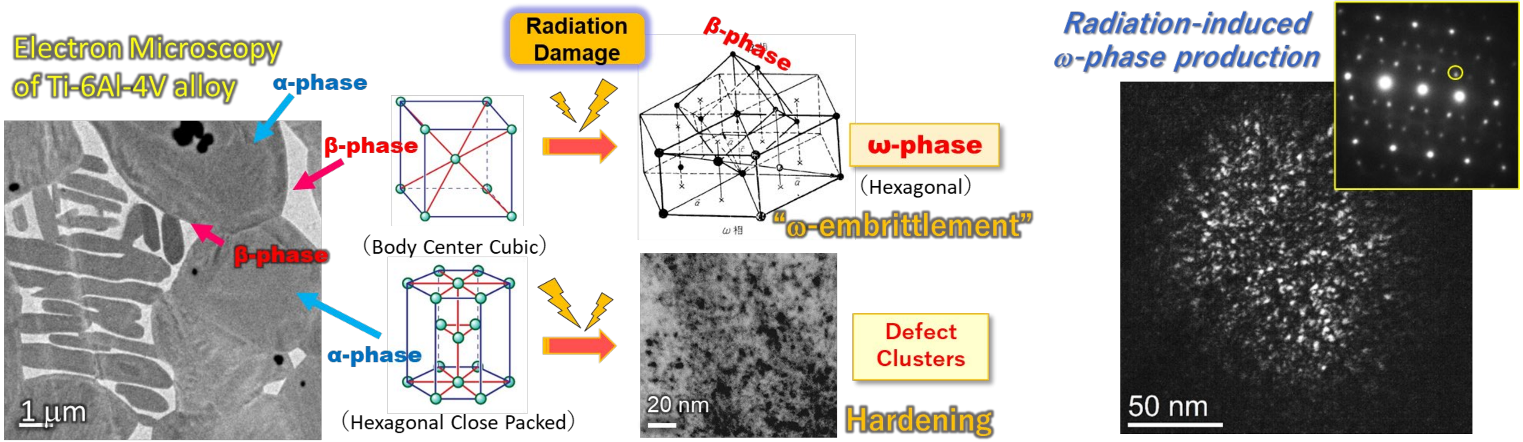
- ✓ Key parameter for exact target/window lifetime prediction **with large uncertainty for high-energy protons**

# Recent DS-Ti1 PIE results

**2020/11/6**
**Press Release**

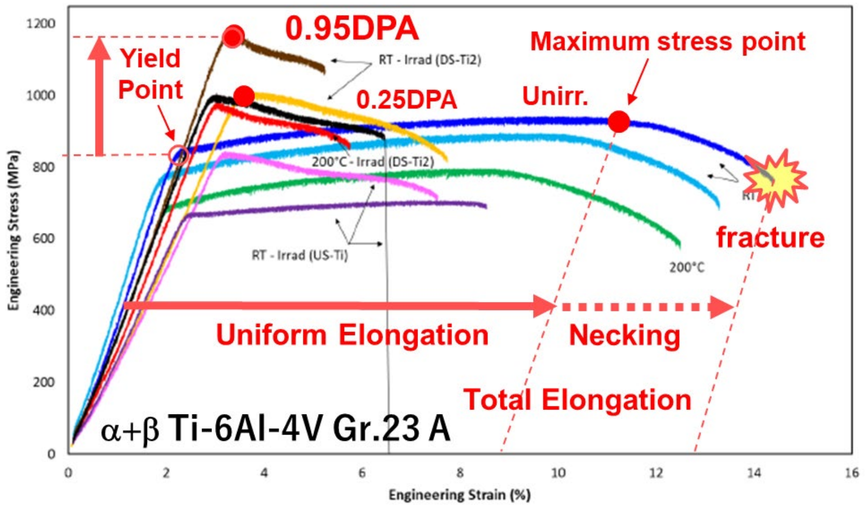
Poster by
T. Ishida

- A high-strength dual α+β phase Ti-6Al-4V (J-PARC/FNAL neutrino facility window) suffers **extreme hardening and loss of ductility from radiation damage after only ~0.1 DPA**
- Advanced microscopy work confirmed high-density of **nanoscale defect clusters** in the primary α phase, further aggravated by high-density nano-sized **radiation-induced ω phase particles** in the β phase **(newly identified by this work)**




T. Ishida, E.Wakai, S.Makimura, D.Senor, A.Casella, D.Edwards et al. Journal of Nuclear Materials 541:152413 (2020)

# DS-Ti2 PIE: Ti-6Al-4V Tensile Test




- Stresses increase beyond Yield Stress (YS), the material elongates uniformly
- The elongation up to the maximum stress point (Ultimate Tensile Strength, UTS) is referred to as "Uniform Elongation (UE)"
  - **It is important to retain UE in a structural material, because it allows for plastic deformation without rapid growth of cracks and sudden failure**
- **Materials are typically hardened after irradiation due to the accumulation of radiation defects**
  - **YS increases, UE reduces**

| RT | DPA | YS | UTS | YS/UTS | UE | TE | ΔYS | ΔUTS | ΔUE | ΔTE |
|---|---|---|---|---|---|---|---|---|---|---|
| Unirr. | 0.00 | 814 | 921 | 88% | 7.05 | 12.10 | | | | |
| T13C | 0.25 | 1002 | 1016 | 99% | 0.20 | 4.85 | 23% | 10% | -97% | -60% |
| T15 | 0.95 | 1169 | 1178 | 99% | 0.10 | 2.37 | 44% | 28% | -99% | -80% |

Ti-6Al-4V loses almost all UE, and stresses decrease immediately after yielding

**"radiation hardening" & "loss of ductility / embrittlement"**



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# Comparison of Ductility between Ti-6Al-4V & Ti-15-3

Poster by
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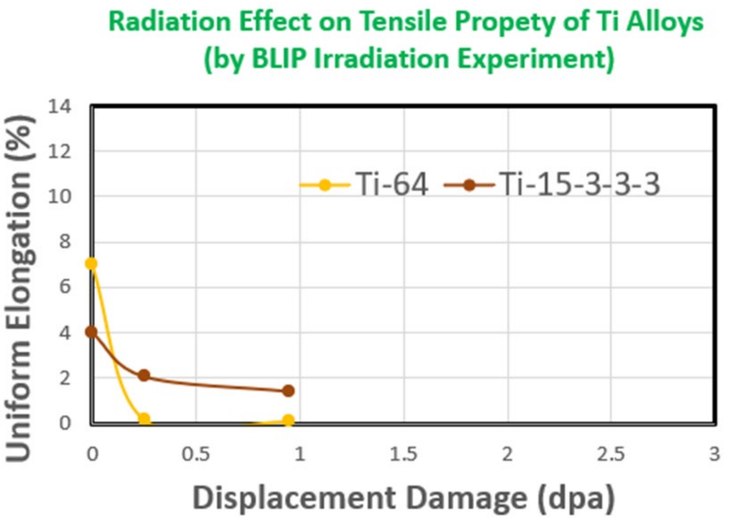


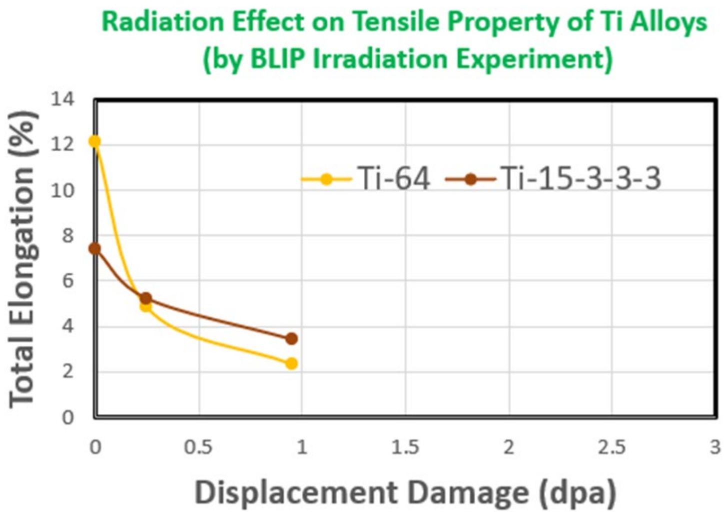


- **Ti-15-3 in STA condition: Higher Strength, Larger Total Elongation (3.4% after irradiation), retain Uniform Elongation**
- **Ti-6Al-4V: Smaller Total Elongation (2.4%), NO Uniform Elongation = deformation only occurs within local region (necking)**



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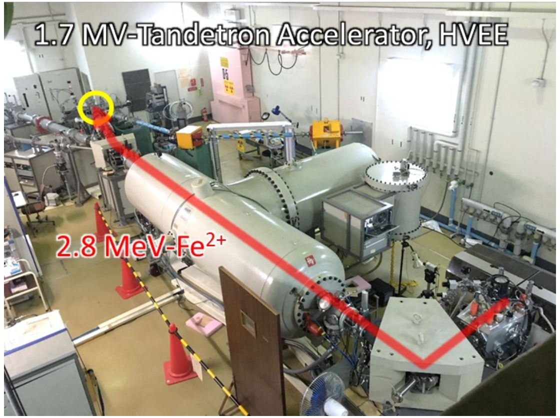
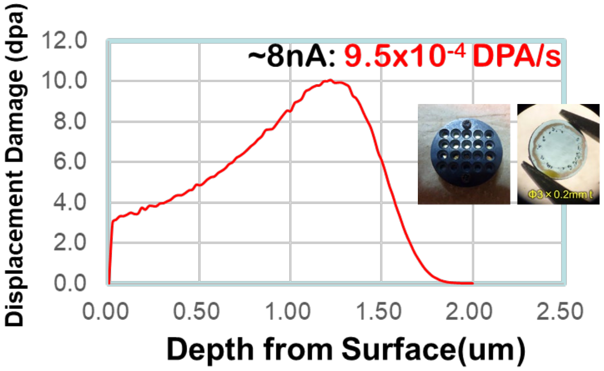
# Low energy $Fe^{2+}$ ion beam irradiation at HIT Facility



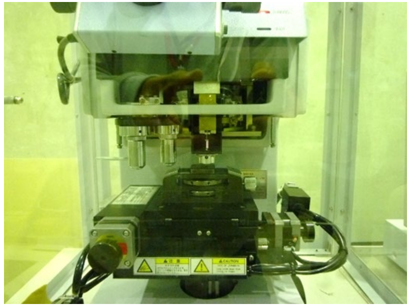

## High Fluence Irradiation Facility at the University of Tokyo

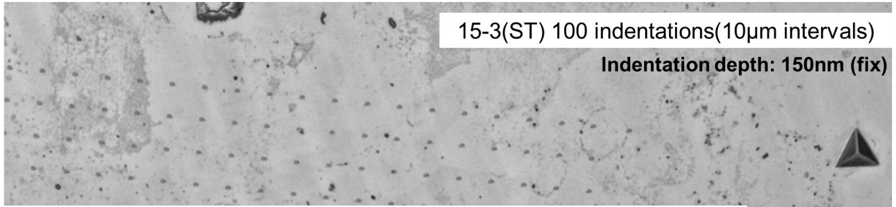



- Accelerated damage rate
  - up to 10 DPA in a few days
- Shorter irradiation time
- No specimen activation

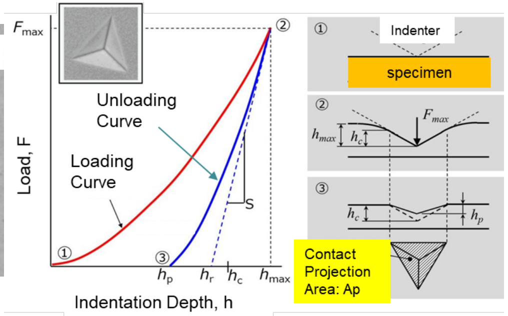
**Effective and fast way to screen materials and to optimize heat treatment at high irradiation dose**

## Nano-indentation hardness test






Hardness is relatively proportional to Yield Strength, but difficult to infer ductility (elongation)



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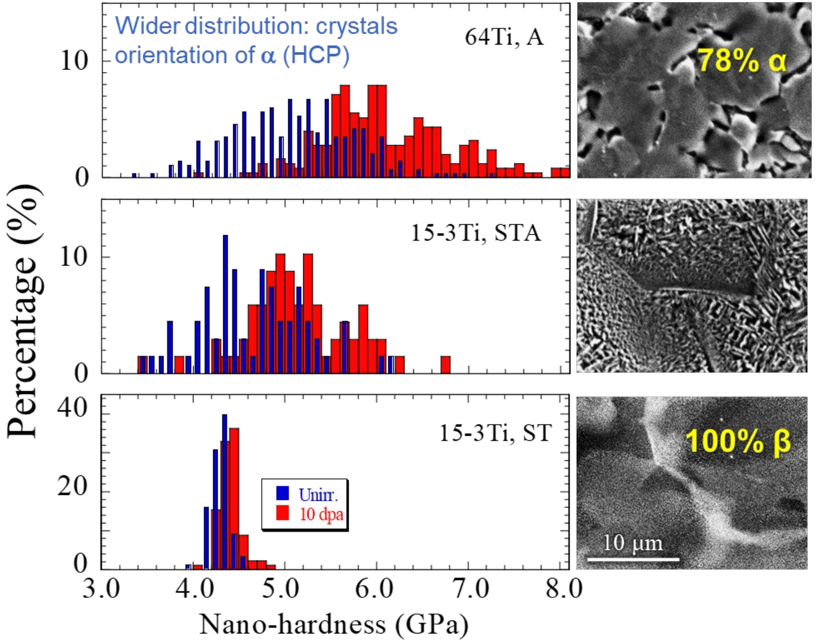
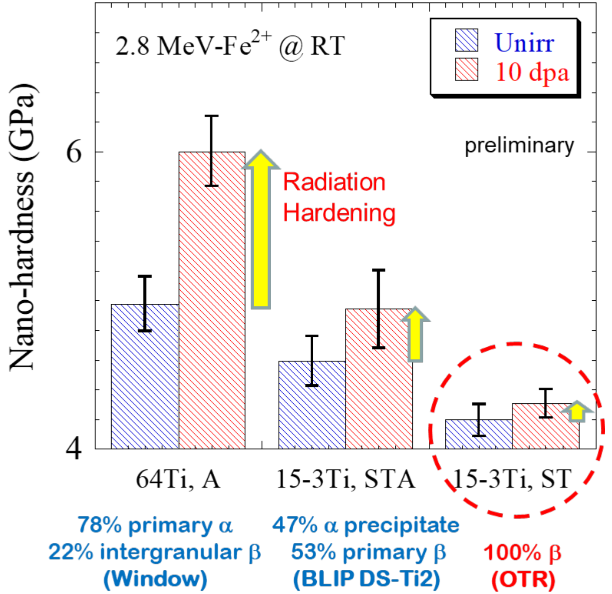
# Titanium Alloy Nano-Indentation Measurements

Poster by
T. Ishida

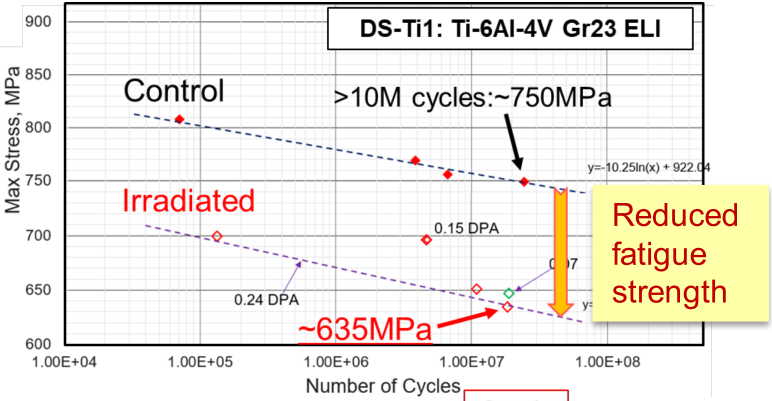


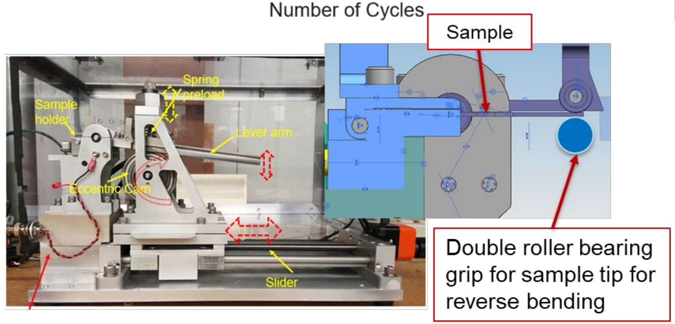

## Contrasting radiation hardening behavior between different titanium alloy microstructures




**Hardness of Ti-15-3 Solution Treatment w/o Ageing (100% metastable β phase) stays the same within the statistical error after the irradiation to 10 DPA**

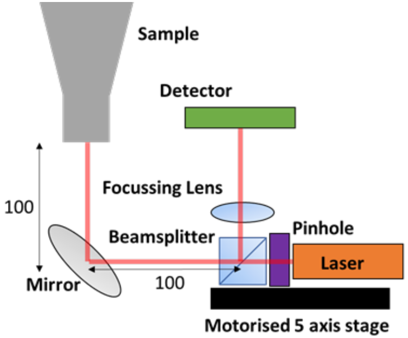
# High-Cycle Fatigue Testing of Irradiated Ti alloys

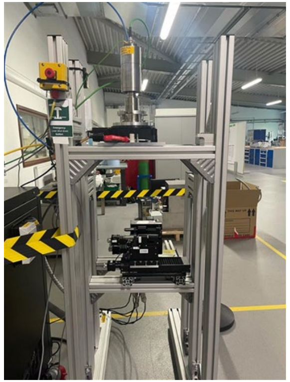


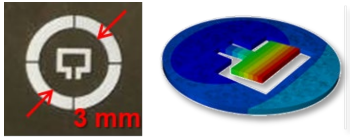

**3rd Generation Fatigue Testing Machine (FTM) under development**

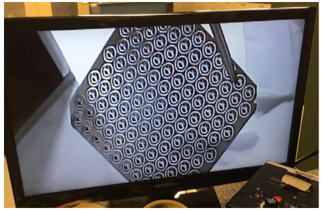
## Ultrasonic mesoscale Fatigue Rig (UFR) at the UKAEA-MRF



20 kHz = $10^8$ cycles in 1.5 h



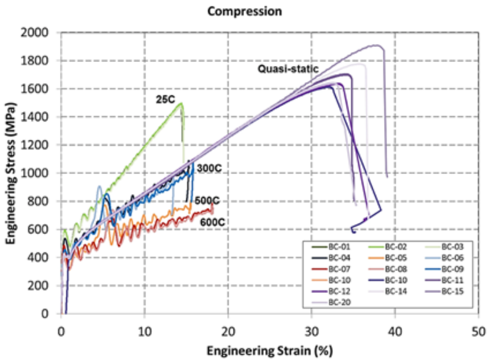
Meso-fatigue foil during extraction in PNNL hot cell

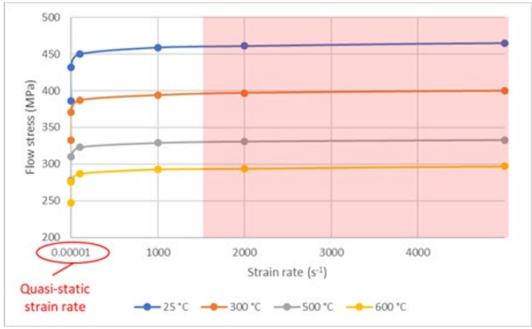
**BLIP 2017-2018 Ti foils will be shipped from PNNL to MRF for testing over the coming weeks**

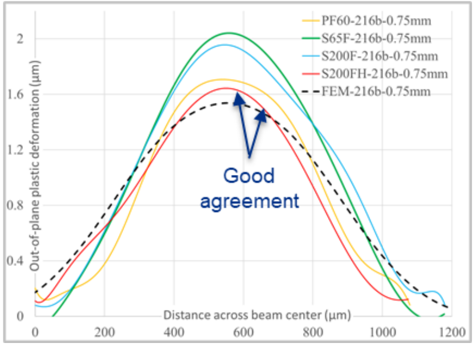
# Thermal Shock Studies at CERN's HiRadMat Facility

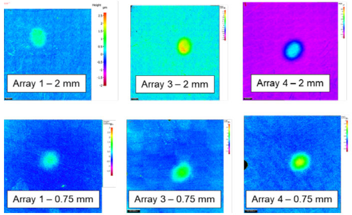
Poster by S. Bidhar

- HRMT-24 experiment successfully carried out in 2015
- PIE and real-time dynamic measurements of temperature, strain and displacement
- Distinct strain responses for several Beryllium grades (beam-induced plastic deformation)
- **Successful validation of Beryllium S-200FH Johnson-Cook strength model**




$$\sigma_Y = \left[A + B(\varepsilon^{p}_{eff})^{n}\right]\left[1 + C\ln\dot{\varepsilon}^{*}\right]\left[1 - T_H^{m}\right]$$

- Flow stress ($\sigma_y$) dependency on strain rate and temperature
- Model parameters A, B, C, m, n empirically determined

K. Ammigan, S. Bidhar, P. Hurh et al., Phys. Rev. Accel. Beams 22, 044501.

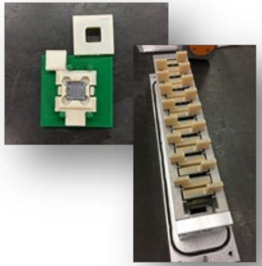
# Thermal Shock Studies at CERN's HiRadMat Facility

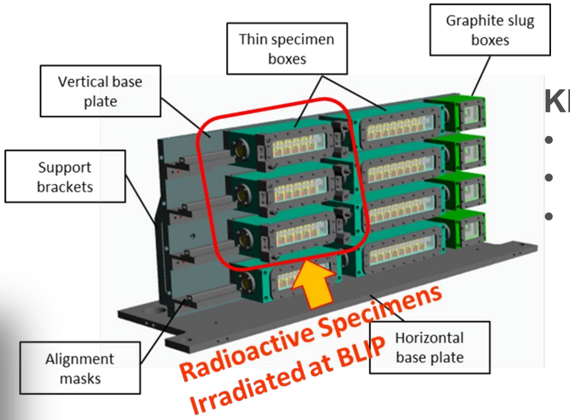
Poster by S. Bidhar

- **World's first thermal shock test** (HRMT-43) of irradiated materials
- Irradiated specimens: Beryllium, Graphite, Silicon, Titanium alloys, SiC-coated graphite, glassy carbon from BLIP


HRMT43 BeGrid2 beam time completed on Oct. 2, 2018, 4 am

| Beam Parameters | |
|---|---|
| Beam energy | 440 GeV |
| Max. bunch intensity | $1.2 \times 10^{11}$ |
| No. of bunches | 1 – 288 |
| Max. pulse intensity | $3.5 \times 10^{13}$ ppp |
| Max. pulse length | 7.2 μs |
| Gaussian beam size | 1σ: 0.1 – 2 mm |






**KEK specimens**
- Ti-6Al-4V (Grade 5)
- Ti-3Al-2.5V (Grade 9)
- SiC-coated graphite

**Next experiment planned in 2022, and will include**
- Irradiated Ti specimens from DS-Ti2 capsule
- Novel materials: NITE SiC/SiC composite and TFGR-W

Titanium alloy and SiC-coated graphite specimens will be shipped from UKAEA-MRF to PNNL for PIE work in the next few weeks

# Measurement of DPA Cross-Section

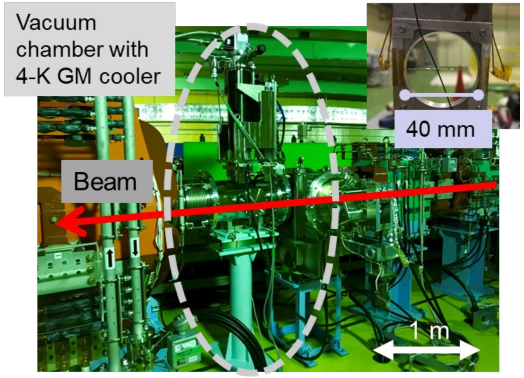
**2020/7/1**
**Press Release**

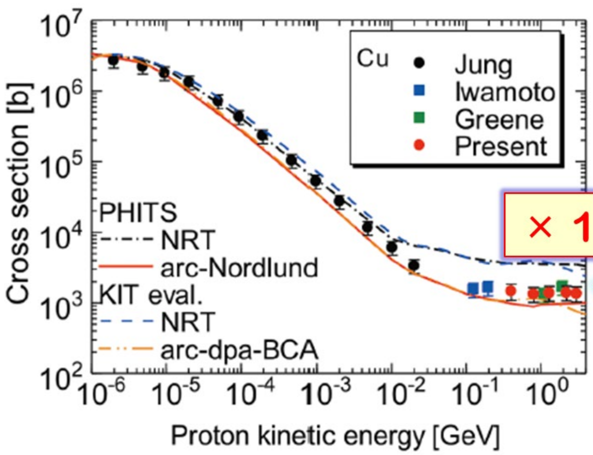
Poster by S. Meigo

- **Displacement Per Atom (DPA)** is used for lifetime estimation of target and beam window
- **Error in DPA calculation fairly large for HE protons**
- Measurements at J-PARC suggest DPA cross section is much smaller than conventional (NRT) model estimation
- This is due to **Athermal Recombination Correction (ARC)** of crystal lattice defects

Under cryo temperature (~ 20 K), displacement cross section($\sigma$) was obtained by increase of resistivity ($\Delta\rho_{cu}$) due to proton irradiation with average flux ($f(E)$)

$$\sigma_{exp}(E) = \Delta\rho_{Cu}/(\overline{\phi(E)}\rho_{FP}),$$




H.Matsuda, S.Meigo, Y.Iwamoto, T.Ishida, S.Makimura et al. J. Nucl. Sci. Tech. 57 (2020) 1141-1151

**Next experiments will be at FNAL FTBF & CERN HiRadMat facility with energy range over 30 GeV**

# Exciting work proposed for FY2022-2024

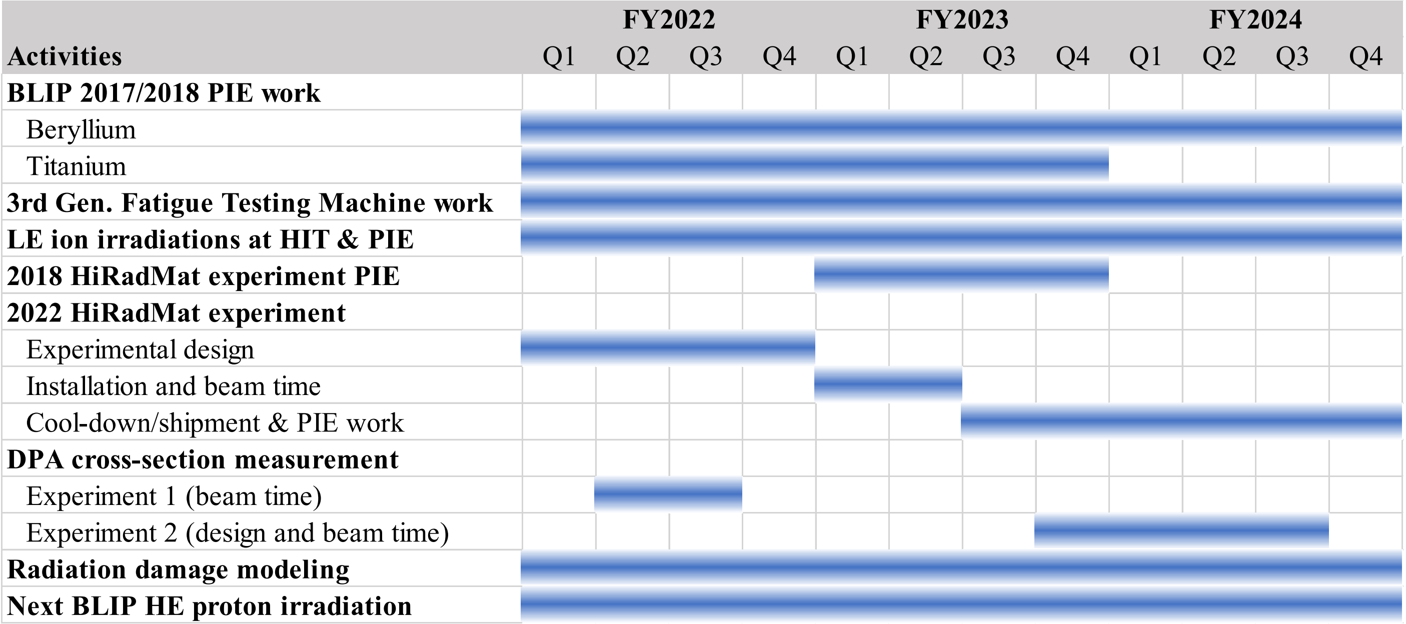
1. **Continue with Titanium Alloy Studies**
   a. **PIE** of DS-Ti2 specimens and 2018 HiRadMat thermal shock DS-Ti1 specimens (PNNL)
   b. **HiRadMat thermal shock test** on DS-Ti2 specimens & novel materials (CERN, 2022)
   c. **Ion beam irradiations** at HIT facility (2021 & 2022)
   d. Next **BLIP irradiation** (BNL, 2023-2024)
   e. **Fatigue strength measurements** (FNAL & UKAEA MRF)

2. **Beryllium Studies**
   a. PIE of specimens from BLIP 2018 irradiation (PNNL)

3. **DPA Cross-Section Experiment (FNAL, CERN)**

4. **Radiation Damage Modeling (PNNL)**

# Schedule for FY2022-2024




**Goal:** Determine optimal material specification (2024/2025) to support the design of next-generation multi-MW windows (2025-2026)

# Summary

- Significant progress has been made in the study of Ti alloys for critical beam window applications
  - Preliminary results already impacting material choices for T2K beamline upgrade and LBNF neutrino beamline components
- Important work over the next few years
  - In-beam tests at BLIP, HiRadMat, HIT
  - Extensive material characterization work

  } *Essential to achieve multi-MW targetry goals!*

➢ **Broad impact of U.S.-Japan target materials project**
  - Together with the RaDIATE collaboration, it has helped establish the methods, technology and procedures necessary for evaluating and developing candidate targetry materials
  - Fostered many beneficial collaborative research activities that benefit the global high-power target community to support future next-generation facilities

Many thanks to KEK and DOE for supporting this project since 2016

# Advanced Materials Studies for High Intensity Proton Production Targets and Windows

## Related Posters

- T. Ishida, Progress of radiation damage studies on titanium alloys as high-intensity proton accelerator beam window materials.
- S. Meigo, Measurement of displacement cross-section at J-PARC, FNAL and CERN.
- S. Bidhar, Extreme beam-induced thermal shock experiment on materials for future high intensity multi-MW accelerator components.

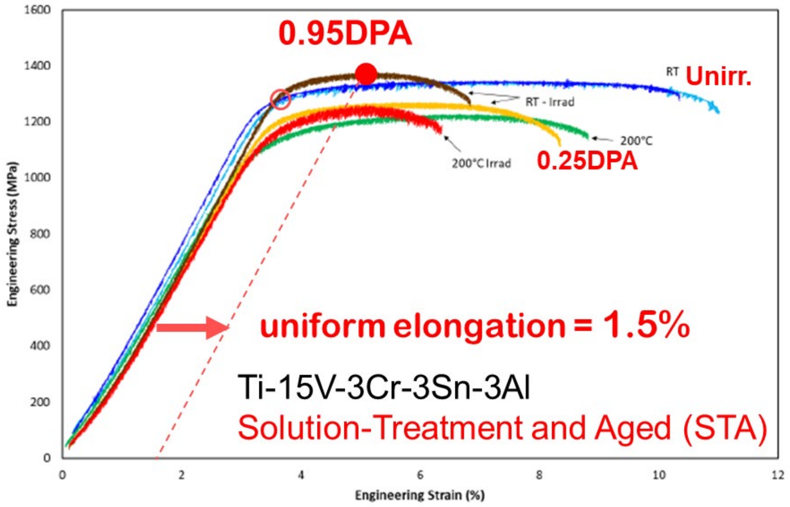
**RaDIATE Collaboration**

**Ra**diation **D**amage **I**n **A**ccelerator **T**arget **E**nvironments

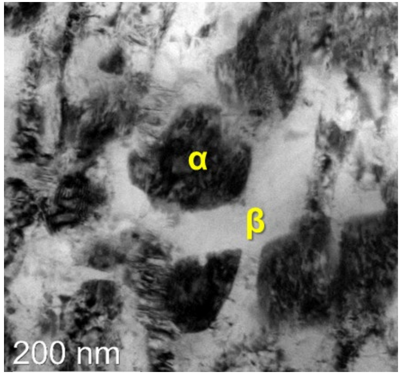
https://radiate.fnal.gov

# Supplemental Slides

# DS-Ti2 PIE: Ti-15-3 Tensile Tests




- The metastable β-phase Ti-15-3 with Solution-Treatment and Ageing (STA) heat treatment
  - Ageing heat treatment typical for this alloy, to produce α-phase precipitation in the mother β-phase (precipitation strengthening)
  - Phase composition/microstructure (53% primary β + 47% coarse α precipitate) and different from OTR foil w/o ageing (100% metastable β-phase)

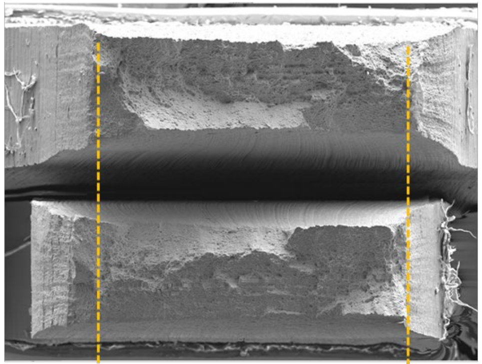



| RT | DPA | YS | UTS | YS/UTS | UE | TE | ΔYS | ΔUTS | ΔUE | ΔTE |
|---|---|---|---|---|---|---|---|---|---|---|
| Unirr. | 0.00 | 1261 | 1350 | 93% | 4.00 | 7.38 | | | | |
| B01C | 0.25 | 1198 | 1271 | 94% | 2.10 | 5.20 | -5% | -6% | -48% | -29% |
| B02 | 0.95 | 1320 | 1379 | 96% | 1.45 | 3.40 | 5% | 2% | -64% | -54% |

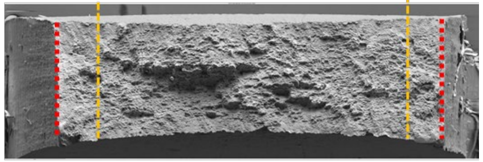
**Almost no radiation hardening, and a few % of uniform elongation remains**

# DS-Ti2 PIE: Ti-6Al-4V Fractography

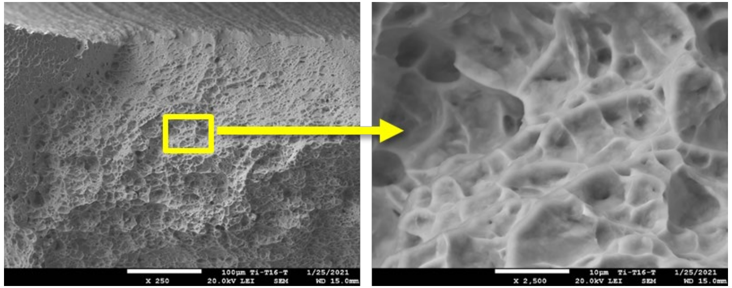
**Unirradiated**

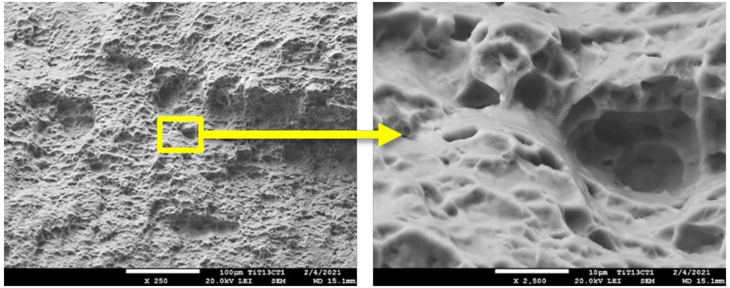
**0.95 DPA**

Reduction in area is much smaller
Fracture surface more planar



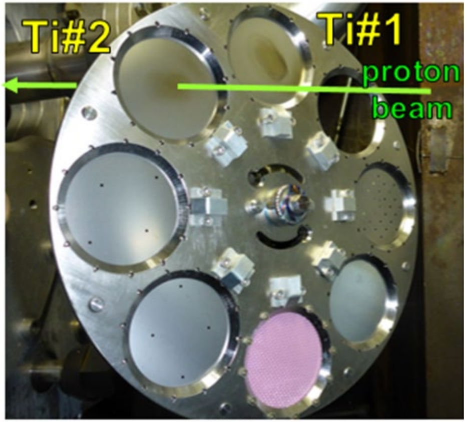
Ductile dimpling is recognized after irradiation
Seemingly smaller and shallower
Larger degree of planar deformation



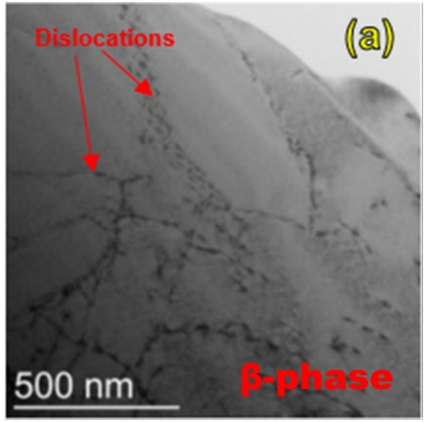
- Ti-6Al-4V exhibited ductile failure mode at both RT and 200 °C, but irradiation reduces the ductility and ability to neck
- **Still no evidence of brittle failure modes at this dose level**

# Metastable β-phase Alloy Ti-15-3

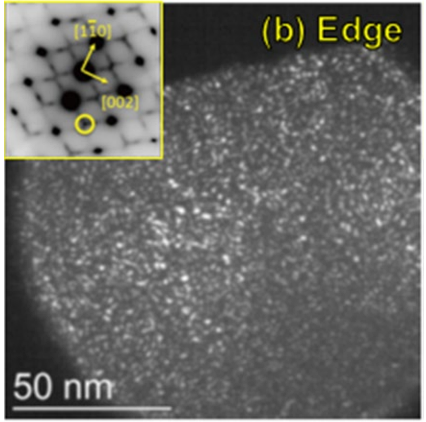
- PIE on irradiated J-PARC OTR foil: 50 μm-thick, single metastable-β (BCC) phase alloy, Ti-15V-3Cr-3Al-3Sn (Ti-15-3) at PNNL




Ti-1: $1.4 \times 10^{20}$pot
0.12 peak DPA




Very low density of defect clusters were formed by irradiation
No significant changes on micro-Vickers hardness

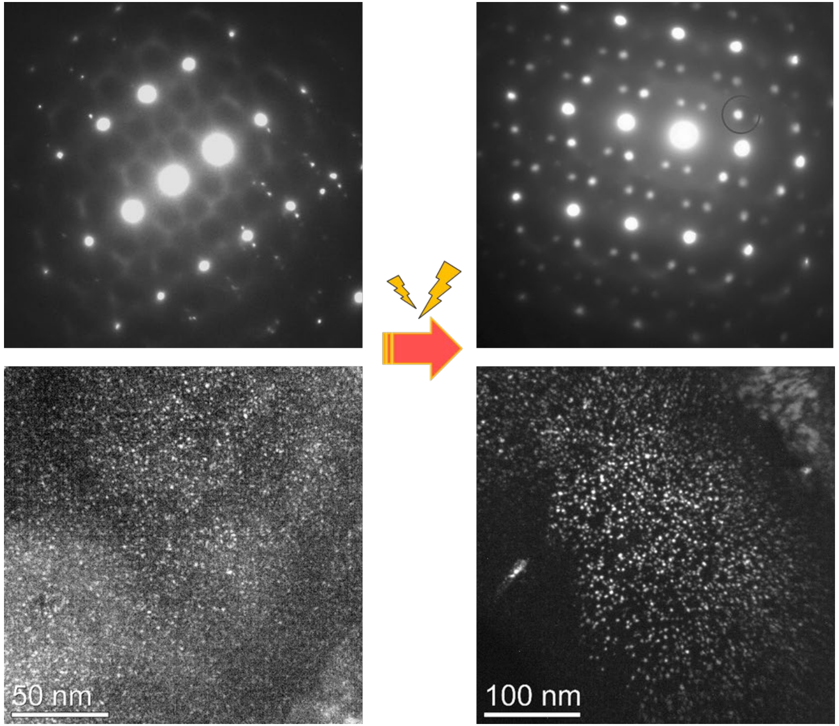



A high density of nano-clusters of athermal w and martensitic α” phases
These were stable after irradiation

Ti-15-3 alloy may have a high resistance for radiation damage due to nanoscale precipitates working as point defect sink sites

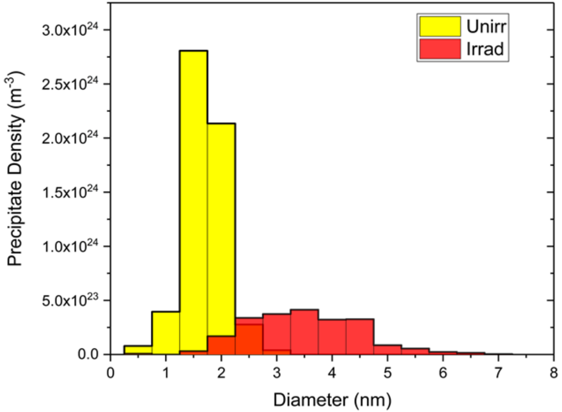
T. Ishida, E.Wakai, D.Senor, A.Casella, D.Edwards et al., Nucl. Mater. En. 15 (2018) 169-174

# DS-Ti2 PIE: Ti-6Al-4V Micro-Structural Studies




Confirmation of radiation-induced ω phase formation, coarsened at higher irradiation dose

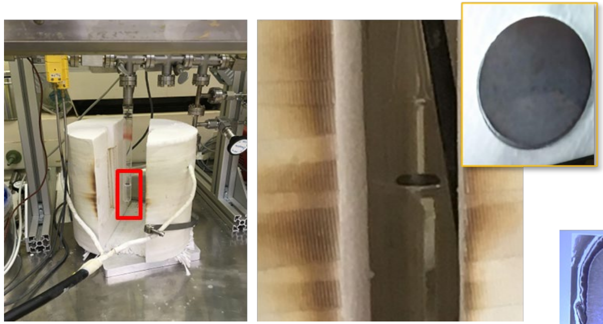
- Diffraction patterns indicate ω-phase is fully formed, discrete diffraction spots instead of diffuse streak, can image small particles of omega phase



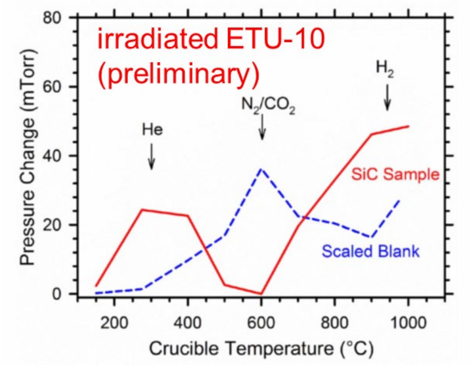

| DPA | Mean Size (nm) | Number density ($m^{-3}$)* | Inter-prt. spacing (nm) | Vol fraction (%) |
|---|---|---|---|---|
| 0.0 | 1.4±0.3 | $5.7x10^{24}$ | 7.6 | 0.8 |
| **0.95** | **↑3.2±1.0** | **↓$2.2x10^{24}$** | **↑8.5** | **↑3.7** |

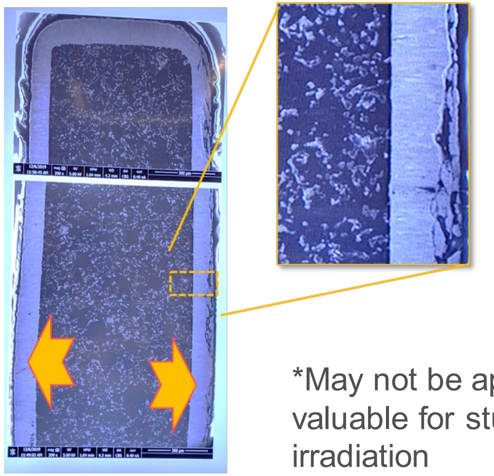
# SiC-coated Graphite Studies

- Thermal Desorption Spectroscopy (TDS) on irradiated specimen and graphite filler to study how SiC-coating prevents rad-produced gas release from graphite



- Release of He: 200~400 C
- Release of H: > 600C
- Agree to reference (SiC
  Crack at SiC surface + very large graphite swelling: 15% after TDS



SiC coating may act as very effective gas confinement barrier

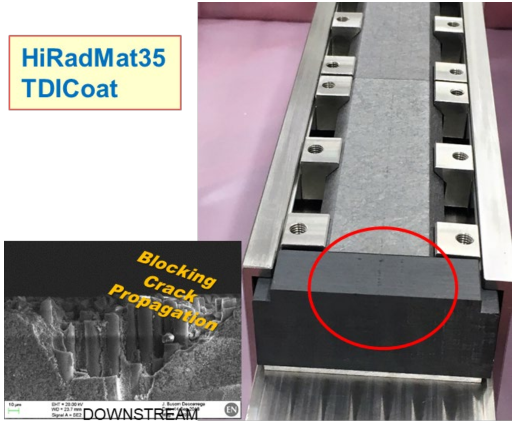
*May not be applicable for target, while very valuable for studies on gas production by irradiation

# New Target Materials tested at HiRadMat

## NITE-SiC/SiC composite

- Density 3.2 g/cc (SiC) → more secondary emission than graphite
- SiC fibers + matrix, control mechanical properties / to replicate ductility

HiRadMat35
TDICoat

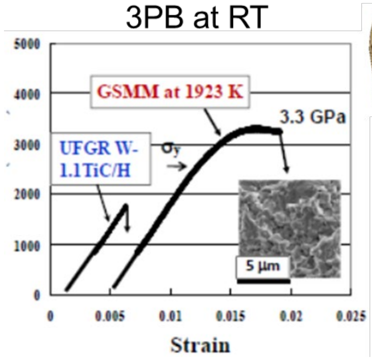


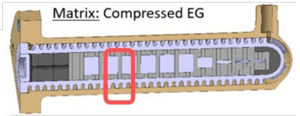

- COMET phase 1 target (u2e)
- Neutrino target to prevent wrong-sign component

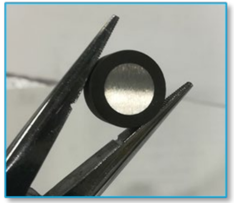
## Highly-Ductile Tungsten

## TFGR W-1.1TiC

- Tungsten: high density/melting point, but become brittle by recrystalization at 1200'C
- 3D MA (FineGrain) → HIP → recrystallization under grain boundary sliding (GSMM): segregation / precipitation of TiC at grain boundary



- COMET phase-2 target (u2e)
- CERN AD / ESS/ MLF-2nd TS target

To be included in the next BLIP2023-2024 irradiation and HiRadMat2022 experiment

Fermilab