



## Novel Materials R&D for Next-Generation Accelerator Target Facilities

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# Outline

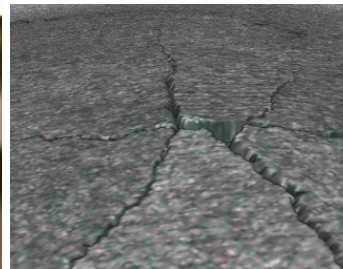
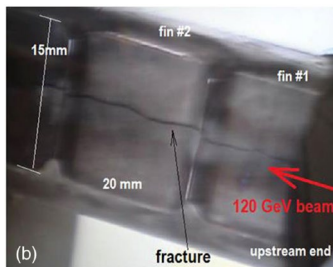
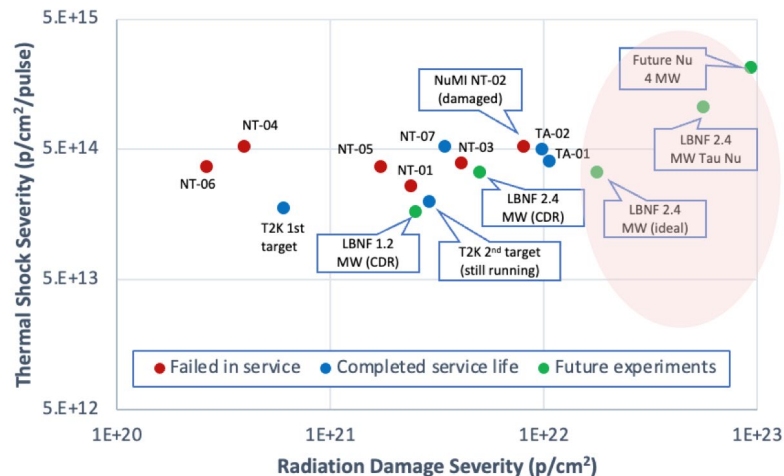
- Introduction and context
- HEA potential benefits
- HEA Compositions
- Characterization studies
- Upcoming irradiation studies
- Brief intro to nanofiber work

# High-power targetry (HPT) overview and challenges

## Neutrino HPT R&D Materials Exploratory Map

Next generation multi-MW accelerators expect proton fluence and power density to increase  $\approx 10X$  over previous facilities

- Target survivability concerns have led several facilities to limit beam power
- Radiation damage and thermal shock are the primary material challenges (RIGHT)
- Novel materials offer promising options to mitigate these effects
  - High-entropy alloys (HEAs) as beam windows
  - Electrospun nanofibers as secondary particle production targets



# Radiation damage and thermal shock effects

## Radiation damage:

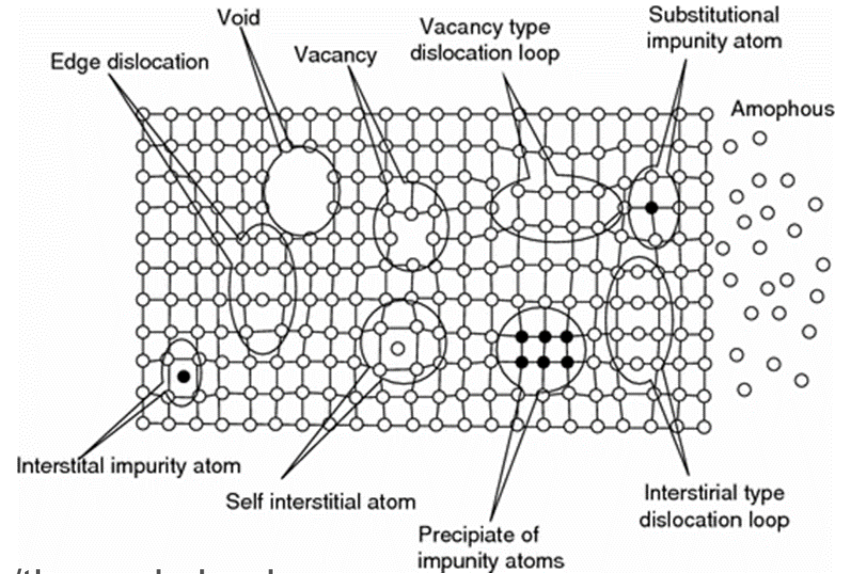
- Displacement of atoms from lattice sites
- Transmutation: interstitials and vacancies + gas

## Material effects:

- Hardening/embrittlement
- Fracture toughness reduction
- Lattice expansion/bulk swelling
- Thermal conductivity reduction
- Thermal expansion coefficient

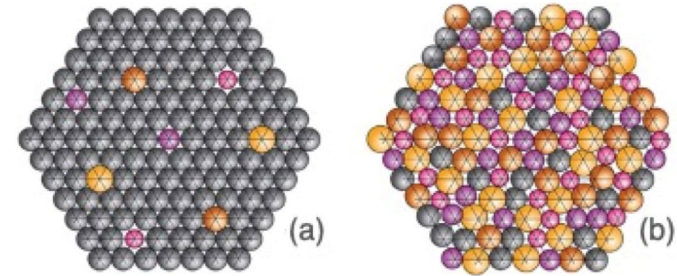
## Beam window properties: mitigate radiation damage/thermal shock

- Resistance to radiation damage effects (embrittlement, swelling)
- High thermal diffusivity (cooling) and specific heat (minimize  $\Delta T$ )
- Low coefficient of thermal expansion
- Good (high T) strength/ductility to survive beam pulse stresses
- Low density to minimize beam energy loss and scattering

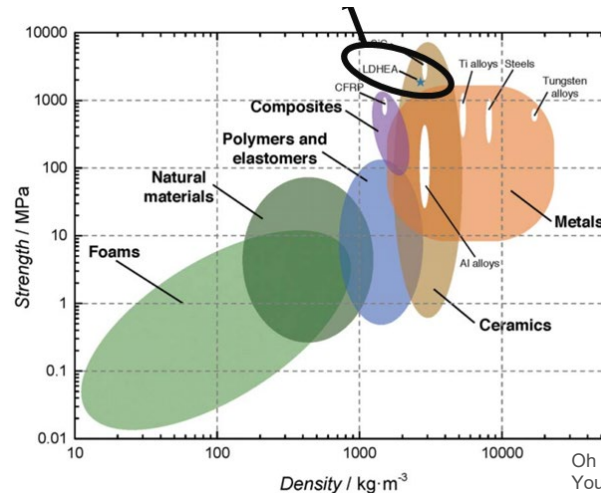


# High entropy alloys (HEAs)

- Alloy with 3+ principal elements
- Near equi-atomic compositions
- Primarily a solid-solution matrix with distorted crystal lattice (atomic size difference)
- Large composition space (adjustment of atomic ratios)



(a) Conventional alloy, (b) High-entropy alloy  
(Miracle & Senkov, 2016)



Many HEAs exhibit:

- Good ductility and high-temperature strength
- High strength to density ratio (specific strength)
- Fatigue, fracture, corrosion, oxidation resistance

Oh et al., Nat Comm. 10, 2090 (2019)  
Youssef et al., Materials Research Letters, 95-99 (2015)



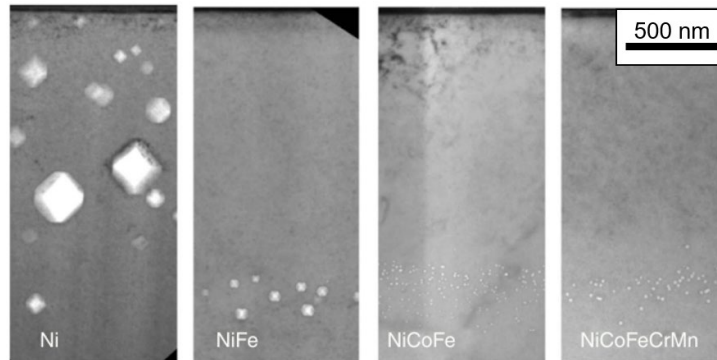
# HEA radiation damage resistance

HEAs: beneficial properties to combat damage

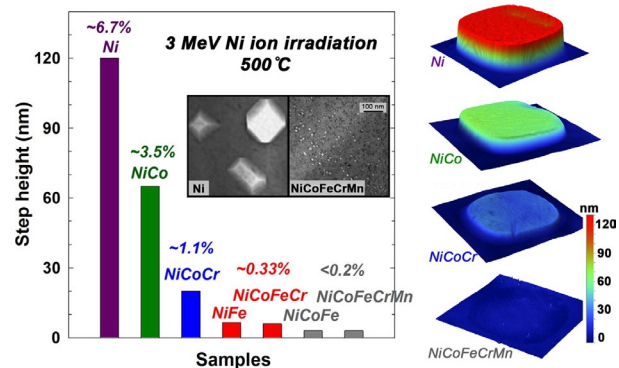
- Sluggish atom diffusion
  - Due to distortion & size mismatch
  - Reduced segregation and defect clustering
  - Increased in-cascade recombination
- Phase stability → reduce grain coarsening and void swelling

Reduced defect segregation/clustering + increased recombination:

- Minimizes void formation & swelling (right, top)
- Reduces bulk swelling effects (right, bottom)
- Increasing # of elements → greater effects (vs. pure materials/traditional alloys)
- Phonon scattering/migration energies



Void swelling shown to be less pronounced in more compositionally complex alloys upon heavy-ion irradiation (3-MeV  $\text{Ni}^+$  ions to  $5 \times 10^{16} \text{ cm}^{-2}$  at 773 K), Lu et al., Nature Com., 2016



Swelling of increasingly complex alloys under ion irradiation, Jin et al., Scripta Materialia 119 (2016)



# 2 generations of compositions

## 4 Gen. 1 HEA compositions

- Varying number of alloying elements:
  - CrMnV: Equimolar with single BCC phase
  - CrMnTiV: Ti as impurity getter
  - AlCrMnTiV: Al to stabilize BCC phase
  - AlCoCrMnTiV: Co for B2 precipitates
- HIP at 1200 °C for 4 hr following arc-melt
- Homogenized at 1200 °C for 48 hr



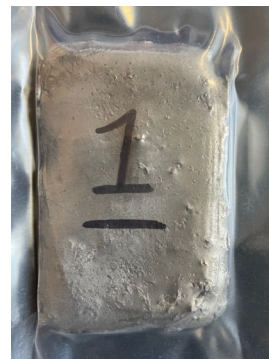
Sectioned arc-melted ingots (UW-Madison)



HEA samples sealed in quartz under vacuum before heat treatment (UW-Madison)

## 8 Gen. 2 compositions from Sophisticated Alloys to study effects of relative concentration

- 5 AlCoCrMnTiV compositions to study:
  - Varied Ti concentration as impurity getter
  - Varied Co concentration to promote secondary B2 phase
- Increased Al content without Cr for BCC phase stability
- Absence of Co: Al as B2 phase enhancer



Gen. 2 plate from Sophisticated Alloys

- 4 hr HIP under Argon
  - 1200 °C, 100 MPa
- Vacuum Anneal
  - 1200 °C for 4 hrs
  - Cool to 650 °C
  - Soak for 36 hr
  - Force cool to < 55 °C in Ar



# Initial pre- and post-irradiation characterization

## Pre-irradiation studies

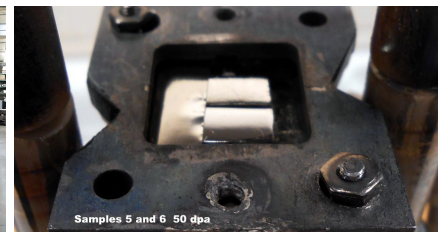
- Elemental composition
  - Homogeneity and impurity segregation
- Grain structure and orientation
  - Grain size distribution
  - Orientation impact on mechanical properties
- Crystal lattice structure and spacing
  - BCC composition with B2 precipitates?
- Mechanical properties
  - Hardness and elastic modulus
  - Yield/tensile strength
  - Fatigue life
- Specific heat capacity
  - Higher to minimize  $\Delta T$  during a pulse
- Coefficient of thermal expansion
  - Minimize to lessen thermal shock effects

## Post-irradiation comparison

- Ti64: baseline performance
- Elemental Segregation
- Grain coarsening
- Crystal lattice expansion
  - Phase stability
- Defect production & dislocation formation
  - Void formation
  - Bulk swelling
- Hardening and embrittlement



Wisconsin Ion Beam Laboratory (IBL)

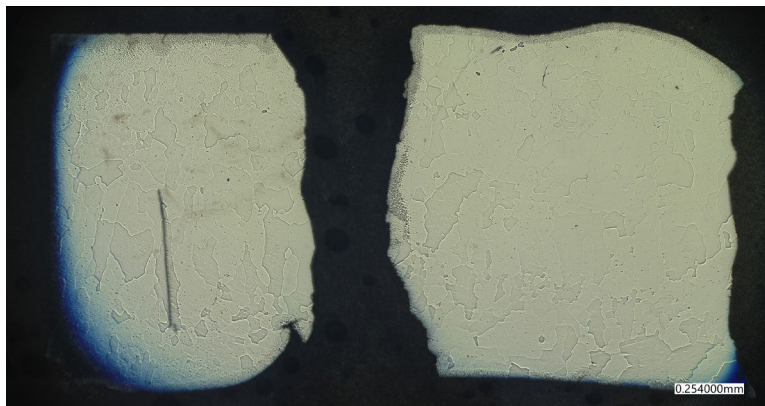


Gen. 1 HEAs for irradiation

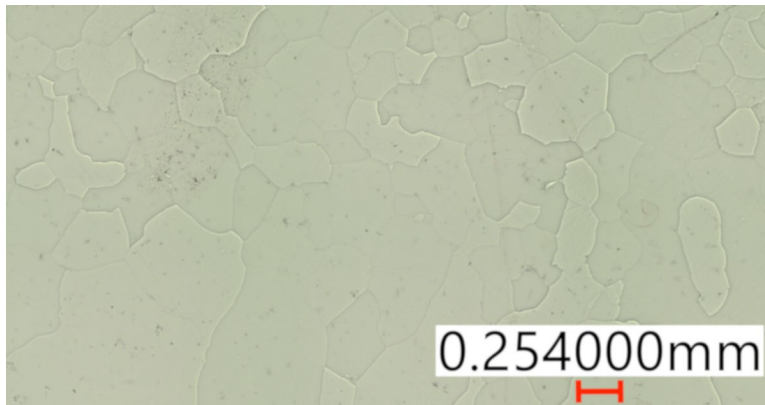
# Pre-characterization

## SEM/nanoindentation specimen preparation

- Sectioning with TechCut 4x low-speed saw
- Grinding and polishing using Metprep3 system
- Optical microscopy inspection with Keyence VHX-7000 microscope
- Preliminary grain size measurements:
  - 150 – 500  $\mu\text{m}$



HEA 2.6, 20X

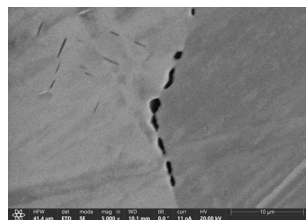


HEA 2.6, 100X

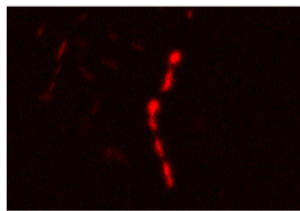
# EDS: homogeneity & impurities

## Energy Dispersive X-ray Spectroscopy (EDS)

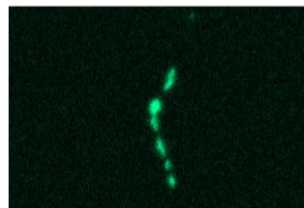
- RIGHT: High degree of homogeneity observed
- BELOW: Ti working well as impurity getter



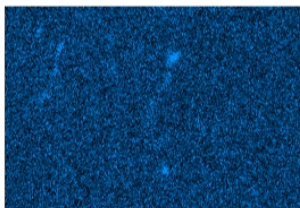
S K $\alpha$ 1



10 $\mu$ m Ti K $\alpha$ 1,2

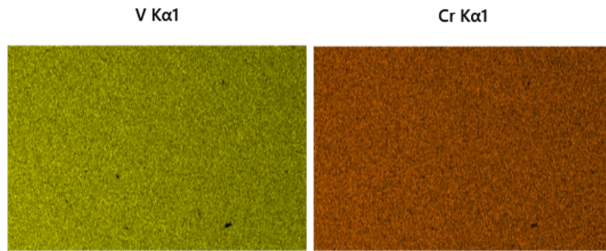


10 $\mu$ m



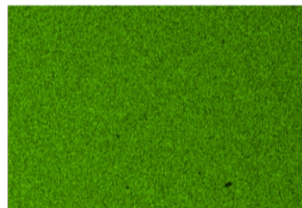
10 $\mu$ m

Ti K $\alpha$ 1

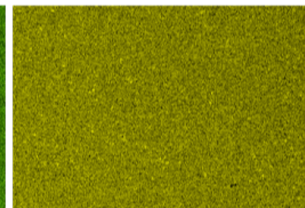


250 $\mu$ m V K $\alpha$ 1

Cr K $\alpha$ 1



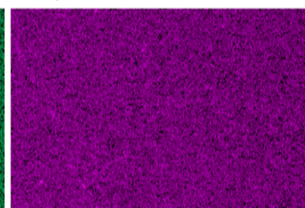
250 $\mu$ m Mn K $\alpha$ 1



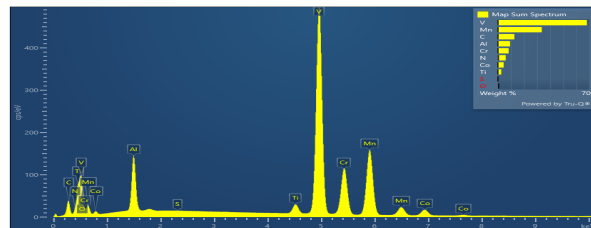
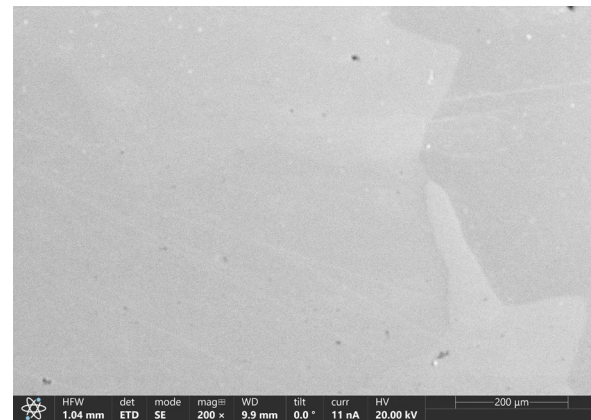
250 $\mu$ m Al K $\alpha$ 1



250 $\mu$ m Co K $\alpha$ 1



250 $\mu$ m Ti K $\alpha$ 1



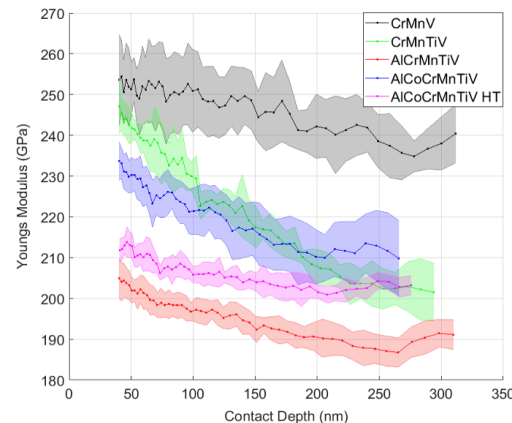
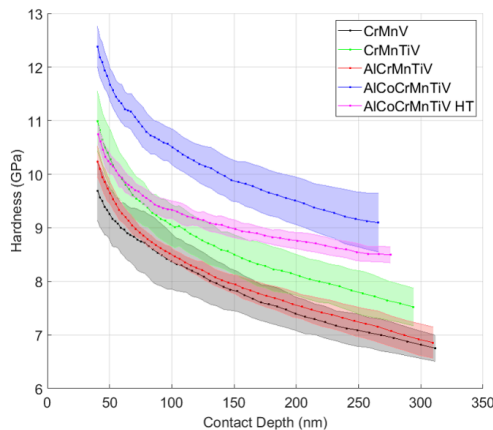
AlCoCrMnTiV Compositions

# Indentation studies

Gen. 1 studies completed

- Significant hardness increase with increased complexity
- Mitigated by heat treatment
- Signs of ductility
- Stiffer than Ti-64, less than Beryllium

Gen. 2 studies ongoing



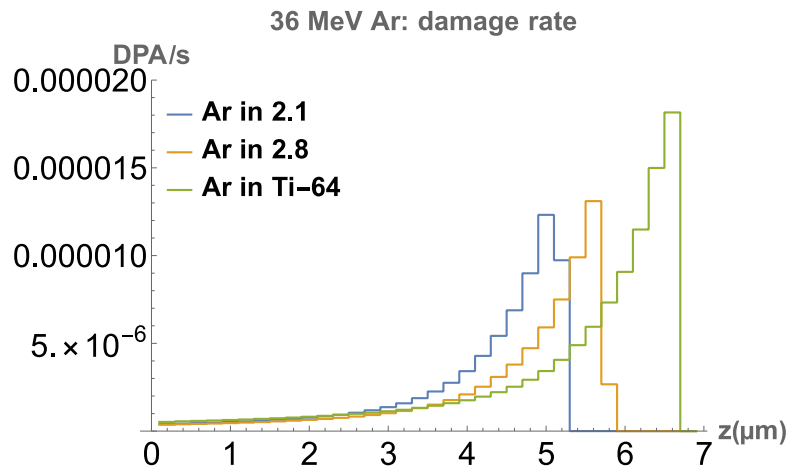
Sample	Vicker's Hardness (Hv)	Modulus of Elasticity (GPa)
CrMnV	390.58	186.93
CrMnTiV	499.80	222.06
AlCrMnTiV	453.06	163.36
AlCoCrMnTiV	608.19	177.29
Ti64	339	110

N. Crnkovich, UW-Madison

# Recent/upcoming ion irradiations

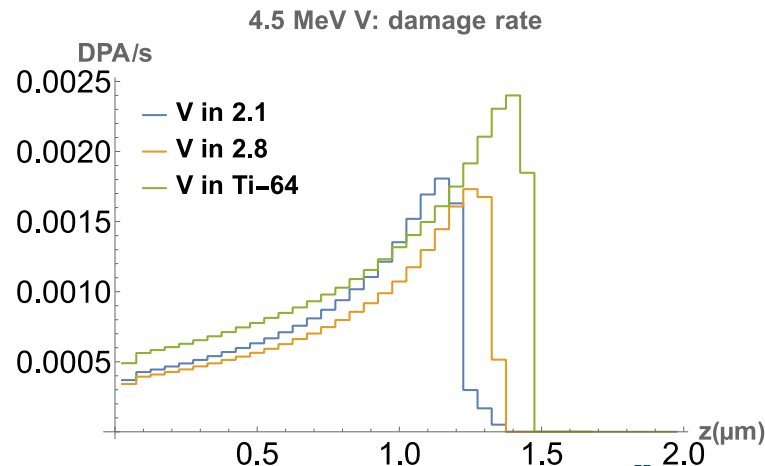
## Argon ion irradiation

- IRRSUD beamline at the GANIL facility, Caen France by UW-Madison collaborators
- $^{36}\text{Ar}^{10+}$ , 1 MeV/A
- Damage: 0.1, 0.3, and 0.4 DPA at
- 550° C ( $\approx$  1/3 melting point  $\rightarrow$  defect mobility)
- Study defect formation, void formation/stability
- Ion irradiation: Fast/inexpensive for screening



## Vanadium ion irradiation

- Ion Beam Lab at UW-Madison with 4.5 MeV  $\text{V}^{2+}$
- ARC-DPA calculations:
  - $\sim$  10 min. (peak) – 40 min. (surf.) for 1 DPA
- Damage levels of 1, 5, and 10 DPA at 550° C and  $\approx$  200° C (estimated peak damage and service temperature of HEA beam window)
- Ti-64 for baseline comparison

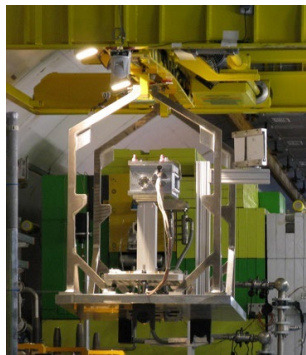
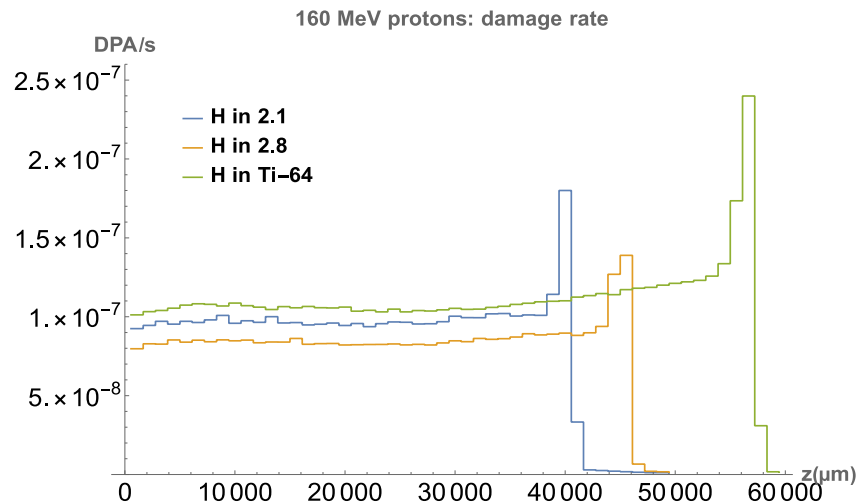




# Future prototypic irradiations

BNL BLIP facility 160 MeV proton irradiation

- Lower damage levels compared to LE-ions (long times)
- Mimics transmutation/gas production
- Increased penetration depth allows for studying irradiation effects on bulk properties

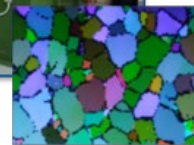


Some 2nd gen. HEAs to be tested at CERN's HiRadMat facility with 440 GeV protons in 2025

- Allows for single-shot pulses to test thermal shock susceptibility (below: iridium target)



Pacific Northwest  
NATIONAL LABORATORY



Increased beam energies result in activated specimens

- Hot-cell work for characterization

# Nanofiber targetry studies

## In-house electrospinner at FNAL

- Electrohydrodynamic production of nanoscale fiber mats
  - Zirconia nanofiber production in place
  - Tungsten nanofibers under investigation
- Irradiation experiments show good resistance to damage/thermal shock (density dependent)
  - CERN's HiRadMat facility (thermal shock)
  - In-situ ion irradiation & TEM at the Argonne National Laboratory – IVEM facility (defects and lattice expansion)

## Inherent resistance to radiation damage/thermal shock

- Radiation tolerance
  - Nanopolycrystalline grain structure → absorb defects
- Thermal shock
  - Discrete at microscale
  - Reduced temperature gradient (only along fiber)
  - Good heat dissipation in gas (high surface area/porosity)
  - Absorb/dampen stress waves

(Bidhar et al., PRAB, 24, 2021)

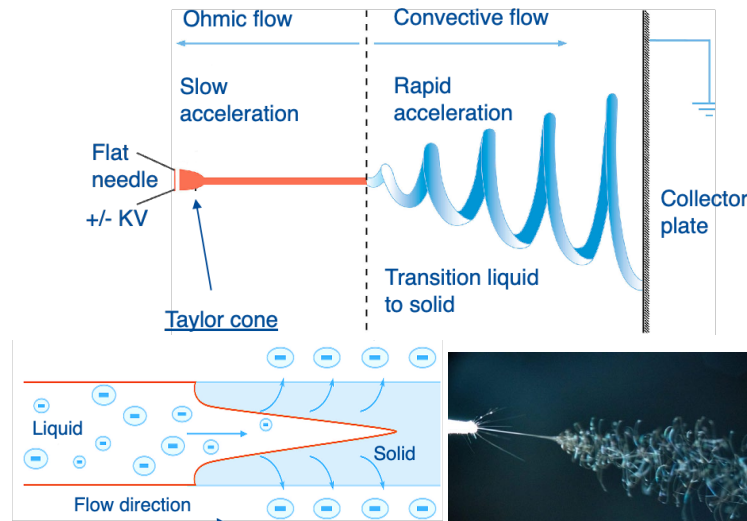
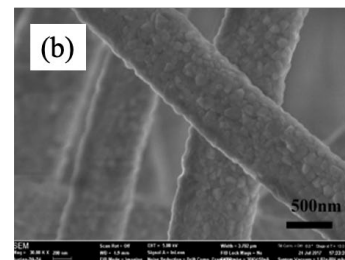
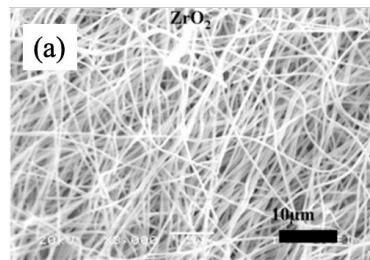


Photo: Reidar Hahn, FNAL



Zirconia nanofibers produced at Fermilab

(a) Bulk nanofiber mat

(b) Single nanofibers revealing polycrystalline grains

# Summary

- Novel materials studies for high-power accelerator windows (HEAs) and particle production targets (nanofibers)
- HEA compositions tailored to mitigate radiation damage and thermal shock susceptibility
- Characterization studies ongoing for two generations of HEAs
- Ion irradiation experiments to determine radiation resistance and property alterations
- Electrospun nanofibers found to have

# Acknowledgements

- UW-Madison group: A. Couet, N. Crnkovich, M. Moorehead, I. Szufarska
- Fermilab HPT group: K. Ammigan, G. Arora, S. Bidhar, F. Pellemoine
- Work being done in the framework of the RaDIATE Collaboration
- Funded by DOE Early Career Award (Kavin Ammigan)



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# Bonus slides



# PIE techniques

Material property	Technique	Probe depth
Elemental/chemical composition	Energy-Dispersive X-Ray Spectroscopy (EDS/EDX)	nms
Crystallographic grain structure/orientation	Electron Backscatter Diffraction (EBSD)	nms
Atomic spacing, lattice defects and spacing	Transmission Electron Microscopy (TEM)	< 1 - 5 $\mu$ m
Nanocrystalline and ultra-fine grain microstructural characterization	Transmission Kikuchi Diffraction (TKD/t-EBSD)	< 1 - 5 $\mu$ m
Atomic structure, lattice parameters	Grazing Incidence X-Ray Diffraction (GIXRD)	ums
Hardness, Elastic modulus	Nanoindentation	nms - ums

High damage rate (surface)

Material property	Technique	Probe depth
Specific heat capacity	Differential Scanning Calorimetry (DSC)	~ 1 mm
Coefficient of thermal expansion	Dilatometer	1 - 5 mm
Elastic modulus, Yield strength, Tensile strength, Elongation	Tensile Tester	~ 1 mm
Fatigue life and endurance limit	Fatigue tester (conventional or mesoscale)	~ 1 mm
Thermal diffusivity/conductivity	Laser Flash Analyzer (LFA)	~ 1 mm

Damage (bulk)

# Pre- and post-characterization

Microstructural studies to be paired with ion irradiation experiments (shallow penetration)

- EDS/EBSD studies of pristine Gen. 2 HEAs ongoing at Fermilab Material Science Lab
- TEM work scheduled for April at UW-Madison Materials Science Lab
- Nanoindentation at Fermilab Target Systems Department Materials Lab

Material property	Technique	Probe depth
Elemental/chemical composition	Energy-Dispersive X-Ray Spectroscopy (EDS/EDX)	nms
Crystallographic grain structure/orientation	Electron Backscatter Diffraction (EBSD)	nms
Atomic spacing, lattice defects and spacing	Transmission Electron Microscopy (TEM)	< 1 - 5 $\mu$ m
Nanocrystalline and ultra-fine grain microstructural characterization	Transmission Kikuchi Diffraction (TKD/t-EBSD)	< 1 - 5 $\mu$ m
Atomic structure, lattice parameters	Grazing Incidence X-Ray Diffraction (GIXRD)	ums
Hardness, Elastic modulus	Nanoindentation	nms - ums

# HEA compositions for study

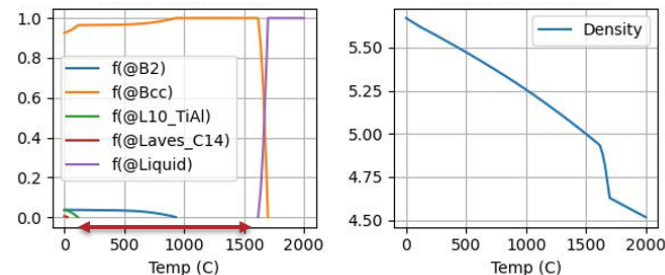
Element	Time to LLW/yr	Element	Time to LLW/yr	Element	Time to LLW/yr
C	99	V	54	Zn	$1.1 \times 10^3$
N	$9.4 \times 10^4$	Cr	40	Y	21
O	$1.1 \times 10^4$	Mn	86	Zr	$> 10^6$
Mg	97	Fe	59	Nb	$2.9 \times 10^5$
Al	157	Co	184	Mo	$8.7 \times 10^5$
Si	58	Ni	$6.6 \times 10^5$	Ta	41
Ti	10	Cu	$1.3 \times 10^3$	W	23

P.J. Barron, A.W. Carruthers and J.W. Fellowes et al.,  
Scripta Materialia 176 (2020) 12–16

M. Gilbert, T. Eade, T. Rey, R. Vale, C. Bachmann, U.  
Fischer, N. Taylor, Nuclear Fus. 59 (7) (2019) 076015

# CALPHAD: HEA Design Refinement

- Systematic compositional space search using high-throughput CALPHAD simulations
  - > 8,500 compositions for optimization of (N. Crnkovich, UW-Madison)
    - B2 phase region
    - Onset of embrittling secondary phases
    - Density, specific heat, CTE
  - > 120,000 compositions were explored (G. Arora, FNAL)
    - New stable single-phase BCC alloys founds



## Compositional search space (at%)

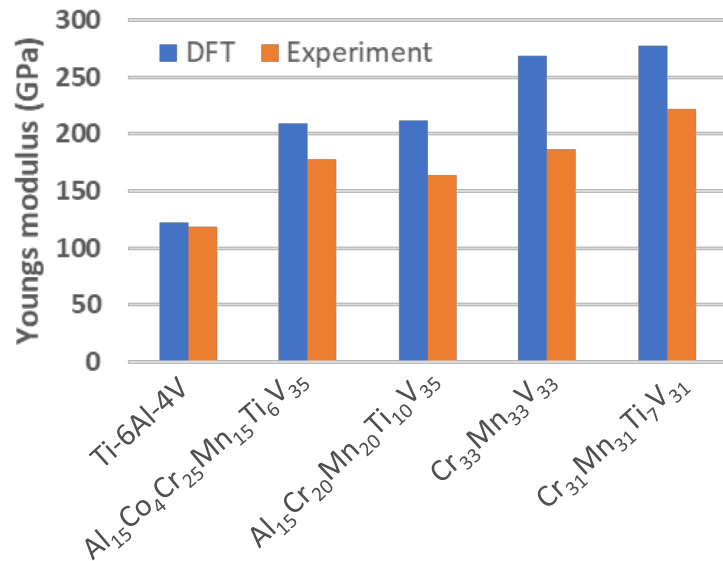
	Al	Co	Ti	Cr	Mn	V
Min	10	0	1	10	1	10
Max	30	5	15	35	30	35
Step	4	1	2	3	Bal	2

N. Crnkovich, UW-Madison

	Al	Co	Ti	Cr	Mn	V
Min	0	0	0	0	0	0
Max	20	5	20	50	50	50
Step	2	1	2	3	3	Bal

G. Arora, FNAL

# Mechanical properties/DFT simulations



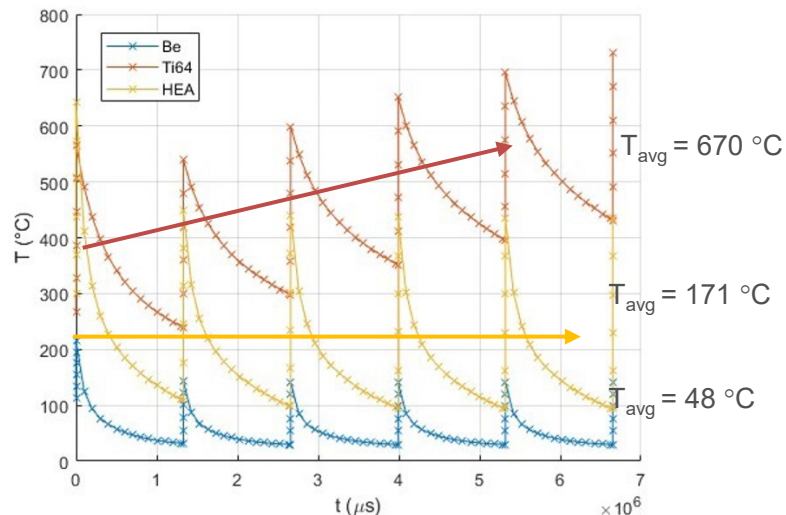
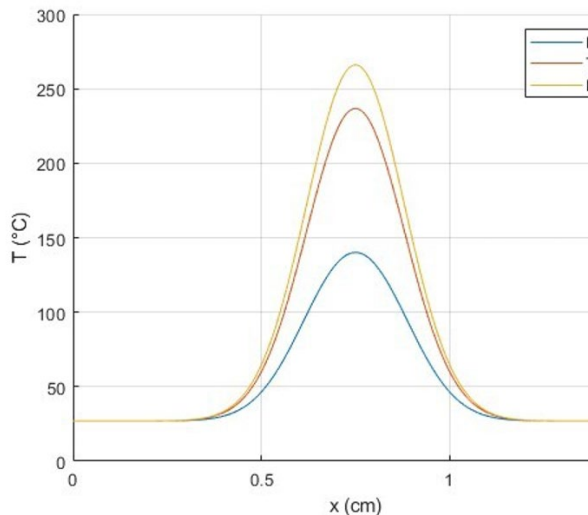
DFT (G. Arora, FNAL)

Measurements (N. Crnkovich, UW-Madison)



# CALPHAD/DFT simulations

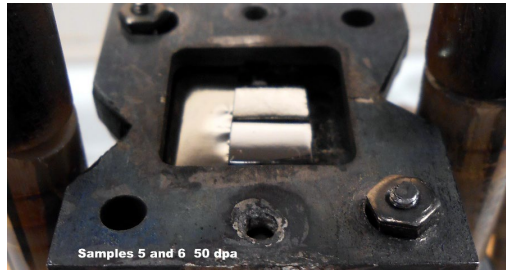
- FNAL NuMI beam line case, based on CALPHAD-predicted properties
  - 120 GeV protons - 1 MW
  - 10  $\mu$ s pulse @ 0.75 Hz repetition
  - 3 cm diameter window, edge held at 300 K



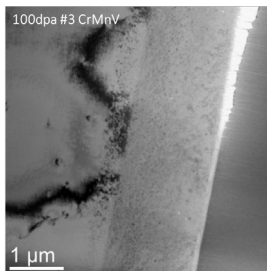
N. Crnkovich, UW-Madison

# Initial In-Beam Tests

$V^{2+}$  ion irradiation 50/100 DPA at 500 °C  
at Wisconsin Ion Beam Laboratory



Two HEAs being prepared for irradiation with a mask

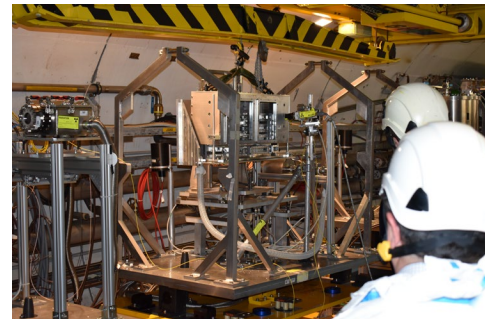


TEM BF image of the irradiated cross-section

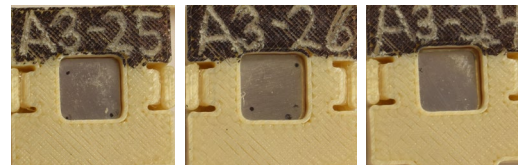
## PIE is ongoing

- What are the dominant irradiation effects?
- Effects visible at low DPA?

Thermal shock test at CERN's HiRadMat facility (2022)



- HEA samples included in HRMT-60 RaDIATE experiment
- Exposed to single-shot  $7 \times 10^{12}$  proton beam ( $\sigma \sim 0.25$  mm)



HEA samples images after beam pulse

- Detailed PIE planned at UKAEA-MRF

# Facilities / beams

- Irradiation condition
  - High damage rate (surface)
  - Damage (bulk,  $E > 22.5$  MeV)
  - Helium implantation / diffusion study
  - Prototypic ("high" energy)

Host Laboratory	Facility	Beam energy	Beam Intensity	Penetration depth ( $\mu\text{m}$ )		
				2.1	2.8	Ti-64
BNL	BLIP - proton	120 - 200 MeV	50-165 $\mu\text{A}$	mm: 24.1 - 58.1	27.3 - 65.7	34.1 - 82.8
	BLIP - HI	2 - 28 MeV/A		He: 22.4 - 1870 V: 10.3 - 222.0 Ar: 9.9 - 262.9 Kr: 11.5 - 172.4 Xe: 12.7 - 141.2	24.8 - 2110 11.56 - 250.1 11.2 - 296.1 12.9 - 380.0 14.3 - 159.0	31.2 - 2650 13.5 - 308.5 13.0 - 364.7 15.1 - 238.2 16.7 - 195.2
University of Birmingham	MC40 - proton	2.7 - 38 MeV	pA to 10's of $\mu\text{A}$	36.3 - 3120	40.7 - 3500	50.9 - 4410
	MC40 - HI	8 - 50 MeV		He: 22.4 - 456 V: 1.9 - 5.7 Ar: 2.2 - 6.8 Kr: 1.9 - 5.4 Xe: 1.4 - 4.8	24.8 - 516 2.2 - 6.4 2.4 - 7.6 2.1 - 6.1 1.5 - 5.4	31.2 - 647 2.5 - 7.3 2.8 - 8.7 2.4 - 6.9 1.8 - 6.2
	Dynamitron - proton	3 MeV	1 mA	41.8	46.8	58.6
	Hyperion - proton	2.6 MeV	30 mA	34	38	47
TRIUMF	ISAC - proton	13 - 500 MeV	Up to 100 $\mu\text{A}$	mm: 0.49 - 258.6	0.55 - 292.5	0.69 - 365.9
University of Michigan - MIBL	Wolverine - proton/HI	1 - 9 MeV, (6 MeV for p)	500 nA	p: 7.6 - 131 He: 1.8 - 26.8 V: 0.4 - 2.1	8.5 - 147 2.0 - 29.8 0.5 - 2.3	10.5 - 184 2.4 - 37.5 0.6 - 2.6
	Maize - proton/HI	Up to 4.5 MeV, (3 MeV for p)	Up to 1 $\mu\text{A}$	p: < 41.8 He: < 9.6 V: < 1.4	< 46.8 < 10.6 < 1.5	< 58.6 < 13.3 < 1.8
	Blue - HI	20 - 400 keV (800 keV for 2+)	Up to 10's of $\mu\text{A}$	He: 0.1 - 0.9, 1.5	He: 0.1 - 1.0, 1.6	He: 0.1 - 1.2, 2.0
J-PARC	TEF-T - proton	400 MeV	0.6 mA	mm: 182	206	258
University of Wisconsin	WIBL - HI	1.7 MeV/q	Up to 100 $\mu\text{A}$	He: 11.1 V: 1.5	12.3 1.6	15.4 1.9
University of Tokyo	HIT - HI	0.4 - 4 MeV	Up to $\sim 1$ $\mu\text{A}$	He: 0.9 - 8.2 V: 0.2 - 1.3	1.0 - 9.0 0.2 - 1.4	1.2 - 11.3 0.2 - 1.6
Kyoto University	DuET (dual beam) - HI	1 - 5.1 MeV	1 $\mu\text{A}$ -1 mA	He: 1.8 - 11.4 V: 0.4 - 1.5	2.0 - 12.5 0.5 - 1.7	2.4 - 15.8 0.6 - 1.9
GANIL	IRRSUD - HI	Up to 1 MeV/A	Up to 3 $\mu\text{Ae}$	Ar: 5.2 Kr: 6.7 Xe: 7.9	6.0 7.5 8.9	6.8 8.5 10.1

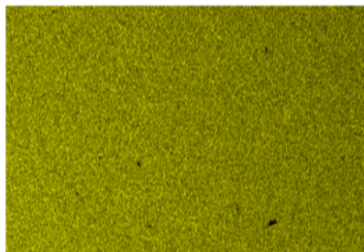
## EDS totals: 2.1B – 2.8

	2.1B			2.2			2.3			2.4			2.5			2.6			2.7			2.8		
Element	Atomic %	Specif.	% diff.	Atomic %	Specif.	% diff.	Atomic %	Specif.	% diff.	Atomic %	Specif.	% diff.	Atomic %	Specif.	% diff.	Atomic %	Specif.	% diff.	Atomic %	Specif.	% diff.	Atomic %	Specif.	% diff.
Al	9.9	10.0	-0.9%	11.7	10.0	16.8%	18.7	16.0	16.8%	18.0	16.0	12.5%	22.4	20.0	12.2%	14.0	12.0	16.7%	20.6	18.0	14.5%	22.3	20.0	11.7%
Ti	1.1	1.0	8.4%	4.3	4.0	6.4%	1.1	1.0	13.7%	1.2	1.0	17.2%	2.1	2.0	6.7%	2.1	2.0	4.8%	2.2	2.0	9.2%	2.0	2.0	1.9%
V	36.4	34.0	7.0%	34.5	34.0	1.5%	26.7	26.0	2.5%	24.3	24.0	1.2%	49.8	50.0	-0.3%	50.7	50.0	1.4%	49.6	50.0	-0.7%	49.5	50.0	-1.0%
Cr	26.5	25.0	6.1%	27.1	27.0	0.4%	25.3	25.0	1.3%	25.1	25.0	0.4%	-----	-----	-----	6.1	6.0	1.3%	-----	-----	-----	-----	-----	-----
Mn	22.1	26.0	-15.2%	18.4	21.0	-12.2%	26.2	30.0	-12.8%	27.4	30.0	-8.6%	24.5	27.0	-9.2%	24.2	27.0	-10.4%	27.6	30.0	-8.1%	24.1	26.0	-7.2%
Co	4.1	4.0	1.5%	4.0	4.0	0.0%	2.0	2.0	1.5%	4.0	4.0	0.2%	1.1	1.0	5.5%	2.9	3.0	-2.1%	-----	-----	-----	2.0	2.0	0.6%
Total	100.0	100.0	0.0%	100.0	100.0	0.0%	100.0	100.0	0.0%	100.0	100.0	0.0%	100.0	100.0	0.0%	100.0	100.0	0.0%	100.0	100.0	0.0%	100.0	100.0	0.0%

- Atomic concentrations compared to casting specifications
- 20 kV, 11 nA
- Above table: spectra fit to specified elements only: Al, Co, Cr, Mn, Ti, V

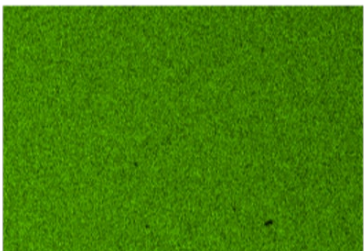
# SEM/EDS: 2.6

V K $\alpha$ 1



250μm

Mn K $\alpha$ 1



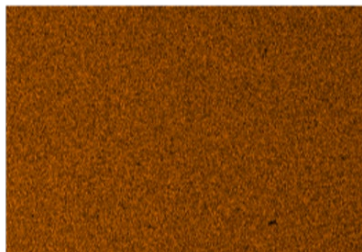
250μm

Co K $\alpha$ 1



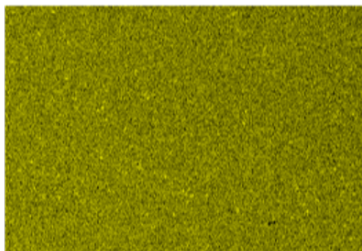
250μm

Cr K $\alpha$ 1



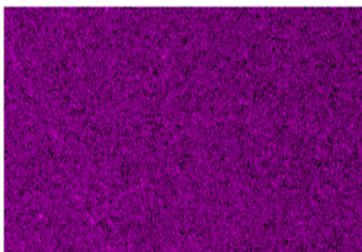
250μm

Al K $\alpha$ 1

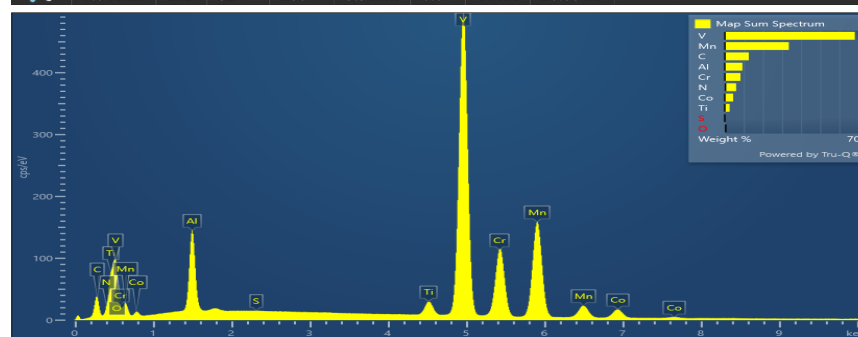
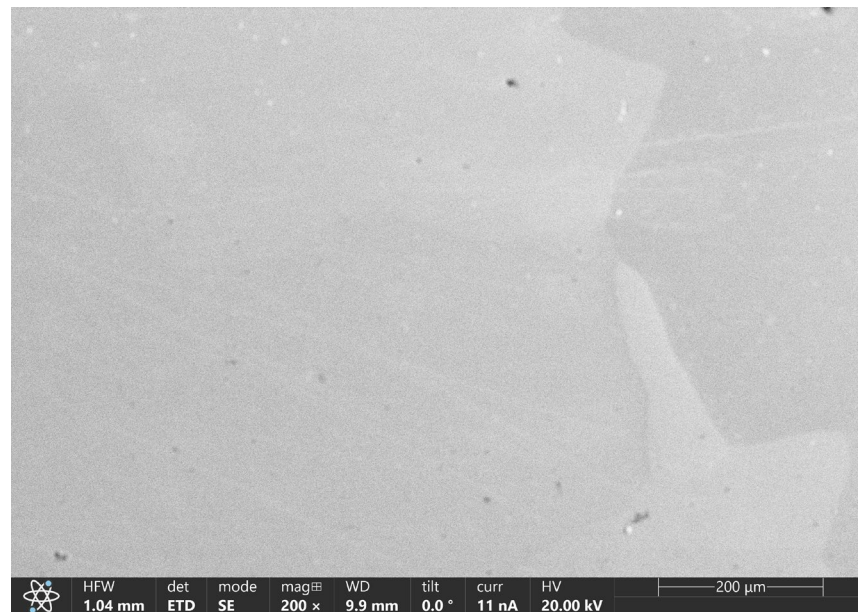


250μm

Ti K $\alpha$ 1

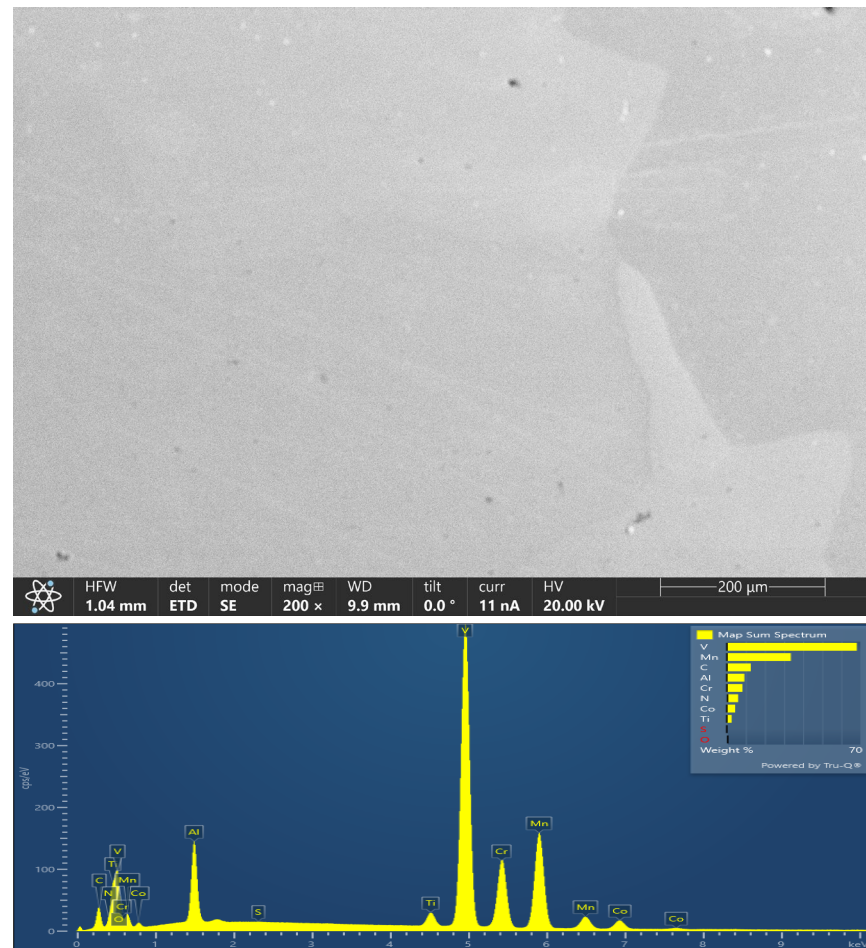
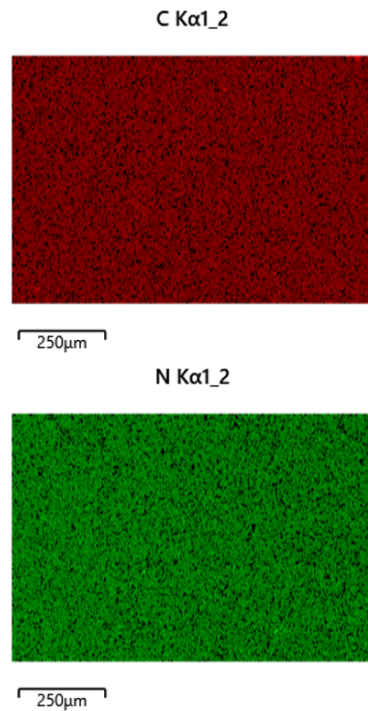


250μm

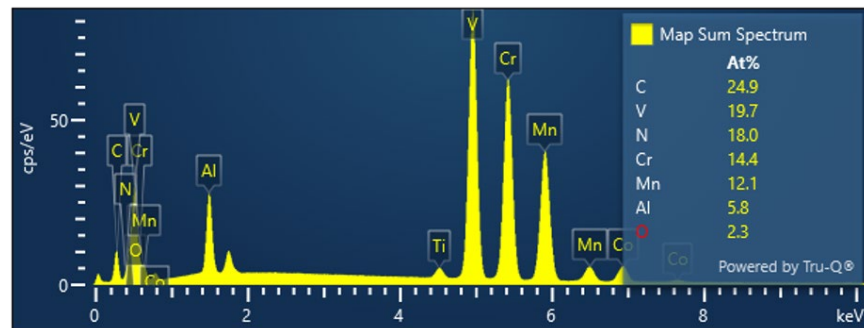
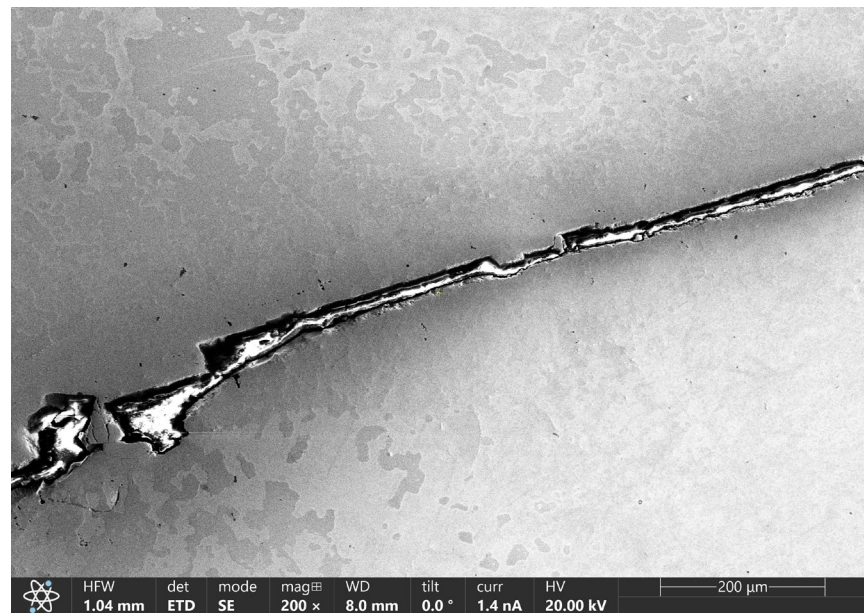
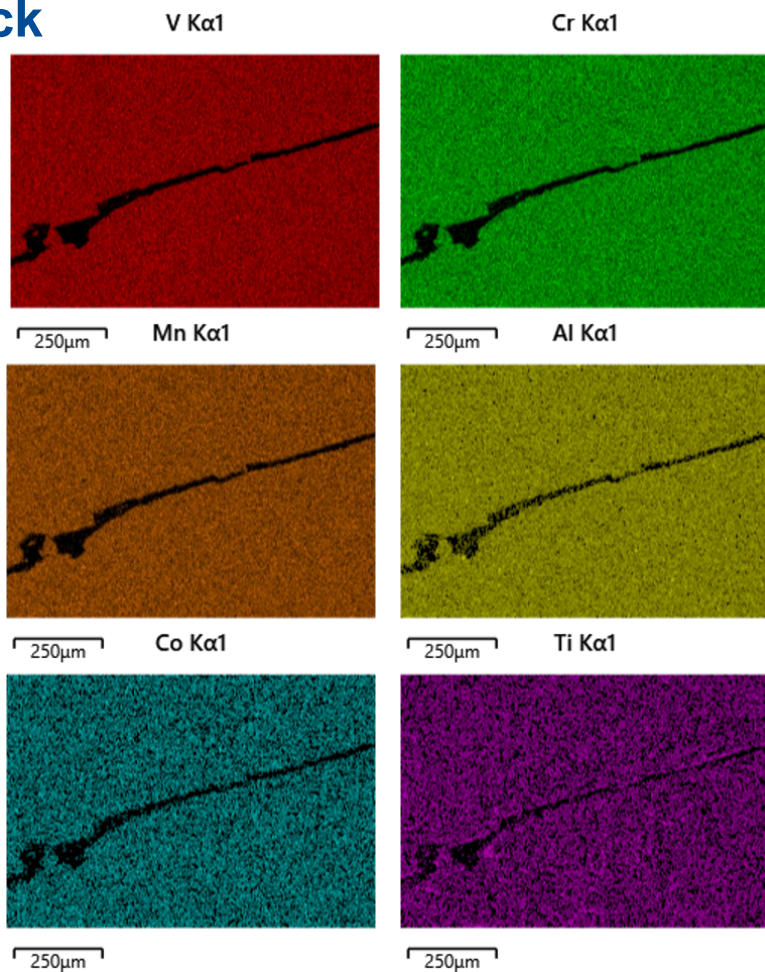




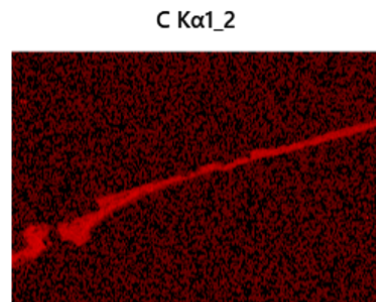
# SEM/EDS: 2.6 - impurities



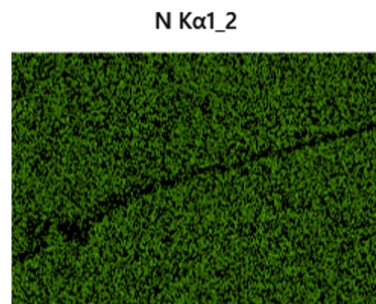
## 2.1B: Crack



## 2.1B: Crack



250µm



250µm

