Powder Metallurgy Fabrication of Molybdenum Target Materials and Assemblies

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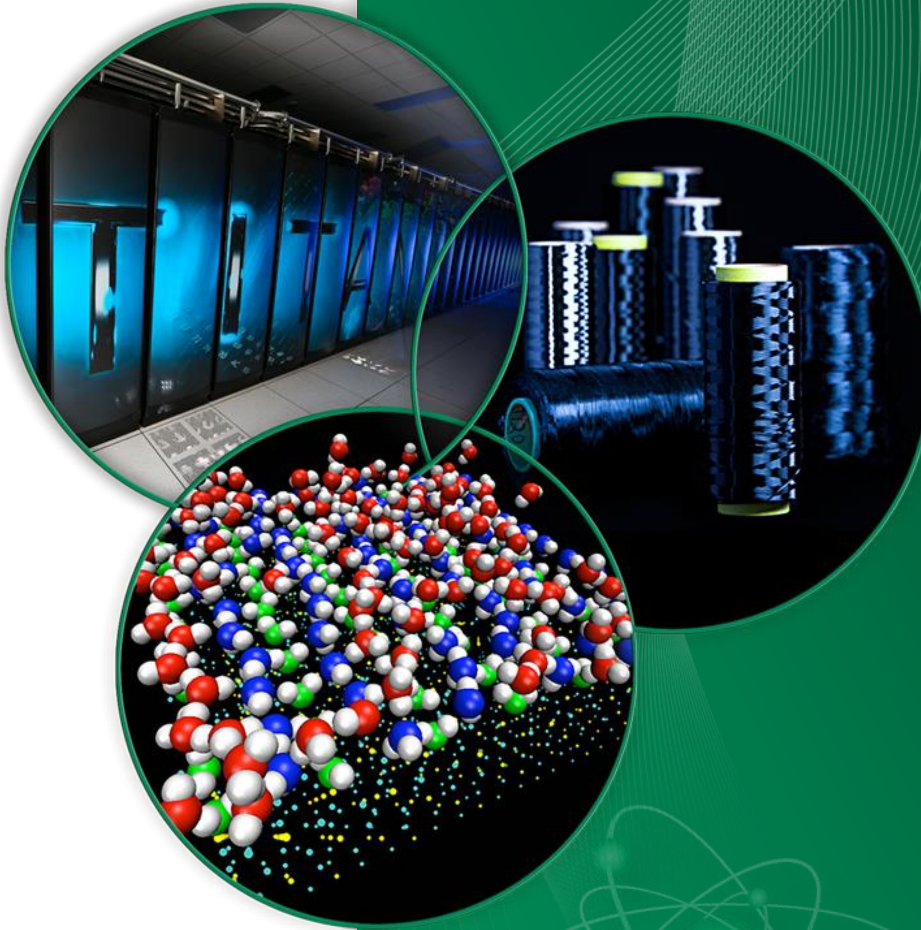


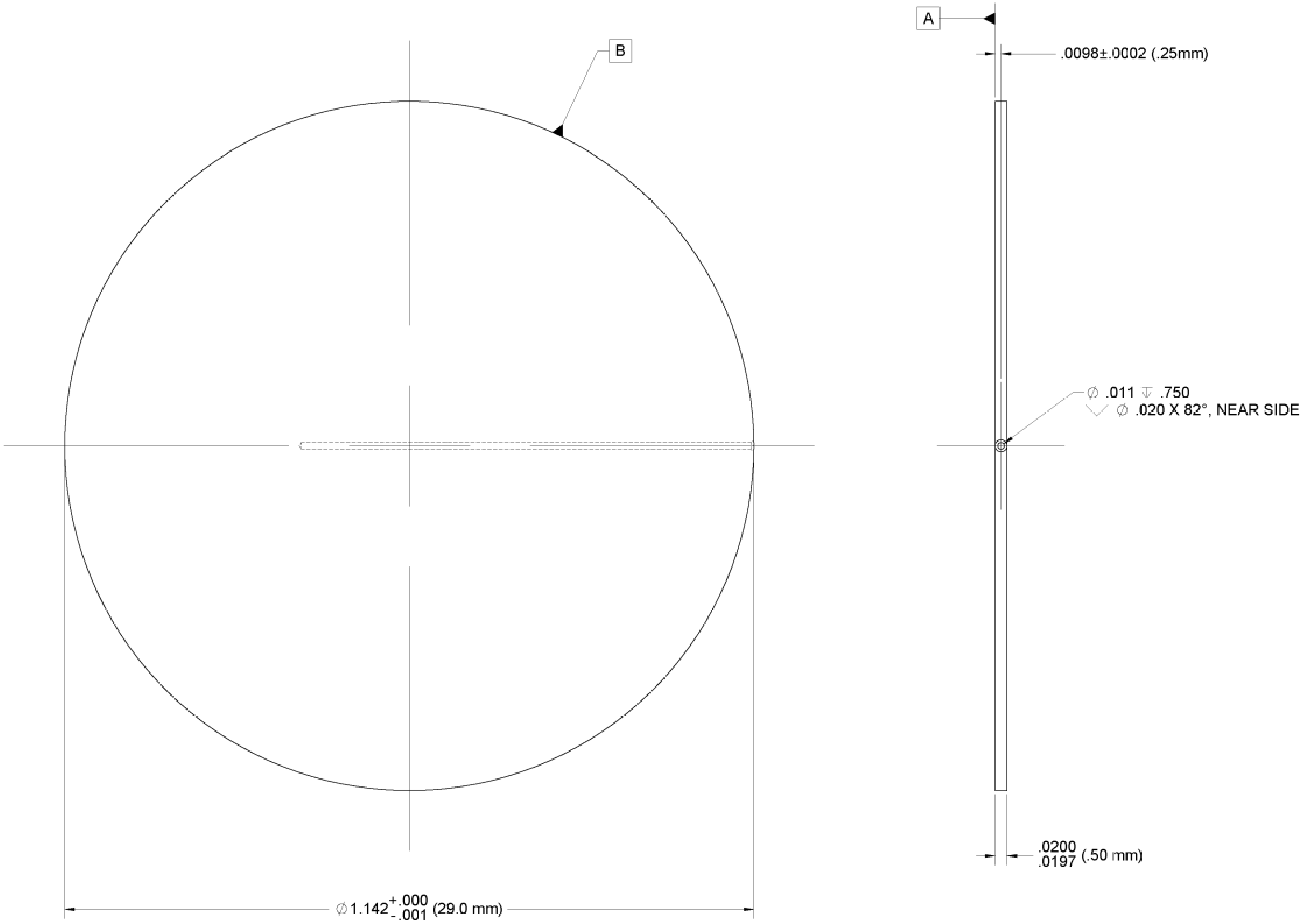
Mo-99 Topical Meeting
Boston, MA
September 3, 2015

Target Disks and Assemblies for the Accelerator Production of Mo-99 Are Being Fabricated Employing Powder Metallurgy Approaches

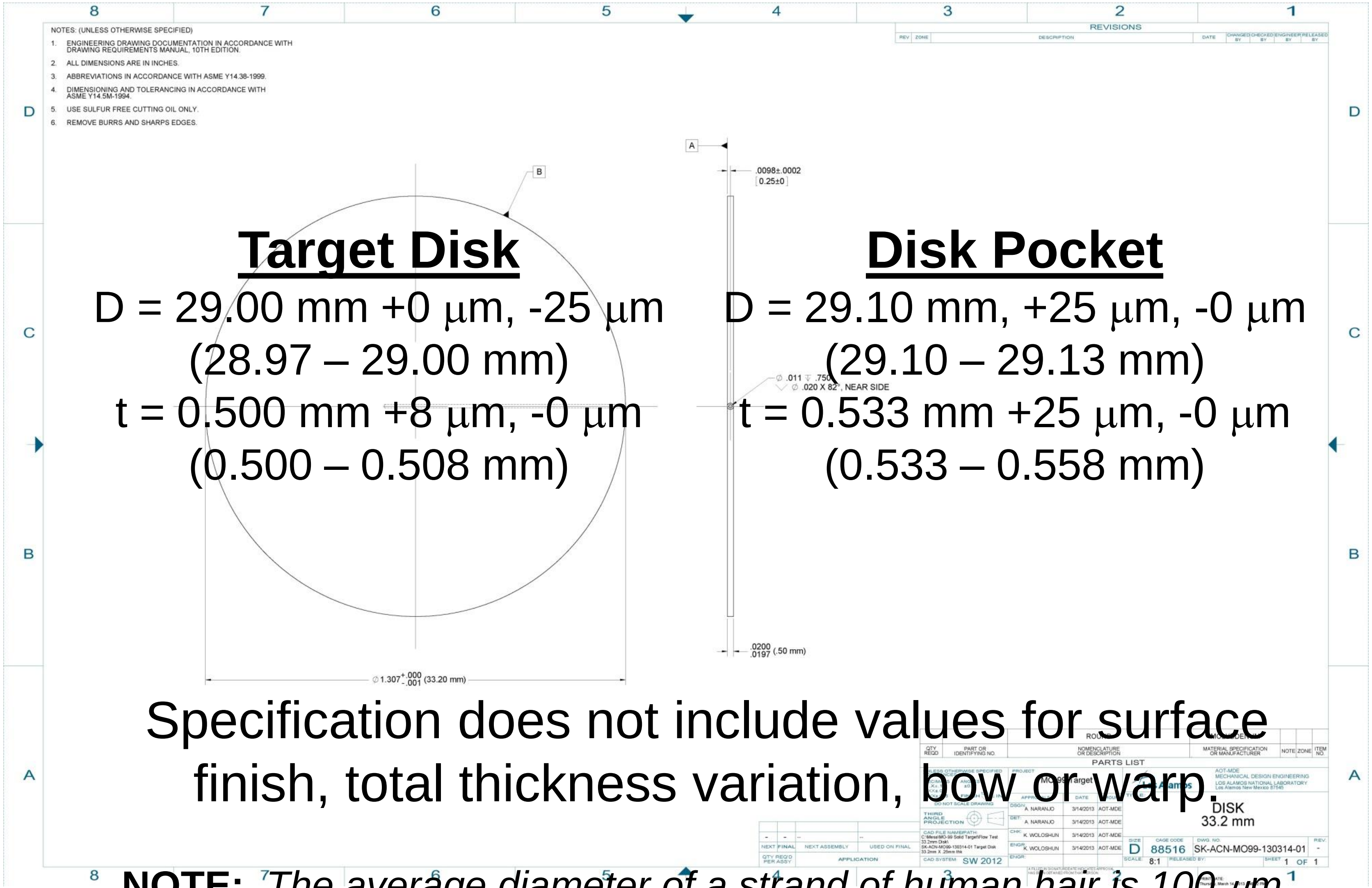
The goals of this effort:

- Understand the **requirements for and fabrication of molybdenum target disks** that will be used in the accelerator production of Mo-99.
- Develop a **process for fabricating accelerator target disks with a density of 90% or greater and acceptable thermomechanical properties**.
- Identify **characteristics that affect the dissolution rate** of target disks.
- Assist in **developing a process for recycling isotopically-enriched molybdenum**.

The Current Target Is a 29 x 0.5 mm Disk



Target Disk and Holder Specifications Are Quite Stringent



Target Disk

D = 29.00 mm +0 μm, -25 μm

(28.97 – 29.00 mm)

t = 0.500 mm +8 μm, -0 μm

(0.500 – 0.508 mm)

Disk Pocket

D = 29.10 mm, +25 μm, -0 μm

(29.10 – 29.13 mm)

t = 0.533 mm +25 μm, -0 μm

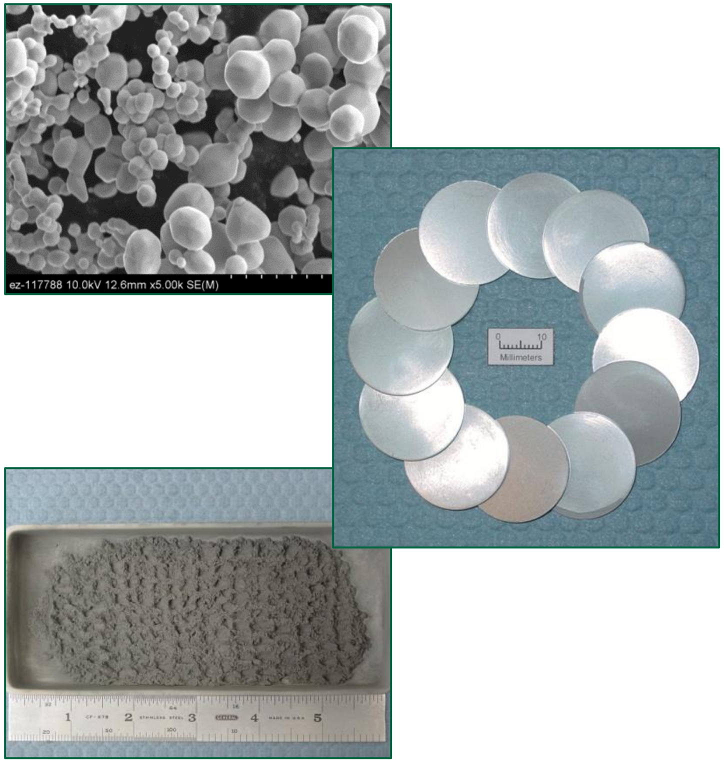
(0.533 – 0.558 mm)

Specification does not include values for surface finish, total thickness variation, bow or warp.

NOTE: *The average diameter of a strand of human hair is 100 μm.*

Powder Metallurgy (PM) Is Being Evaluated for the Production of Accelerator Targets

- **Powder Metallurgy** is a method of producing components by pressing or shaping metal powders which are subsequently heated to create a dense, coherent object.
- Advantages
  - **Near-net shape**
  - **No waste**
  - **Controlled porosity**
- Disadvantages
  - High cost of tooling and powder
  - Powders can be difficult to handle
  - Geometric and size limitations
  - Density variations



Most If Not All Molybdenum Powders Were Pressed and Sintered to the Desired Density

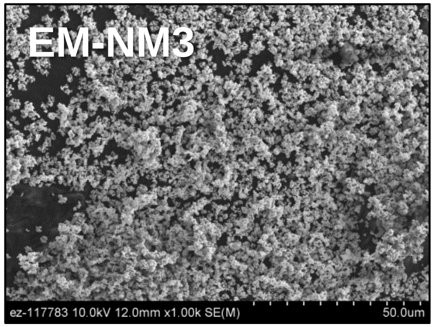
Powder	Compact Press. (ksi)	%TD (green)	Sintering (°C/h)	%TD (sintered)	Open Porosity	Diameter (mm)	Thickness (mm)	Shrinkage (%)		Bow
								D	t	
33 mm disks										
NPA	100	80	1500/1	90	NM	32.0	0.59	3.9	1.7	mixed
NPA	100	80	1550/1	91	NM	NM	NM	NM	NM	severe
NPA	150	85	1500/1	92.5	NM	32.5	0.56	2.0	0	unknown
NPA	200	87	1500/1	94	NM	32.8	0.53	1.0	0	unknown
NPA	100/150	81	1200/1	91	NM	32.6	0.54	0.9	3.2	unknown
NPA	100/200	86	1400/1	93	NM	32.6	0.52	1.2	0	unknown
29 mm disks										
NPA	100	80	1500/1	**89**	**10**	29.2	0.50 ± 0.03	3.4	4.1	mixed
NPA	100	80	1550/1	93	4	**29.0**	**0.50 ± 0.02**	4.0	1.9	mixed
EM-NM3	100	43	1500/1	98	0	22.9	0.76	24.6	25	severe
LB1	100	65	1500/1	89	5	27.6	0.57 ± 0.02	8.4	9.6	moderate
LB1	100	65	1550/1	91	< 1	27.4	0.56 ± 0.02	9.4	11.4	severe
LB1-M	100	65	1500/1	88	~ 5	27.7	0.58 ± 0.03	8.3	7.3	moderate
LB1-S	100	67.5	1500/1	87	~ 9	28.0	0.56 ± 0.02	7.2	7.8	severe

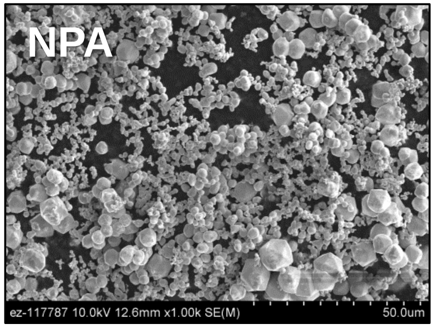
However, most if not all disks were severely distorted, i.e. warped and bowed.

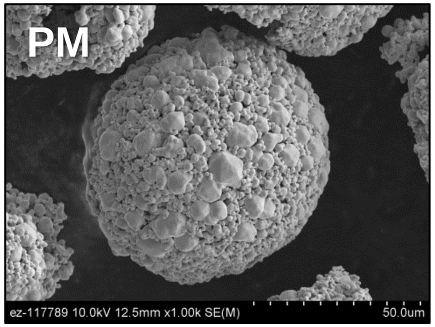
Powder Must Flow to Uniformly Fill the Die Cavity

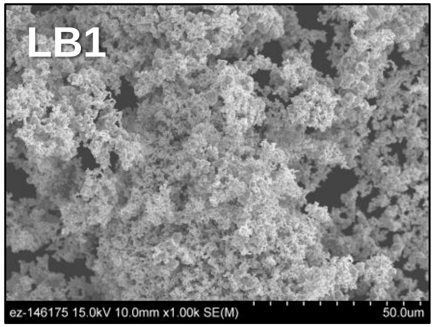
Molybdenum Supplier	Grade	Purity (% Mo)	Max. Oxygen (ppm)	Particle Size	BET (m^2/g)	Hall Flow (sec/50 g)
Climax Molybdenum	EM-NM3	99.9	1400	0.7 – 1.5 μm	2.83	No flow
Climax Molybdenum	NPA	99.95	1000	4.0 – 4.8 μm	0.45	No flow
Climax Molybdenum	PM	99.9	2000	-200/+325 mesh (spray-dried)	NM	< 45
Large-batch reduction	LB1	NM	> 5000	4.8 ± 1.4 μm	0.46	No flow

NM = not measured

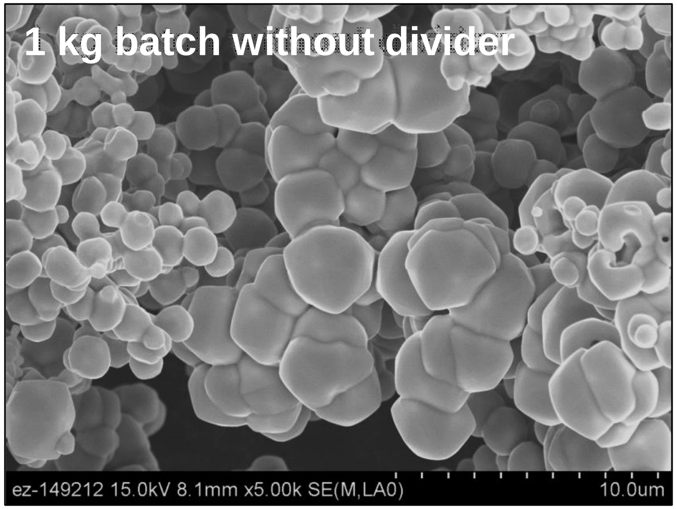


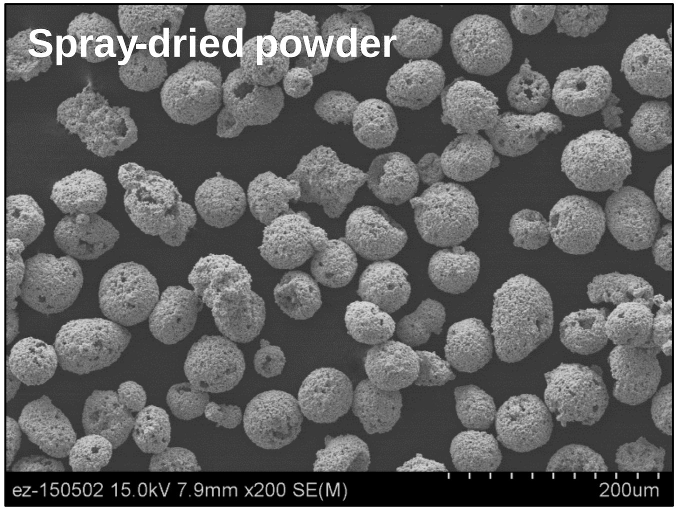




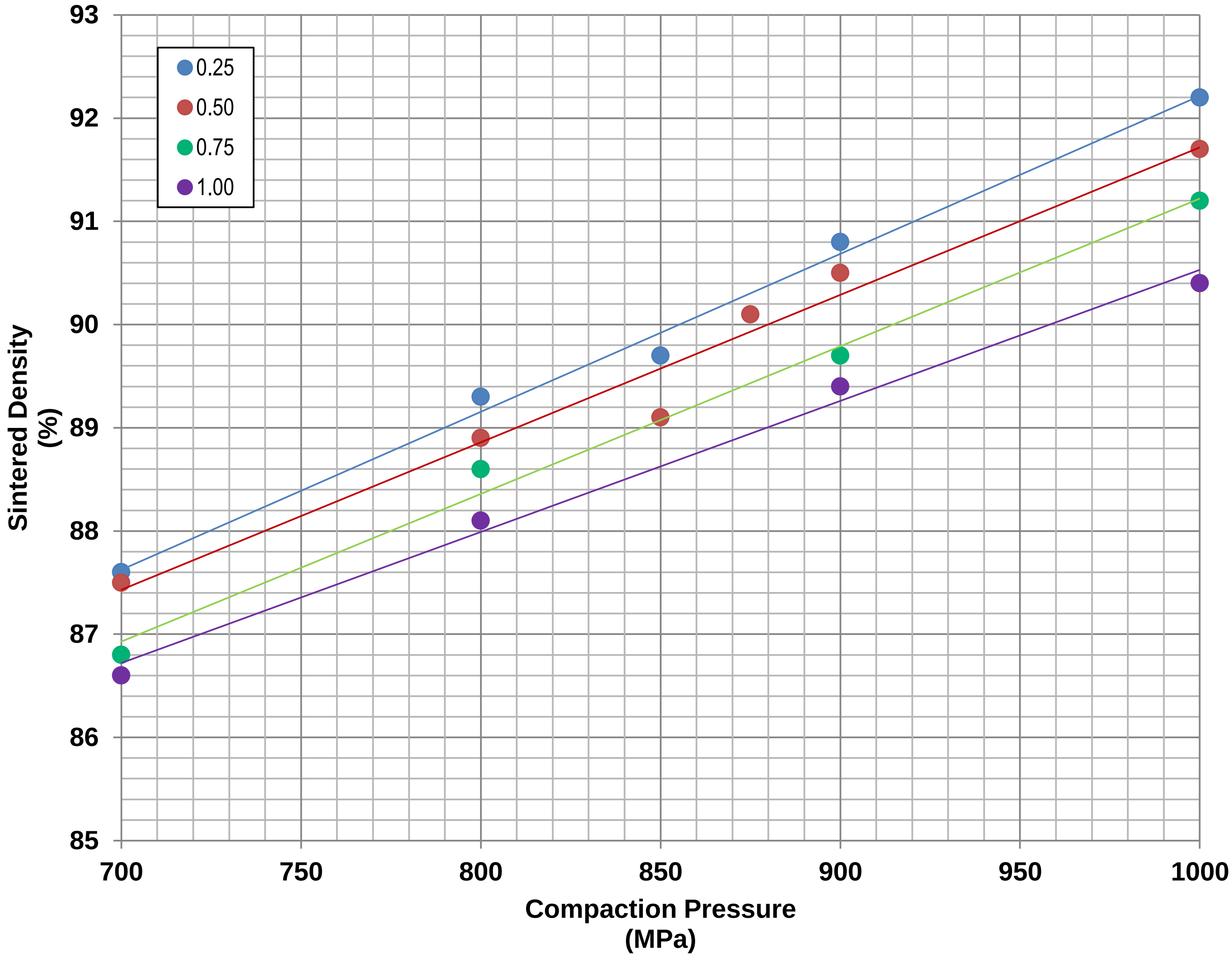


Reduced Powder Can be Milled and Spray-Dried to Produce "Flow-able" Feedstock

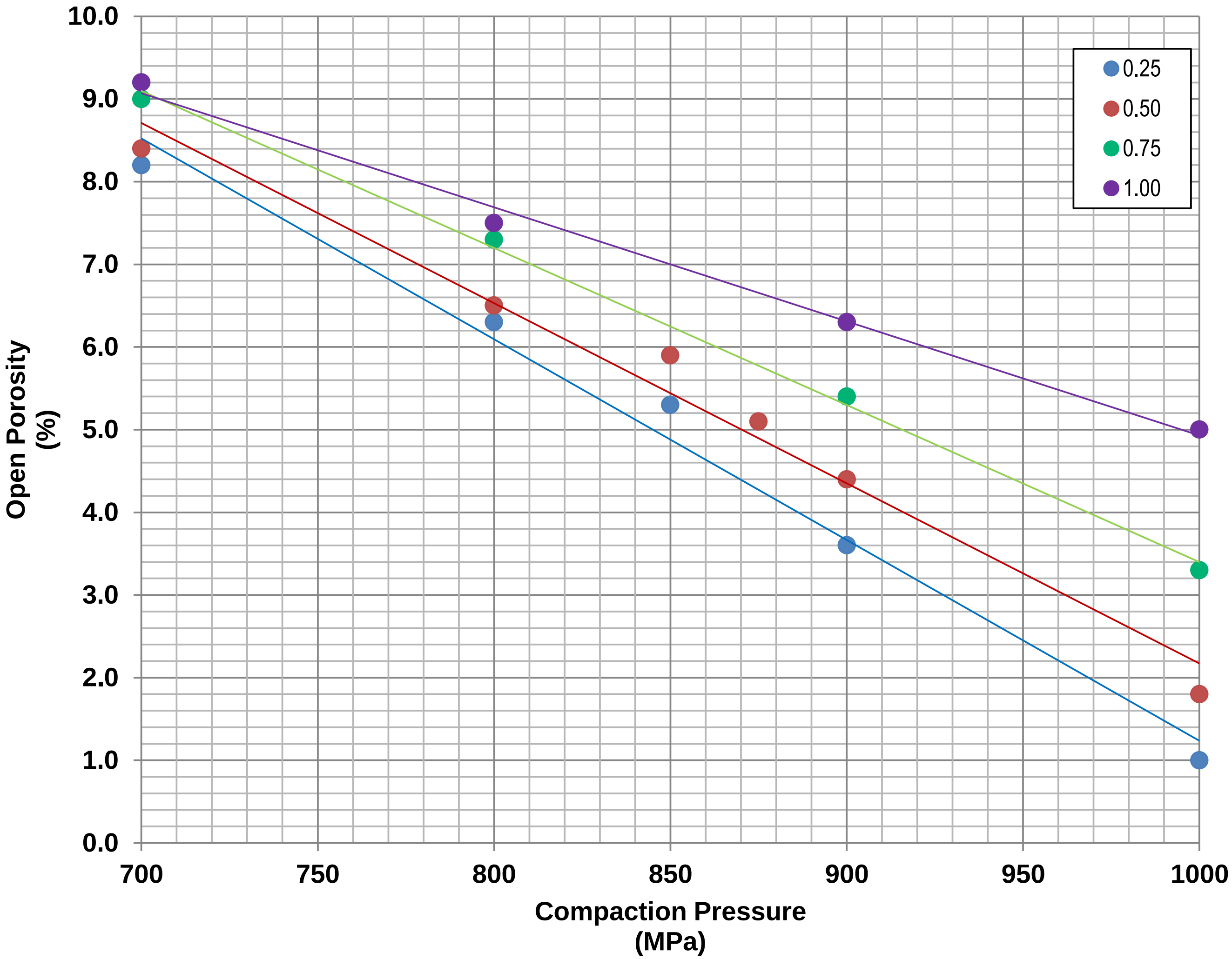




Powder Characteristics and Compaction Influence Sintering Behavior



A Factor That Affects Dissolution Rate is Open Porosity



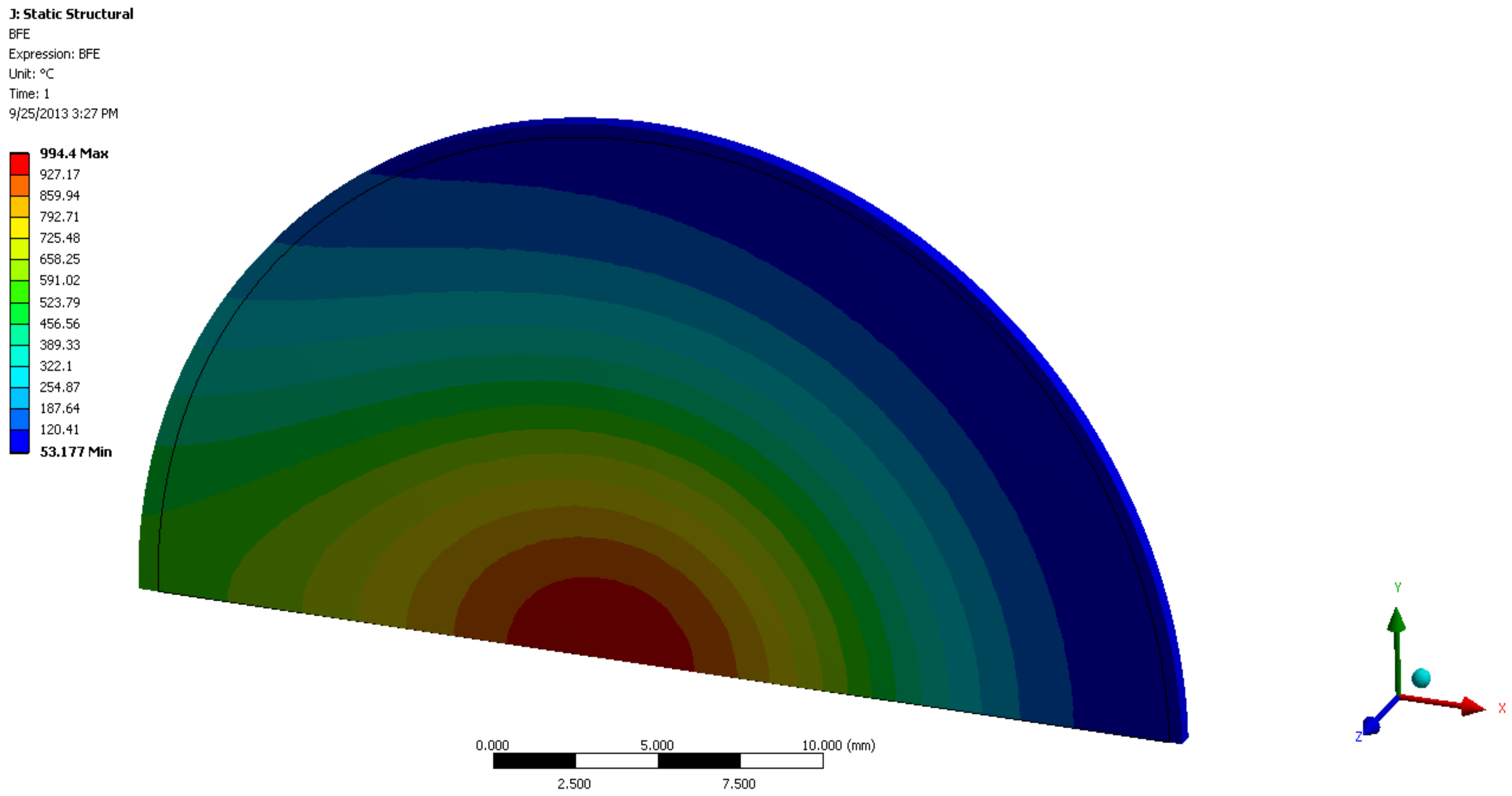
The Addition of Lubricants Improves Uniformity and Reproducibility

EBS Lube (wt%)	Comp. Press. (MPa)	Green Density (%)	Sintered Density/ Open Porosity (%)	Average Diameter (mm)	Thickness (mm)		
					Average	All Disks	Each Disk
0.25	850	79	90/5	29.3	0.52	± 0.002	± 0.006
0.50	875	79	90/5	29.3	0.52	± 0.005	± 0.010
0.75	900	80	90/5	29.4	0.52	± 0.004	± 0.010
1.00	1000	80	90/5	29.4	0.51	± 0.003	± 0.010

3.1 g of SD powder pressed at given pressure and sintered at 1600°C for 4 h

Shrinkage of ~ 4% in all directions!

Target Disks Will be Subject to Non-Uniform Heating During the Irradiation Process



Disk #14

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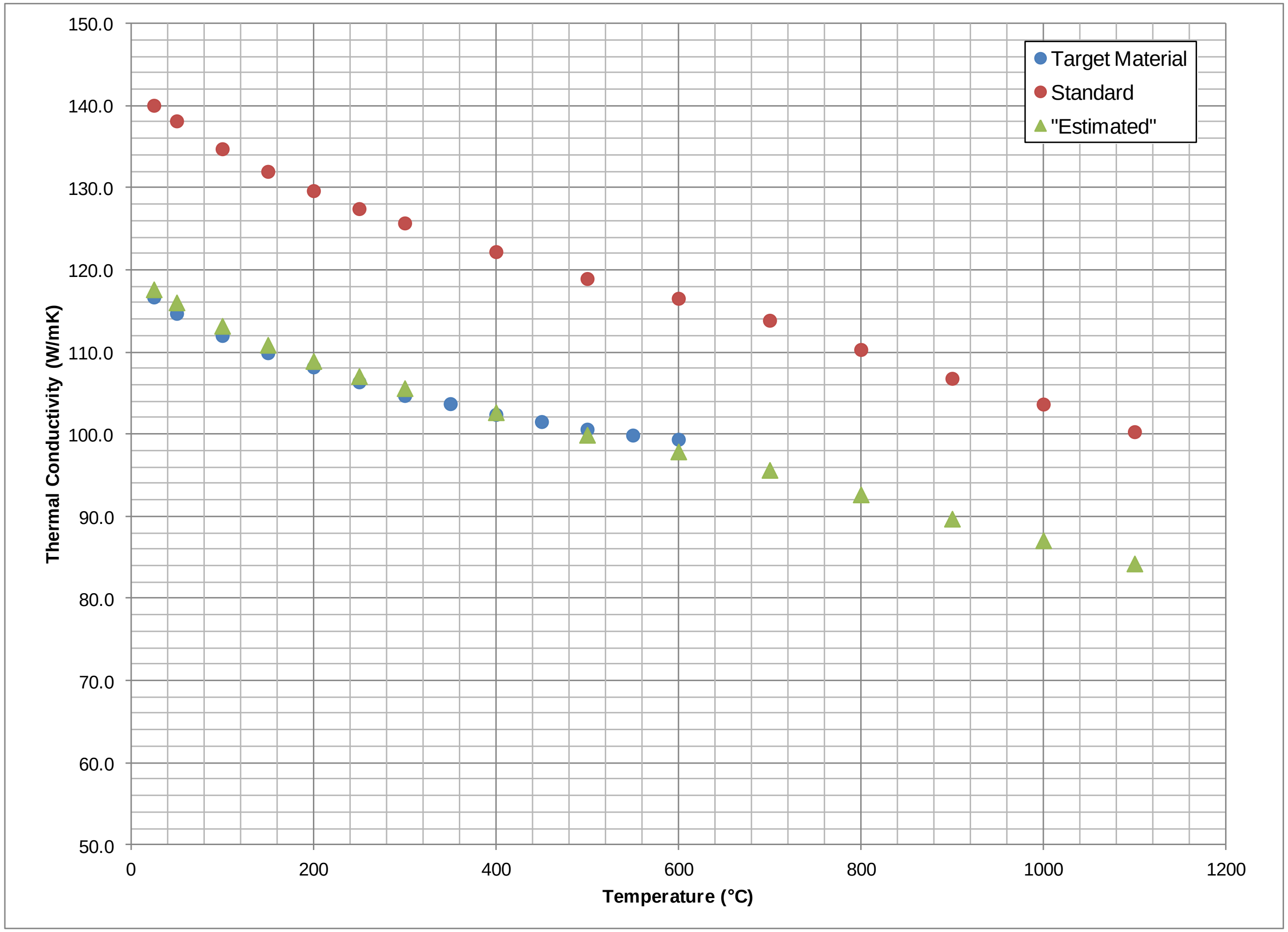
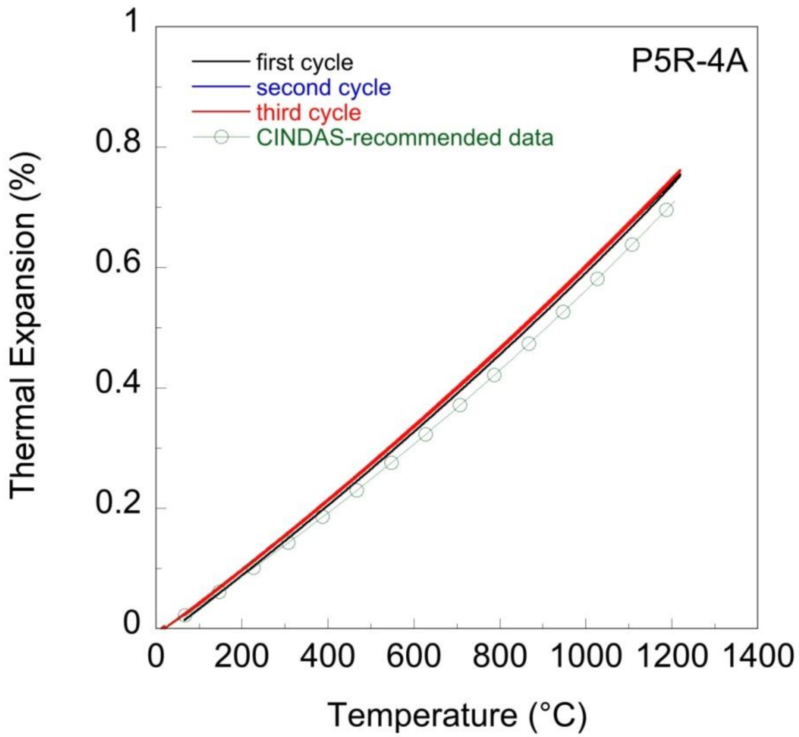
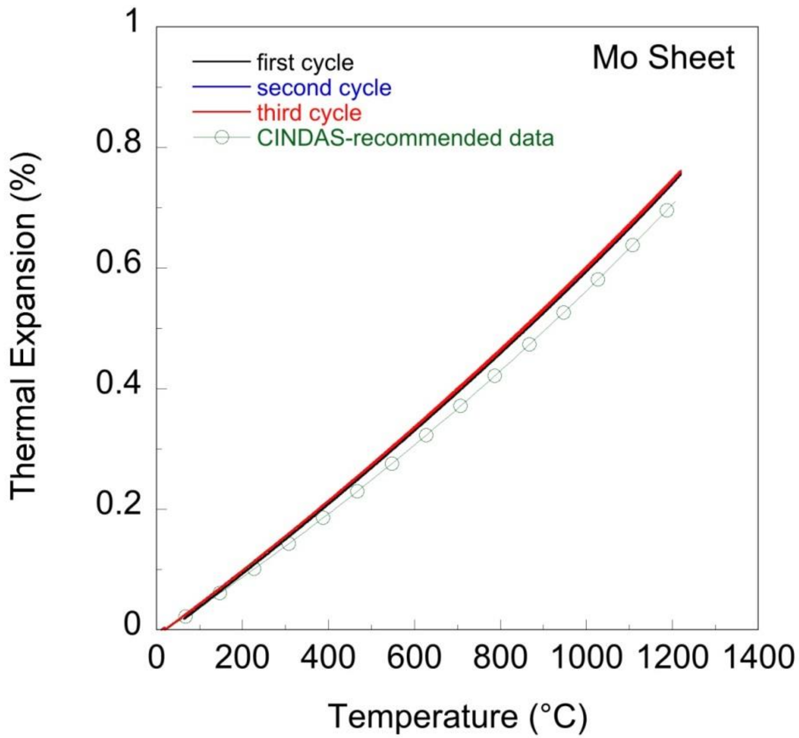
Thermal Conductivity of the PM Mo Was ~ 85% of the Values Measured for Dense Molybdenum



Results for 90% dense Mo with 5% open porosity. Values above 600°C for the PM Mo were estimated by comparison to trends for dense material.

Thermal Expansion Was Identical to the Values Measured for Dense Molybdenum Sheet Product

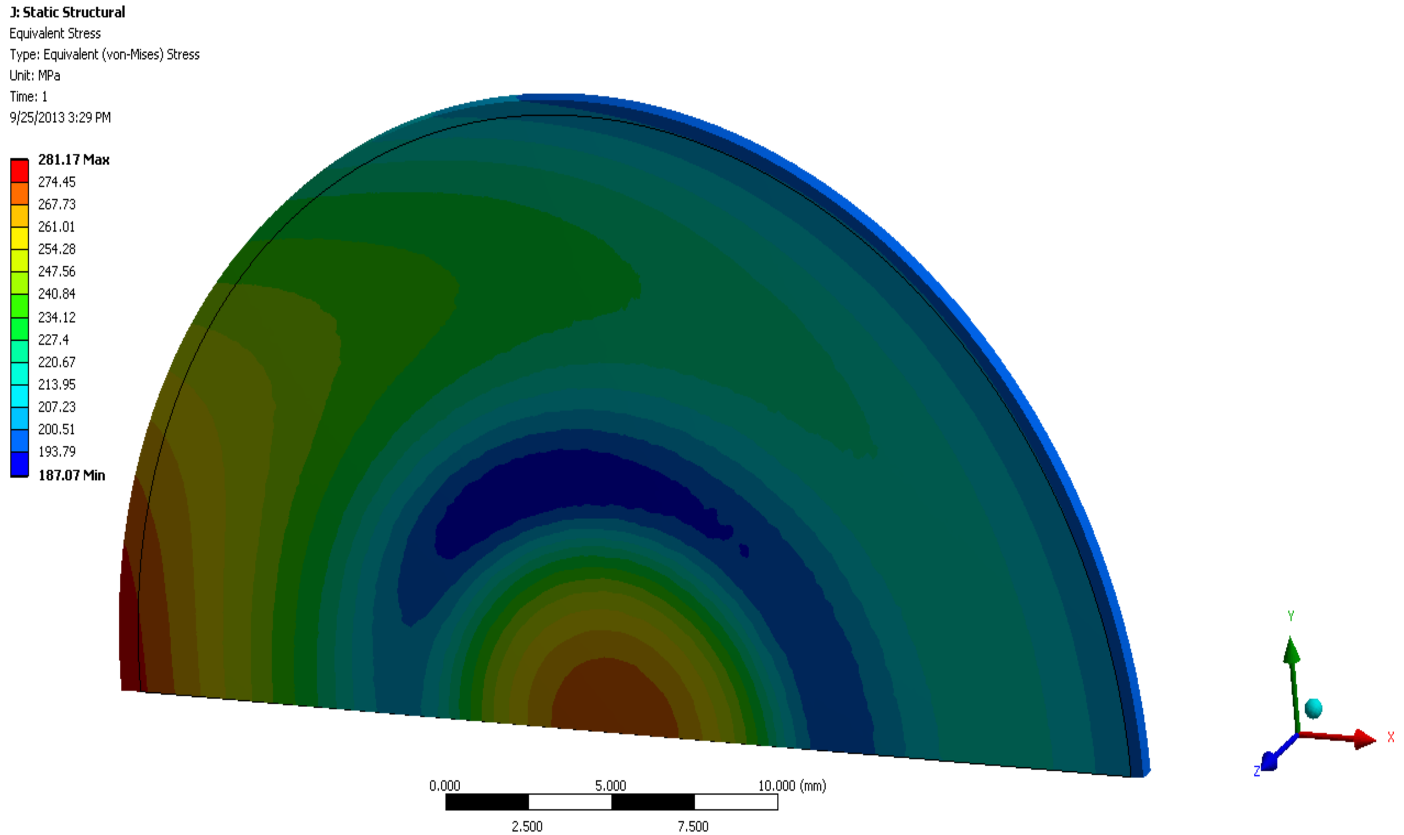




Results for 90% dense Mo with 5% open porosity.

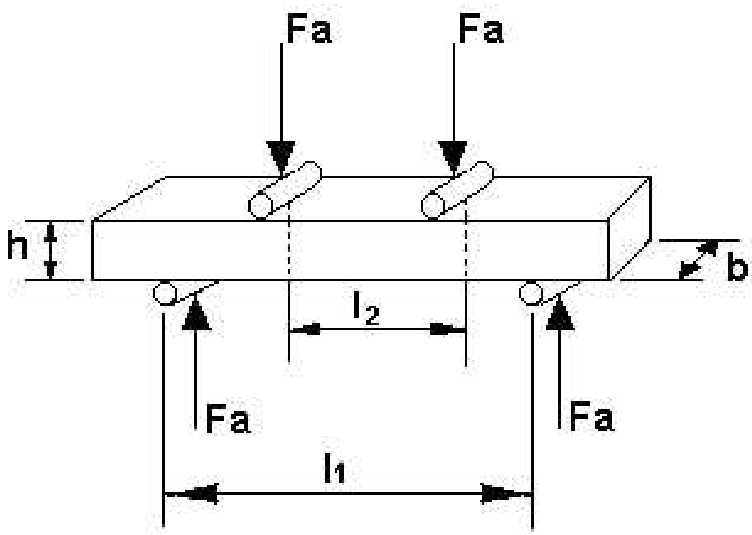
The Hot Center Expands While the Cold Periphery is Constrained Resulting in Non-Uniform Stress Across the Disk

Resulting stress:

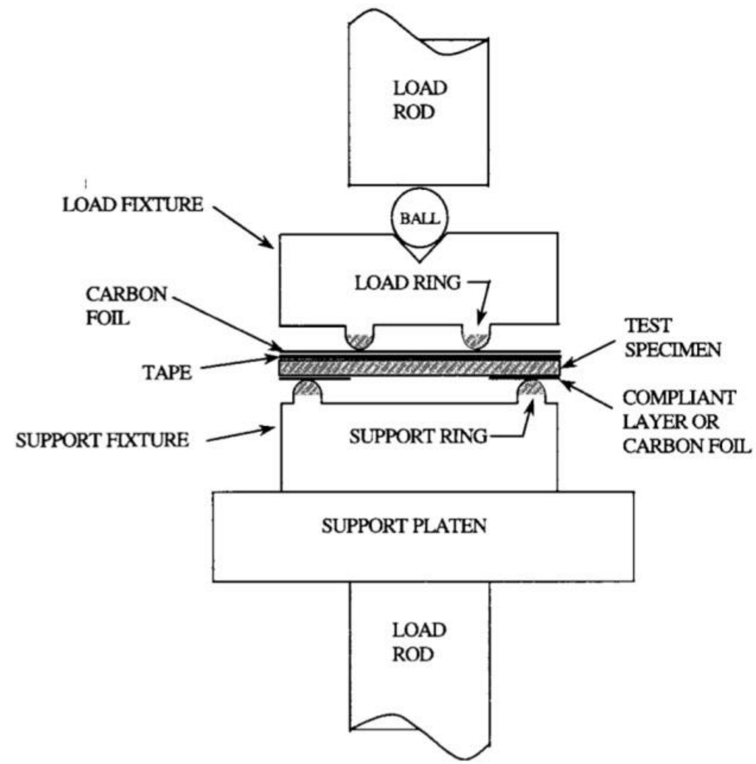


Disk #14

Mechanical Properties Were Measured Employing Two Flexure Test techniques



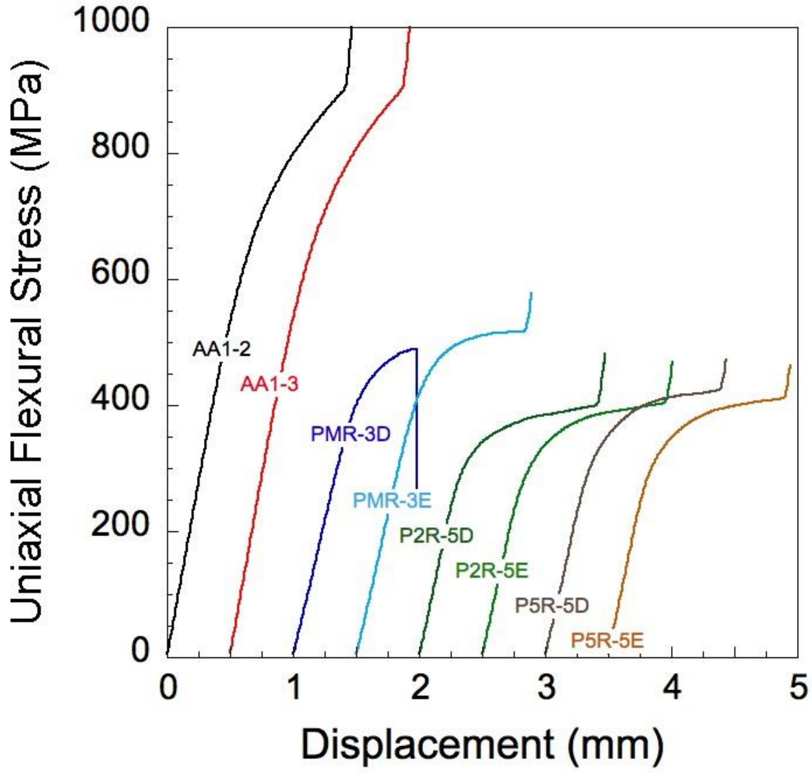
Four-Point Flexure



Biaxial Flexure

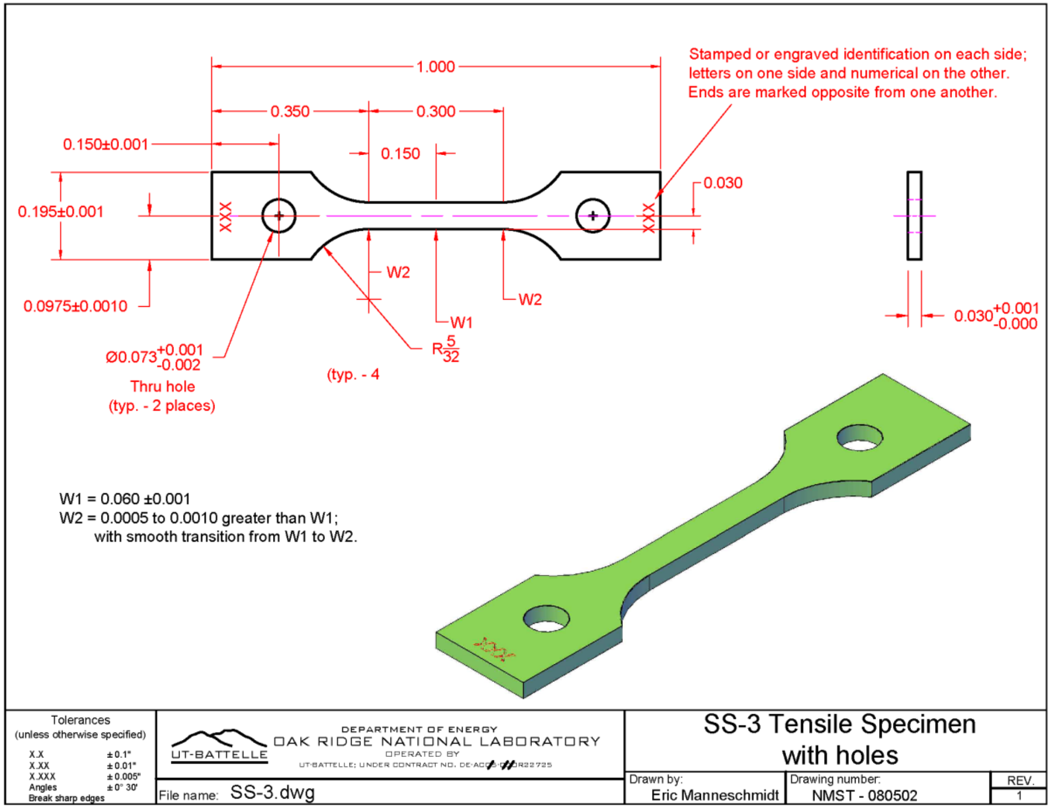
It was assumed the PM Mo would be brittle.

The PM Mo Target Materials Exhibited "Ductile" Behavior



Tests conducted by A. Marques and R. R. Lowden, ORNL

Tensile Properties Are Being Measured Using Sub-Scale "SS-3" Specimens

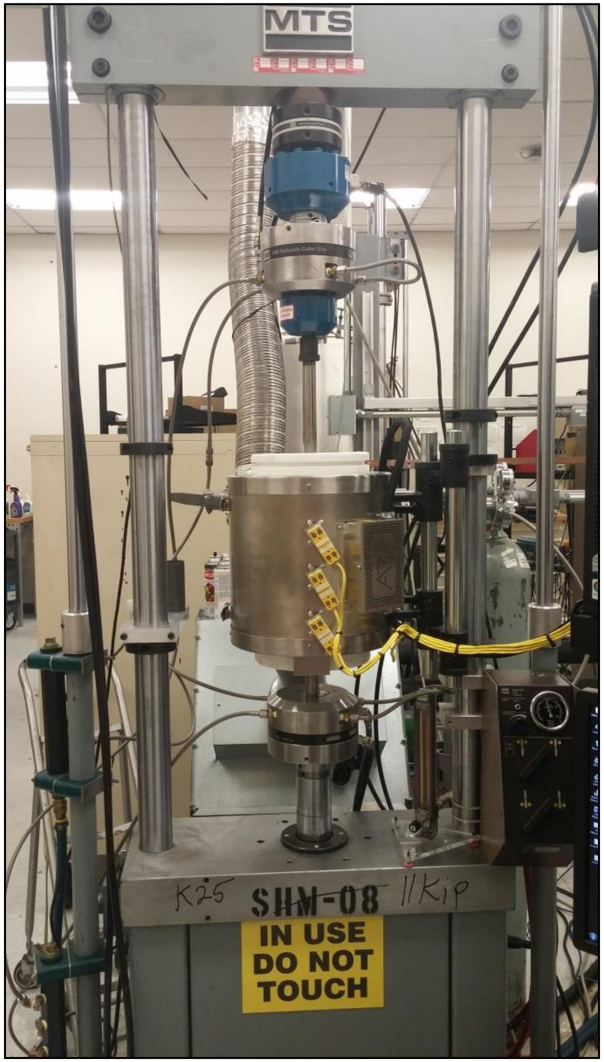


- Modulus (E)*
- Yield stress (YS)
- Ultimate stress (US)
- Total elongation (TE)
- Uniform elongation (UE)

* Due to small size of specimens, measured modulus (E) is not representative of true modulus.

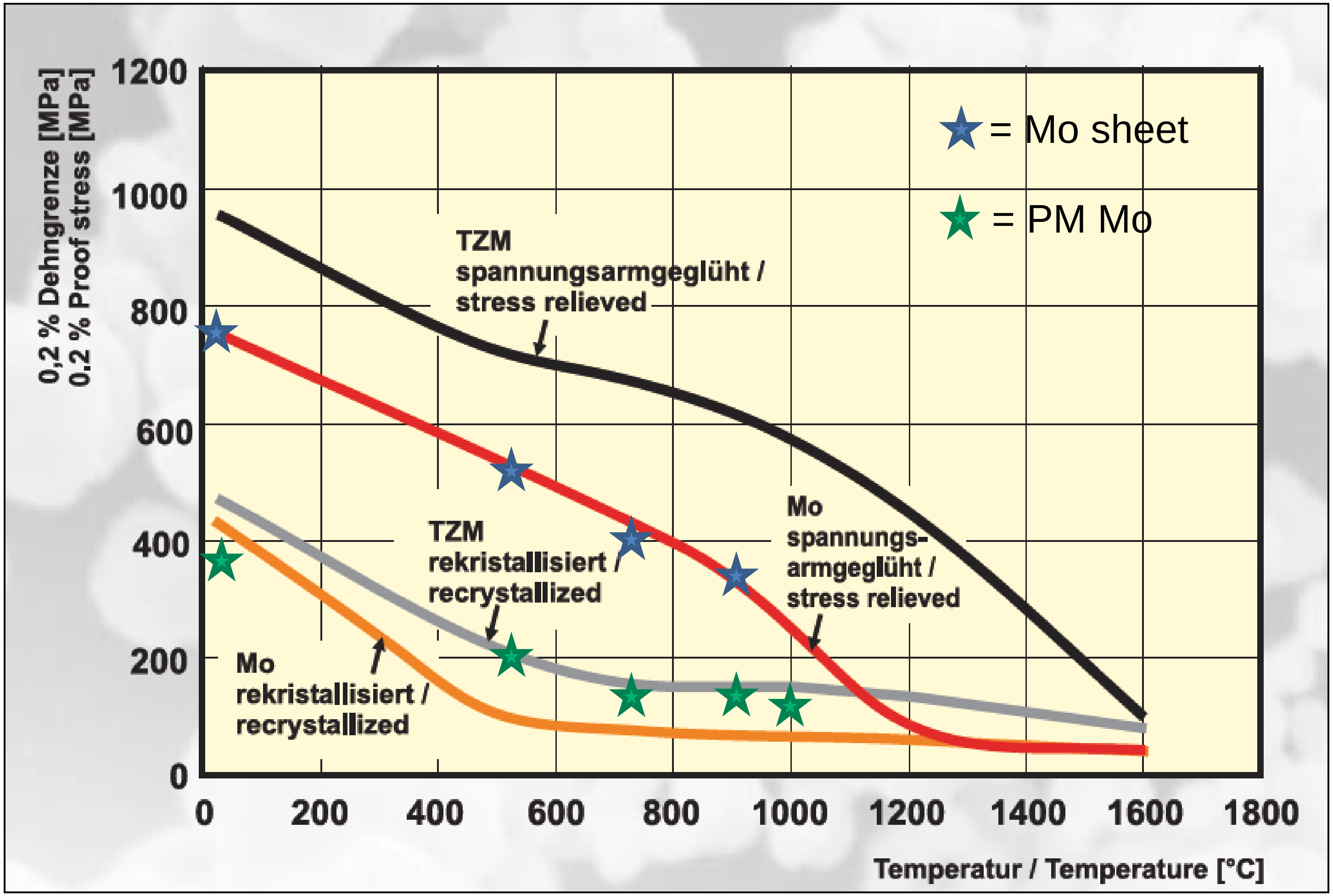
The small size of the SS-3 allows test specimens to be cut from target disks.

Tensile Properties Are Being Measured to 1000°C in an Inert Atmosphere



- 0.5 mm/min loading rate
- Load versus crosshead displacement (no extensometer)
- Test temperatures: room, 525°C, 725°C, 900°C and 1000°C
- > 20°C/min heating rate
- ~ 10°C/min cooling rate
- Flowing mixture of argon with 4% hydrogen

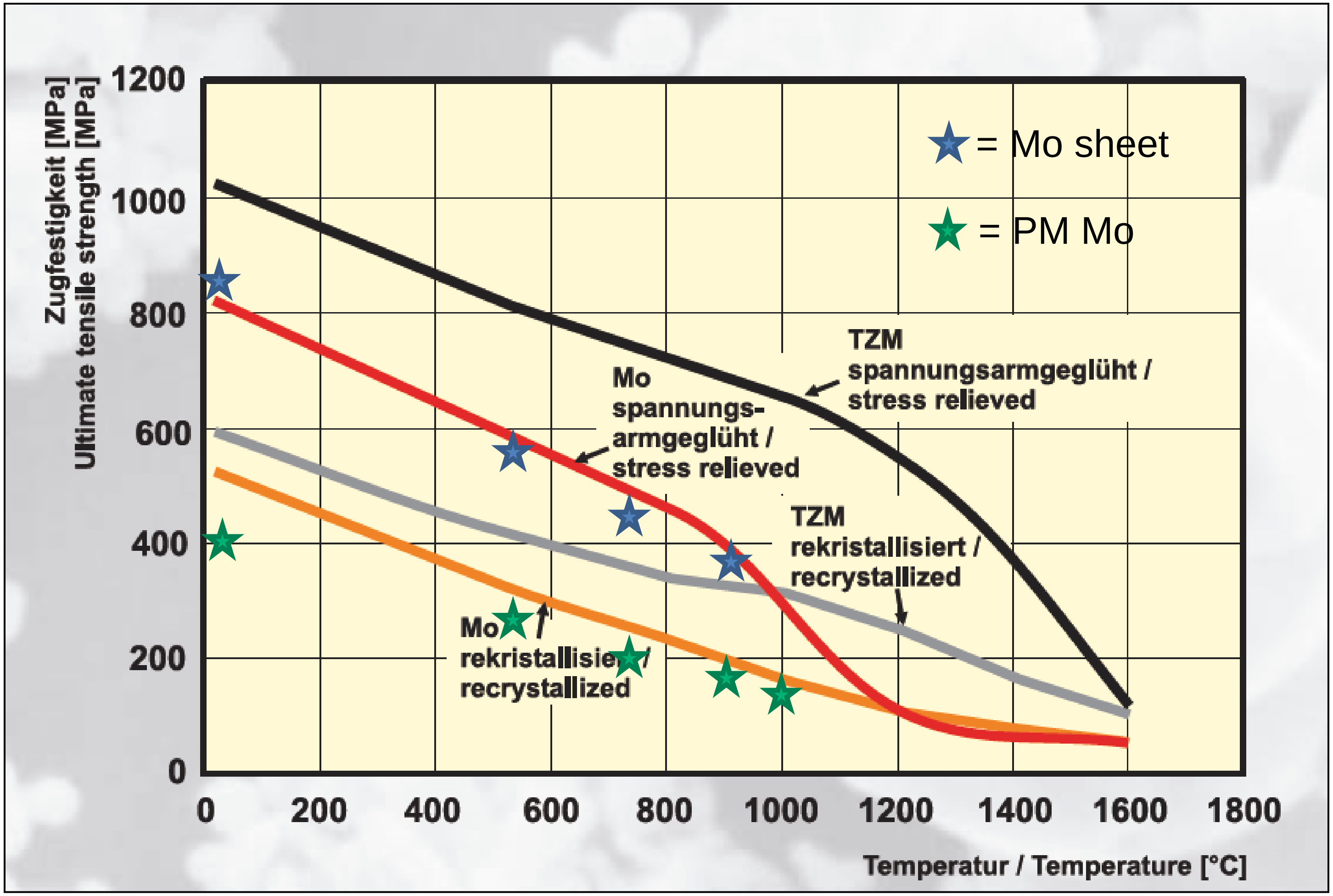
The PM Mo Target Material Behaves Much Like Recrystallized Sheet



0.2% Off-Set or Yield Stress (YS)

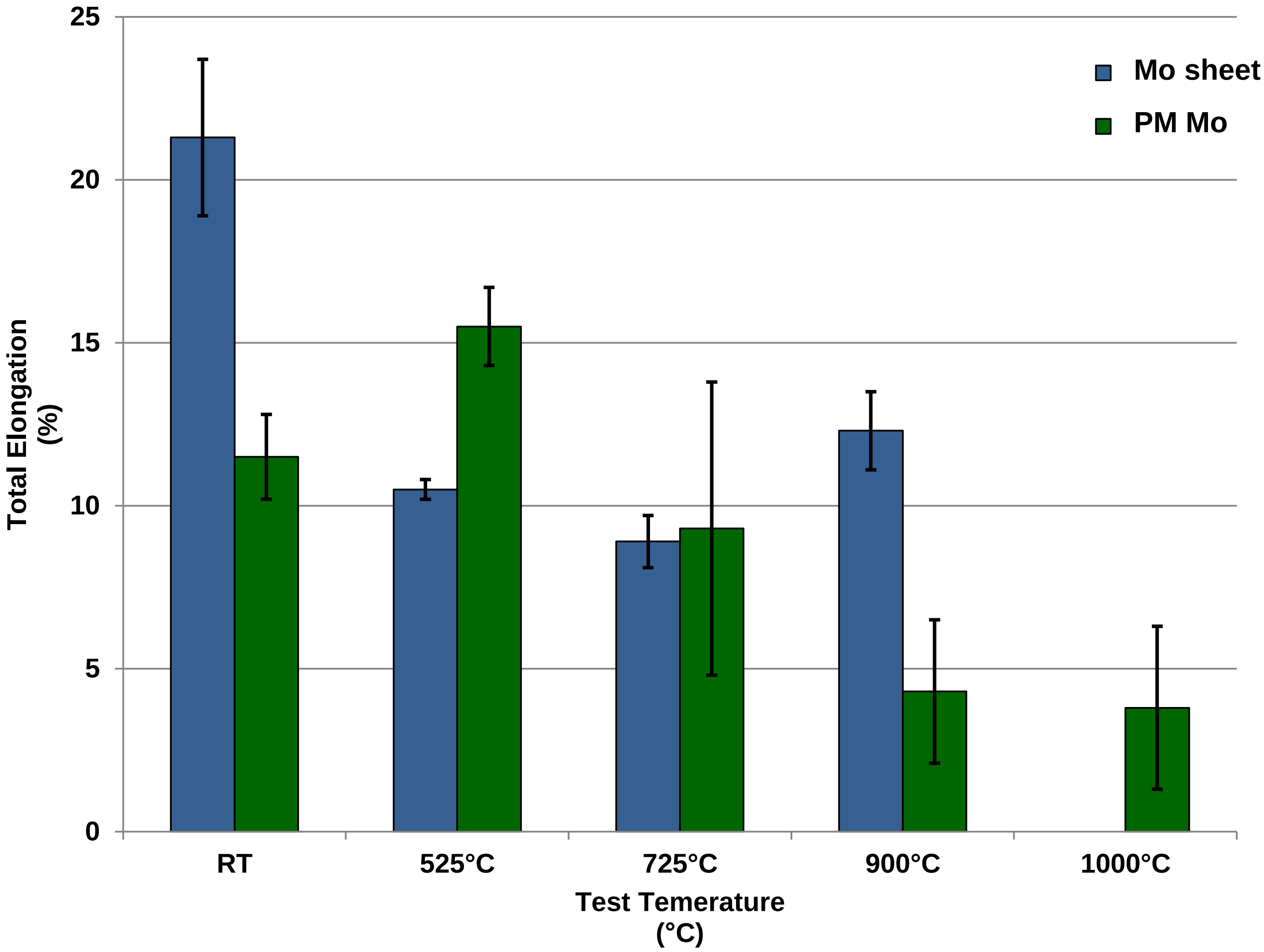
From Plansee, *Molybdenum: Material Properties and Applications*

Again, the PM Mo Target Material Behaves Much Like Recrystallized Sheet



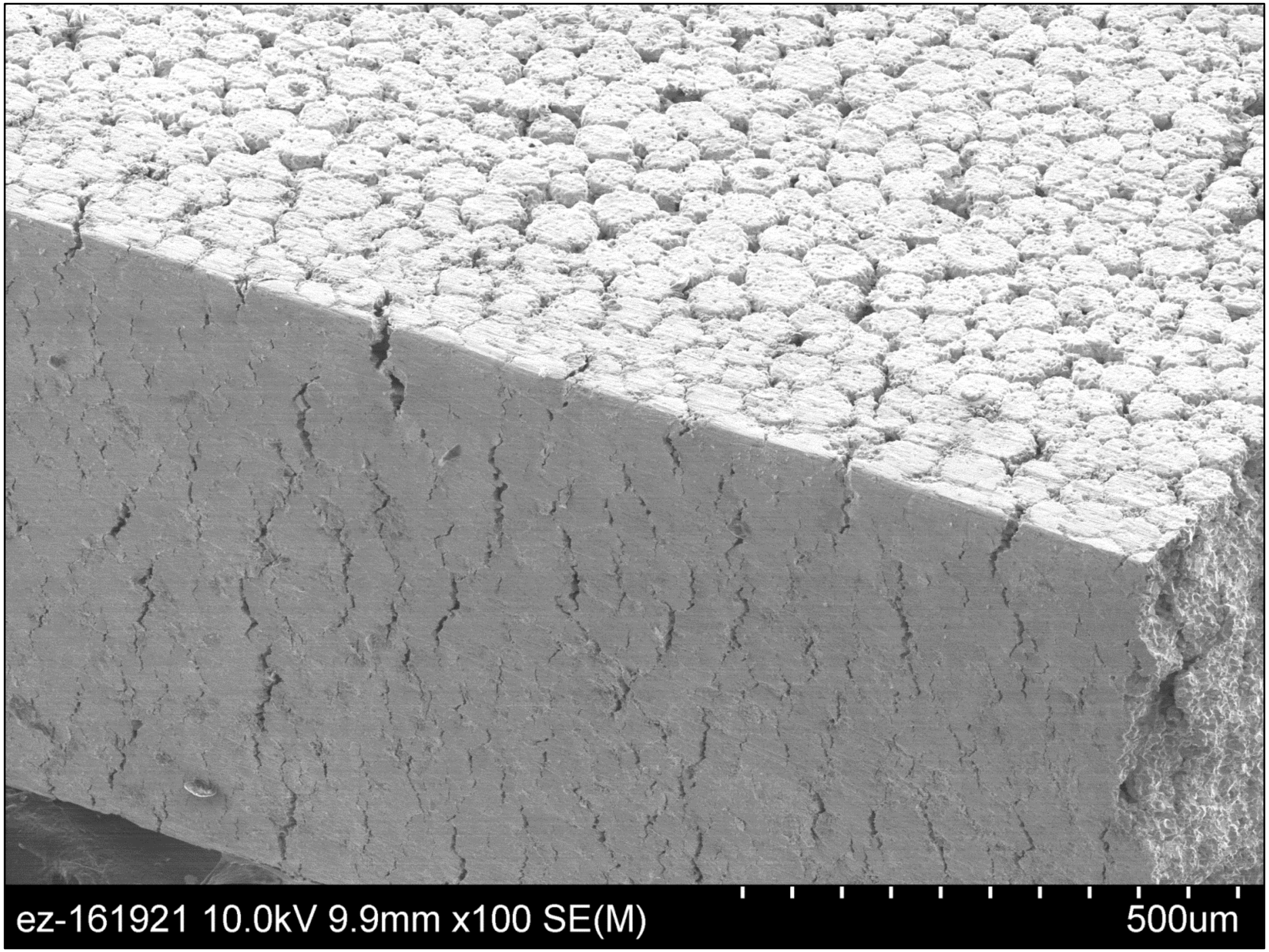
Ultimate Stress (US)

Total Elongation* Is Used to Asses “Ductility”

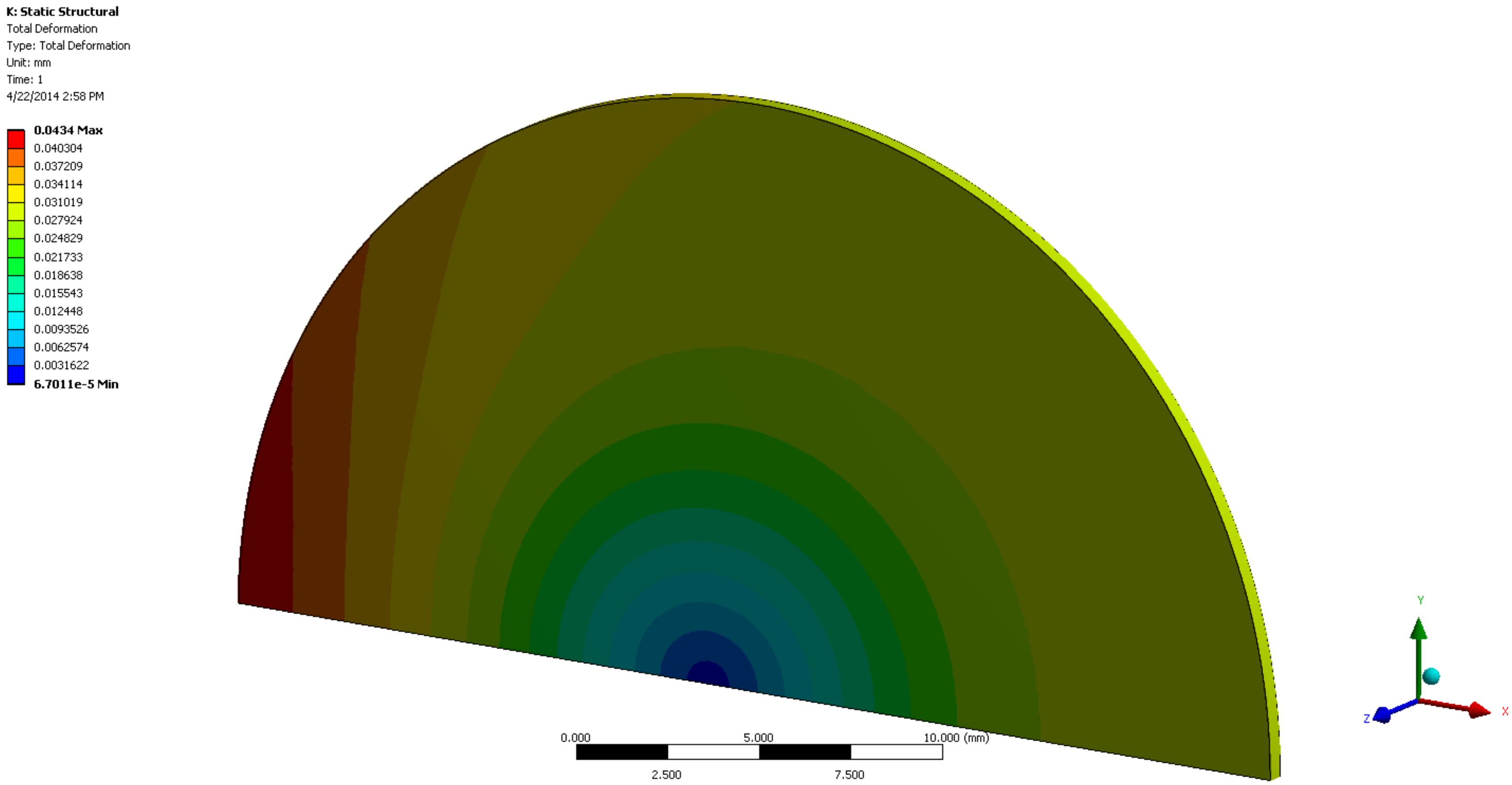


**** A measure of how much the specimen stretches before it breaks.***

The PM Mo Does Not Exhibit Traditional Ductile Behavior – No Necking

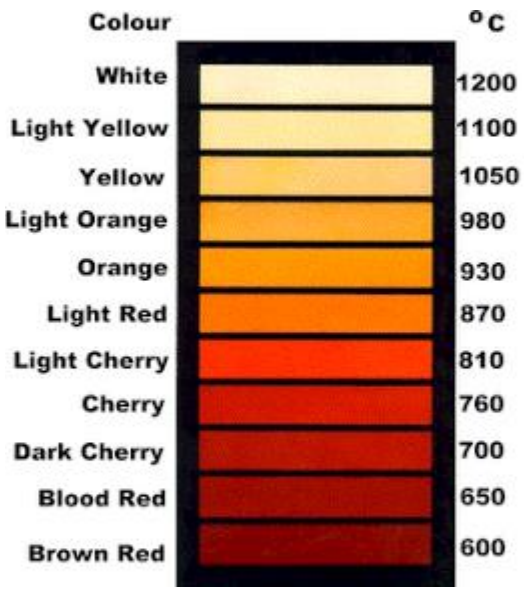


Thermomechanical Models Predict Disk Distortion Due to the Thermal Gradients



Disk #14

The Center of Target Disks Were Heated in an Inert Atmosphere Using a 350W Laser



Observed color was used to determine temperature. Disks achieved temperatures of ~ 950° to 1050°C.

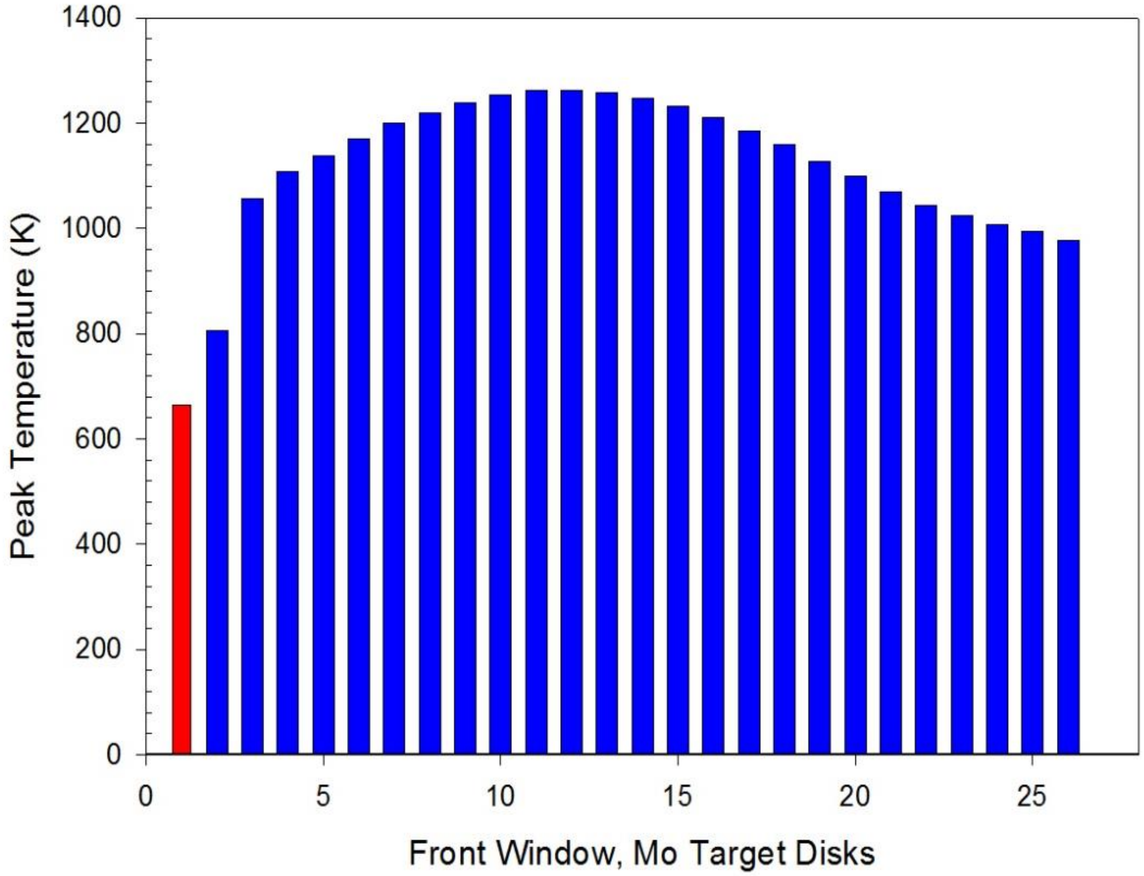
All Disks Were Distorted After Heating

Specimen	Power (W)	Pulse (msec)	Frequency (Hz)	Time (sec)	Δ (μm)
SM-1	92	0.5	100	30	+18
SM-2	120	0.5	100	30	+70
SM-3	200	1	50	60	+60
PMA-28	120	0.5	100	30	+6
PMA-29	193	1	50	30	+10
PMA-30	193	1	50	60	+16
PMA5-22	200	1	50	75	+17
PMA-26	200	1	50	150	+20
PMA5-21	250	1.5	50	30	+40

Δ = increase in bow, SM = sheet metal

Target Materials Were Heat Treated to Simulate "Long-Term" Exposure in an Accelerator

Center temperature of disks, typ.



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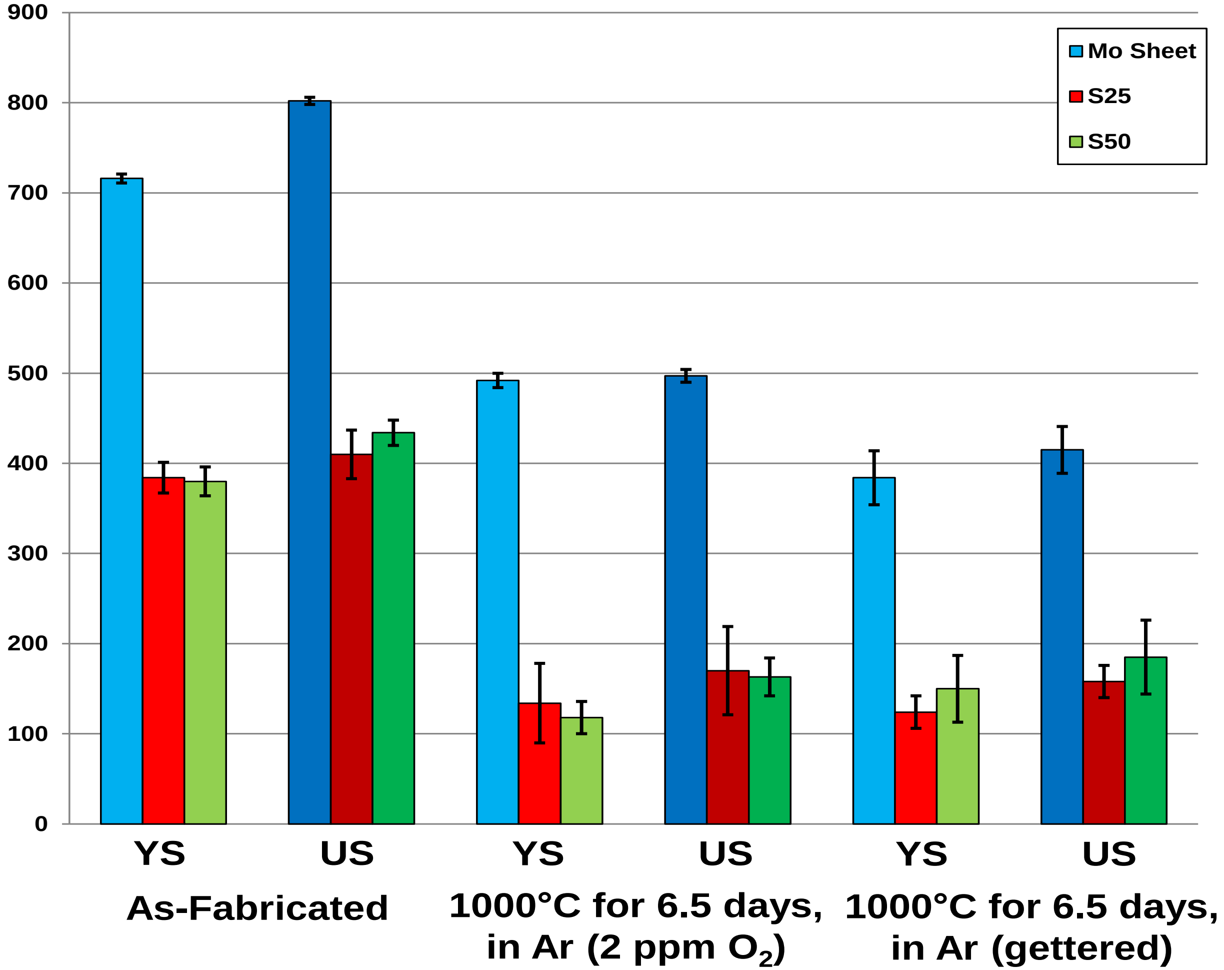
6.5 days (156 h) at 1000°C in flowing argon

Room Temperature Properties Were Degraded by the Heat Treatment



- **Mo sheet**
  - MO361, 99.95% Mo, as-rolled, 0.65 mm thick
- **S25**
  - Starck SD with 0.25 wt% EBS, 88%TD and 7% open porosity
- **S50**
  - Starck SD with 0.5 wt% EBS, 90%TD and 5% open porosity

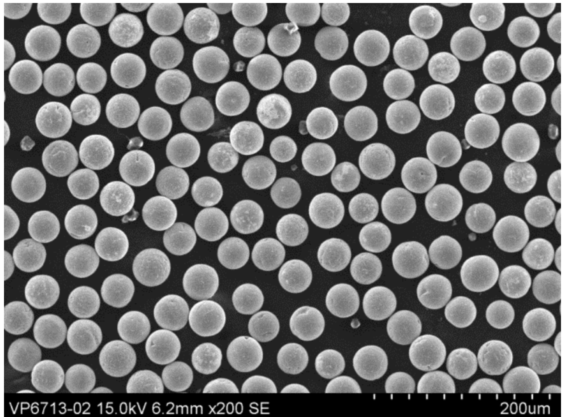
ALL PM materials exhibited brittle behavior with TE < 1%.

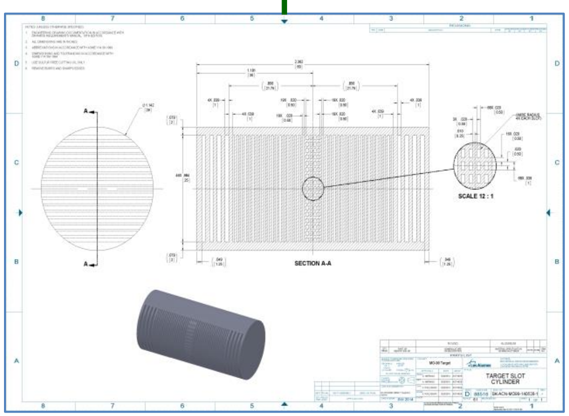
The Powder Metallurgy Fabrication of Target Disks for the Accelerator Production of Mo-99 Continues to Progress

- Powder metallurgy can be used to produce accelerator target disks with minimal waste.
- All molybdenum powders can be pressed and sintered to densities greater than or equal to 90% of theoretical; however, a combination of lubricated, spray-dried powders and good tooling has been used to produce the most uniform disks.
- Thermophysical and thermomechanical properties of the powder metallurgy Mo materials have been measured. Room and elevated temperature mechanical properties are similar to recrystallized Mo sheet.
- ALL disks bowed and/or warped upon spot heating with a laser.
- The mechanical properties of the PM Mo are degraded by long-term exposure to accelerator conditions.
- Sub-scale, 12 mm x 1 mm disks are to be included in an accelerator test to evaluate in-process performance.

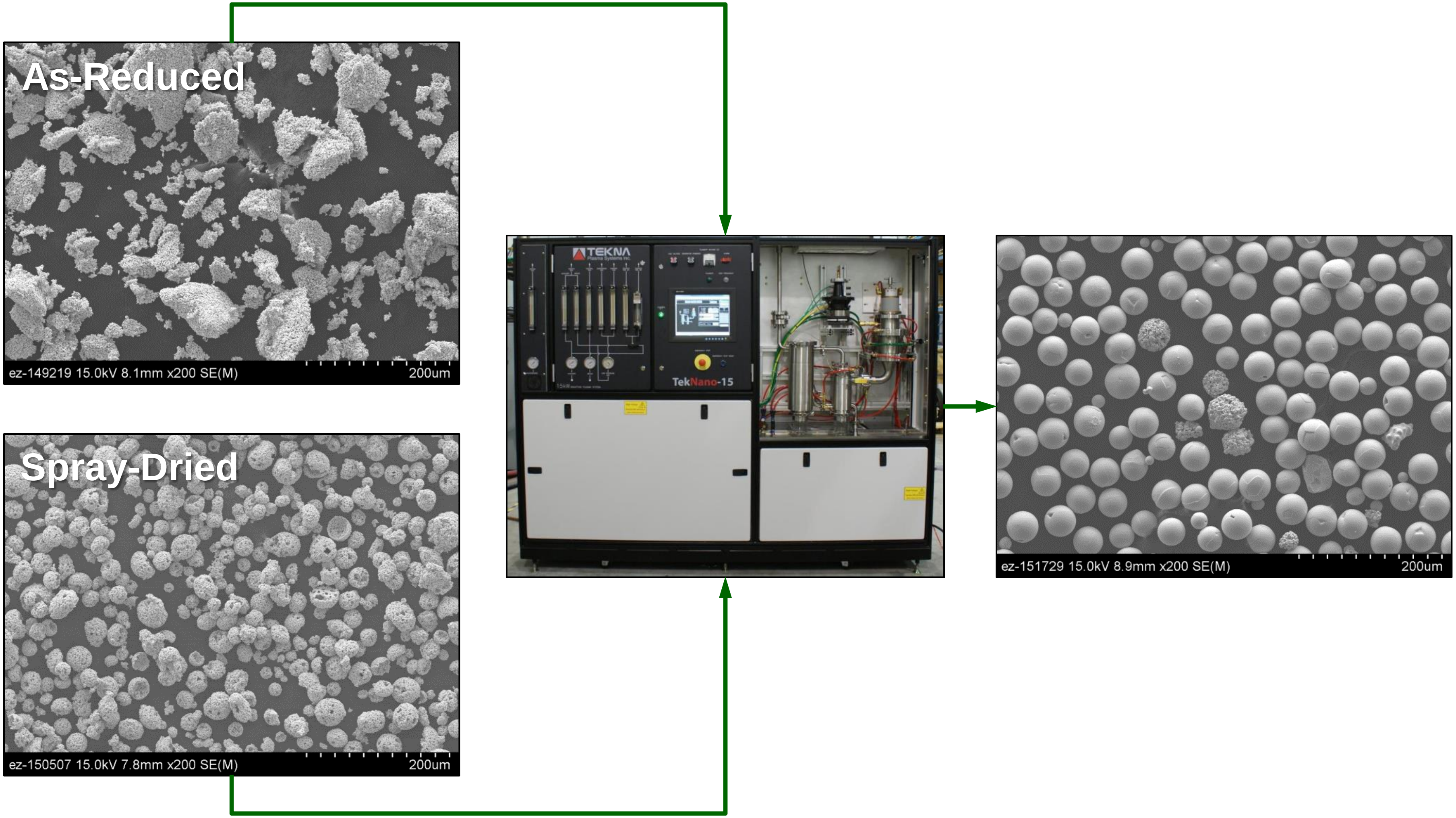
What's next?

Target Assemblies Are To Be Fabricated Employing 3D Selective Laser Sintering

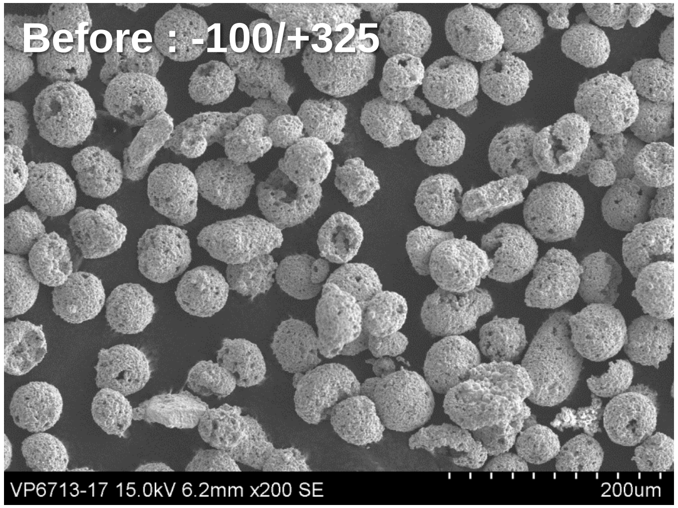


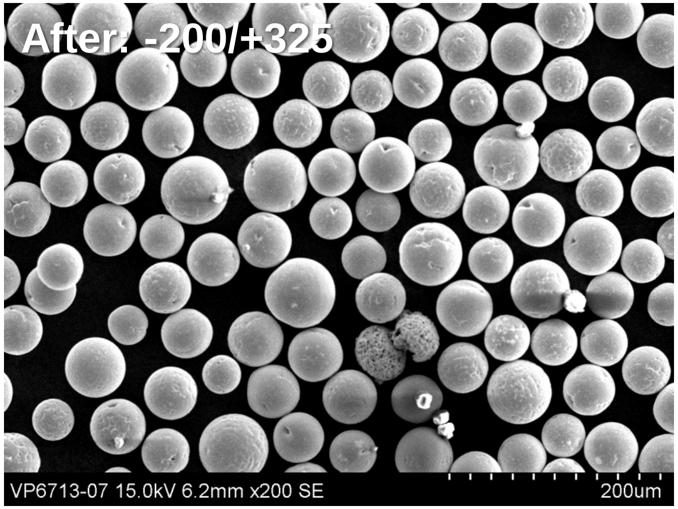


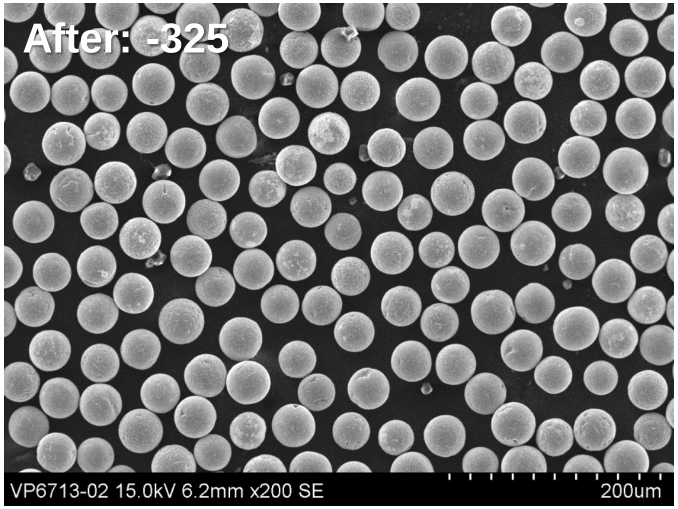
As-Reduced And Spray-Dried Powders Are Being Spheroidized for Use in Printing Targets



Who Says, “You Can’t Polish a Horse Turd?”







Any Questions or Comments?