RaDIATE Collaboration for Target Material Studies

Kavin Ammigan on behalf of Frederique Pellemoine & the RaDIATE Collaboration

Topical Workshop on RADiation effects in SUperconducting Magnets (RADSUM 2025)

16 January 2025

RaDIATE collaboration created in 2012, with Fermilab as the leading institution. The collaboration has grown up to 20 institutions over the years.

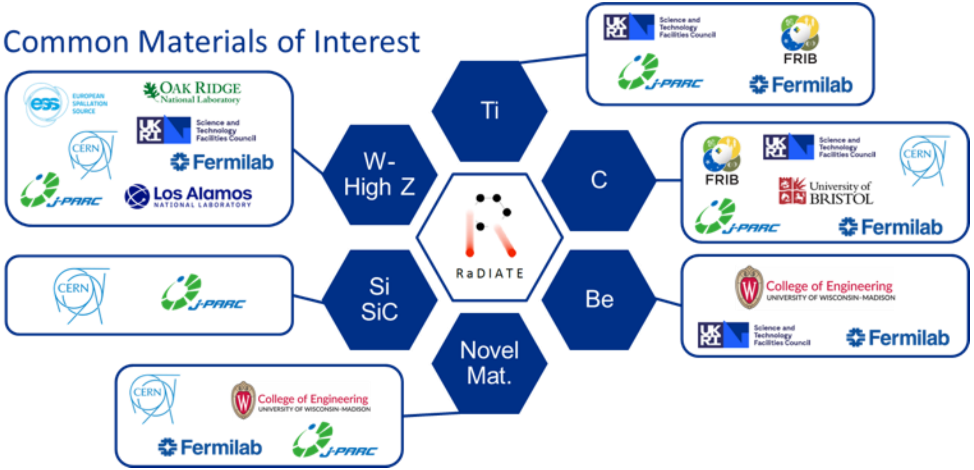
Objectives

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

https://radiate.fnal.gov

Activities include

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



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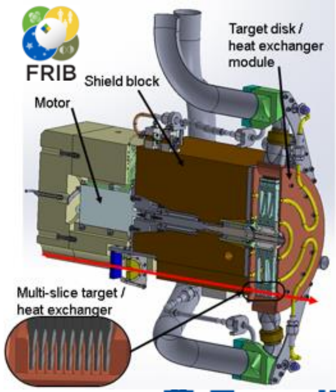
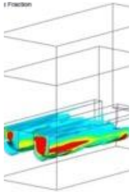
High Power Targetry Context

- Beam Intercepting Devices (BIDs) are continuously bombarded by high-energy high-intensity particle beams
  - Primary beam and target containment windows
  - Secondary particle-production targets (eg. neutrino targets)
  - Collimators, absorbers, dumps
- Few target examples
  - Neutrino production at Fermilab (LBNF-DUNE)
    - Graphite target: ~ 1MW of 120 GeV pulsed proton beam
    - Future up to 2.4 MW
  - Rare isotope production at MSU-FRIB
    - Graphite rotating target
    - 400 kW of heavy ion beams (from O to U) at a minimum of 200 MeV/u
  - Spallation Neutron Source at Oak Ridge National Lab
    - Liquid mercury target: 1.4 to 2 MW of 1.3 GeV pulsed proton beam



High Power Targetry Challenges

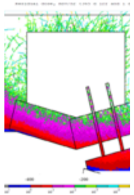


Heat removal

Thermal shock

Physics performance

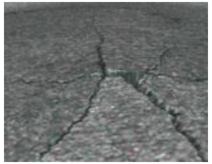
Radiation damage



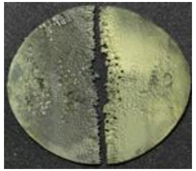
Operational safety

Storage and disposal

Thermal Shock and Radiation Damage identified as most cross-cutting challenges of high-power target facilities

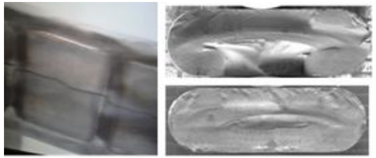


Be window embrittlement (FNAL)



Target containment vessel cavitation (ORNL - SNS)

Horn stripline fatigue failure (FNAL)



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

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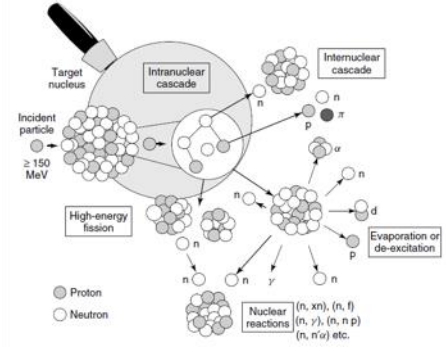
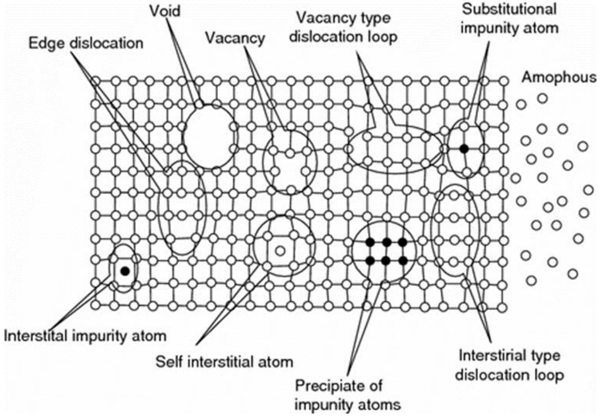
Radiation Damage

Disruption of lattice structure of the material upon irradiation

- Atomic displacements (cascades)
  - Average number of stable interstitial/vacancy pairs created
  - Expressed as Displacement Per Atom (DPA)
- Creation and agglomeration of point defects
- Creation of transmutation products (H and He production)



From D. Filges, F. Goldenbaum, in:, Handb. Spallation Res., Wiley-VCH Verlag GmbH & Co. KGaA, 2010, pp. 1–61.



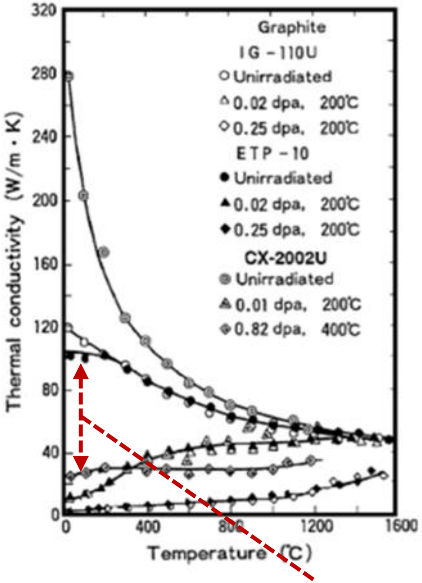
From V. Verma, K. Katovsky, Radiation Damage and Development of a MC Software Tool, in: Spent Nuclear Fuel and Accelerator-Driven Subcritical Systems, Springer, 2019.

Radiation Damage Effects in Materials

- Phase change
- Hardening and embrittlement (loss of ductility)
- Creep and swelling
- Reduction of fracture toughness
- Reduction of thermal conductivity
- Change in coefficient of thermal expansion and modulus of elasticity
- Accelerated corrosion
- Transmutation products: H, He gas production can cause void formation and embrittlement

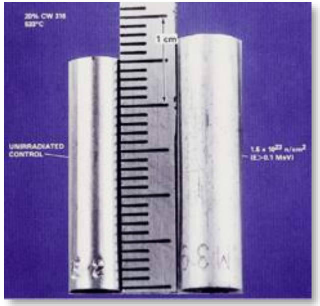
Radiation damage effects very dependent upon material and irradiation conditions (temperature, dose rate)

N. Maruyama and M. Harayama, Journal of Nuclear Materials, 195, 44-50 (1992)



Factor of 10 reduction in thermal conductivity of graphite after 0.02 DPA

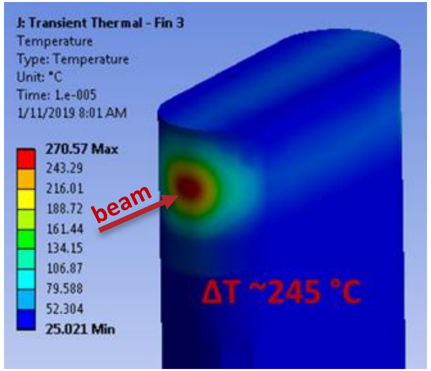
D.L. Porter and F. A. Garner, J. Nuclear Materials, **159**, p. 114 (1988)



Void swelling in 316 stainless steel tube exposed to reactor dose of 1.5 x 10^{23} n/cm^2

Thermal Shock and Thermal Fatigue

- Induced by sudden energy deposition from pulsed beam
- Differential expansion creates localized area of compressive stress and generates stress waves
  - 1 MW target: 250 K in 10 μs (2.5 x 10^7 K/s)
- Stress waves move through the target at sonic velocities and can cause plastic deformation and induce cracks
- Cyclic loading environment progressively damages the material's microstructure leading to fatigue failure



Iridium cylinder tested at CERN's HiRadMat facility

Heavy dependence on material properties **but material properties dependent upon radiation damage**

Initial stress wave amplitude ⇨ $\sigma = \sqrt{\rho E} \cdot \alpha \cdot L \cdot \frac{\Delta T}{\Delta t}$

Elastic wave speed ⇨ $c = \sqrt{\frac{E}{\rho}}$

Radiation Damage in Accelerators

Irradiation Source	DPA rate (DPA/s)	He gas production (appm/DPA)	Irradiation Temp (˚C)
Mixed spectrum fission reactor	3 x 10^{-7}	1 x 10^{-1}	200-600
Fusion reactor	1 x 10^{-6}	1 x 10^{1}	400-1000
High energy proton beam	6 x 10^{-3}	1 x 10^{3}	100-800

- Use of data from nuclear materials research is limited, cannot be directly utilized but give us some insight of radiation damage trends
- Could develop some methods to overcome issues and challenge to simulate protons with neutrons or other alternative methods

n ≠ p
1-14 MeV   100+ MeV

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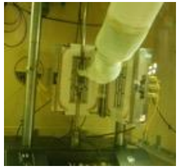
Radiation Damage In Accelerator Target Environments



In-Beam Studies

Evaluate in-beam performance of candidate materials

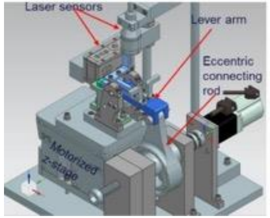
- **Irradiated material studies:** high energy proton irradiation at BNL-BLIP
- **Thermal shock testing:** HiRadMat facility at CERN
- **Analyze beamline components**

Alternative Methods

Emulate high energy proton irradiations for accelerated and cost-effective material screening

- **Low energy Ion:** for assessing radiation damage effects (dpa + transmutation)
- **Electron beam** for thermal shock and fatigue testing
- **Novel testing methods** (fatigue studies)

RaDIATE



Novel Target Materials

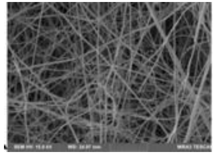
New materials with enhanced thermal shock and radiation damage resistance

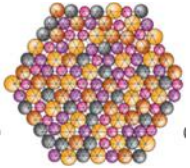
- **High-Entropy Alloys**
- **Nanofibers**
- **New W alloys**

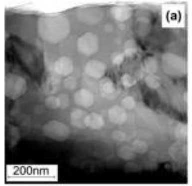
Develop Modeling

Prediction of fundamental response of various materials to irradiation and thermal shock

- **Helium gas bubbles** formation and segregation
- **Radiation damage effects**
- **Heat transfer mechanism** in nanofiber media



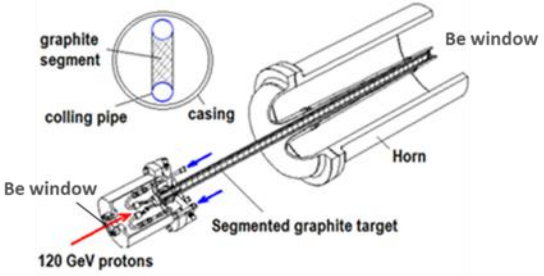


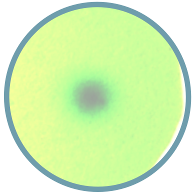


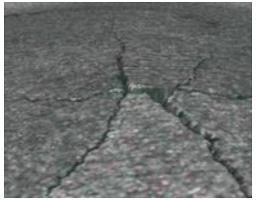
Some Examples of RaDIATE Collaboration Studies

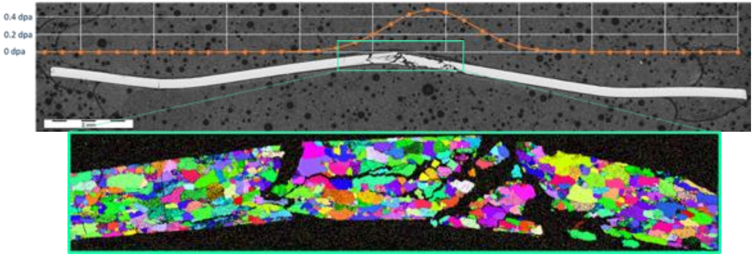
Study Components Failed in Service: NuMI Be Window Analysis

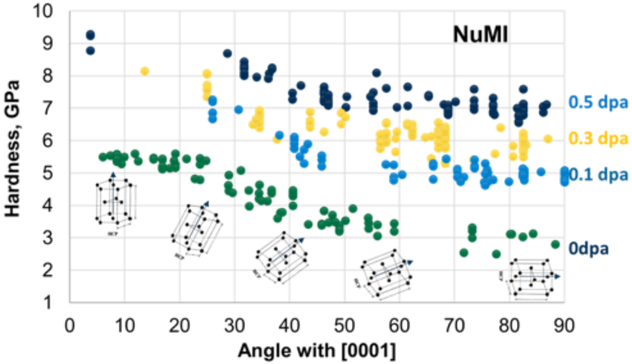
120 GeV proton beam, 1.54 x 10^{21} POT (0.5 peak DPA), T ~ 50 °C











- Observed transition from transgranular fracture to grain boundary/mixed mode fracture in irradiated Be

UK Atomic Energy Authority

- Significant hardening even at 0.1 DPA
- Hardness of irradiated Be less anisotropic
- Increased hardness means less ductility (more brittle)

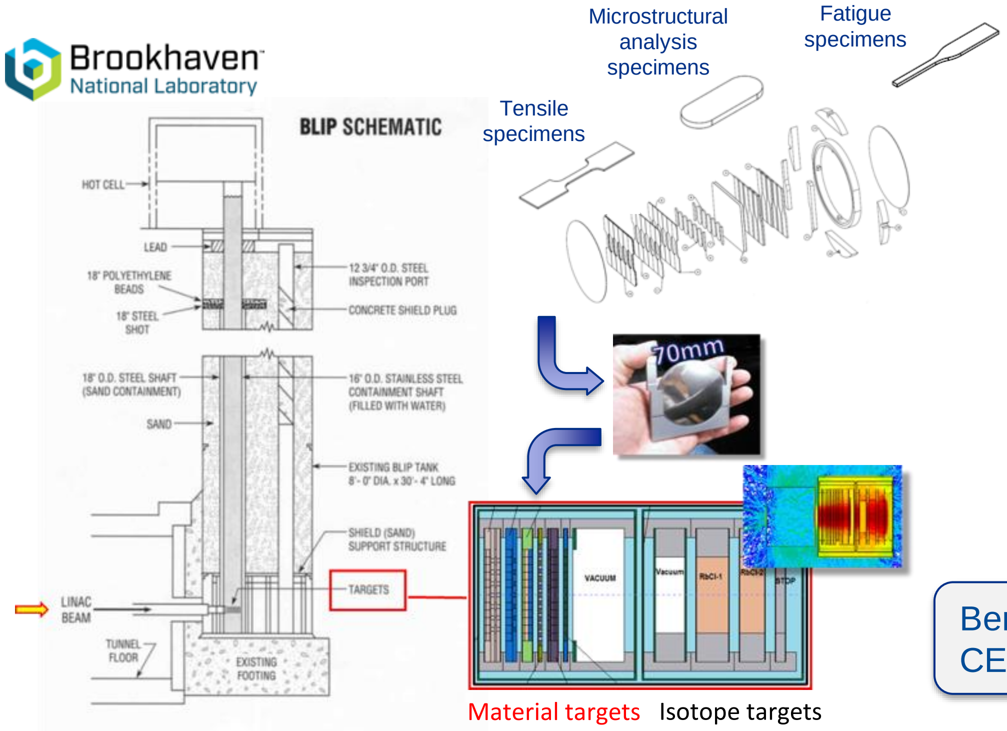
V.Kuksenko et al. J. Nuclear Materials, 490, pp.260-271 (2017)

Near-Prototypic Irradiation Testing

- High-energy proton irradiation of material specimens at BNL-BLIP facility in partnership with the RaDIATE collaboration
  - Irradiation campaign completed in 2017/2018
- Post-Irradiation Examination (PIE) conducted at participating institution equipped with hot-cell facilities (PNNL)
- In-beam thermal shock experiments at CERN's HiRadMat facility that included both pre-irradiated (BLIP) and non-irradiated specimens
  - Completed experiments in 2015, 2018 and 2022
  - Next experiment scheduled in 2025

High Energy Proton Irradiation at BNL's BLIP Facility

- Unique facility for material irradiation in tandem with medical isotope production
- High energy protons: 66 – 200 MeV with 165 µA peak current

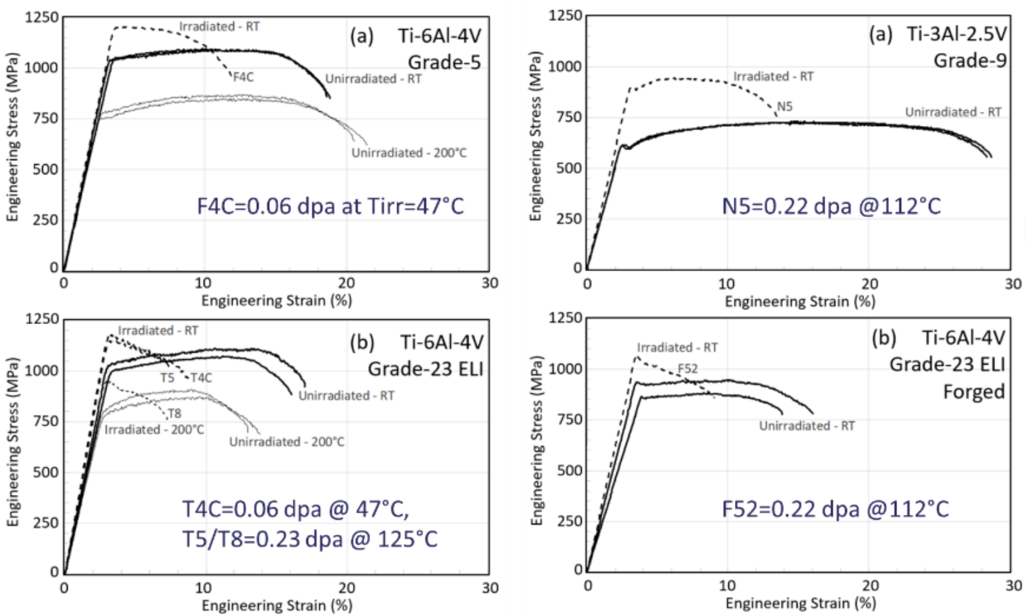


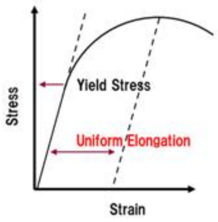
- RaDIATE multi-material irradiation campaign
- 181 MeV p irradiation for 8 weeks
  - Over 200 specimens from 6 RaDIATE collaborators
- Participants: BNL, PNNL, FRIB, ESS, CERN, J-PARC, STFC, Oxford, FNAL
- Completed irradiation in 2018
  - 4.5 x 10^{21} accumulated protons on target
  - Peak DPA: 0.95 (Ti alloy)
- Post-Irradiation Examination still ongoing
  - Mechanical/Thermal testing
  - Microstructural analysis

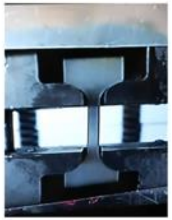
Benefit to many facilities including LBNF, T2K, BDF at CERN, FRIB, and HL-LHC collimators

BLIP Ti Alloy Tensile Testing

Stress-strain curves for Ti-6Al-4V and Ti-3Al-2.5V







Evidence that Ti-3Al-2.5V alloy is more radiation-resistant

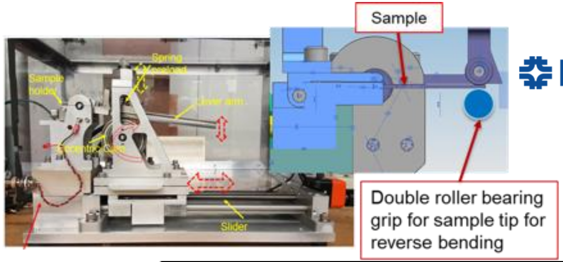
T. Ishida et al., J. Nuclear Materials 541 (2020) 152413

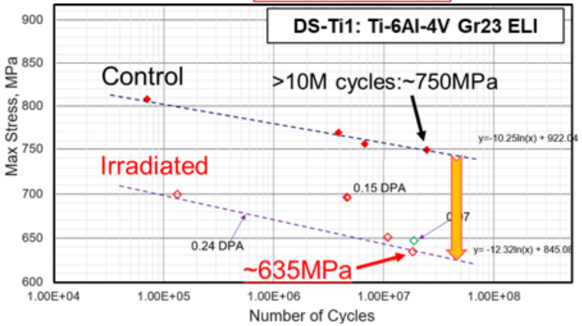
Testing done in PNNL hot cell

- Ti-6Al-4V (Grade 23) loses almost all of its uniform elongation (UE) after irradiation
  - Important to retain UE in a target material as it allows for plastic deformation without rapid growth of cracks and sudden failure

High-Cycle Fatigue Testing of Irradiated Ti Alloys

- Proton-irradiated fatigue life data crucial in evaluating component lifetime

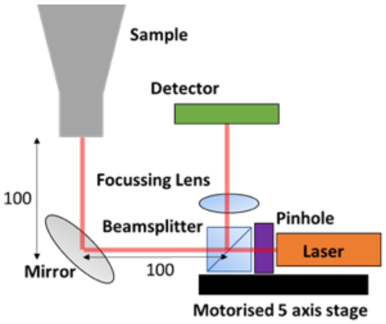




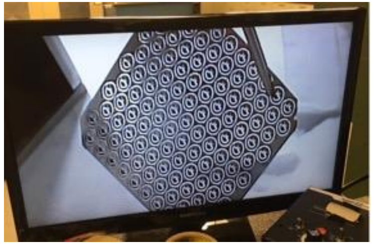
S. Bidhar, 6th RaDIATE Collaboration Meeting, 2019

First high-cycle fatigue testing of irradiated Ti at Fermilab

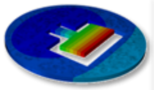
Ultrasonic mesoscale Fatigue Rig (UFR) at the UKAEA-MRF



Extraction of meso-fatigue foil from BLIP capsule in PNNL hot cell



20 kHz = 10^8 cycles in 1.5 h



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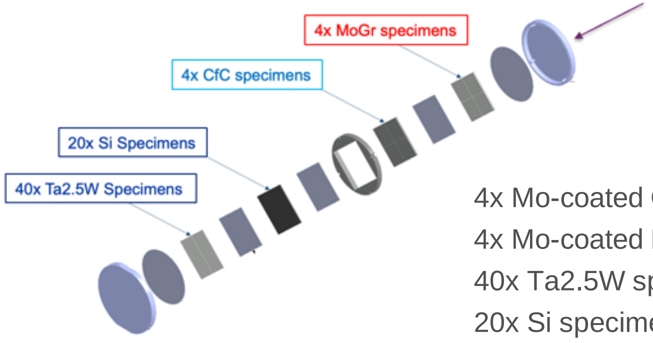
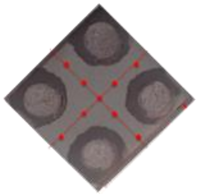
CERN BLIP-Irradiated Specimens Analyzed by Framatome



4x Mo-coated CFC specimens
4x Mo-coated MoGr specimens
40x Ta2.5W specimens
20x Si specimens

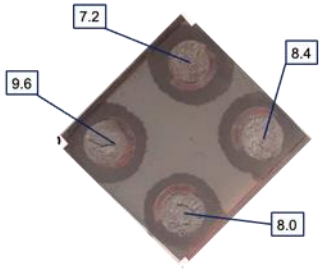
Micro-indentation of coating and bulk

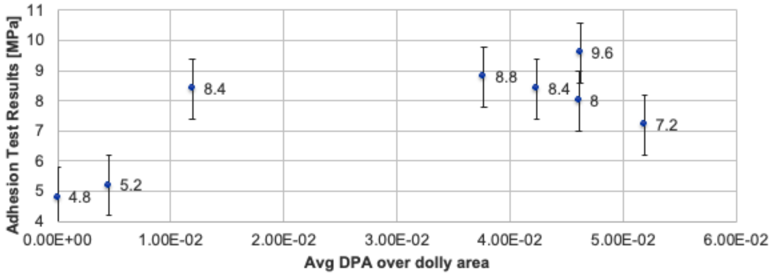


- Goal: Gain insight into the mechanical behavior of coating and bulk
- Observed decrease of indentation depth associated with increase in hardness for both coating and bulk

Adhesion tests

- Observed fracture always localized in the bulk





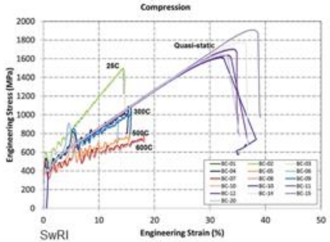
N. Solieri, 8th RaDIATE Collaboration Meeting, 2023

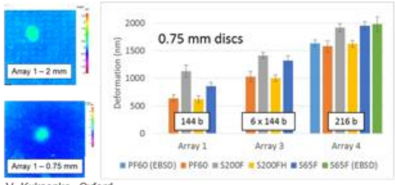
Thermal Shock Tests at CERN's HiRadMat facility

HRMT24 - 2015

- Observed distinctive thermal shock response for **various beryllium grades**
- Detected plastic strain ratcheting from multiple beam pulses
- Successful validation of Be S200FH Johnson-Cook strength model

Profilometry to measure plastic out-of-plane deformations and Johnson-Cook strength model validation

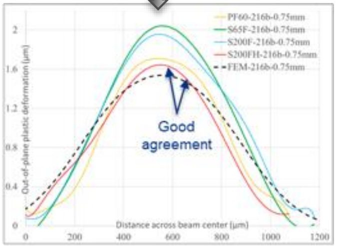




V. Kuksenko, Oxford

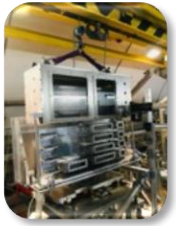
$$\sigma_Y = \left[A + B(\varepsilon_{eff}^{p})^n\right][1 + C ln\dot{\varepsilon}^*][1 - T_H^m]$$

Ammigan et al., Phys. Rev. Accel. Beams 22, 044501, 2019.



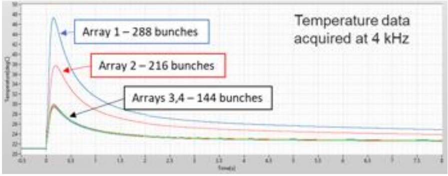
S. Bidhar, FNAL

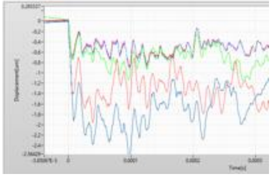
HRMT43 - 2018

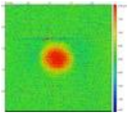


- **First and unique test with pre-irradiated material specimens** (Be, C, Ti, Si)
- First test on nanofiber electrospun fiber mats and metal foam (SiC, ZrO, Al_2O_3, RVC)
- Dynamic online measurements of graphite cylinders

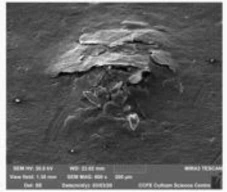
Online measurements and benchmarking of graphite cylinders



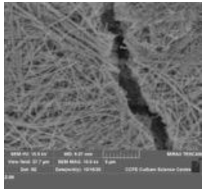


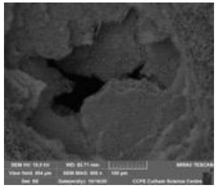


Beam-induced damage



Sigraflex, F. Nuiry, CERN





ZrO2 nanofiber, S. Bidhar, PRAB 24, 123001, 2021

HRMT-60 RaDIATE Experiment (2022)

- **~120 material specimens tested:**
  - Graphite, Ti alloys, Beryllium, Nanofiber mats, High-Entropy alloys, Sigraflex, TFGR tungsten, SiC/SiC composite
  - Understand single-shot thermal shock response and limits
  - Explore novel advanced materials
  - Assess the performance of various grades of conventional materials
  - Compare the behavior of non-irradiated to pre-irradiated materials
  - Directly measure beam-induced dynamic effects to validate simulation codes

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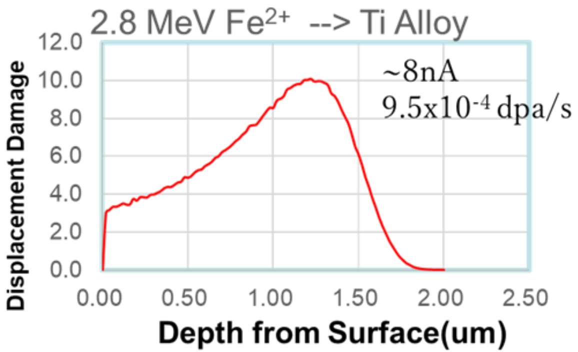
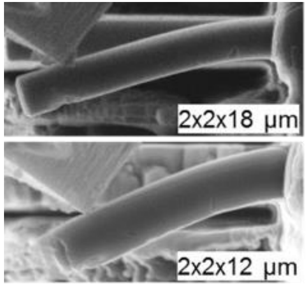
J-PARC
CERN
Pacific Northwest NATIONAL LABORATORY
UK Atomic Energy Authority

Alternative to High Energy Proton Beam Tests

- **High energy proton irradiation**
  - Highly activated material (handling/tests in hot cells)
  - Low damage accumulation rate (lengthy irradiation)
  - Costly

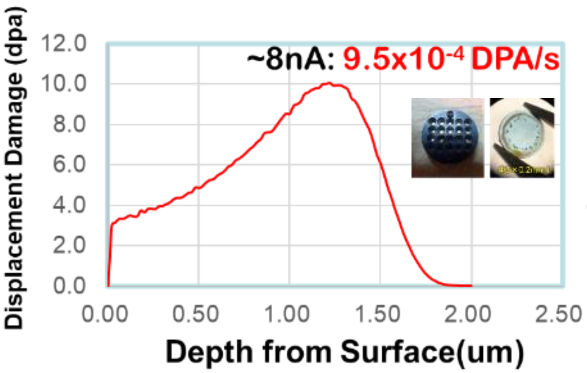
- **Alternative radiation damage method**
  - **Low-energy ion irradiation**
    - Lower cost, high dose rate (without activation)
    - Narrow penetration depth
      - Micro-mechanics and meso-scale testing
    - Doesn't reproduce transmutation gas production
      - He/H implantation with dual or triple irradiation

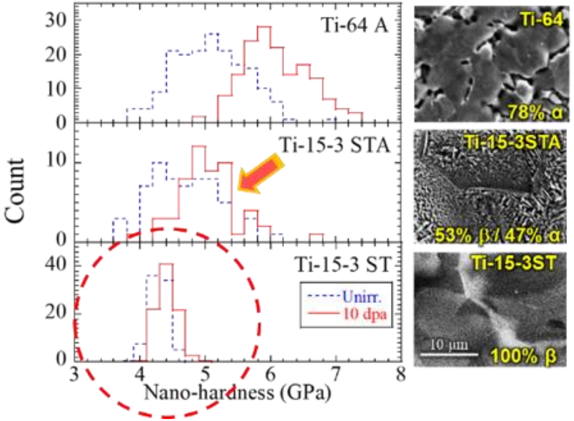


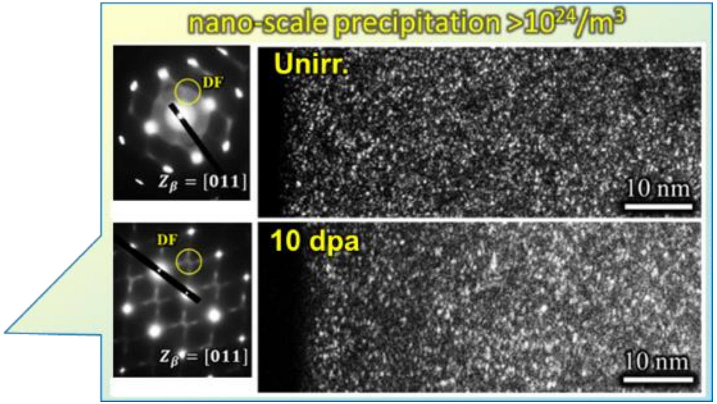


Low Energy Ion Beam Irradiation to High DPA

High Fluence Irradiation Facility at the University of Tokyo – 2.8 MeV Fe^{2+}







- Ti-15-3 (STA) is typically aged at ~500 °C to precipitate α-phase that enables higher temperature operation
  - Clear irradiation hardening (but less than Ti-64 A)
- The single metastable β phase Ti-15-3 (ST) alloy exhibits high radiation damage tolerance, that does not undergo irradiation hardening up to 10 dpa at room temperature

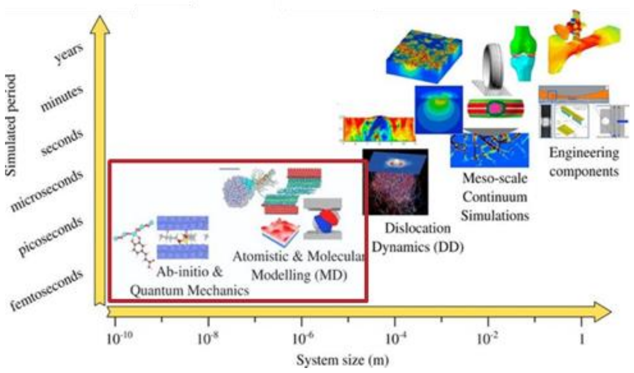
> Evidence that Ti-3Al-2.5V alloy with specific heat treatment is more radiation-tolerant

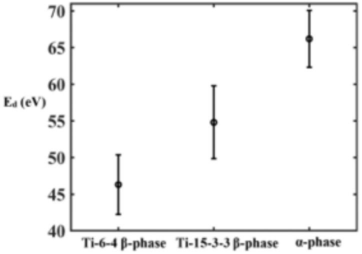
Radiation Damage Modeling

- Molecular dynamics simulations of Ti alloys at PNNL
  - Ti-15-3-3 is more radiation damage resistant than Ti-6-4 due to the higher displacement threshold energy of its β-phase

- Collaboration with Computation Materials Group at University of Wisconsin – Madison
  - Modeling of He gas bubbles in Beryllium
    - Simulated shape of He bubble in Be is consistent with earlier experimental observations
  - Modeling of radiation damage in High Entropy Alloys

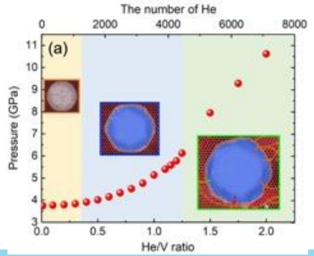
- Need more experimental data from irradiation station (proton, heavy ion, …) with controlled parameters to validate our models

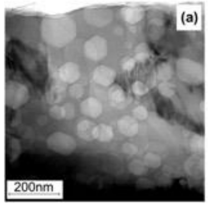




Displacement threshold energies in the α and β-phases of Ti-6Al-4V and Ti-15V-3Cr-3Al using molecular dynamics

A Roy et al., J. Nuclear Materials 586 (2023) 154695





J. Xi et al., J. Nuclear Materials 576 (2023) 154249

M. Klimenkov et al., J. Nuclear Materials 443 issues 1-3 (2013) 409-416

College of Engineering
UNIVERSITY OF WISCONSIN–MADISON

Fermilab

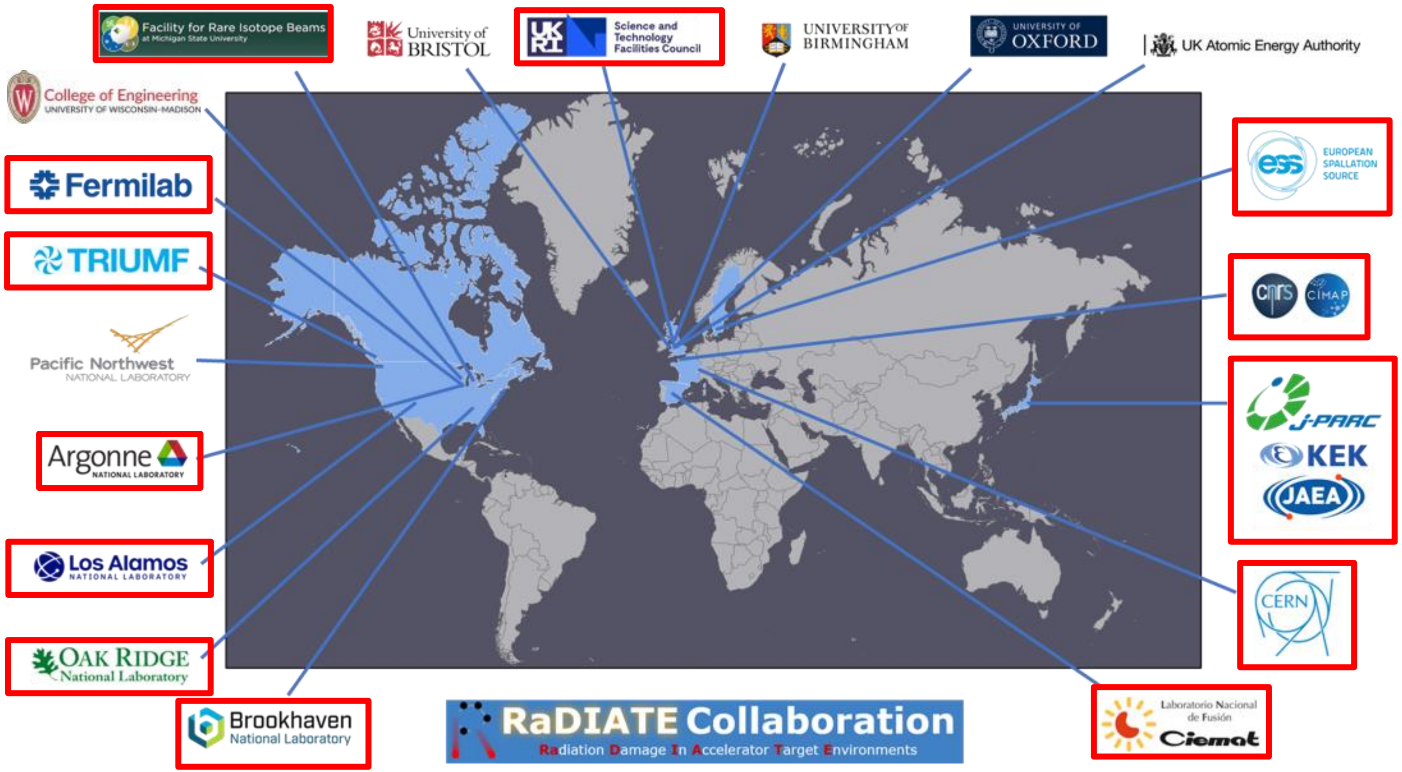
Pacific Northwest
NATIONAL LABORATORY

RaDIATE Participating Institutions

Accelerator facilities, irradiation stations, PIE institutions

R a D I A T E Collaboration

Radiation Damage In Accelerator Target Environments



Facility/Accelerator

RaDIATE Collaboration

Radiation Damage In Accelerator Target Environments

Heavy Ion Irradiation station

Proton Irradiation station

Proton Beam

- Intermediate energy
  - University of Birmingham (up to 40 MeV)
- High energy
  - CERN-HiRadMat : 440 GeV.
    - Single pulse for thermal shock
  - Los Alamos-LANSCE (100 MeV) and Brookhaven-BLIP (up to 200 MeV)
    - BUT dedicated for isotope production and not a user facility

RaDIATE Collaboration

Radiation Damage In Accelerator Target Environments

Post Irradiation Examination

Post Irradiation Examination (as user facility)

- Most of them have limited capabilities
- Few of them are user facilities
- Advantageous to have the PIE close to the irradiation station
- Commercial PIE services: Framatome, Westinghouse, etc.)

PIE capabilities of RaDIATE Collaboration Institutions

PNNL as a user facility

- Reduced sample size to accommodate PIE in fume hoods or bench tops
- 3 types of hot cells dedicated for numerous PIEs
  - Sample preparation
  - Mechanical and physical testing
  - Thermo-physical characterization
  - Microstructural analysis (optical, SEM, TEM, AFM)
  - Mass spectrometry (APT, TIMS, ICP-MS, HR mass spec for H and He isotopes)
  - Analytical chemistry
  - Surface science (AES, XPS, SIMS, FTIR)

UKAEA-MRF as a user facility

- Multiple hot cells dedicated for numerous PIEs
  - Sample preparation
  - Mechanical testing (nanoindentation, load frame for static and dynamic tests, small push testing, DIC,…)
  - Thermo-physical characterization (laser flash analyzer, dilatometer, physical properties measurement, calorimetry…)
  - Microstructural analysis (FIB, SEM, TEM, Raman spectroscopy, XRD, …)

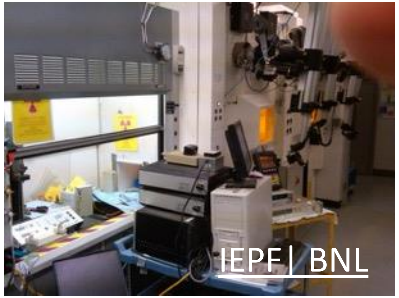
https://www.pnnl.gov/radiochemical-processing-laboratory

https://mrf.ukaea.uk/equipment/

More PIE capabilities…

Hot cells

ANL (Irradiated Materials Lab), ORNL (Irradiated Materials Examination and Testing Laboratory), TRIUMF, LANL, BNL (Isotope Extraction and Processing Facility), CERN (ISOLDE)



Facilities for low activation materials

ORNL (Low Activation Materials Development and Analysis), University of Wisconsin (MADCOR)

For more details:

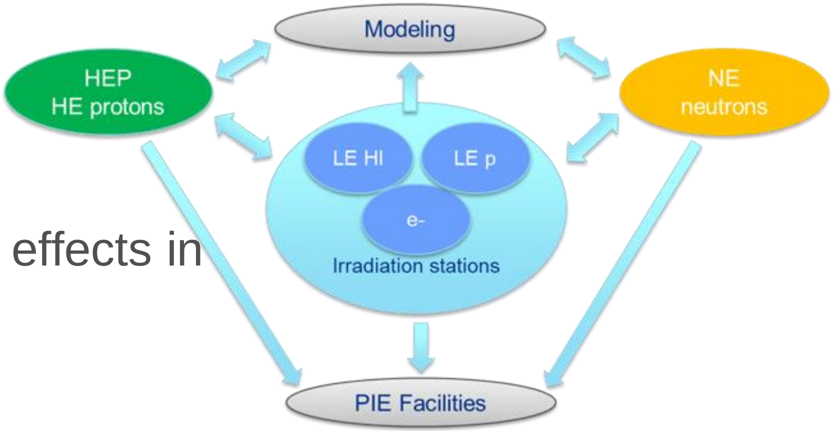
- Irradiation Stations and Alternatives workshop
- Irradiation Facilities and Irradiation Methods for High Power Target whitepaper

Synergy with superconducting magnet development

- Superconducting magnet environment
  - Secondary particle interaction
  - Radiation damage and heat deposition effects on performance

- High Power Targetry environment
  - Much higher level of damage with direct beam interaction

- Tools and approach to study radiation damage effects in materials are similar
  - Irradiation stations
  - Post Irradiation Examination techniques
  - Existing data in the literature (neutron irradiation)



Questions?