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State of Knowledge and Research Activities on RPV Materials in UK

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Outline

- Background
- Evolution of current understanding of irradiation embrittlement of RPV materials
  - Properties and Physical Changes in Neutron-Irradiated Steels/Welds
  - Emphasis on model development (empirical and fundamental understanding) for predicting behavior / trends
  - Where are we now?
- Research activities in the UK (Irradiation-induced degradation of structural materials)
- Future

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Irradiation Damage and Embrittlement of RPV Steels

- Mid- 1960's onwards: Unexpected shifts in ΔT_{41J} in weld surveillance specimens. Welds had been fabricated using Cu-coated weld wire. (SP Grant, 1968)
- PRE-DB development: CVN ΔT_{41J} data analysis leading to Reg. Guide development (and revisions)
- **UK (CEGB and UKAEA): Efforts on Magnox Steels and related welds (C-Mn steels not A533B/A302B or A508 Gr3)**
- **UK Dose-Damage relationships developed**

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Irradiation Damage of Low Alloy Steels/Welds: Evaluation

- Late 1960's: Reports (S.P. Grant) of embrittlement in Cu-containing (~0.2-0.3 wt.% Cu) RPV Beltline weld specimens – concern for RPV structural integrity
- 1977: Development of US NRC Reg. Guide 1.99 (Cu and P)
  - Rev. 2 (Guthrie and Odette) for prediction of irradiation embrittlement (ΔT_{41J}) issued in 1988.
- **1987 UK: Fisher and Buswell: Correlation of irradiation embrittlement data (ΔT_{41J}), steel composition and hardness**
  - **Assumed irradiation-induced Cu precipitation in ferrite**
  - **SANS data interpreted in terms of Cu precipitation + vacancies**
  - **Yield strength increase due to irradiation; linked to hardness increase.**

$$\Delta Hv_{irrad} = \Delta Hv_{matrix} + \Delta Hv_{Cu\ precipitation}$$

- **1986/88 UK: Williams *et al.:* Ni enhanced "Cu precipitation" in promoting embrittlement.**

Explanations call for justification based on microstructural characterisation

Irradiation Damage/Embrittlement of RPV Steels (continued)

Research in 1970's+: Evaluating Cu effect on embrittlement (NRL and industry). Speculation of mechanisms. Conventional analytical techniques unable to identify the "changes" in material responsible for degraded properties. *(older methods lacked necessary resolution)*

- 1960's-1970's+: Analysis of materials/fundamental radiation damage studies/ "Russell-Brown model: Cu precipitation in Fe".
- 1983: Odette- Proposed formation of Cu-coated microvoids in welds (SANS);
- MTR irradiations (high Ni-Cu (Mn) welds); Odette: SMD, UMD, CRPs
- 1983-86: Burke, Brenner, Grant: first AP-FIM analysis of surveillance-irradiated high Cu (0.23 wt.%) and high Cu – high Ni welds; identified Cu-Ni-Mn-enriched solute clusters in welds (not visible by TEM or FIM).
- Subsequently numerous examples of irradiation-induced Cu-Ni-Mn(–Si)-enriched clusters from AP data from French (Rouen/EdF), US, UK, Japan (CRIEPI) during the 1990's-present.

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Irradiation Damage of Low Alloy Steels/Welds: Microstructural Characterisation

Microstructural Analysis and empirical studies on irradiated commercial welds, steels and model steels needed complementary techniques (bulk and nanoscale analyses)

- TEM unable to detect features/defects in RPV surveillance steels/welds. *CIEMAT: demonstrated that defects could be imaged in model binary and ternary Fe-Mn-Ni, but not in commercial steels or welds.*
- **1980's-present:** AP-FIM and (1990's+) APT: continued research
- **Late 1990's – 2008:** Emphasis on combined techniques for characterization
  - APT/SANS/TEM/PIA-PALA-Hv of A508 Gr4N low Cu steels and welds (Burke *et al.)*
  - Ringhals weld studies (APT/SANS/HRTEM/PIA-Hv)
- **2013+:** Advanced ATEM providing new insights/assess variability/etc
  - Neutron-irradiated Welds/Steels
  - Ex-service components (pressurizer weld from Ringhals)

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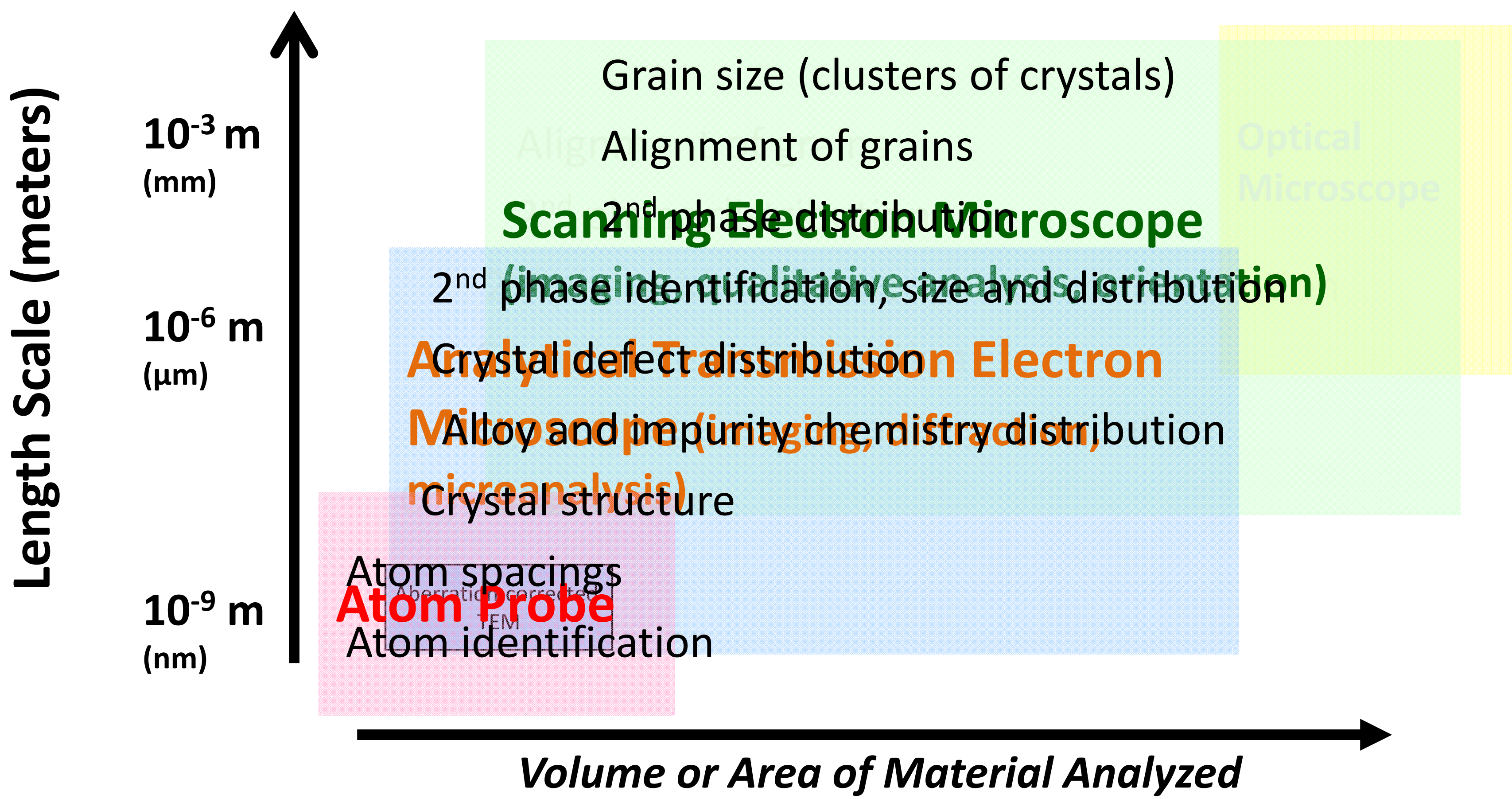
Different Instruments are Required to Resolve Fine and Coarse Microstructural Features



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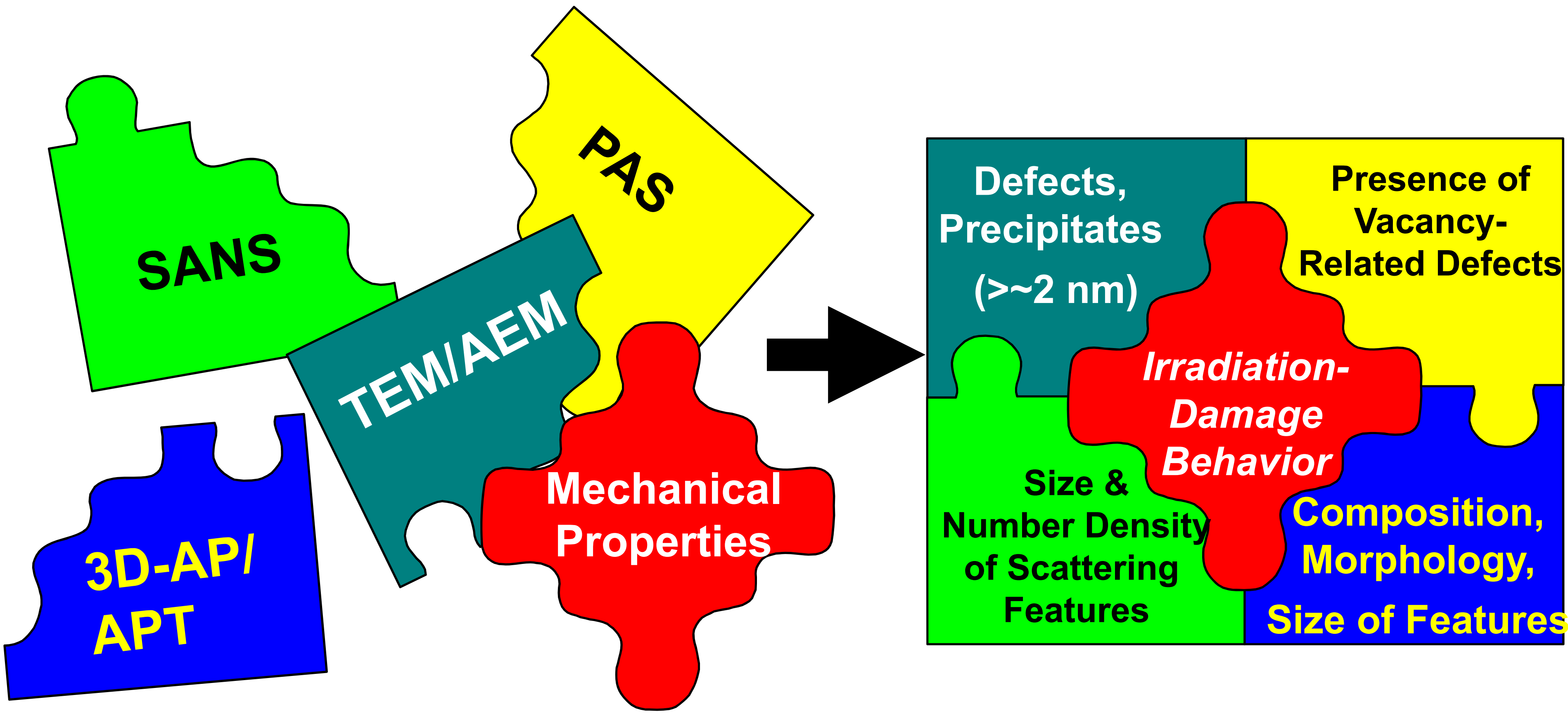
Irradiation Embrittlement of RPV Steels:
➔ Identifying the Physical Changes



A508 Gr4N Research into Irradiation Damage Mechanisms (1998-2003)

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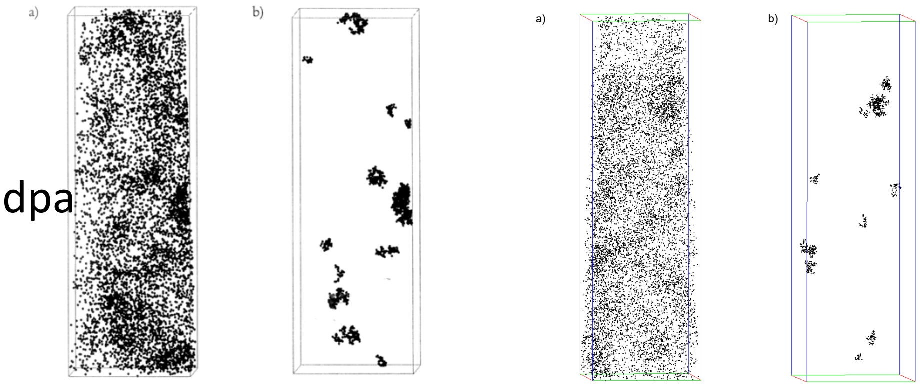
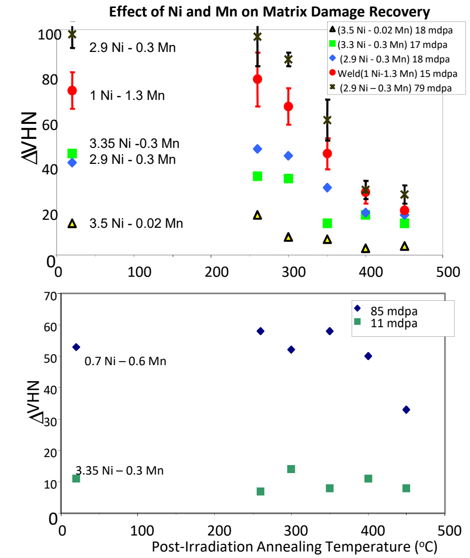
Atom probe data show solute clustering following hardening trend

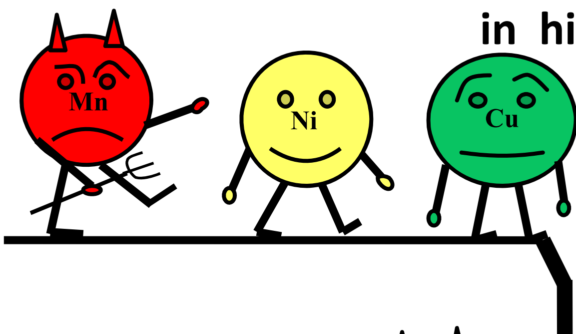
0.79 dpa

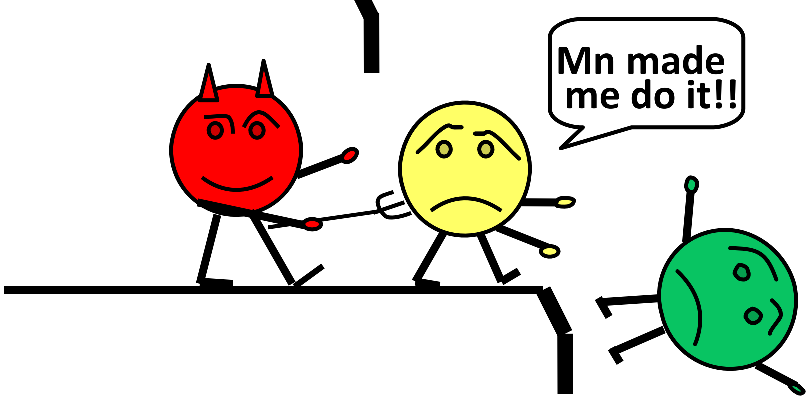


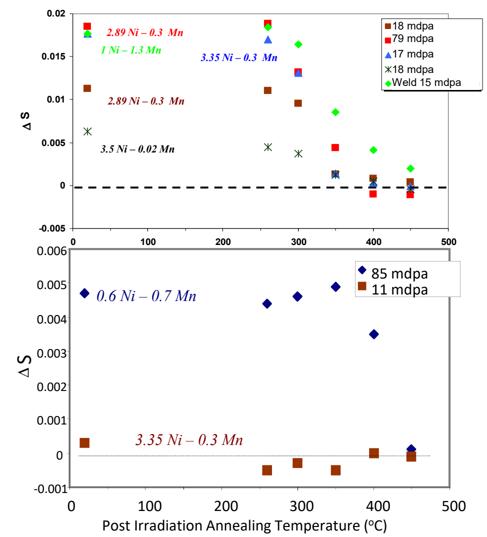
0.79 dpa + PIA (remove Vacancy damage)



Mn controls the "Ni Effect" in enhancing embrittlement in high Ni steels

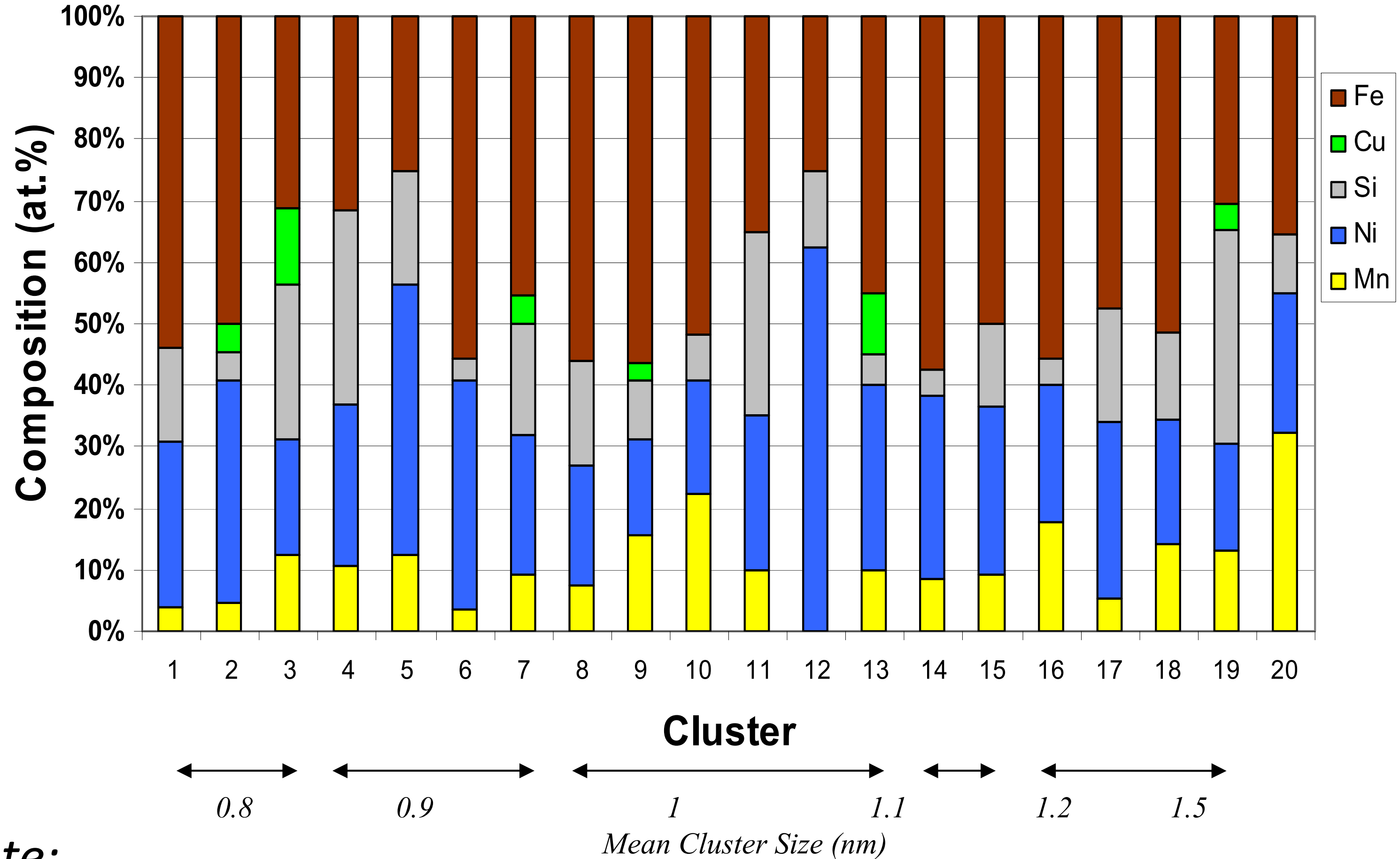






Mate

1 Ni – 1.3 Mn - 0.03 Cu Weld: 95 mpda
APT Solute Cluster Composition (at.%)



Note:

- All solute-enriched clusters contain Ni and Si, all but 1 contain Mn.
- Only ~30% of the clusters contain Cu
- No effect of size on composition

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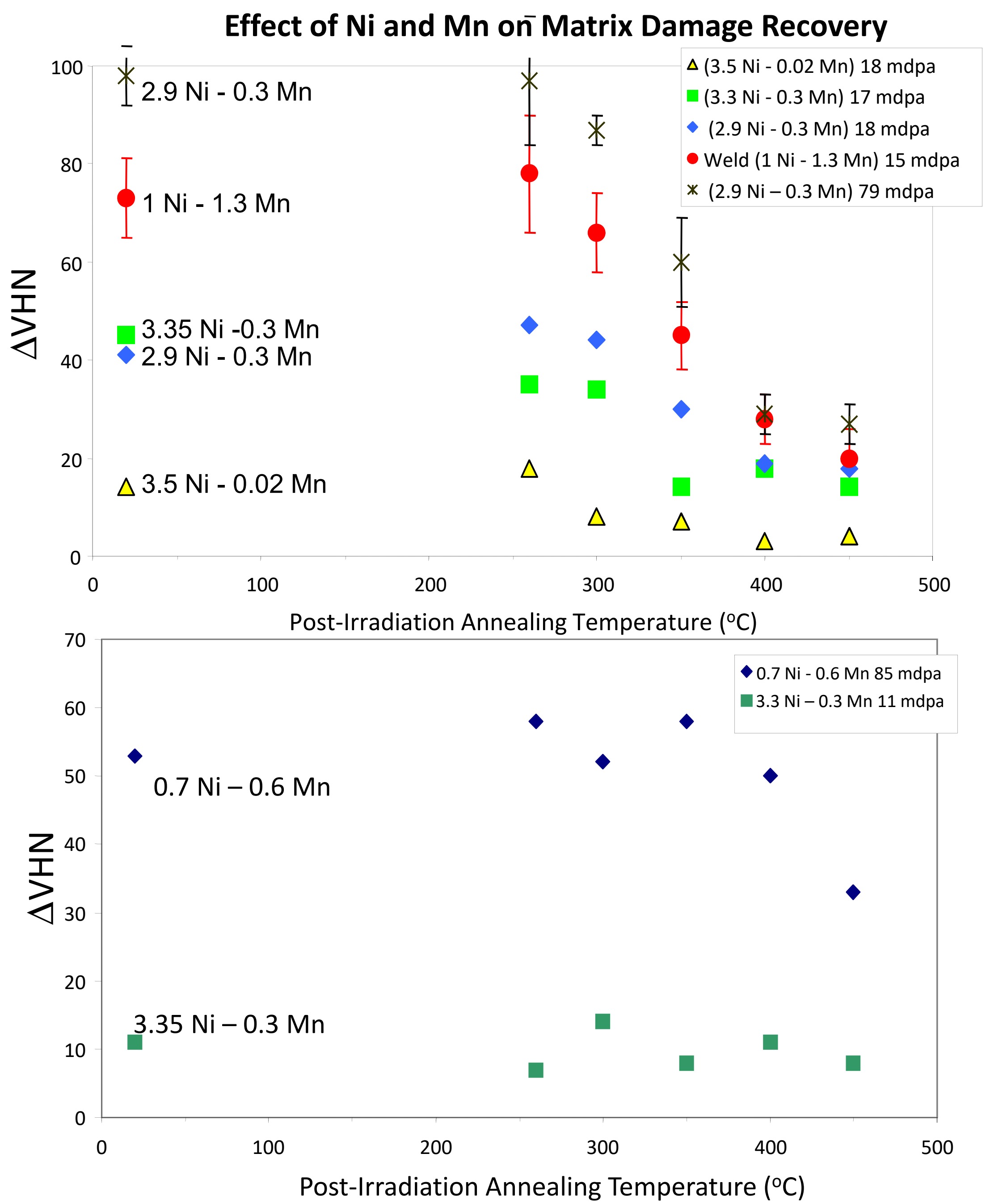
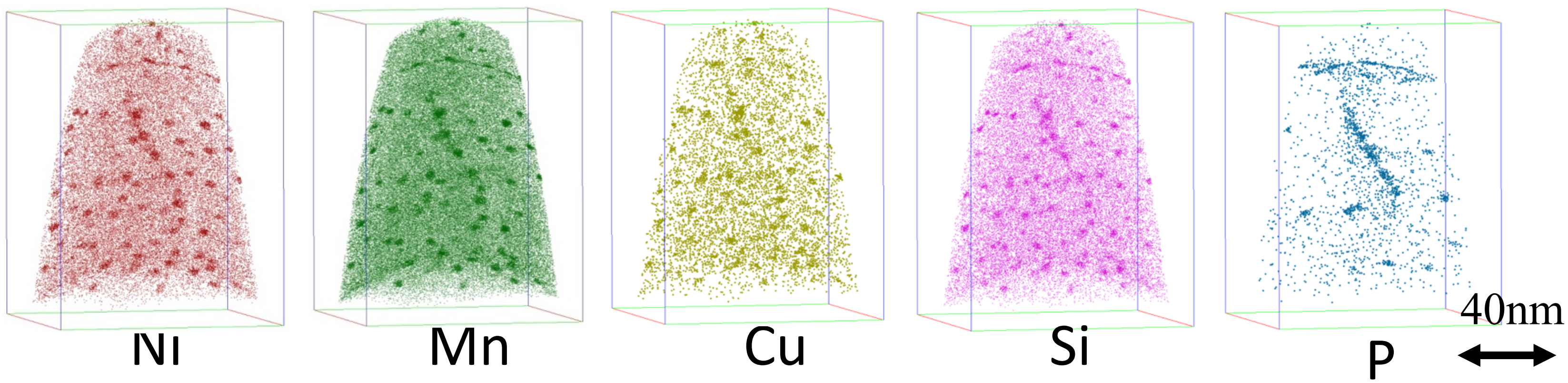
Comparison of High and Low Flux Irradiation Damage on ΔVHN

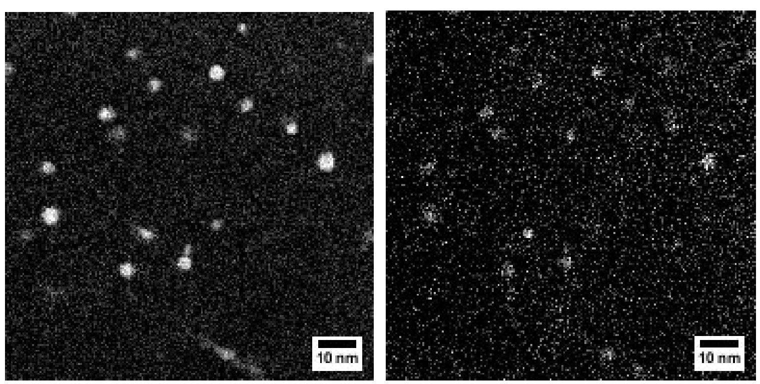
Note "less" irradiation-induced hardening damage in low flux 3.35 Ni – **0.3 Mn** sample and "less" hardening in 3.5 Ni - **0.02Mn** high flux sample



Ringhals 1.6 Ni – 1.4 Mn – 0.05 (and 0.08) Cu Welds Surveillance-Irradiated (0.073 dpa)

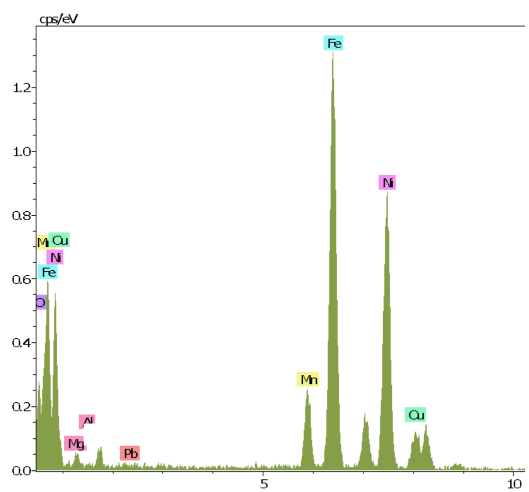
- 3-4 nm irradiation-induced Ni-Mn-Si-(Cu)-enriched clusters
- Solute-decorated dislocations

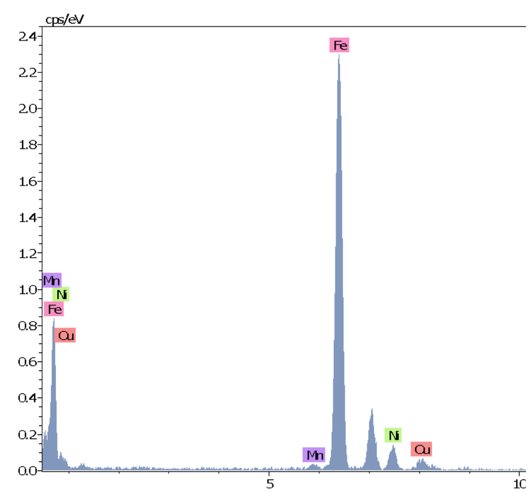




Mn L edge

Ni L edge

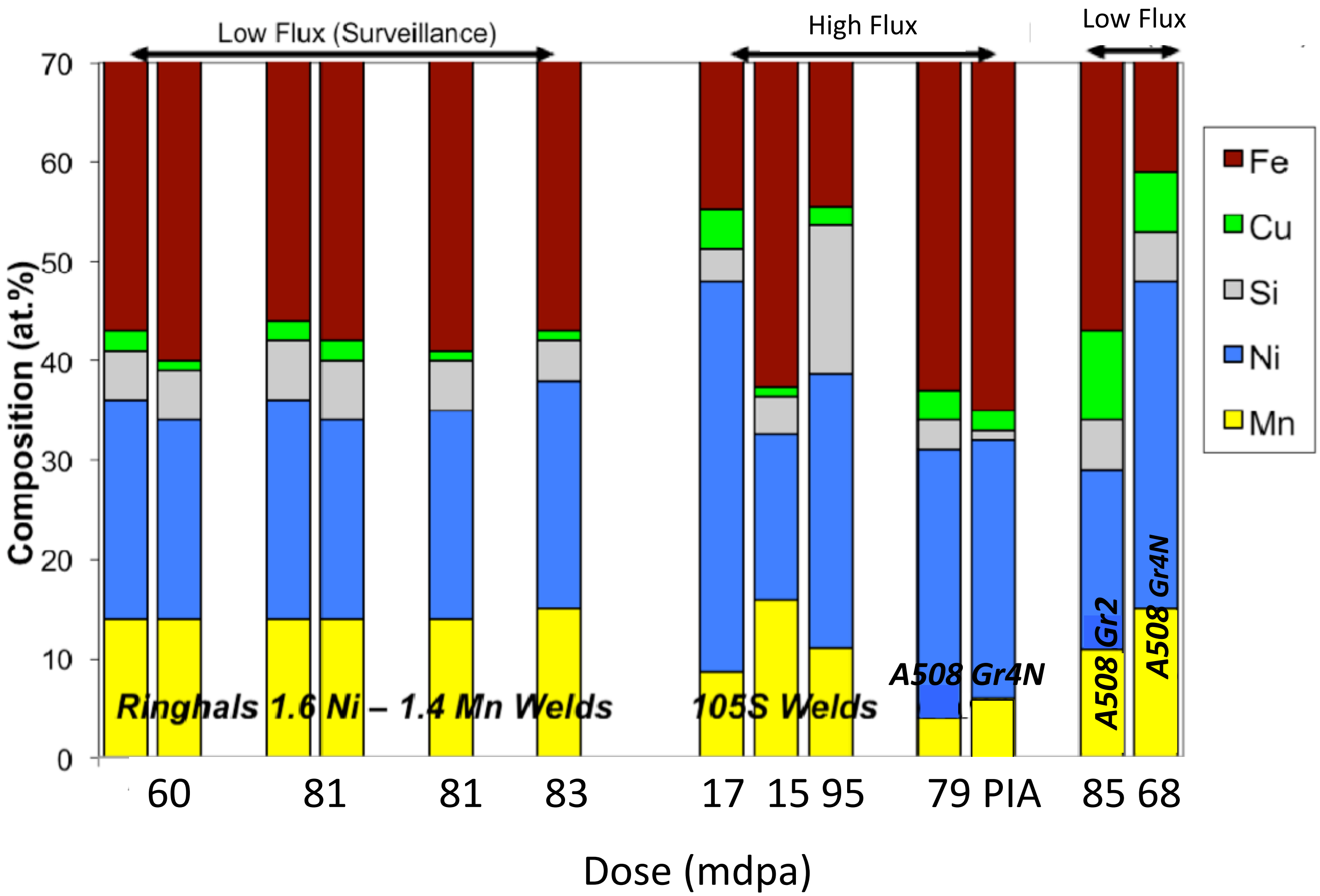




Summary of 3D-AP Data

High Ni Welds and Ni Steels (Low and High ϕ) show consistent solute clusters

Average Cluster composition (at.%)



Aberration-Corrected TEM
Irradiated Weld: 6.7 $X10^{19}$ n/cm^2

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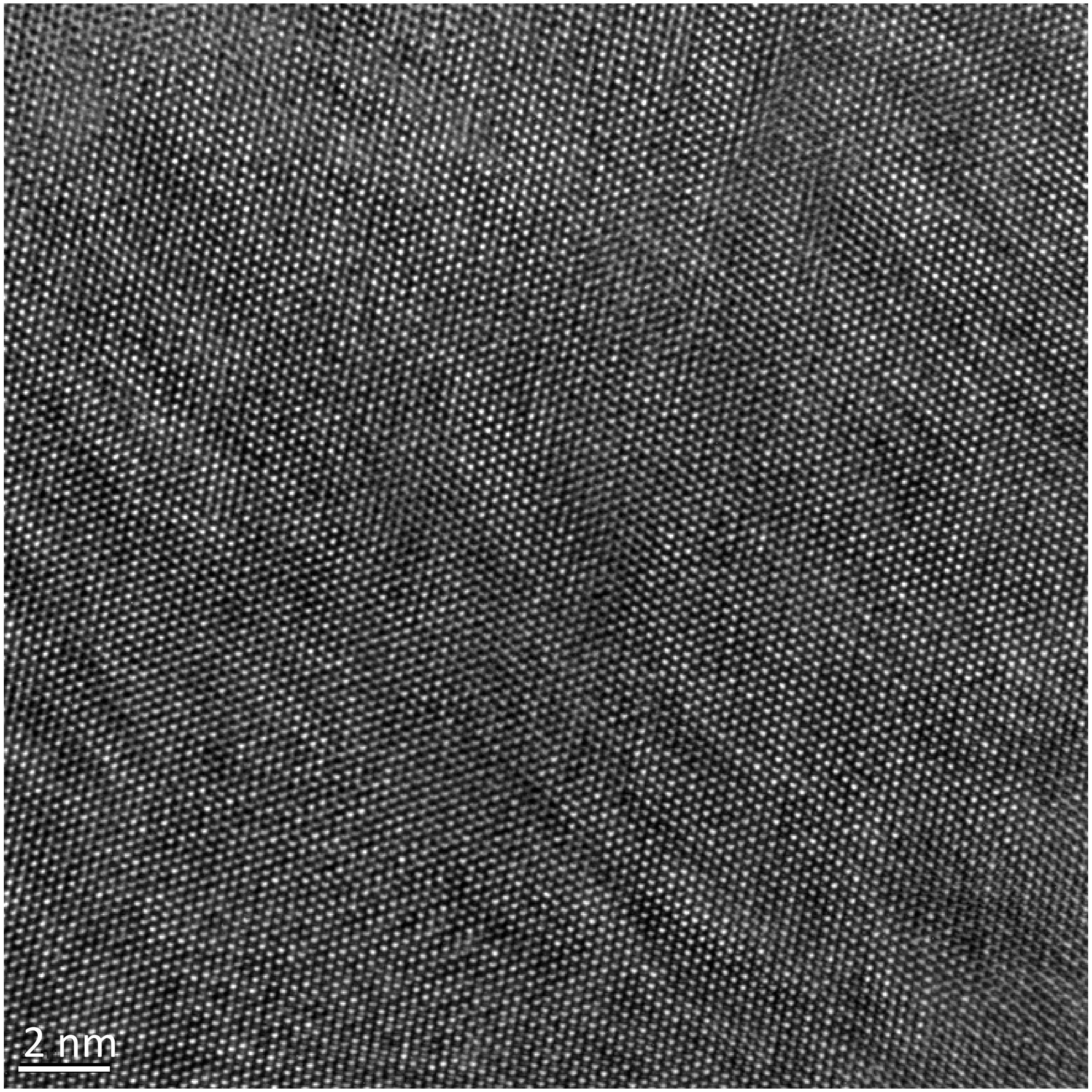


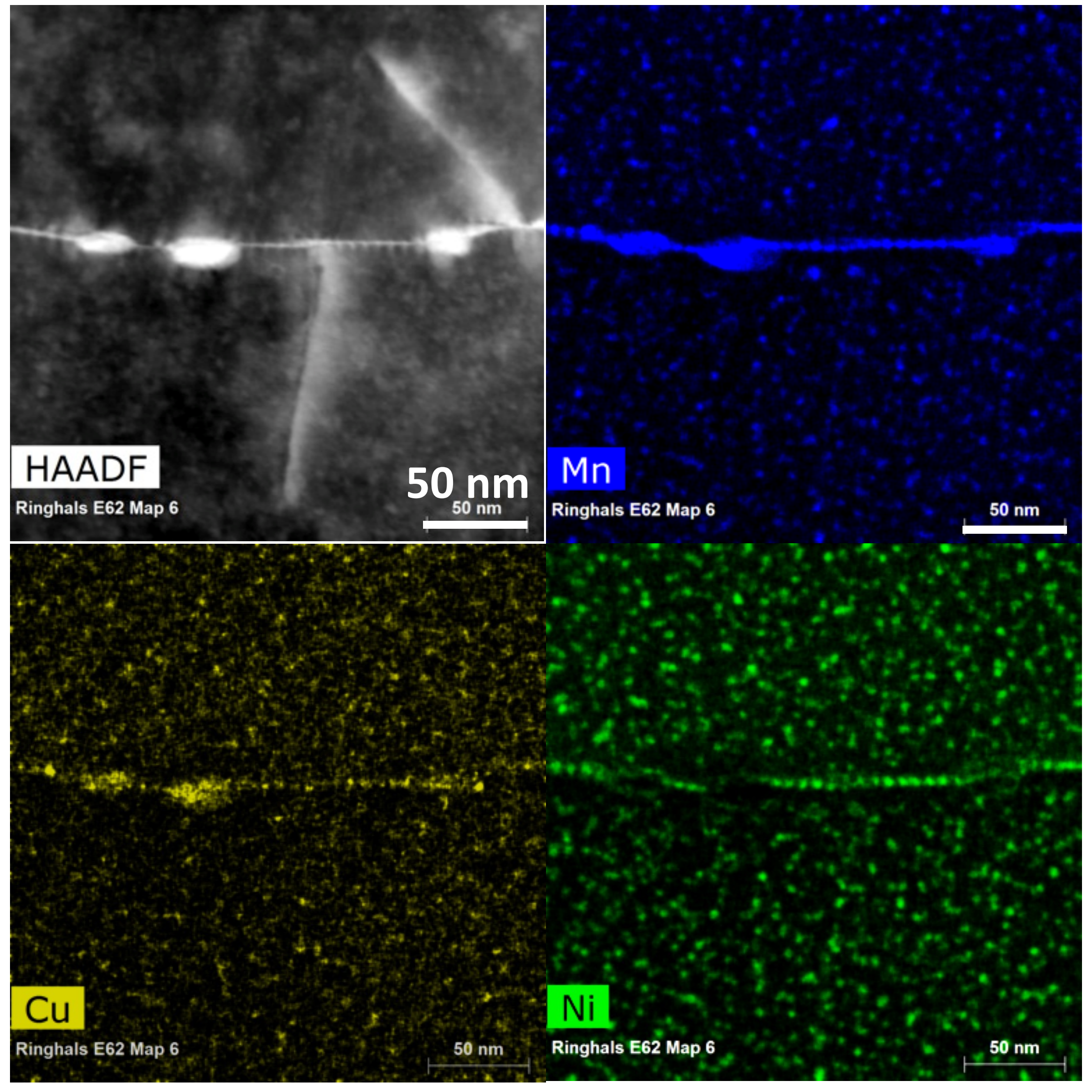
[111] high resolution TEM image

No clearly visible precipitates

"New" Technique: Advanced Analytical S/TEM

Ringhals 1.6 Ni – 1.4 Mn – 0.08 Cu Weld (0.073 dpa)

- 2-5 nm irradiation-induced Ni-Mn-Si-(Cu)-enriched clusters
- Cu-Mn ε precipitates
- Large areas of analysis
- STEM-EDX SI – all elements collected permitting subsequent reanalysis
- Excellent agreement with APT data



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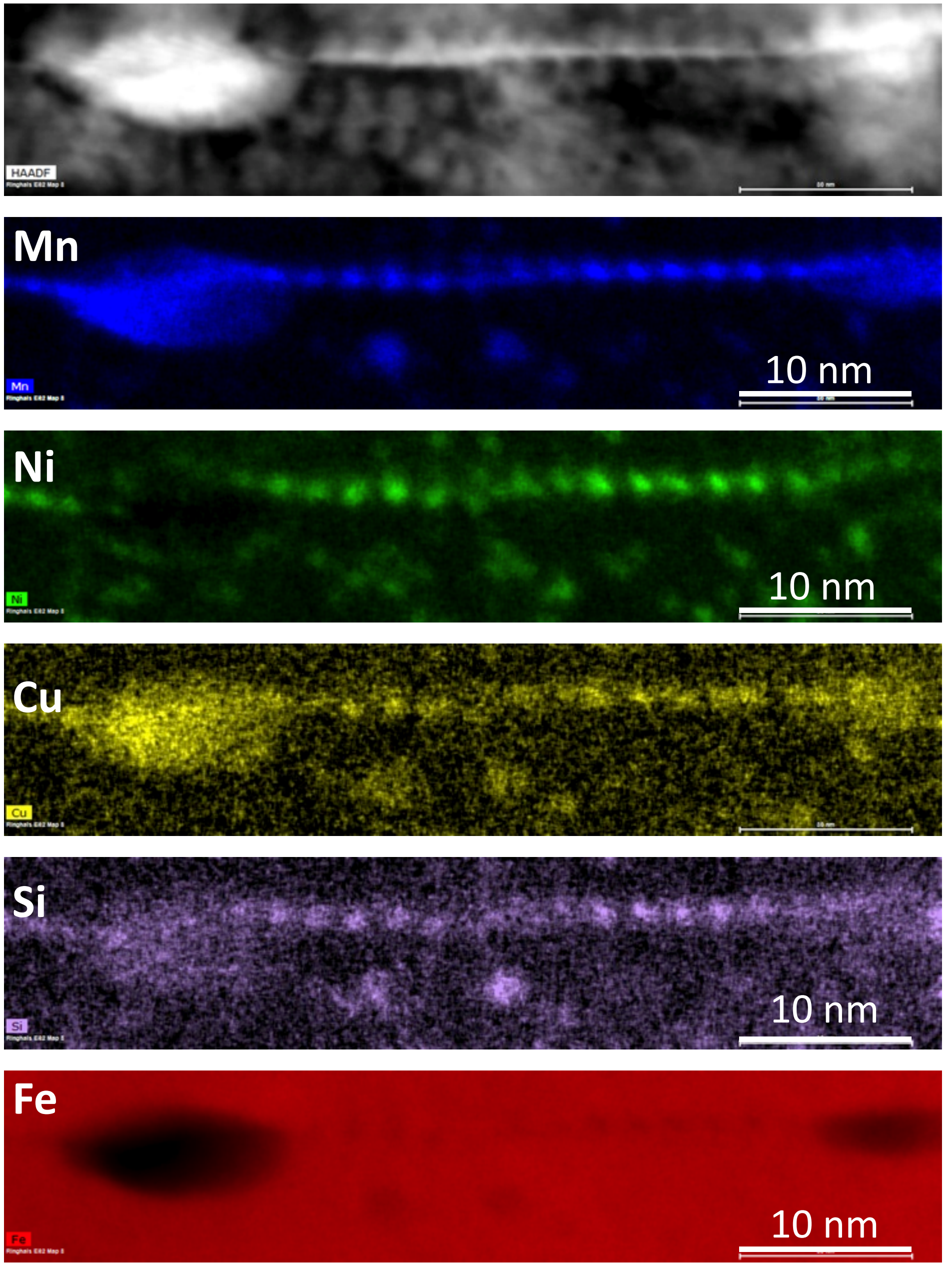
"New" Technique: Advanced Analytical S/TEM

Talos STEM-EDX Spectrum Image Data

Cu-Mn (epsilon Cu) – like precipitates consistent with previous studies on Cu-containing RPV welds

Note discrete solute-enriched clusters along boundary and within matrix



EDX elemental maps

RPV Steels and Welds

→ *Continuum of solute-related hardening features in contrast to single Cu precipitation term.*

From Irradiation Damage Mechanism studies of high Ni steels and welds

$$\Delta Hv_{irrad} = \Delta Hv_{vac(PALA)} + \Delta Hv_{sol(AP)}$$

Data consistent with Russell-Brown hardening (for measured solute-related hardening)

No need to invoke so-called "Late Blooming Phases"/ Ni-Mn precipitates/ phases/etc.

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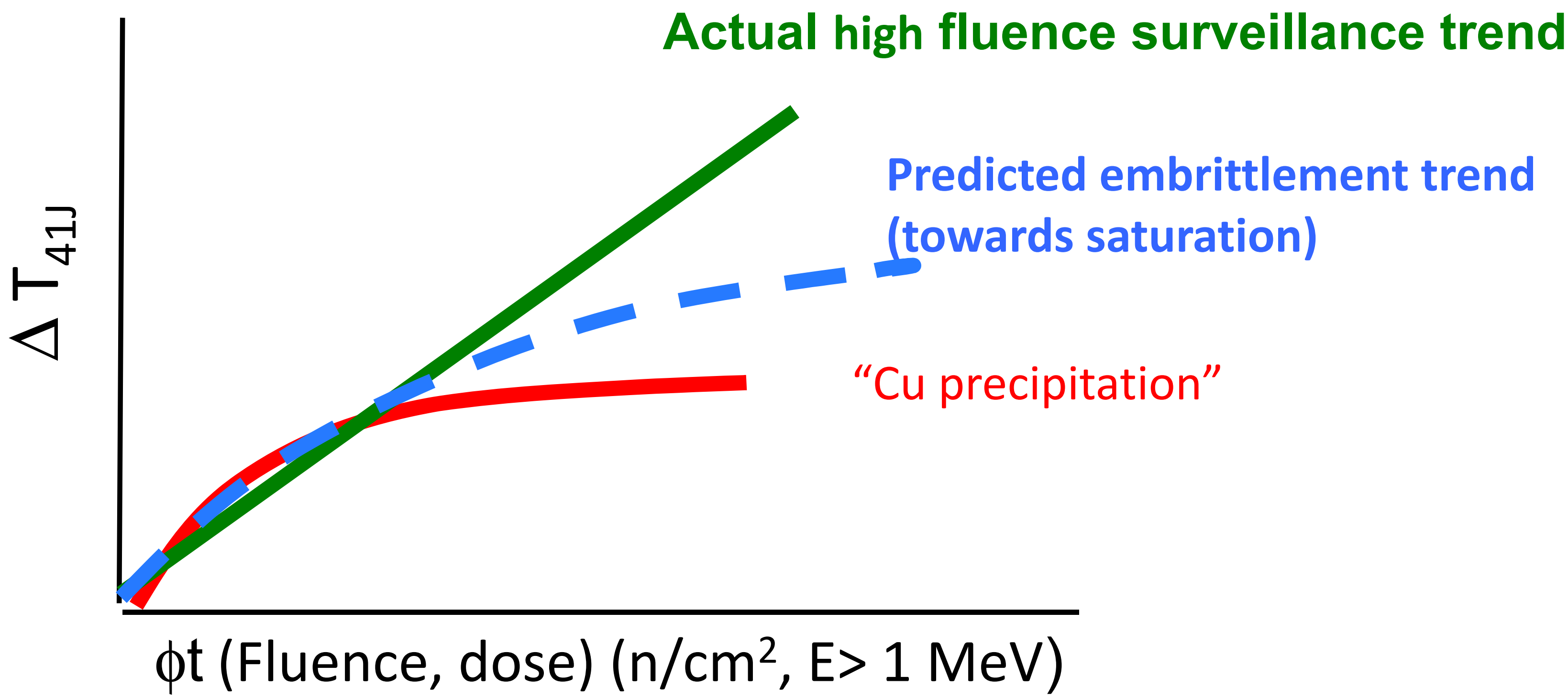
Irradiation embrittlement can be explained by the evolution of solute-enriched clusters…



- Presence of Mn and Ni (- Si) in clusters from inception – presence of Fe is not an artefact.
- No need to invoke precipitation and Cu exhaustion, i.e. LBPs, to explain high fluence trends

Neutron-irradiated (embrittled) steels/welds

↓

Microstructural Characterisation (APFIM/APT, SANS, etc.)
Solute "Features" containing Cu, Mn, Ni, Si, Fe..
No/very few visible defects (TEM)

↓

- Continuum of solute-enriched "features" that develop with increasing dose;
- Consistent with embrittlement trends and supported by thermal ageing and modeling studies

vs

Interpreted as CRPs;
Presence of Fe is artifact

+

Embrittlement continues to increase after Cu is depleted (pptn stops)

↓

New Feature to explain greater than predicted embrittlement trend → LBPs

So….. The appears to be a consistent picture concerning irradiation-induced "solute-related" hardening "features" based on numerous RPV and LA Steels/Welds …

- Can these observations be predicted from atomistic modelling?
- Can atomistic modelling help to interpret our microstructural / microchemical observations – particularly with respect to solute-enriched hardening features and vacancy-related hardening?
- EU coordinated programme activities in PERFORM 60, SOTERIA and national programmes

EU PERFORM60 & LONGLIFE Activities

- Successfully developed interatomic potentials necessary to address the complex Cu-Mn-Ni-Si-Fe interactions (*Becquart, Malerba and colleagues*)
- Demonstrated vacancy-Mn (and other significant solute interactions) binding; independent confirmation of experimentally-measured irradiation damage behaviour of production high Ni steels with/without Mn (A508 Gr4N)
- Demonstrated experimentally and via modeling the formation of irradiation-induced defects (effect of alloying on TEM imaging/ visibility) from binary/ternary model alloys to RPV steel. (*Mayoral, Malerba*)
- Demonstrated that very long-term ageing at 365°C can produce CuMnNiSiFe “precipitates” (thus lending support for the irradiation-induced solute-enriched clusters formed due to neutron irradiation *(Styman et al.)*

On-going Research

- Issue: Degradation of materials in reactor environments….
  - Changes in material properties as a function of:
    - * neutron irradiation
    - * environment (non-neutron contribution)
- Identification of physical changes in material associated with degradation in materials performance…
  - Detailed characterisation of the microstructure to identify those changes that are associated with changes in material properties/performance
- Development of models to predict material performance
- Development of improved materials/processing/heat-treatments and component design for existing and new reactors

UK National Nuclear Users Facilities (NNUF)

UK Government investment in facilities for nuclear R&D

NNL Central Laboratory (Sellafield) *(high activity work)*
UKAEA Materials Research Facility (Culham)* *(lower activity)*
Culham Centre for Fusion Energy (Culham).
Dalton Cumbrian Facility for Proton and Ion Irradiation
University of Huddersfield MIAMI-1 and MIAMI-2 TEMs for *in situ* ion irradiations (Microscope and Ion Accelerator for Materials Investigations)

Linked with Nuclear Advanced Manufacturing Research Centre (U Sheffield) and the new Henry Royce Institute for Advanced Materials (active laboratories for Graphite and Fuels at the University of Manchester)

Lack of MTR facilities….

Can proton irradiation be used to produce similar damage forms in low alloy steels

Potential for Research Applications?

Inexpensive

Fast

Materials can be readily handled (no hot cell required)

X Protons are not neutrons!

X Limited penetration depth (~15-~40 microns)

UK Dalton Cumbrian Facility for Ion/proton irradiation

Variables: dose and T_{irrad}

Ongoing Research into High Toughness A508 Gr4N Forging Steels

- High Ni (~3.3-3.7 wt%) and low Mn (**0.02** to 0.3 wt%)
- Martensitic-Lower Bainitic Steels – As-cooled + Tempered
- Outstanding toughness
- Irradiation Damage Behavior?
  - Comparable to lower Ni steels – No "enhanced embrittlement"
  - Extensive multi-technique research program (1998-2003) (APT, TEM, HRTEM, SANS, PALA, PIA) demonstrated the **critical role of Mn** in the development of stable, neutron-irradiation-induced hardening
- Can we generate similar hardening and structures using proton irradiation?

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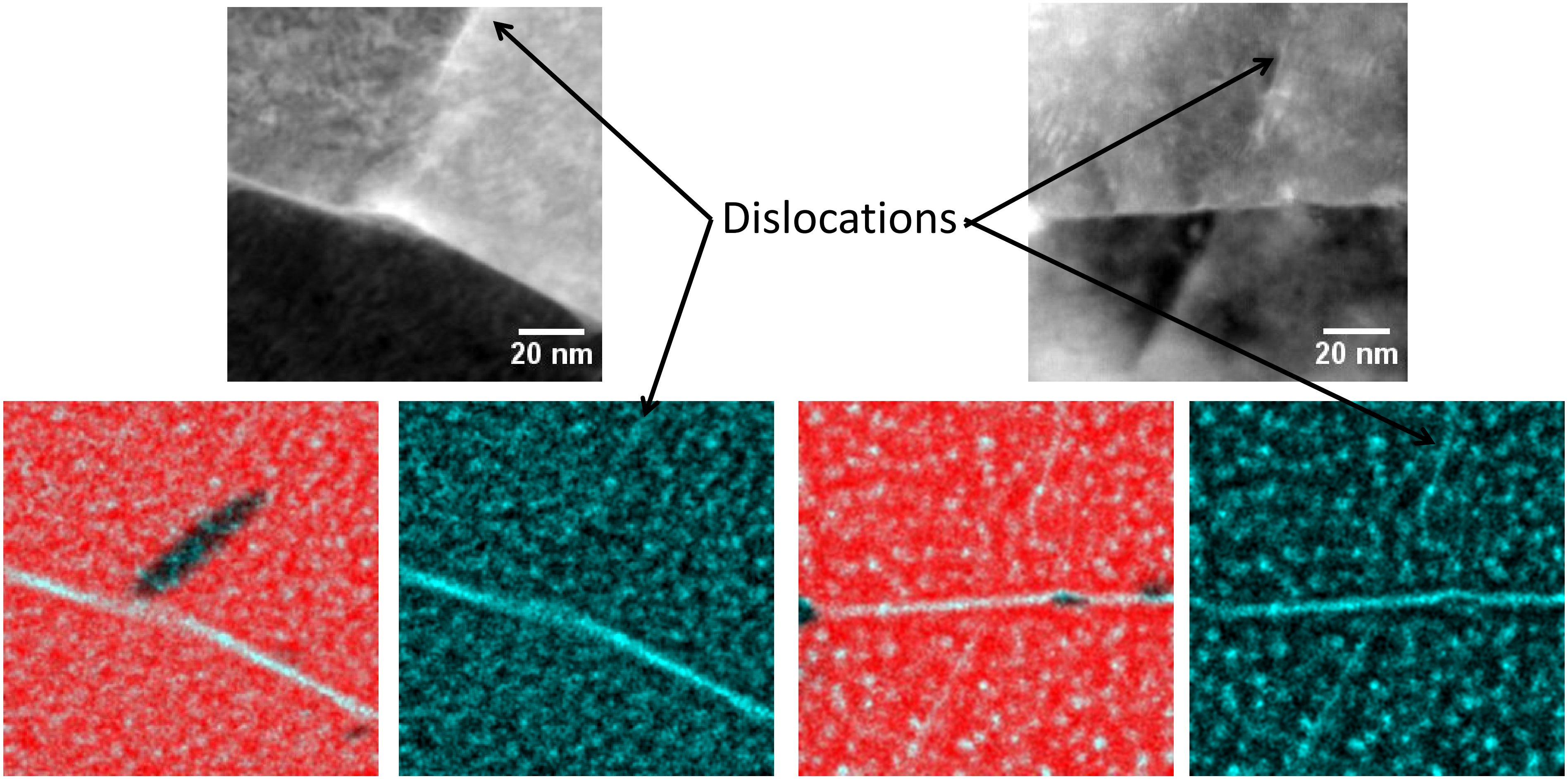
A508 Gr4N Steel
Neutrons vs. Protons (~225°C)

Neutron Irradiation 0.092 dpa | **Proton Irradiation 0.4 dpa**



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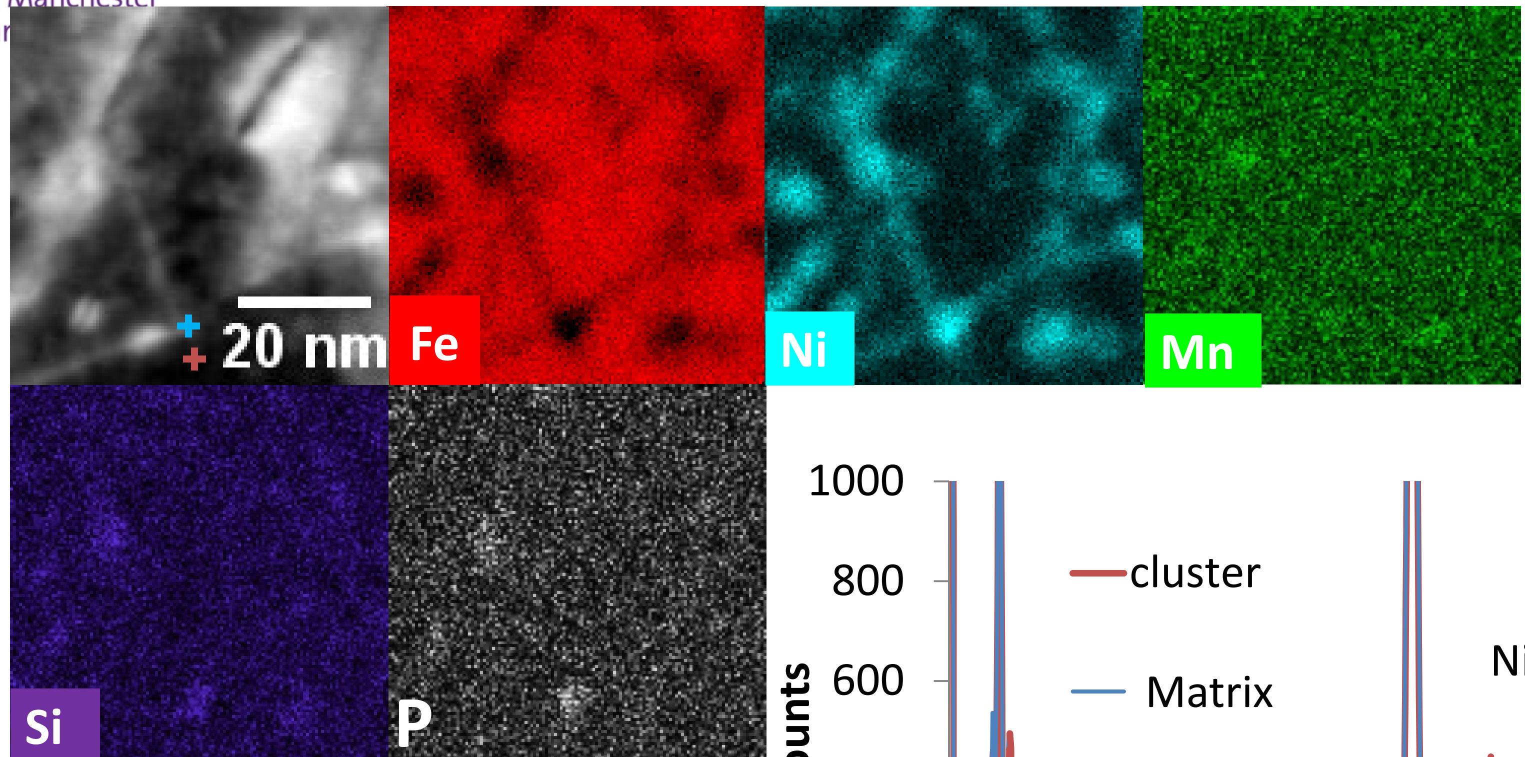
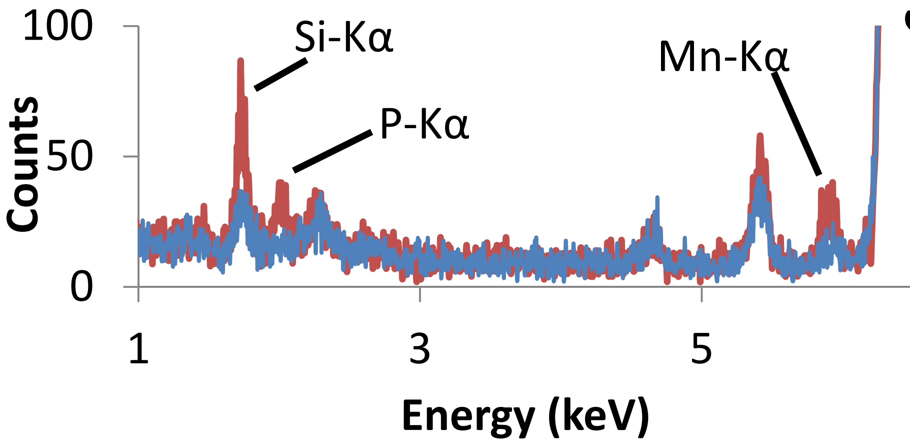
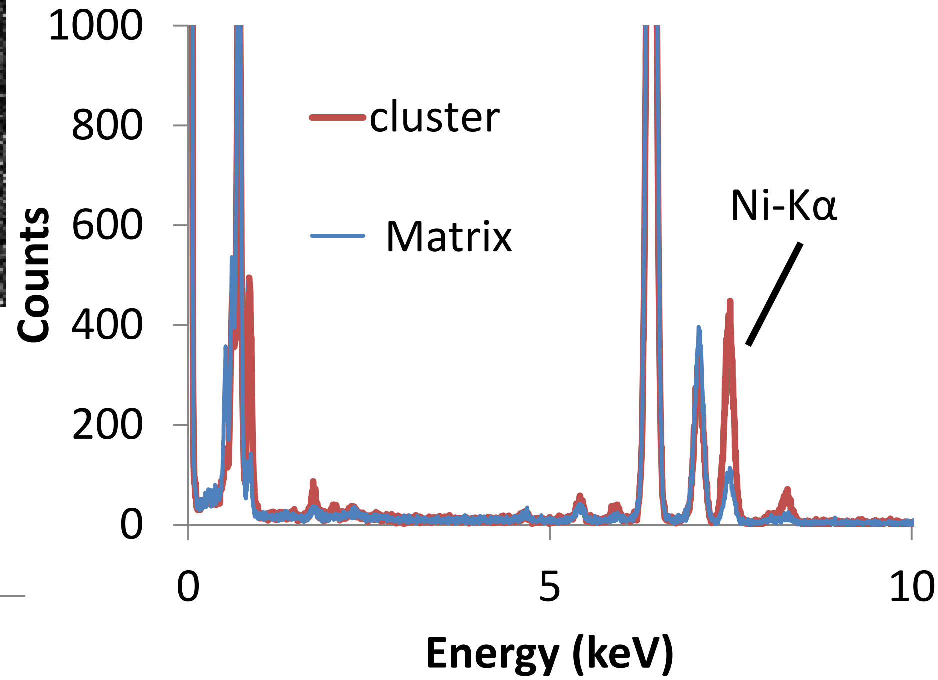
Proton Irradiation at 320 ºC: 1.1 dpa







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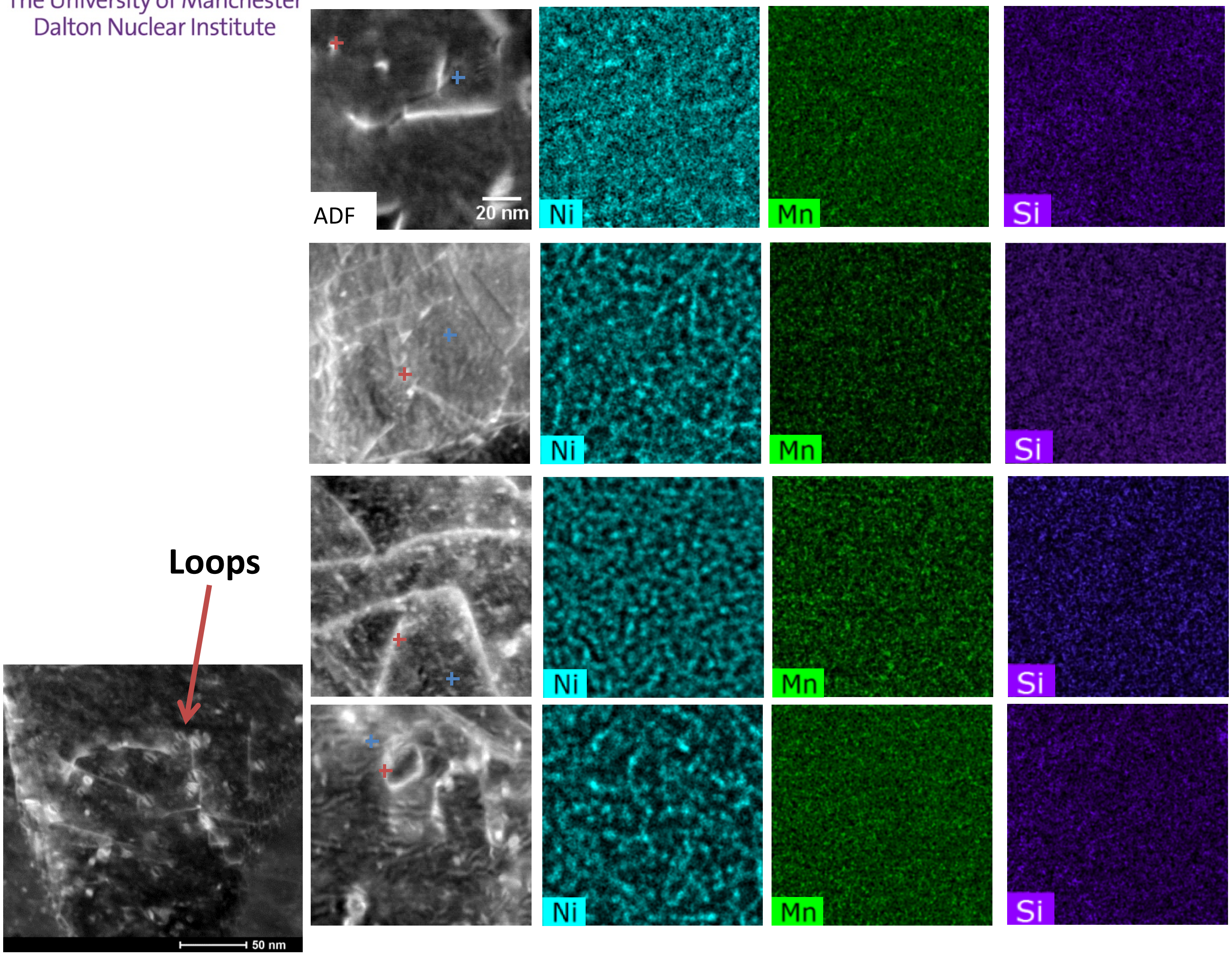
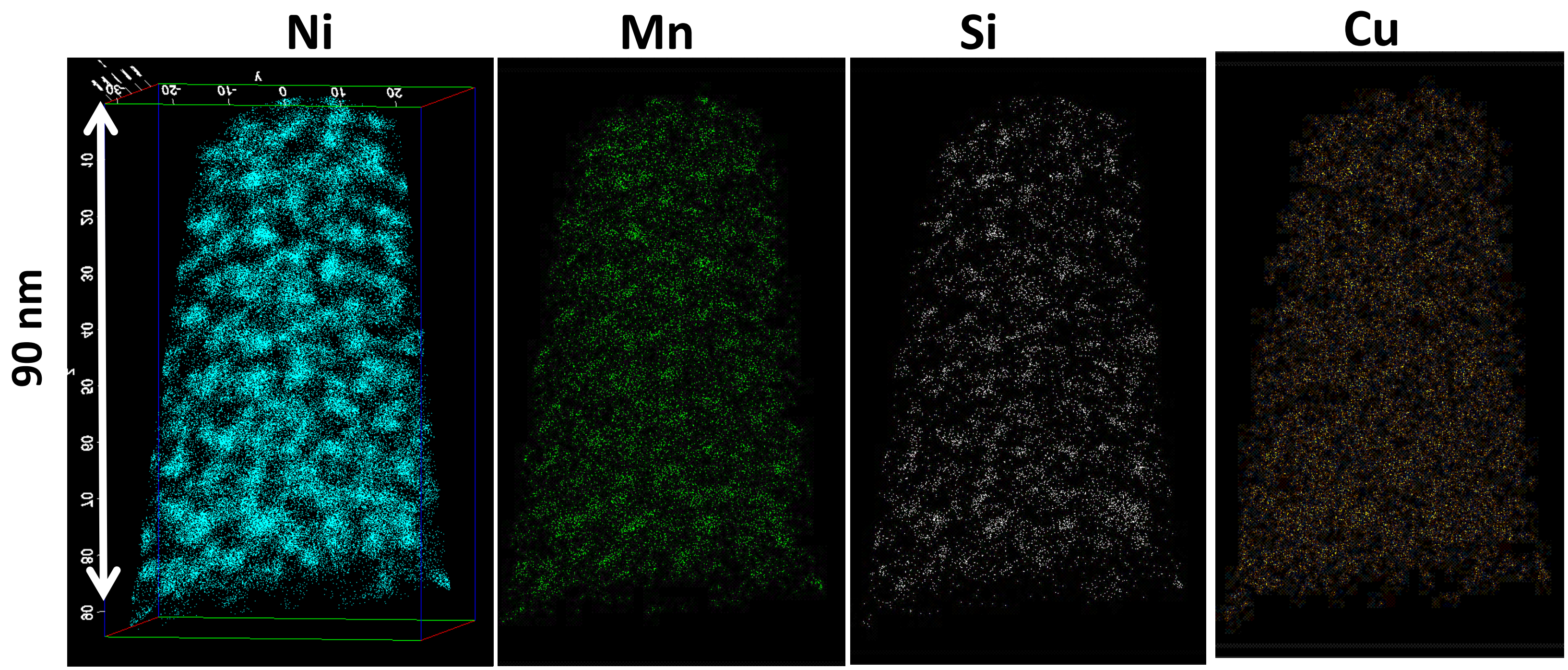
Proton Irradiation: Effect of Dose and T_{irrad}



T (°C)	dpa
225	0.09
225	0.4
225	1.1
320	1.1

But – how do Advanced ATEM STEM-EDX SI data compare with APT data?

0.4 dpa – 225°C Proton-Irradiated A508 Gr4N APT Results



Collaboration with James Douglas/Michael Moody (Oxford)

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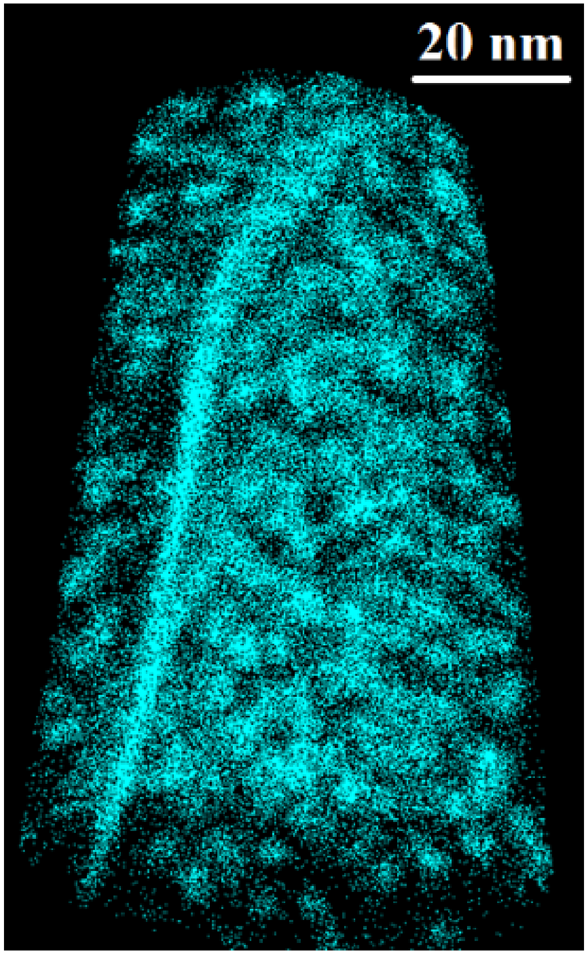
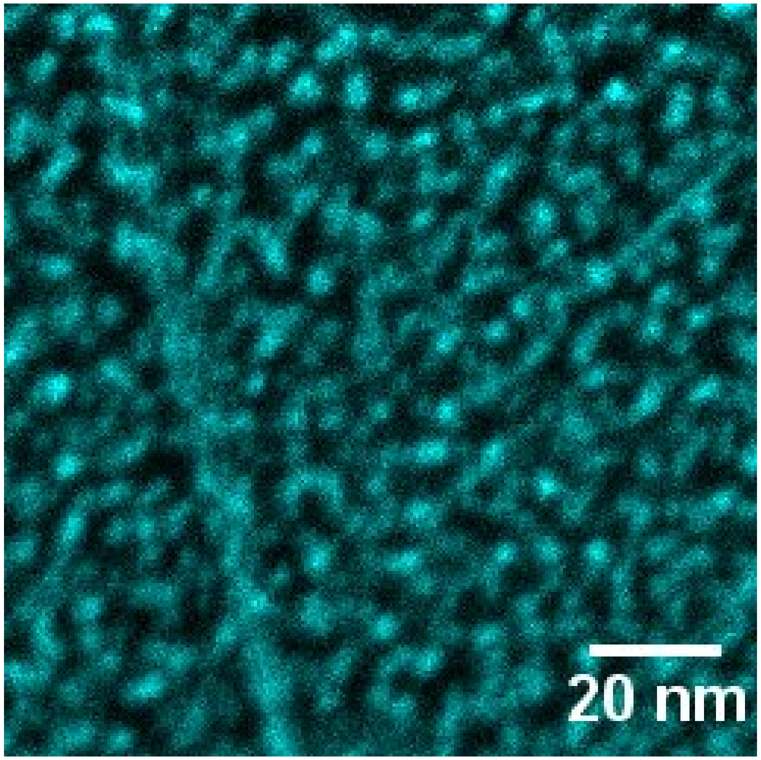
Solute Clustering (Ni) in A508 Gr4N Steel Induced by Proton Irradiation
Comparison of APT and Talos STEM-EDX SI Data

0.4 dpa at 225° C





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Thermal Contribution to Neutron-Irradiated RPV Steels/Welds? (Current Research - Vattenfall)

- Understanding role of fluence (dose) and flux (dose rate) on microstructural development/evolution of solute-enriched hardening "features" (clusters)
- Thermal contribution?
- Pressurizer: Higher temperature (~345^{o}C) than RPV (~300-310^{o}C) for 28 years
- **No increased hardening in base steel**....increase in hardness for weld and increase in ΔT_{41J} ***(less than neutron-irradiated)***

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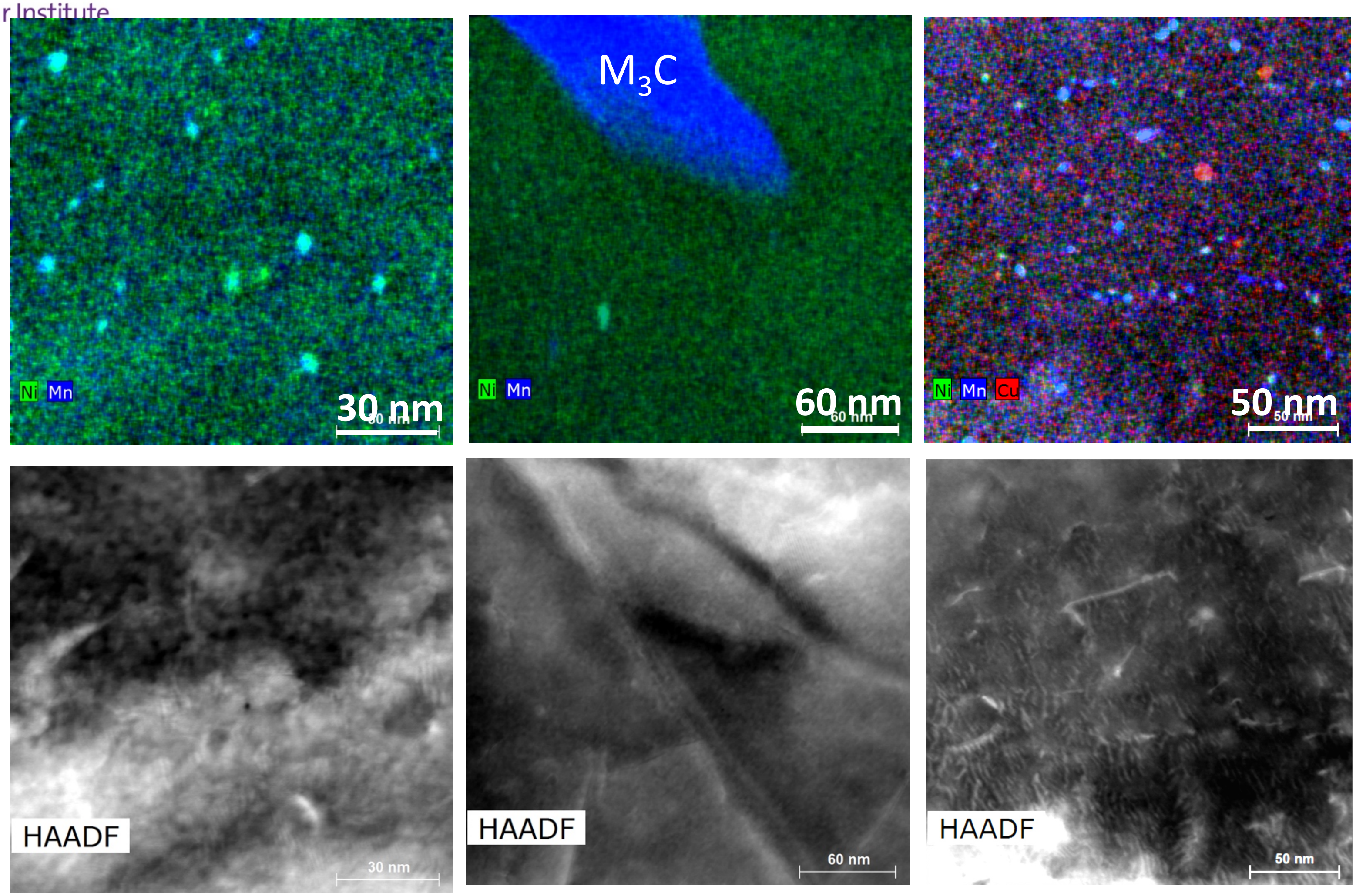
Pressurizer Weld – ~28 years at 345° C
FEG-STEM EDX SI Datasets



- **Non-uniform** nanoscale precipitation enriched in (Ni-Mn-(Cu))
- Preferential sites: GBs, dislocations

Lim, Burke, Efsing, To be published

Status

Multi-Scale Data Enables Fundamental Understanding and Mechanistically-based Predictive Models

- Explains Hardening/Mech Properties
- Key "Features" Identified: Size/Composition – Vacancy-related Defects ("Open Volume Defects")
- ***Assessing Variability – Importance of "Baseline" Properties*** (Microstructural Variability Documented)
- **Data Analyses must be SELF-CONSISTENT and CONSISTENT** with independent Complementary Analyses (Multi-Technique approach)
- **More Microstructure data needed for model development**

Route Forward? Coordinated/Collaborative Programs

Future Needs

- **Extracting data from real components to assess performance:**

Contributions of thermal ageing (pressurizer weld/pressurizer – note that T is ~40 C-deg higher than operation, but same/similar materials. Note : No shift for ex-service plate but weld showed hardening and a measurable shift. (As T decreases, thermal contribution is decreased).

- **SMR Development/Materials Validation:** Need for lower T data (K and ΔT_{41J})

How to generate?

Existing support structures?

Low flux and low T_{irrad}. Need both mech property data and analysis of material (determine physical changes in microstructure) for input to predictive model(s).

- Benefit of IGRDM (originally started with US NRC guidance)

Need for collaborative, jointly-sponsored international research programs.

Acknowledgements

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UK National Nuclear Laboratory

ROLL-ROYCE

VATTENFALL

Horizon 2020

Many colleagues in the US and UK (MRF, MPC, Oxford University)

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