Development of Low Temperature Spray Process for Manufacturing Fuel Cladding and Surface Modification of Reactor Components

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Project Team

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- Oak Ridge National Laboratory (Dr. David Hoelzer)
- Los Alamos National Laboratory (Dr. Stuart Maloy)
- Oxford University, UK (Dr. Patrick Grant, unpaid international collaborator)

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Presentation Outline

- Introduction, Motivation, and Project Overview
- Brief Review of Cold Spray Deposition Process
- Development of Cold Spray Process for Oxide Dispersion Strengthened (ODS) Steel
- Cold Spray Manufacturing of ODS Fuel Cladding Tubes

Introduction, Motivation, and Project Overview

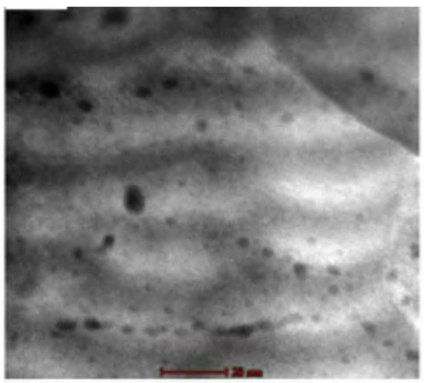
Broad Objectives of the Project

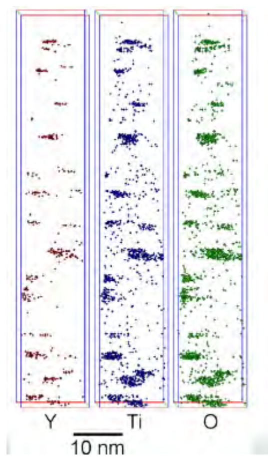
Develop low temperature, solid-state (*no melting involved*) powder spray deposition process (cold spray process) as a:

- Rapid, near-net shape manufacturing of oxide dispersion strengthened (ODS) steel cladding tubes
- High deposition rate coating technology for corrosion and wear protection, and repair of nuclear reactor components

Oxide Dispersion Strengthened (ODS) Steels

- ODS steels are ferritic (BCC) and contain fine dispersion of nanometer-sized oxide particles (Y-Ti-O) [0.2 to 0.3 wt.%]





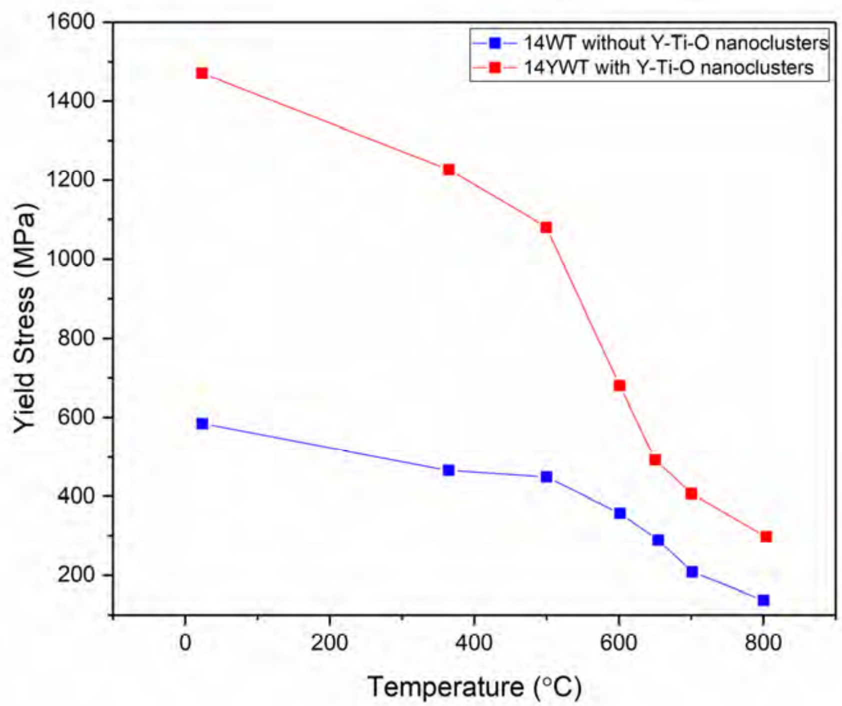
TEM image showing nano-scale oxide particles in ferritic matrix of ODS steel [1, left] and atomic probe tomography (APT) image of nanoparticles [2]

- Has the low radiation-induced swelling of conventional ferritic steels
- High temperature strength superior to conventional ferritic steels
- **Regarded as a cross-cutting material for multiple reactor concepts**

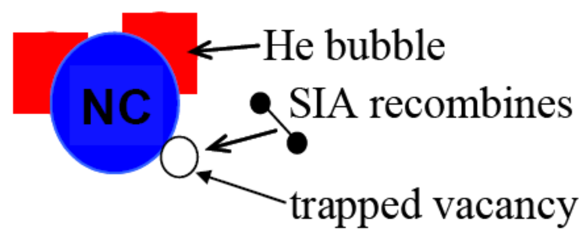
[1] Sridharan et al., UW-Madison
[2] courtesy Dr. Hoelzer, ORNL

Role of Oxide Nanoparticle (nanoclusters) in Ferritic Steels

Increase in High Temperature Strength



Enhanced Radiation Damage Tolerance

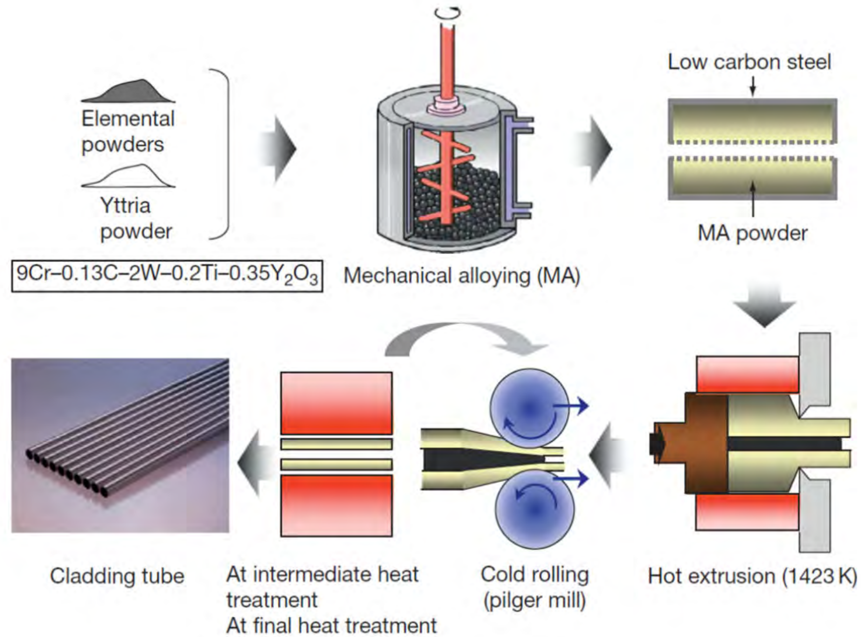


NC: Oxide Nanoclusters

"Influence of Particle Dispersions on the high Temperature Strength of Ferritic Allloys", D. Hoelzer, *Journal of Nuclear Materials*, 2007.

Conventional Manufacturing of ODS Cladding Tubes – Slow and Expensive Process

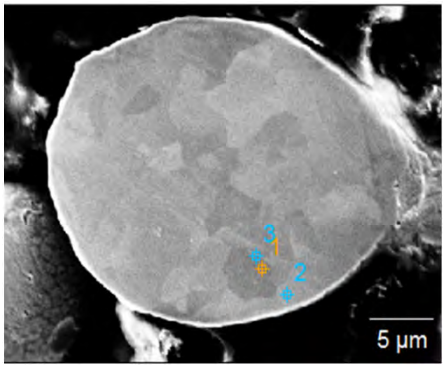
- Melting processes cannot be used as they lead to upward stratification of oxide nanoparticles
- Milled powders are canned and degassed at 400°C and subjected to multiple hot/ warm extrusion steps (8 -10 steps) at temperatures > 1000°C and lower temps.
- Low strain rate extrusion processes not conducive to large-scale manufacturing
- May lead to grain anisotropy, and anisotropy in mechanical properties

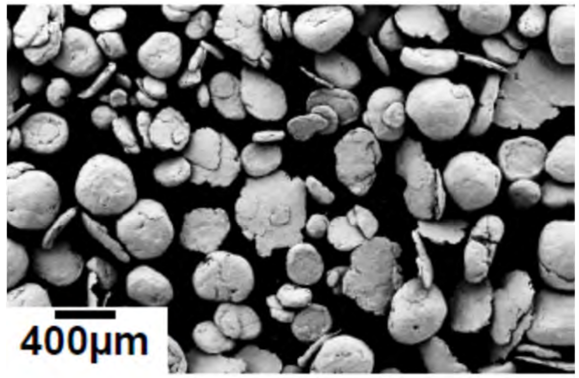


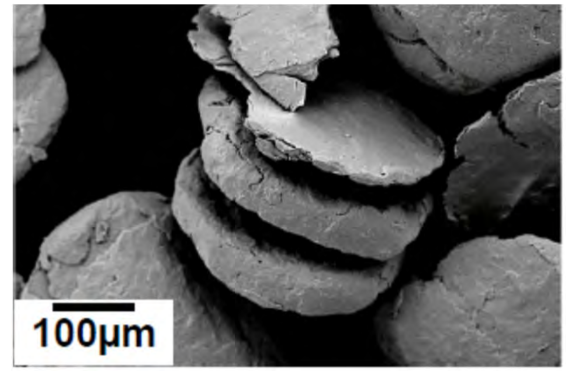
Conventional fabrication of ODS steel tubes requires multiple extrusion steps [3]

[3] "Recent Developments in Irradiation-Resistant Steels", G.R. Odette, et al, *Annu. Rev. Mater Res.*, 2008, 28, p. 47.

Powders for ODS for Conventional Manufacturing of Cladding





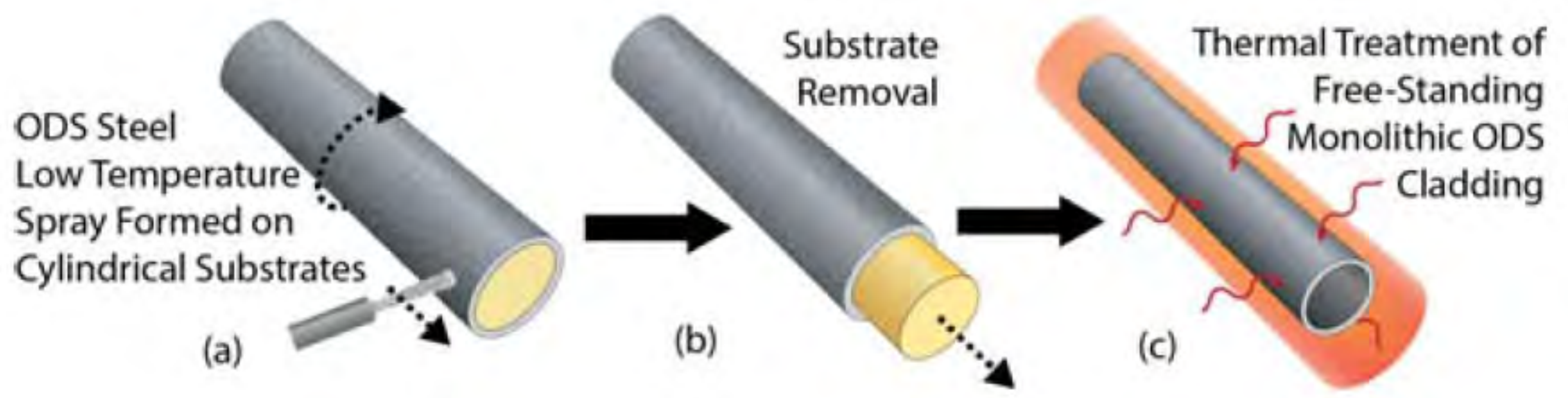


ODS powders *after* ball milling (powder from Dr. Maloy, LANL)

- Gas atomize ferritic steel powders with Y, Ti, O
- Atomization does not fully solutionize Y, Ti, and O therefore ball milling is done with some added FeO to achieve full solutionizing (mechanical alloying)
- During high temperature consolidation treatments, oxide nanoclusters come out of solution

Concept of Manufacturing ODS tube via Cold Spray Process

- **Three major steps for cold spray manufactured ODS cladding tube**

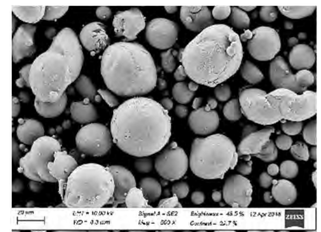
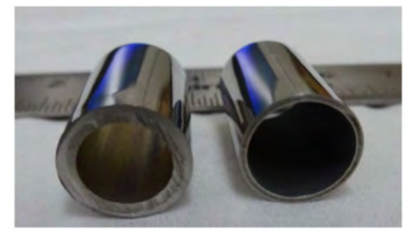
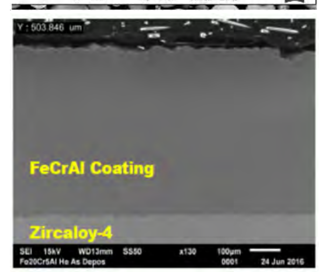


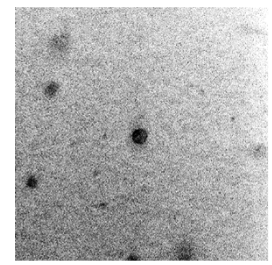
Potential Benefits:

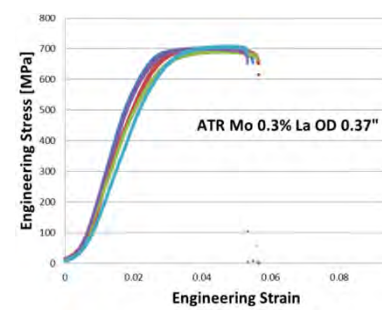
- **Eliminates multiple extrusion steps**
- **May eliminate ball milling mechanical alloying**
- **Fabrication process faster and cheaper**

Key Objectives and Milestones

1. **Optimization** of the powder spray process for the manufacture of ODS cladding tubes
2. Post-deposition **thermo-mechanical treatments**
3. **Characterization and testing** of ODS steel cladding tubes produced by cold spray process
4. **Surface modification and coatings** by cold spray process for addressing corrosion and wear in reactor components, (i) single material coatings, (ii) compositionally-graded coatings, and (iii) multi-layered coatings.
5. **Bench-marking** and **alternative** novel approaches

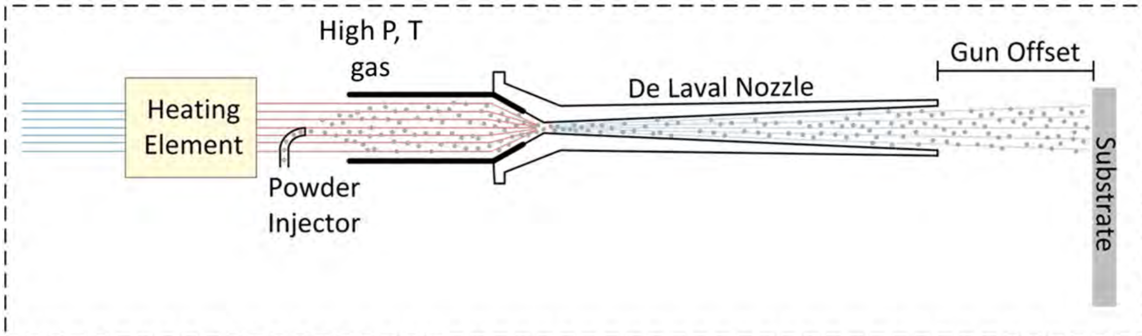


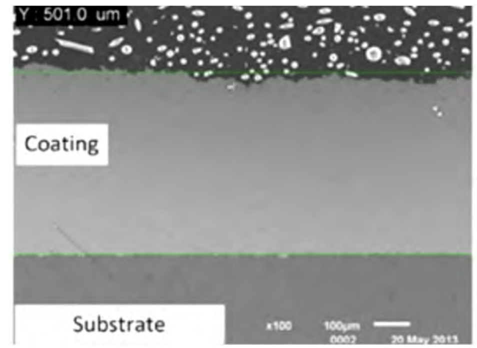


Brief Overview of Cold Spray Deposition Process

Cold Spray Process

[Courtesy UW-Madison]





Zn cold spray coating on steel substrate

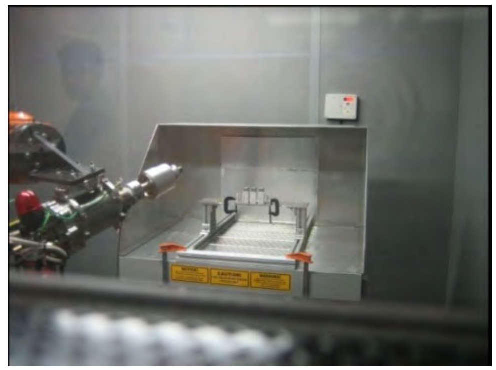
- **Powder particles of the coating material propelled at supersonic velocities by a gas onto the surface of a part to form a coating or deposit**
- **Particle temperature is low – particles are not melted and deposition occurs in solid state**
- **Coating/deposit formation occurs by particle deformation and an associated adiabatic shear mechanism**

Cold Spray Process – Attributes as a Manufacturing Process

- Performed at ambient temperature
- Performed at atmospheric pressure
- High deposition rates – fast manufacturing
- Supports factory and field fabrication
- High technology readiness level

Cold Spray Laboratory at University of Wisconsin, Madison (est. 2012)

Robot for pre-programmed movement of spray gun



Sample stage and dust collector (below that)

Sound-proof spray booth

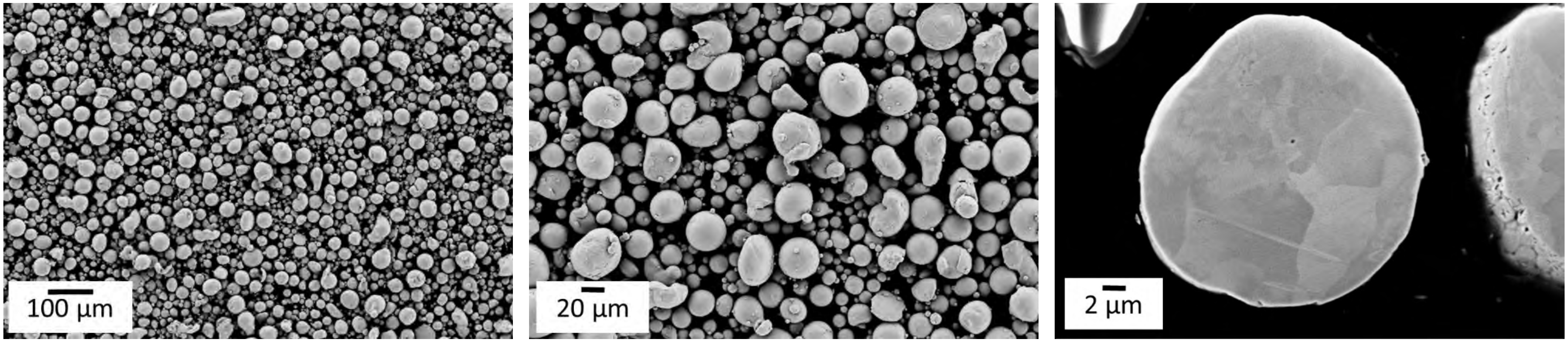
- **4000-34 KINETIK System, from ASB Industries/CGT-GmBH**
- **Spray booth from Noise Barriers**
- **Robot controlled (Nachi system, from Antennen)**

Nitrogen/helium gas cylinders

Robot controls (left) and spray gun control (right)

Development of Cold Spray Process for Manufacture of ODS Steel

Feedstock Powder for Cold Spray Process

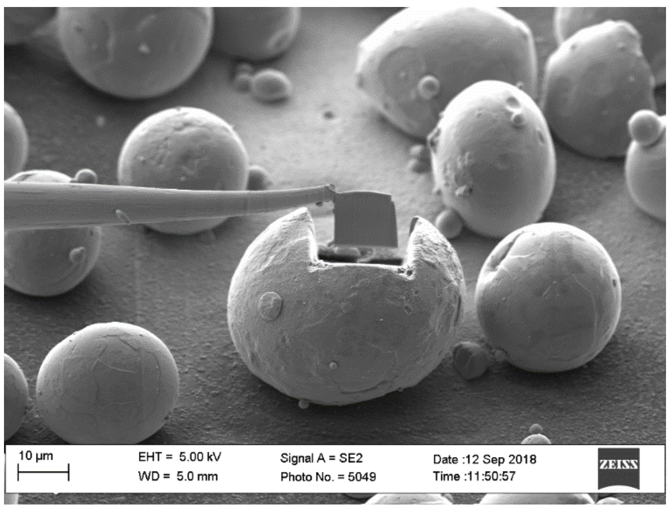


- Received from Oak Ridge National Laboratory
  - 14YWT (Fe-14Cr-3W-0.4Ti-0.2Y-0.25O)
  - Gas-atomized, spherical powder
  - Size less than 44 µm
  - Large grain size (4 to 8 µm)

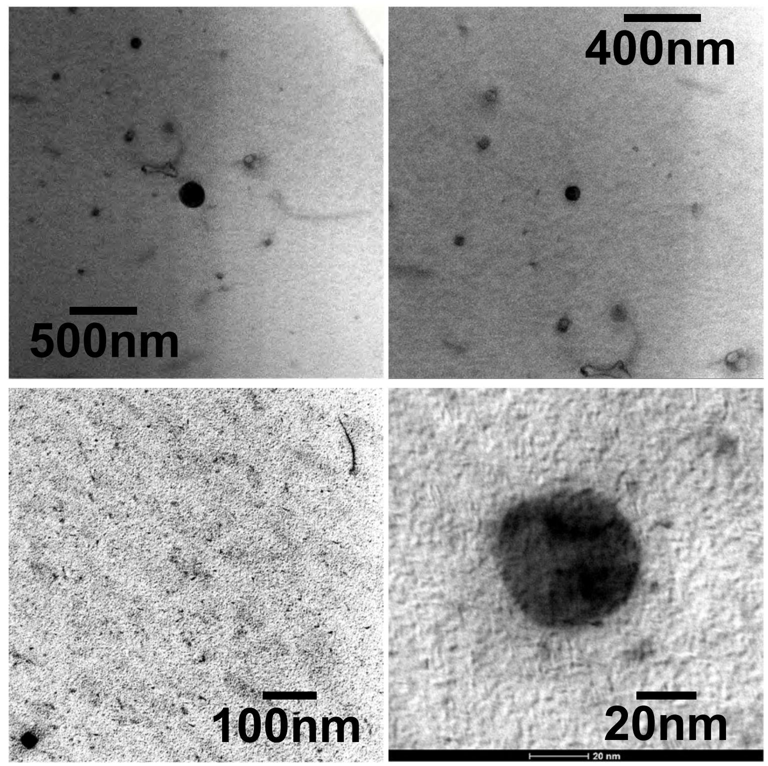
Nanoparticles in Feedstock Powder

- **Y and Ti -rich nanoparticles in size of 10 nm to 200 nm are dispersed in the ferritic steel matrix**

Transmission Electron Microscopy (TEM) lamellae prepared from a particle



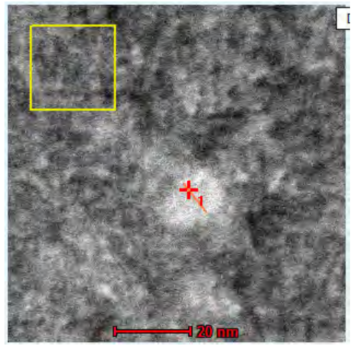
TEM images showing fine particles in the matrix

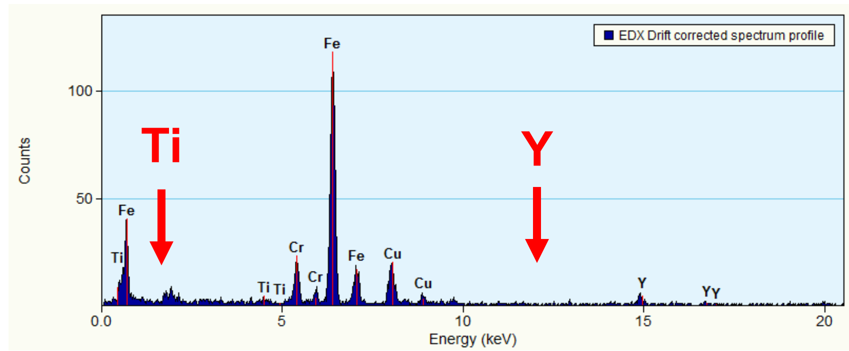


Y and Ti Detection in the Fine Nanoparticles in 14YWT Powder

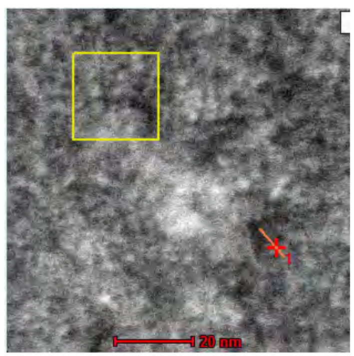
HAADF Image | **EDS spectra**

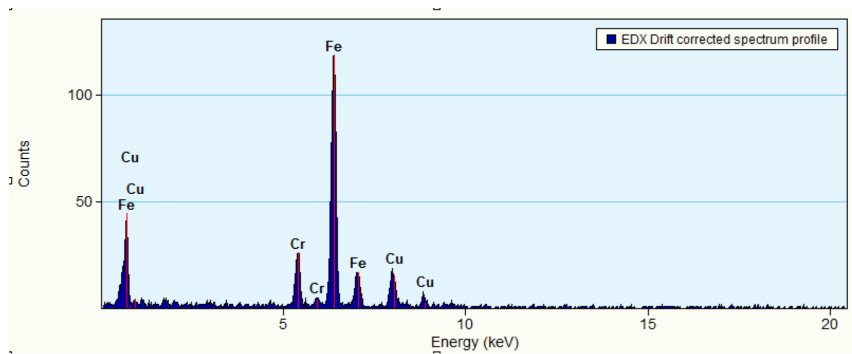
Inside nanoparticle





Outside nanoparticle



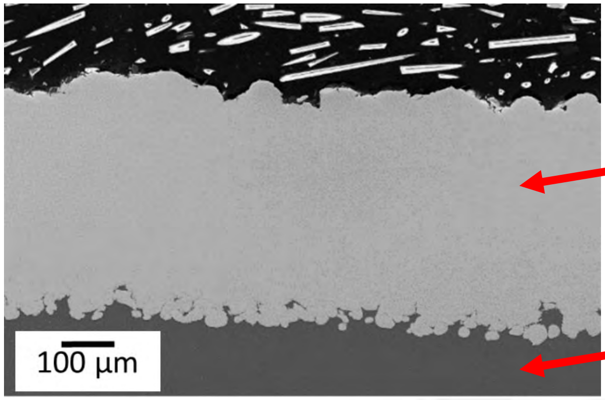


Atomic %	**Fe**	**Cr**	**O**	**W**	**Ti**	**Y**
Inside particle	73.4	11.5	12.0	0.5	too low	**2.2**
Outside particle	72.5	15.1	11.9	0.5	-	-

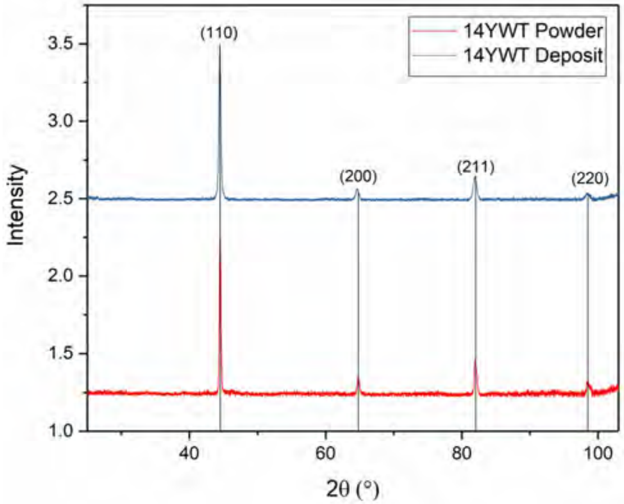
Thick and Dense ODS Cold Spray Deposit on Flat Substrates

Before spray

After spray



- 14YWT powder was successfully deposited onto 6061-T6 aluminum flats
- Deposits were very dense with negligible porosity
- XRD confirmed no phase change or oxide inclusions of powder during deposition



XRD peaks for the powder and the deposit

Initial Parametric Investigation of Cold Spray Process for Optimal Parameter

Three different gas conditions were investigated

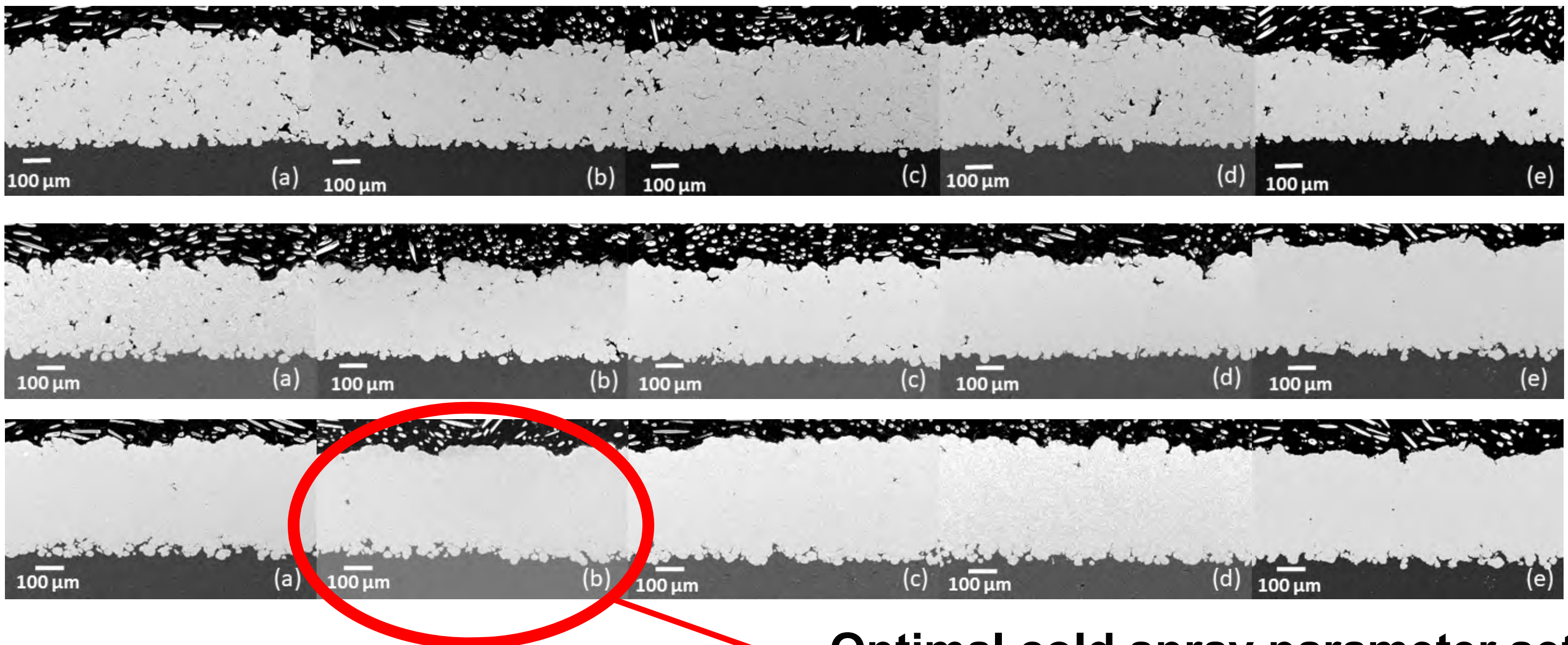
- 100% nitrogen
- Helium/nitrogen mixture A
- Helium/nitrogen mixture B (more helium gas)

Five substrates were investigated

- Annealed 6061
- 6061-T6
- 7075-T6
- Two different cold rolled 6061-T6 substrates

Parametric Investigation of Cold Spray Process (Propellant Gas and Substrate Effect)

	Annealed 6061	6061-T6	40% CR 6061-T6	80% CR 6061-T6	7075-T6
100% N_2					
Mixture A					
Mixture B					



Hardness range of substrates from 60HV – 170HV

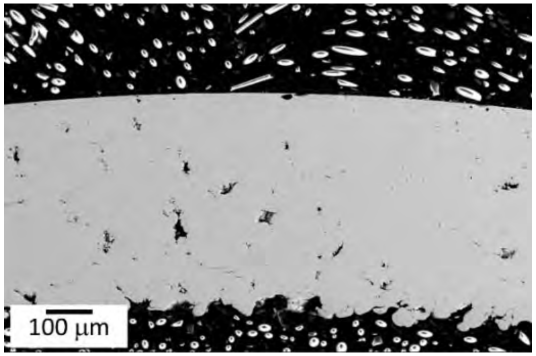
- No significant affect seen from cold spray process in terms of deposition thickness

Optimal cold spray parameter set with 6061-T6 substrate because it is readily available and can be easily dissolved

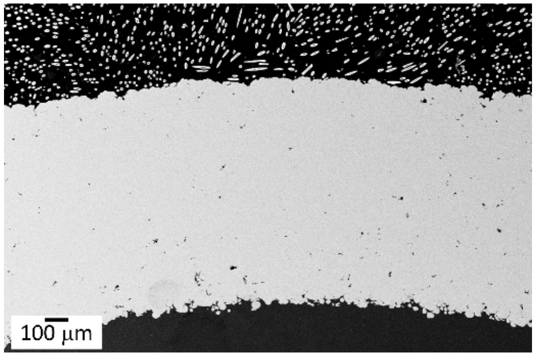
- Showed the least amount of porosity

Further Parametric Investigation of Cold Spray Process (powder size effect)

ODS tube with 25-44 µm powder

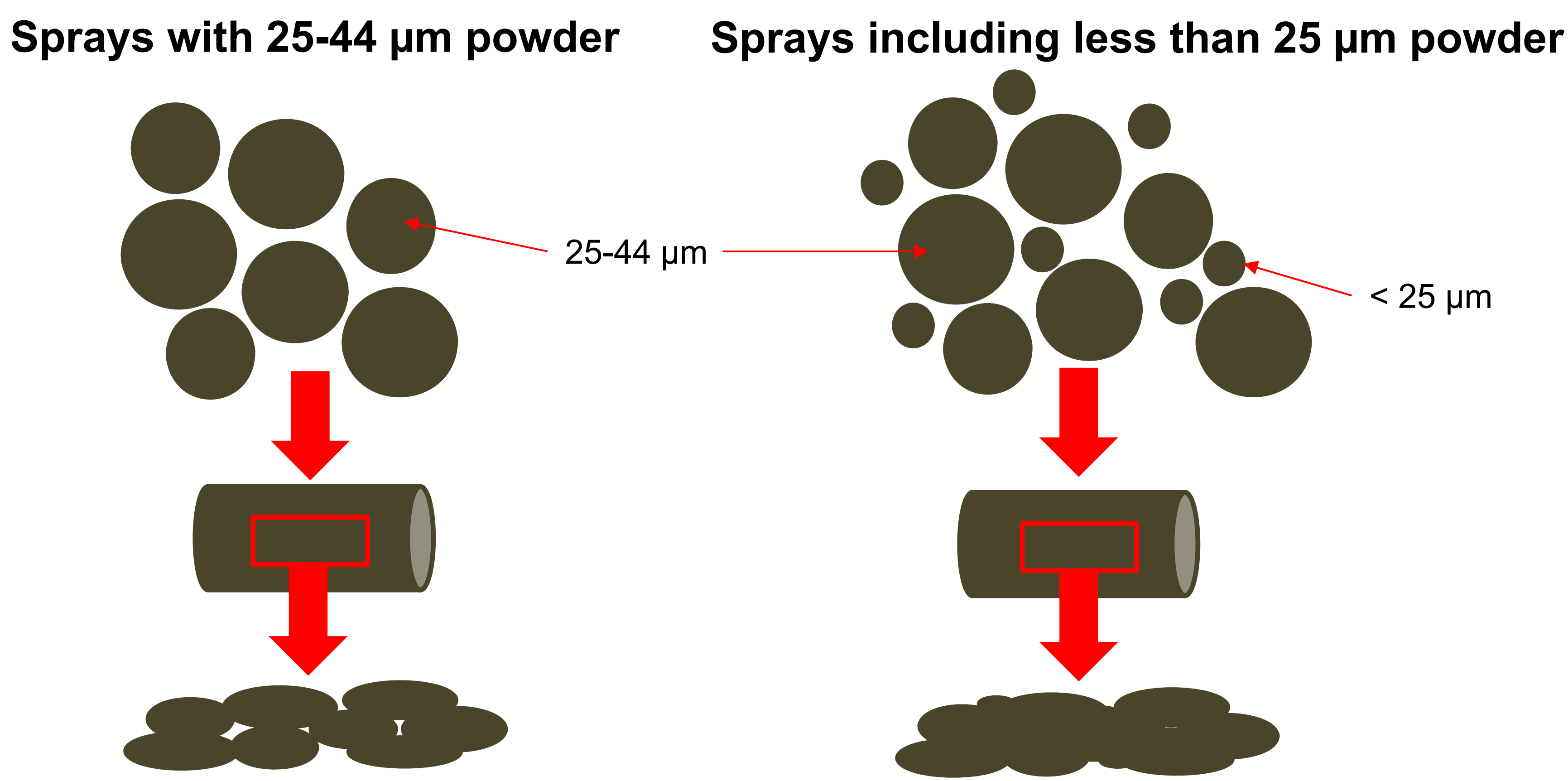


ODS tube including < 25 µm powder



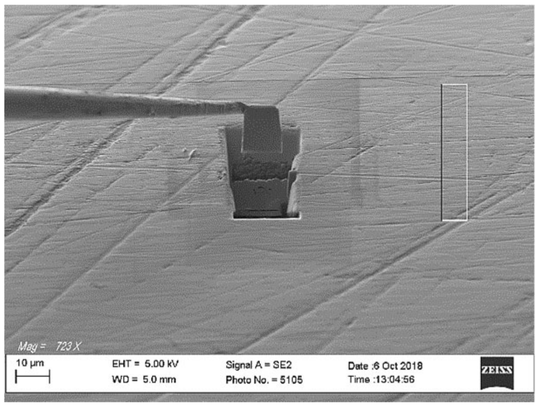
- Two different powder size distributions investigated:
  - 25-44 µm size powder
  - -44 um size powder that included powder sizes less than 25 um
- The -44 um sized powder increased both the deposit density and thickness

Why is there an increase in deposition thickness and density?

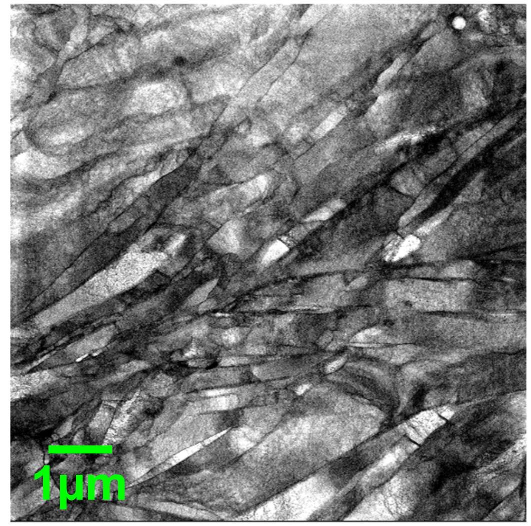


- Smaller particle sizes were able to deform more readily (higher velocity) and could deposit and fill in pores between the larger particles increasing the deposit thickness and decreasing the porosity

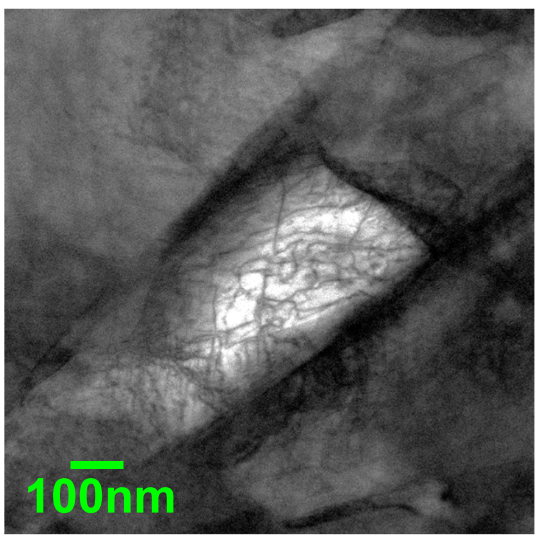
Microstructure of as-deposited 14YWT



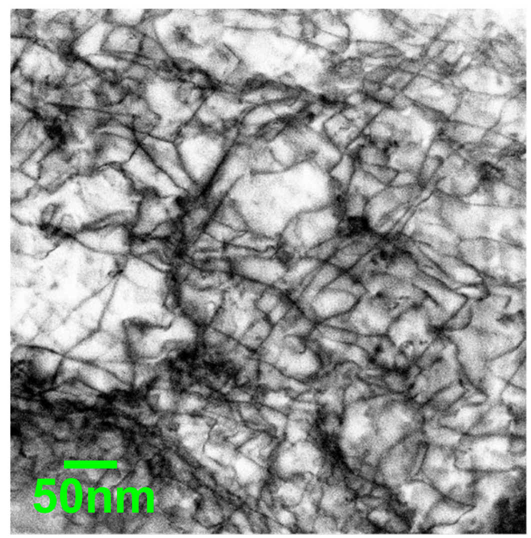
FIB lift-out technique



Grain refinement (grain size is less than 1µm)

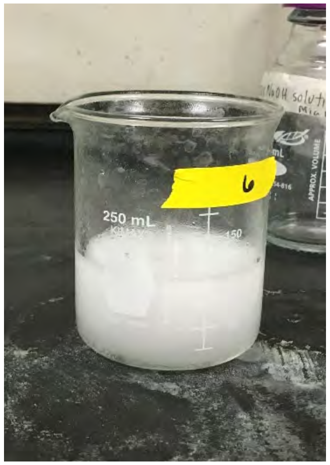


High density of line dislocations in the small grains

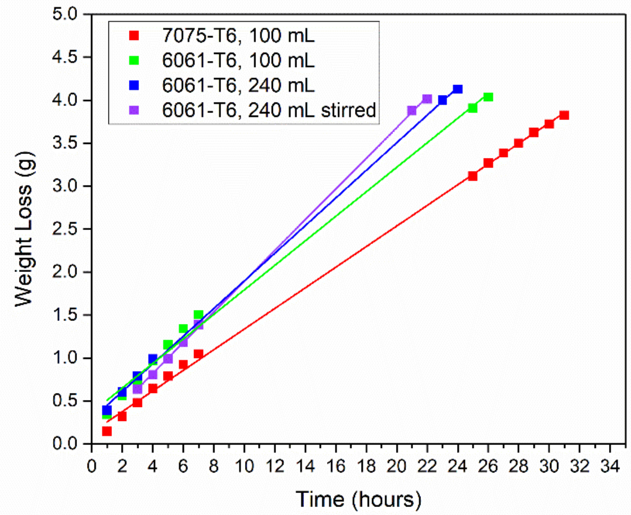


Dislocation forest and disappearance of the nanoparticles

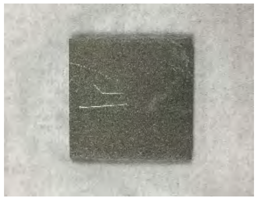
Dissolution of Aluminum Mandrel



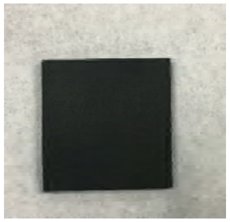
20% NaOH solution dissolving aluminum flats



6061-T6 after immersion in 20% NaOH solution

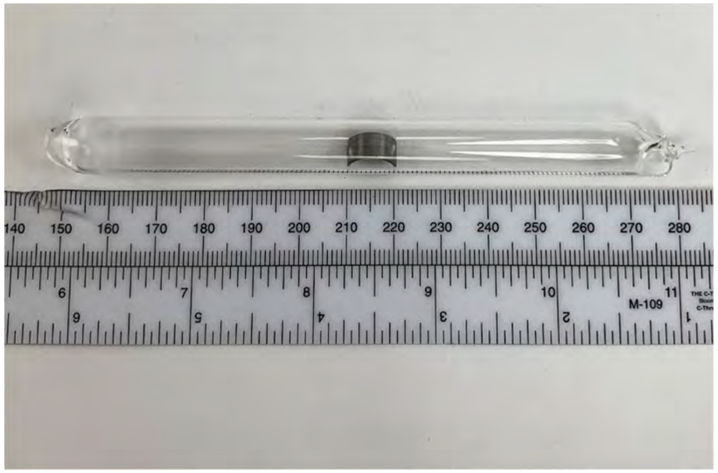


7075-T6 after immersion in 20% NaOH solution



- Dissolution studies on both 6061-T6 and 7075-T6 aluminum flats were performed in 20% NaOH solution
- 6061-T6 dissolved faster than 7075-T6
- Of course, stirring increased dissolution rate

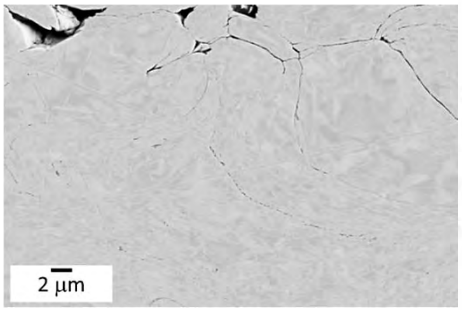
Post-heat Treatment of ODS Flats/Tubes

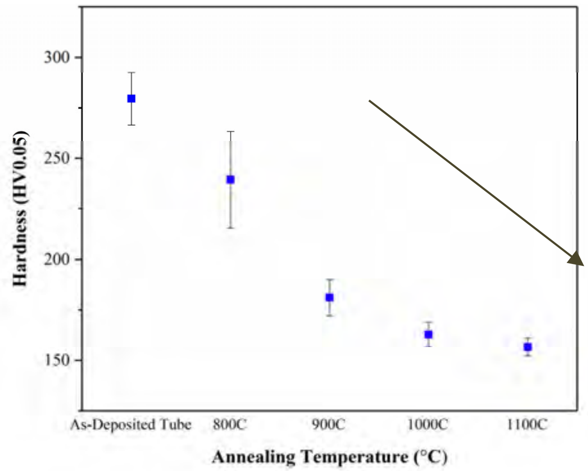


- Annealing experiments were conducted for precipitation the Y-Ti-O nanoclusters and improve ductility
- Flats/Tubes were annealed in a quartz tube furnace at 800°C, 900°C, 1000 °C and 1100 °C for 1 hour

Microstructural Evolution during Post-heat Treatment (SEM Images & Hardness)

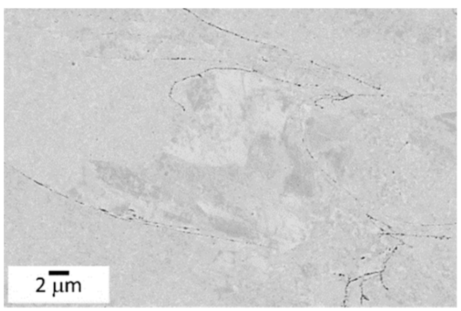
As-deposited



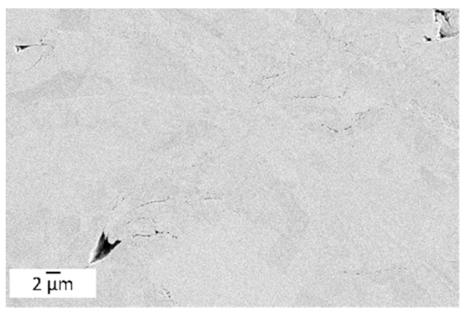


Done on un-optimized microstructure (porosity in the matrix, which has now been now addressed)

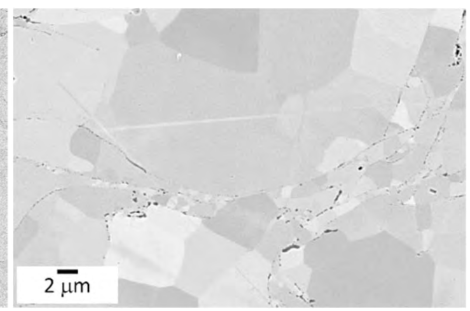
800°C



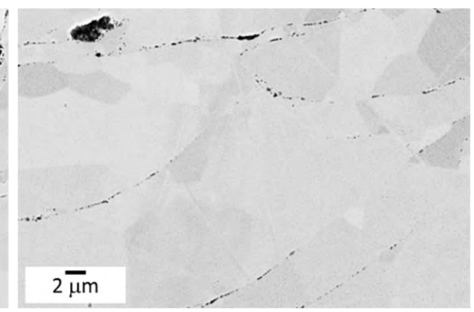
900°C



1000°C



1100°C

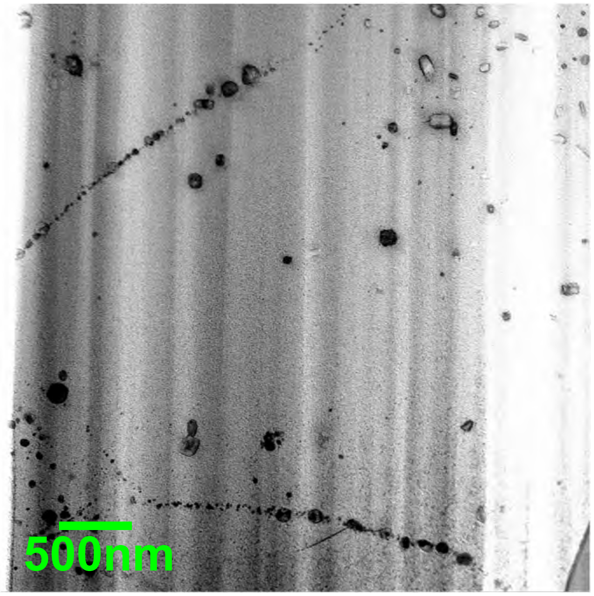


- Grain growth with increasing annealing temperature
- Annealing studies were performed on un-optimized ODS tube deposit

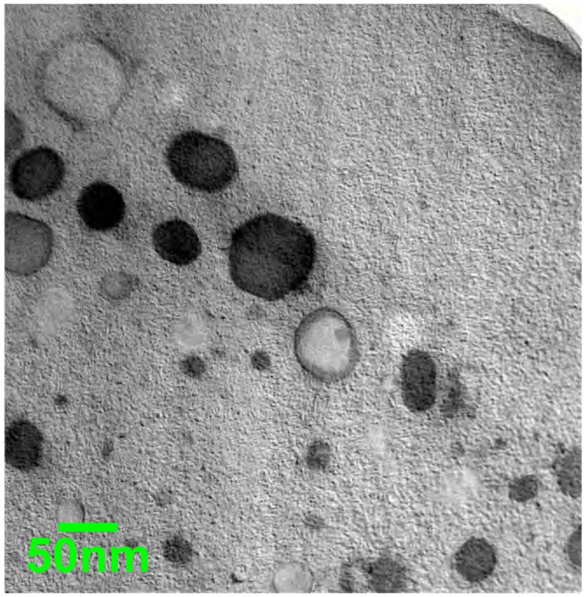
Microstructural Evolution during Post-heat Treatment (TEM Images)

- The heat treatment (1000 ºC) induced strain relaxation, recrystallization, and *reprecipitation* of nanoparticles or nanoclusters

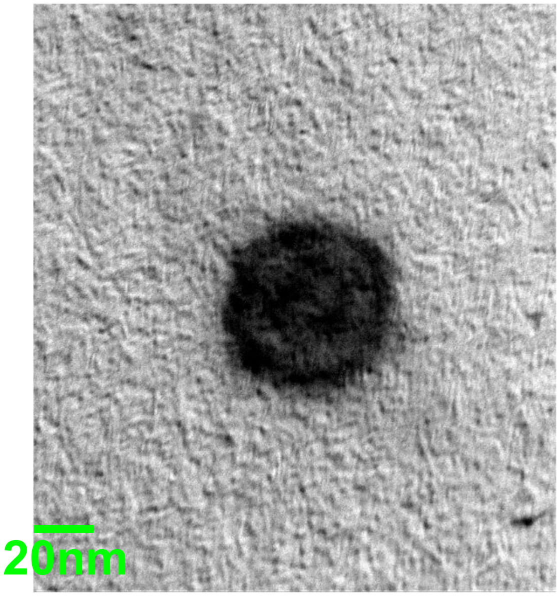
Nanoparticle dispersed in the grain



Annealed out dislocation forest



Fine nano-particles (~ 40 nm)

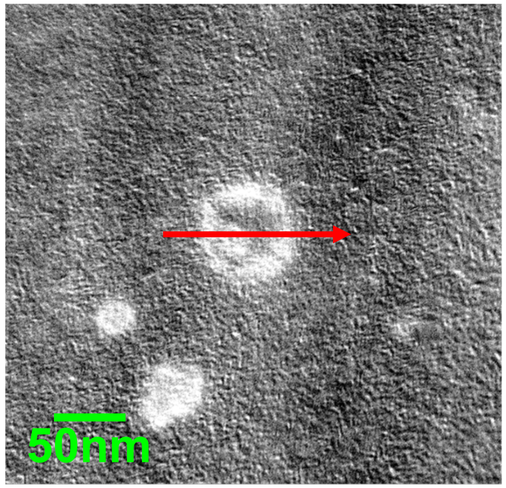


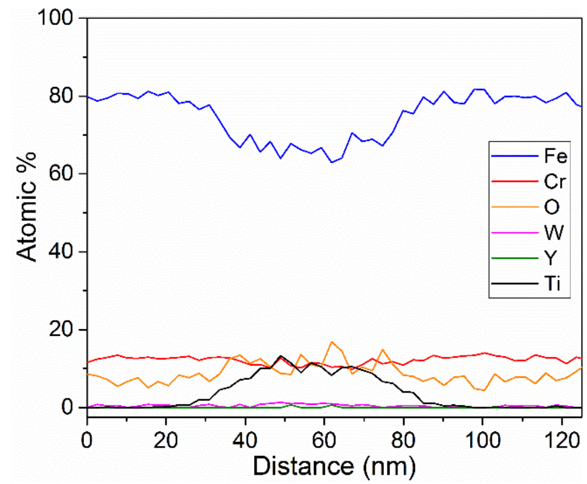
Chemical Composition of Nanoparticles in Heat Treated ODS Deposit

HAADF Images

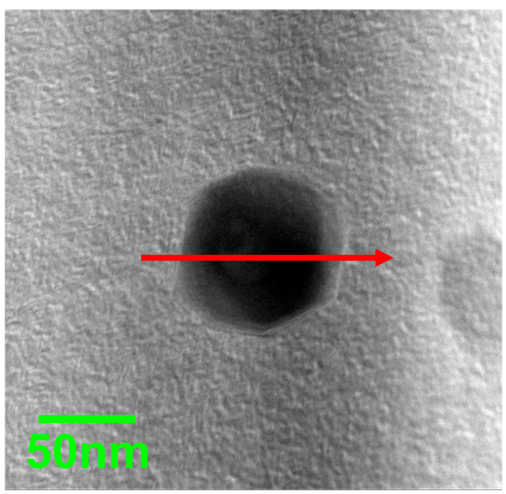
EDS line profile

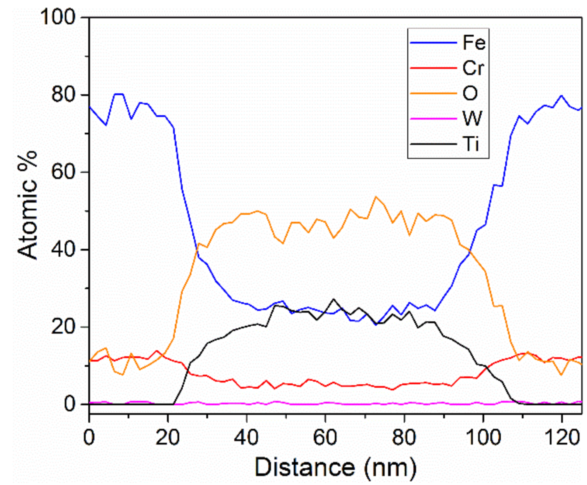
Ti and Y-rich and low oxygen particle





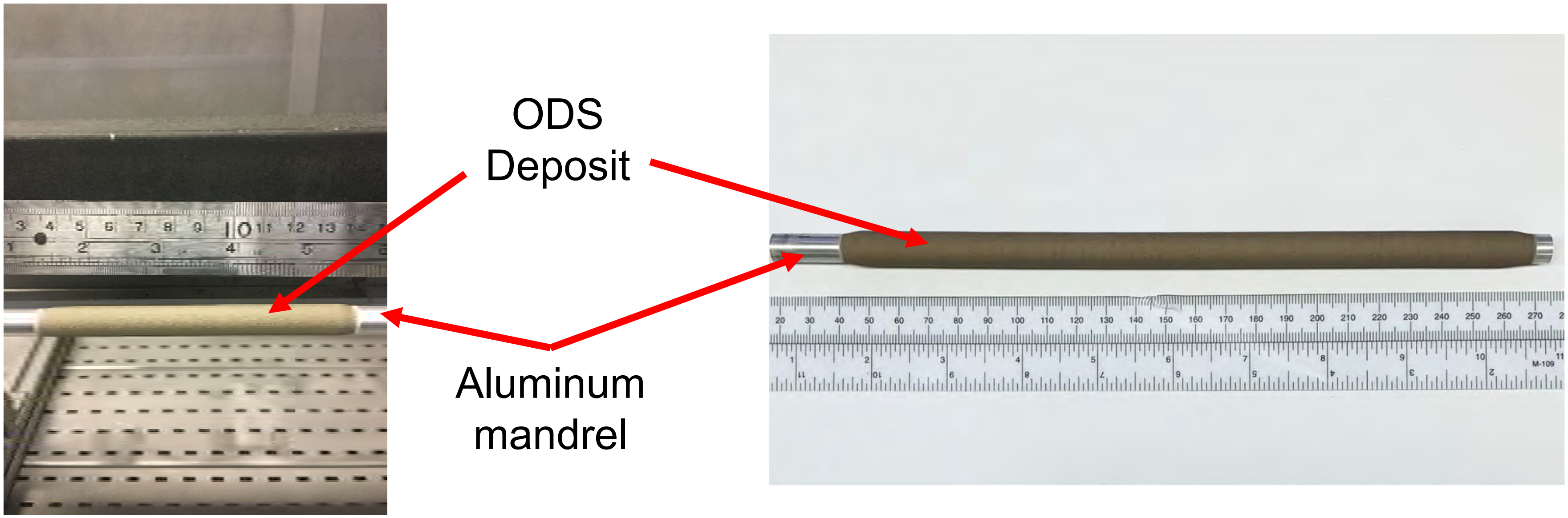
Ti-rich and high oxygen particle





Cold Spray Manufactured ODS Fuel Cladding Tubes

Step 1: ODS Cladding Tube Fabrication: Cold Spray Deposition



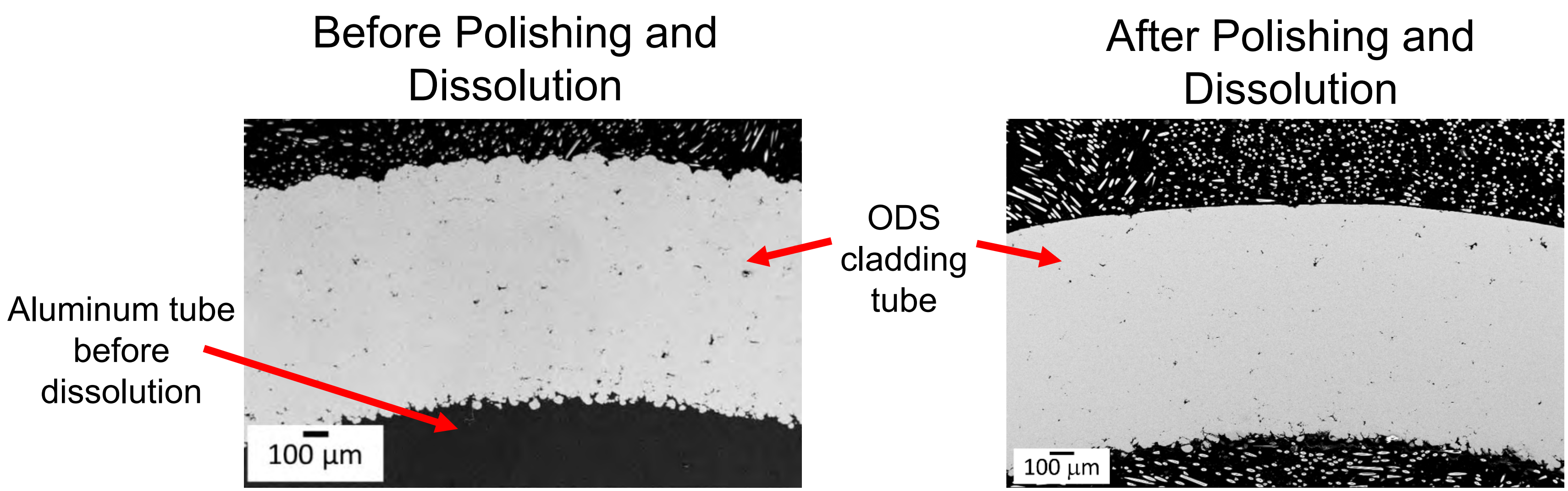
- Deposition was performed on a 6061-T6 aluminum tube mandrel (0.375” OD) while rotating
- Cold spray parameters were adjusted to achieve the highest quality deposit using 4” long tubes
- A longer 10” tube was then produced to show scalability for full length cladding tube

Deposition of ODS on 6061-T6 Aluminum Tube Mandrel

5" tube formed in ~ 60 seconds

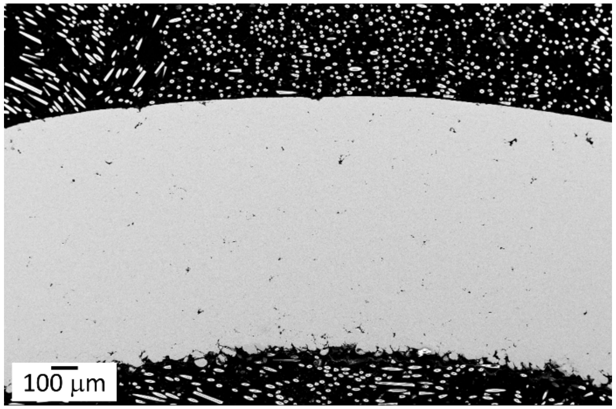
Thickness: 1 mm for < 44 μm size particles

Step 2: Dissolution of Aluminum Tube Mandrel to leave free-standing ODS Tube

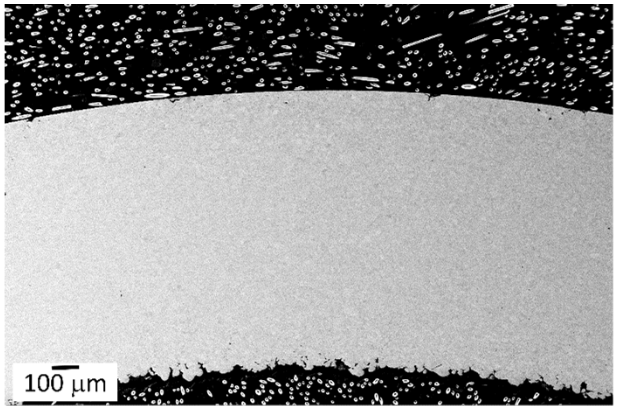


- Dissolution of the aluminum tube mandrel in 20% NaOH solution
- Final polishing of the ODS cladding tube was done to improve surface finish

Step 3: Vacuum Heat Treatment of ODS Tube



Before vacuum heat treatment

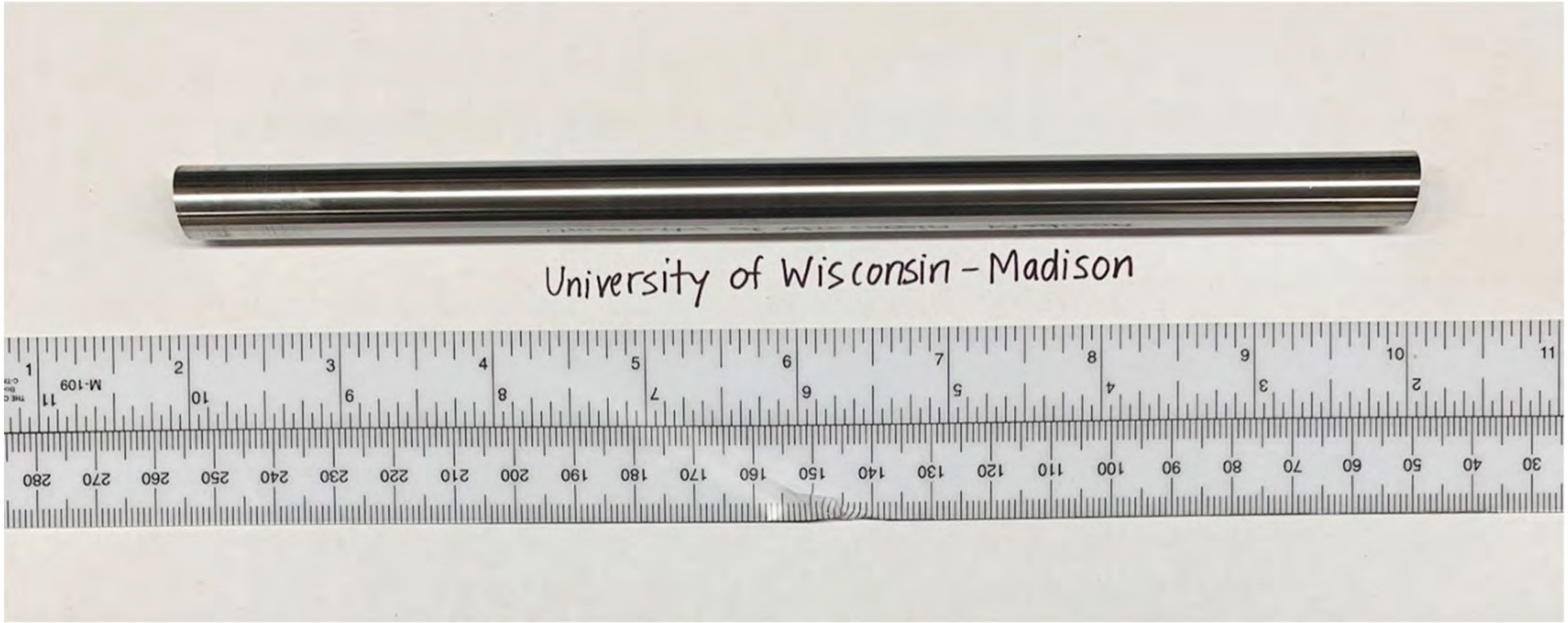


After vacuum heat treatment

- Tubes were vacuum heat treated at 1085°C for 1 hour to eliminate all residual porosity in the cladding tube
- Vacuum heat treatment serves to both precipitate oxide nanoclusters and promote densification

Vacuum heat treatment performed at Thermal Spray Technologies (TST)

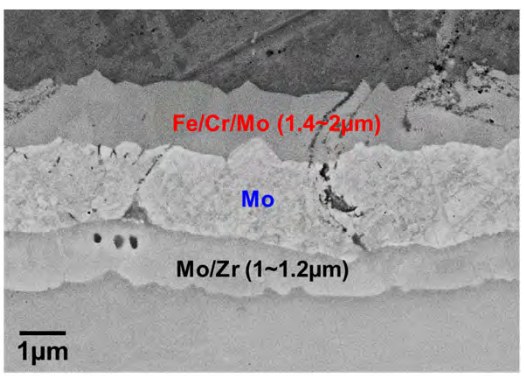
Final Free-Standing ODS Cladding Tube

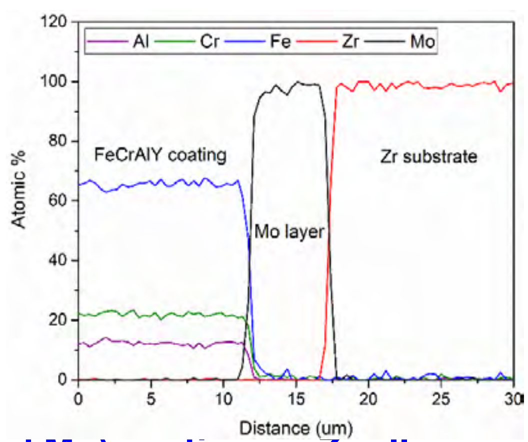


8” ODS cladding tube

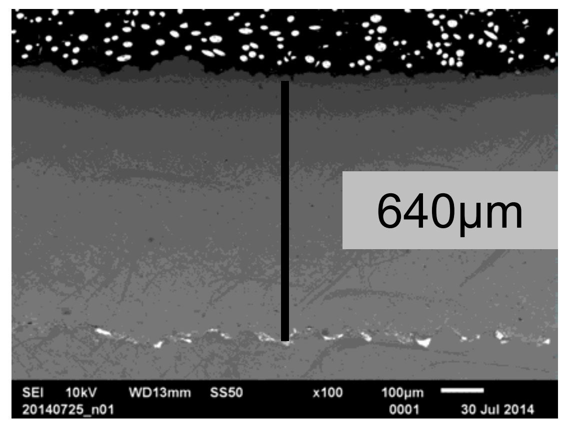
Immediate Future Work

- Further optimization of the manufacturing process including microstructural engineering (e.g., grain size, nanocluster size/density)
- Mechanical testing of ODS cladding tubes
- Heavy ion irradiation experiments on ODS cladding tubes
- Seeking future opportunities for neutron irradiation experiments and associated post-irradiation examination research (not a part of NEET program)
- Surface modification and coatings for corrosion and wear resistance (e.g., multi-layered coating and compositionally graded coatings)





Dual-layered (FeCrAl and Mo) coating on Zr-alloy



IN600 coating on steel

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