Advanced Materials Studies for High Intensity Proton Production Targets and Windows

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*Lead Principal Investigators
Motivation from Past Experience

• 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
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)

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
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
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)

https://radiate.fnal.gov
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

<table>
<thead>
<tr>
<th>Material</th>
<th>Capsule position</th>
<th>Capsule</th>
<th>Irrad. Phases</th>
<th>Peak DPA/p (NRT)</th>
<th>POT</th>
<th>Total DPA</th>
</tr>
</thead>
<tbody>
<tr>
<td>DS Ti1 (KEK)</td>
<td>6</td>
<td>titanium</td>
<td>1</td>
<td>3.38E-22</td>
<td>7.30E+20</td>
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<tr>
<td>DS Ti2 (KEK)</td>
<td>6</td>
<td>titanium</td>
<td>3</td>
<td>3.38E-22</td>
<td>2.81E+21</td>
<td>0.95</td>
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<tr>
<td>US Ti (KEK)</td>
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<td>titanium</td>
<td>1, 2, 3</td>
<td>3.34E-22</td>
<td>4.57E+21</td>
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<td>Si (CERN)</td>
<td>3</td>
<td>SiC</td>
<td>1, 2</td>
<td>7.03E-23</td>
<td>1.76E+21</td>
<td>0.12</td>
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<td></td>
<td></td>
<td>Graphite</td>
<td>1, 2</td>
<td>2.52E-23</td>
<td>1.76E+21</td>
<td>0.04</td>
</tr>
</tbody>
</table>

181 MeV rastered beam
3 cm diameter footprint with
154 µA avg. current x 55 days

4.6 x 10^{21} POT

Past tensile data on titanium alloys only up to 0.3 DPA

Comparable to MW facility operation
Titanium alloys irradiated

<table>
<thead>
<tr>
<th>Phase</th>
<th>Grade</th>
<th>Heat Treatment</th>
</tr>
</thead>
<tbody>
<tr>
<td>α + β</td>
<td>Ti-6Al-4V Gr-5</td>
<td>Annealed</td>
</tr>
<tr>
<td>α + β</td>
<td>Ti-6Al-4V Gr-5</td>
<td>Ultrafine grain</td>
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<tr>
<td>α</td>
<td>Ti-3Al-2.5Sn Gr-6</td>
<td>Annealed</td>
</tr>
<tr>
<td>β</td>
<td>Ti-15V-3Al-3Cr-3Sn</td>
<td>Solution Treated Aged</td>
</tr>
<tr>
<td>α</td>
<td>Pure Ti – Gr-2</td>
<td>Annealed</td>
</tr>
<tr>
<td>α + β</td>
<td>Ti-3Al-2.5V Gr-9</td>
<td>Annealed</td>
</tr>
<tr>
<td>α + β</td>
<td>Ti-6Al-4V Gr-23 ELI</td>
<td>Solution Treated Aged</td>
</tr>
<tr>
<td>α + β</td>
<td>Ti-6Al-4V Gr-23 ELI</td>
<td>Annealed</td>
</tr>
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</table>
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 Fe²⁺ 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

- 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)

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

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

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

“radiation hardening” & “loss of ductility / embrittlement”
Comparison of Ductility between Ti-6Al-4V & Ti-15-3

- **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)
**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

**Nano-indentation hardness test**

Hardness is relatively proportional to Yield Strength, but difficult to infer ductility (elongation)
Titanium Alloy Nano-Indentation Measurements

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.

Contrasting radiation hardening behavior between different titanium alloy microstructures.

[Graph showing nano-hardness distribution and hardness values for different conditions and microstructures.]
High-Cycle Fatigue Testing of Irradiated Ti alloys

Reduced fatigue strength

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

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

Meso-fatigue foil during extraction in PNNL hot cell
Thermal Shock Studies at CERN’s HiRadMat Facility

• 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

\[ \sigma_Y = [A + B(e_{eff}^P)^n] [1 + Cln\dot{e}^*][1 - T_H^m] \]

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

Thermal Shock Studies at CERN’s HiRadMat Facility

- 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

<table>
<thead>
<tr>
<th>Beam Parameters</th>
<th>Value</th>
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<tbody>
<tr>
<td>Beam energy</td>
<td>440 GeV</td>
</tr>
<tr>
<td>Max. bunch intensity</td>
<td>1.2 x 10^{11}</td>
</tr>
<tr>
<td>No. of bunches</td>
<td>1 – 288</td>
</tr>
<tr>
<td>Max. pulse intensity</td>
<td>3.5 x 10^{13} ppp</td>
</tr>
<tr>
<td>Max. pulse length</td>
<td>7.2 μs</td>
</tr>
<tr>
<td>Gaussian beam size</td>
<td>1σ: 0.1 – 2 mm</td>
</tr>
</tbody>
</table>

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

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

HRMT43 BeGrid2 beam time completed on Oct. 2, 2018, 4 am
Measurement of DPA Cross-Section

- 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 ($f(E)$)

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


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

<table>
<thead>
<tr>
<th>Activities</th>
<th>FY2022</th>
<th>FY2023</th>
<th>FY2024</th>
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<tbody>
<tr>
<td></td>
<td>Q1</td>
<td>Q2</td>
<td>Q3</td>
</tr>
<tr>
<td>BLIP 2017/2018 PIE work</td>
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<tr>
<td>Beryllium</td>
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<tr>
<td>Titanium</td>
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<tr>
<td>3rd Gen. Fatigue Testing Machine work</td>
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<tr>
<td>LE ion irradiations at HIT &amp; PIE</td>
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<tr>
<td>2018 HiRadMat experiment PIE</td>
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<tr>
<td>2022 HiRadMat experiment</td>
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<tr>
<td>Experimental design</td>
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<td>Installation and beam time</td>
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<tr>
<td>Cool-down/shipment &amp; PIE work</td>
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<td>DPA cross-section measurement</td>
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<tr>
<td>Experiment 1 (beam time)</td>
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<tr>
<td>Experiment 2 (design and beam time)</td>
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<td>Radiation damage modeling</td>
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<tr>
<td>Next BLIP HE proton irradiation</td>
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</table>

**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

➢ 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

Essential to achieve multi-MW targetry goals!

Many thanks to KEK and DOE for supporting this project since 2016
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
Radiation Damage In Accelerator Target Environments
https://radiate.fnal.gov
Supplemental Slides
Almost no radiation hardening, and a few % of uniform elongation remains

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

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

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

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

<table>
<thead>
<tr>
<th>DPA</th>
<th>Mean Size (nm)</th>
<th>Number density (m⁻³)</th>
<th>Inter-prt. spacing (nm)</th>
<th>Vol fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>1.4±0.3</td>
<td>5.7x10²⁴</td>
<td>7.6</td>
<td>0.8</td>
</tr>
<tr>
<td>0.95</td>
<td>↑3.2±1.0</td>
<td>↓2.2x10²⁴</td>
<td>↑8.5</td>
<td>↑3.7</td>
</tr>
</tbody>
</table>
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: > 600°C
  - 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

Highly-Ductile Tungsten
TFGR W-1.1TiC
- Tungsten: high density/melting point, but become brittle by recrystallization at 1200°C
- 3D MA (FineGrain) → HIP → recrystallization under grain boundary sliding (GSMM): segregation / precipitation of TiC at grain boundary

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

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

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