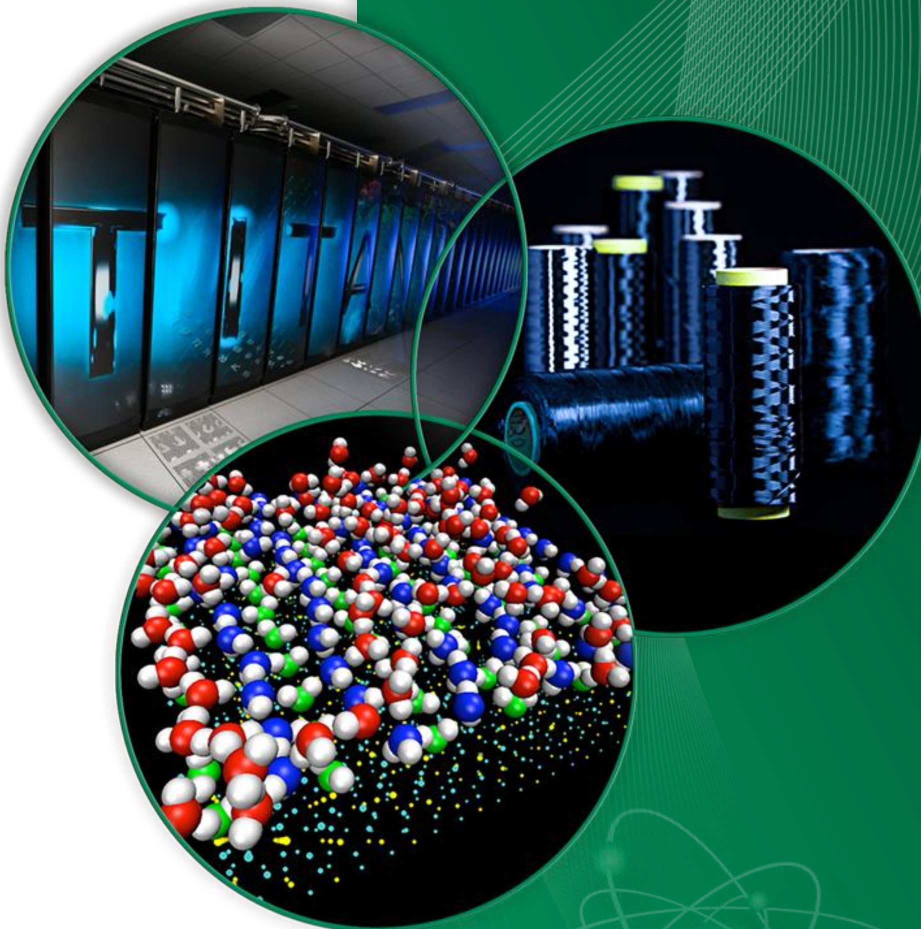
Powder Metallurgy Fabrication of Thin, Flat, Molybdenum Disks



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Mo-99 Topical Meeting
Washington, DC
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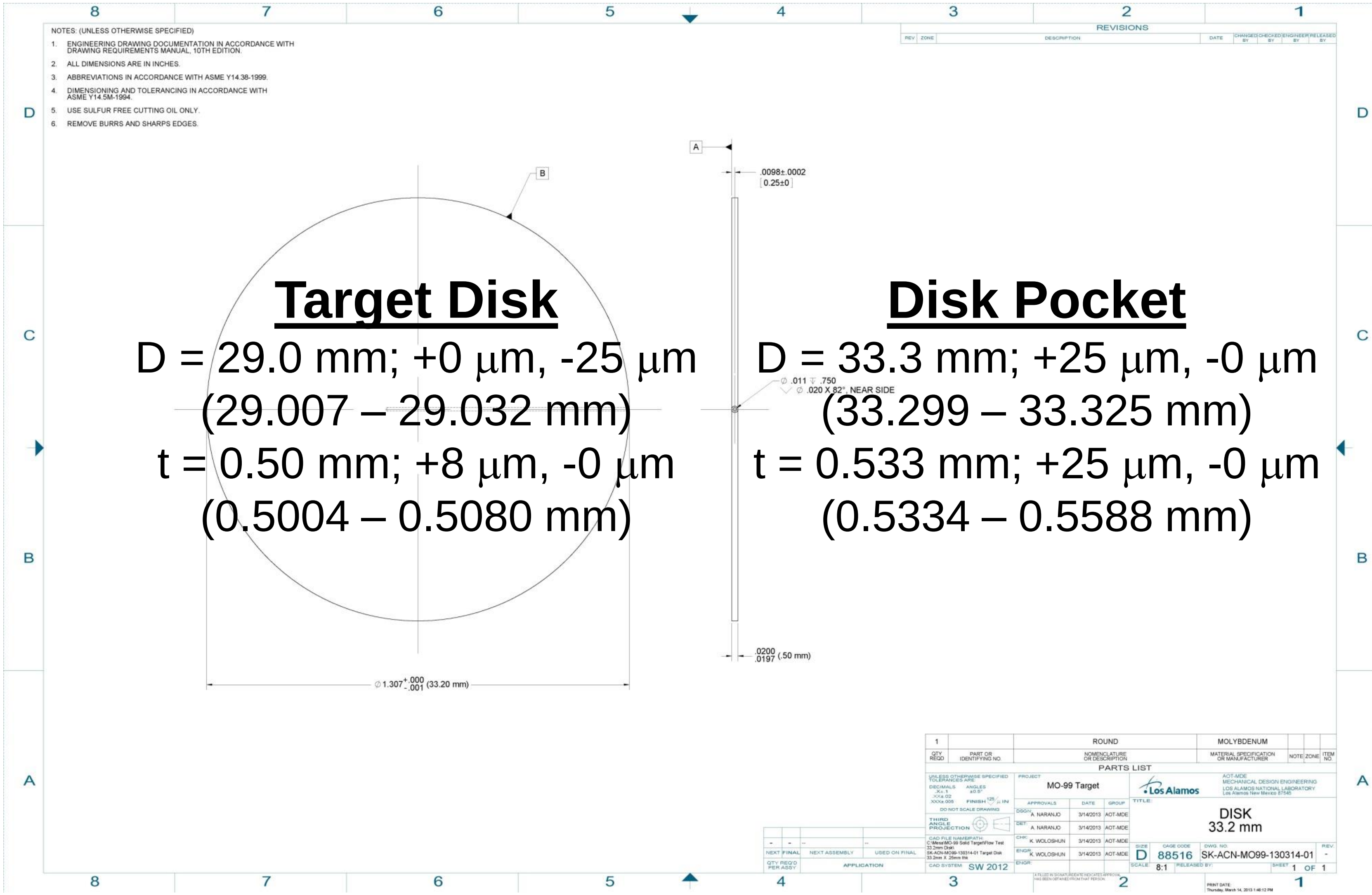
Target Disks for the Accelerator Production of Mo-99 Are Being Fabricated Employing Powder Metallurgy Approaches

The goals of this effort:

- Understand the requirements for 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 and subsequently control characteristics that affect the dissolution rate of target disks.
- Assist in developing a process for recycle and re-use of isotopically-enriched molybdenum.

Target Disk and Holder Specifications Are Quite Stringent



Target Disk

D = 29.0 mm; +0 μm, -25 μm

(29.007 – 29.032 mm)

t = 0.50 mm; +8 μm, -0 μm

(0.5004 – 0.5080 mm)

Disk Pocket

D = 33.3 mm; +25 μm, -0 μm

(33.299 – 33.325 mm)

t = 0.533 mm; +25 μm, -0 μm

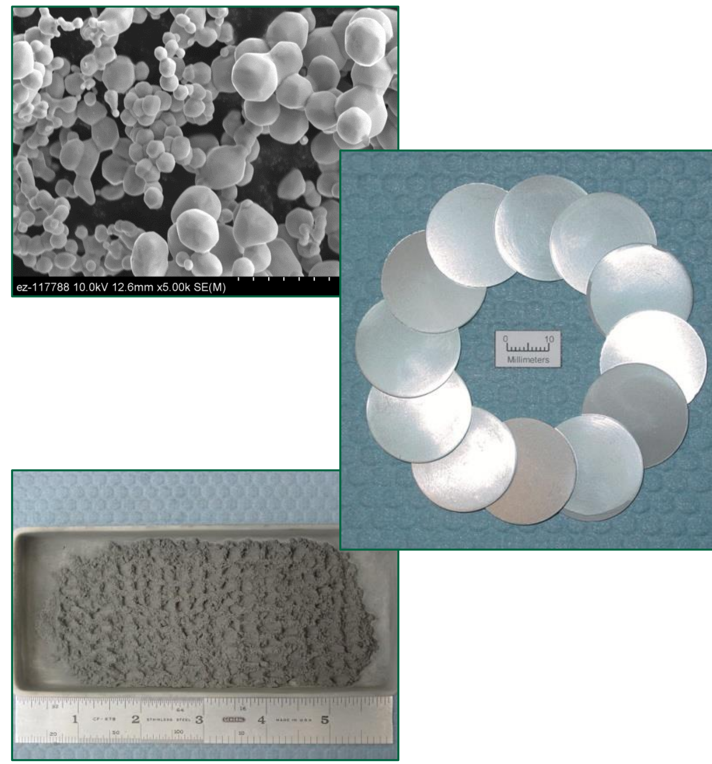
(0.5334 – 0.5588 mm)

Thin, Disk-Shaped, Molybdenum Parts Are Usually Punched From Sheet and Ground and/or Lapped to Final Dimensions

Because of the high cost of isotopically-enriched powder, scrap and waste must be eliminated from the process thus the typical approaches are not viable.

Powder Metallurgy (P/M) 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 P/M Production Approaches Are Not Designed to Produce High-Precision Parts

Pressed and Sintered Tolerances

	Practical		Possible	
	mm	(in.)	mm	(in.)
Length (+/-)	0.190	.0075	0.130	.005
Inside diameter (+/-)	0.100	.004	0.050	.002
Outside Diameter (+/-)	0.100	.004	0.050	.002
Concentricity	0.150	.006	0.100	.004
Flatness on ends	0.130	.005	0.100	.004
Parallelism of ends	0.130	.005	0.100	.004

P/M is Typically Not Just Pressing & Sintering

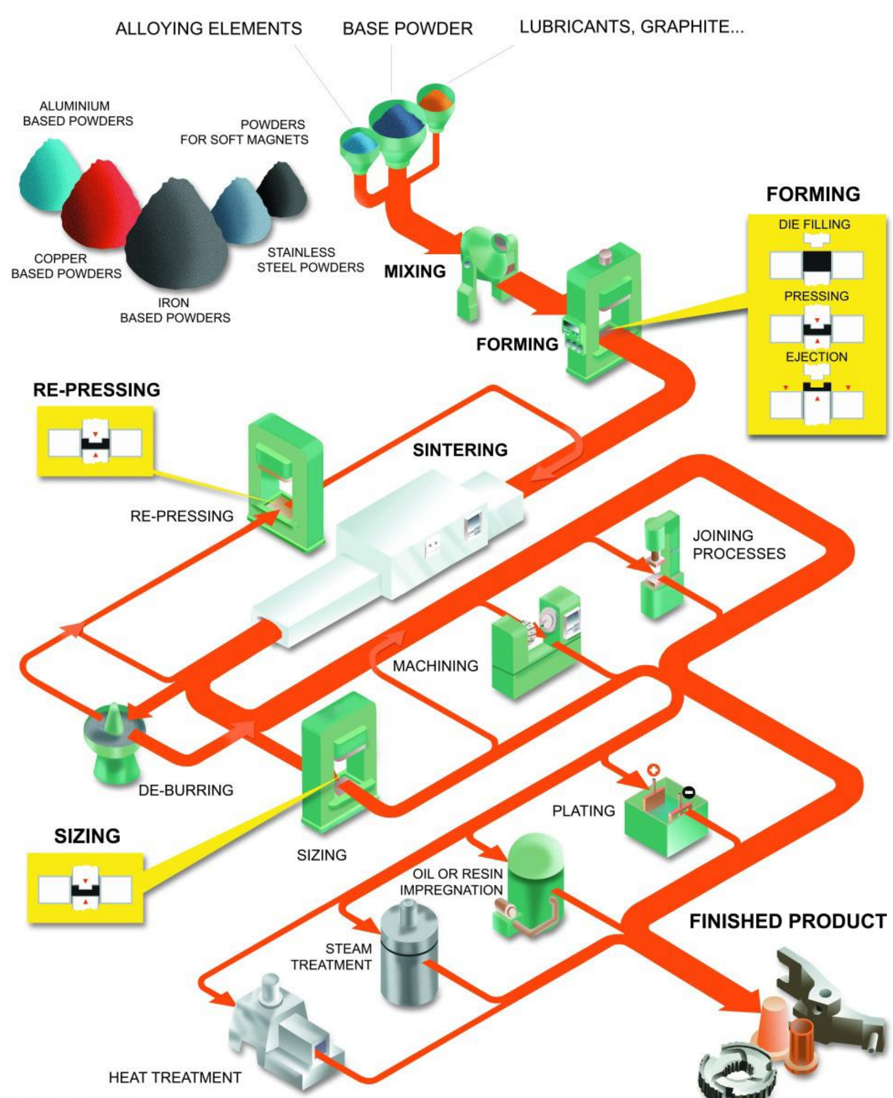
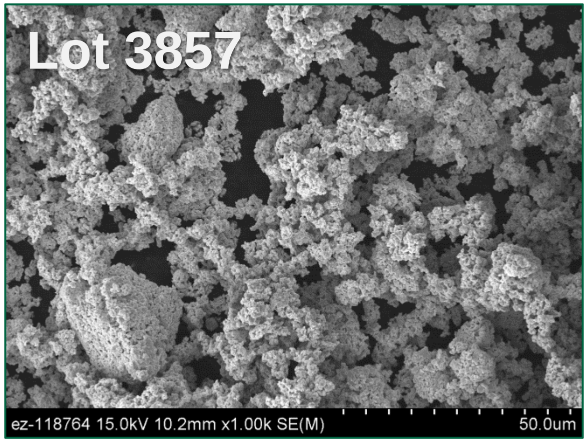
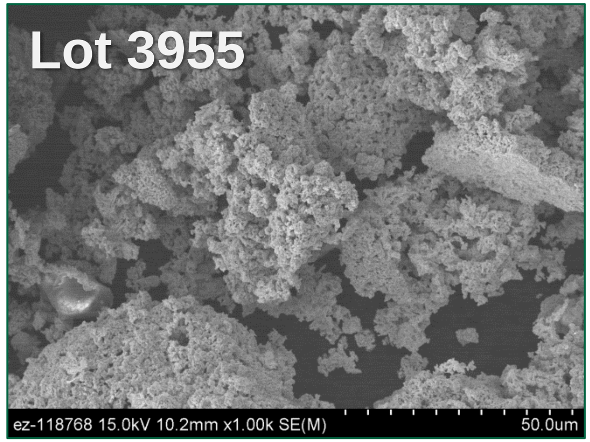
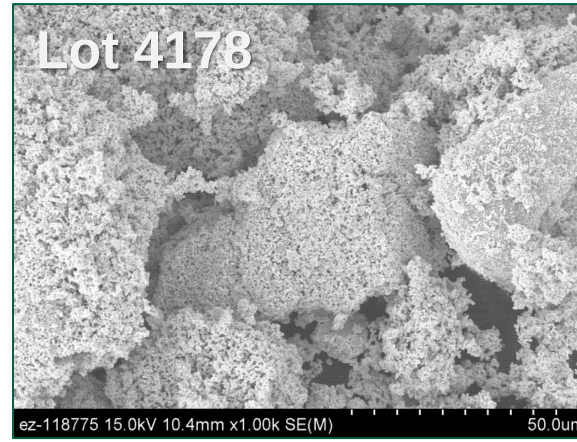


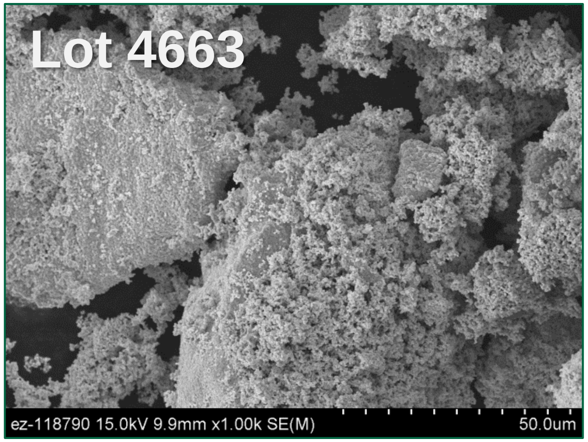
Chart courtesy of EPMA

P/M Processing Begins With Characterization of the Feedstock Material AKA Powder Metal



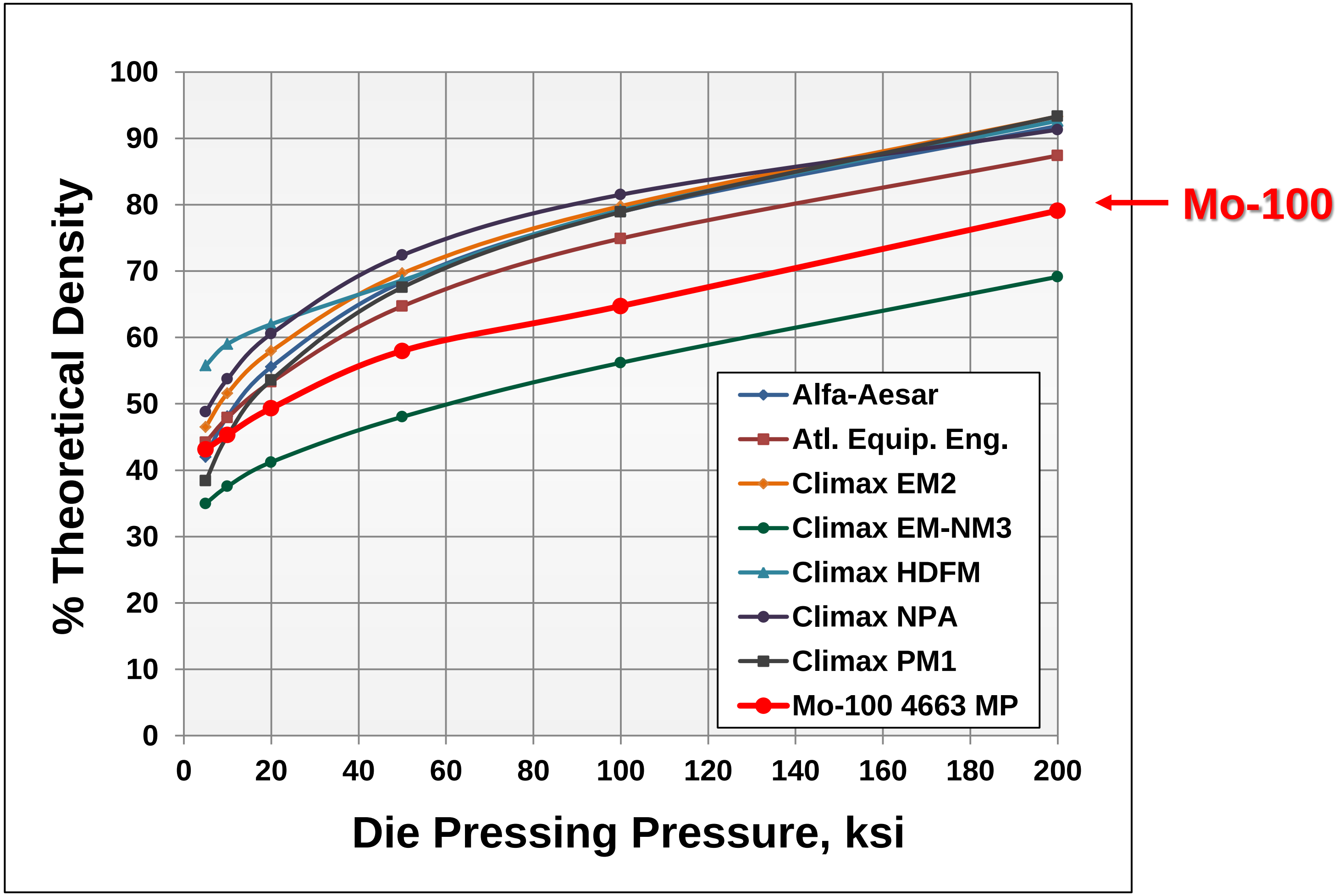




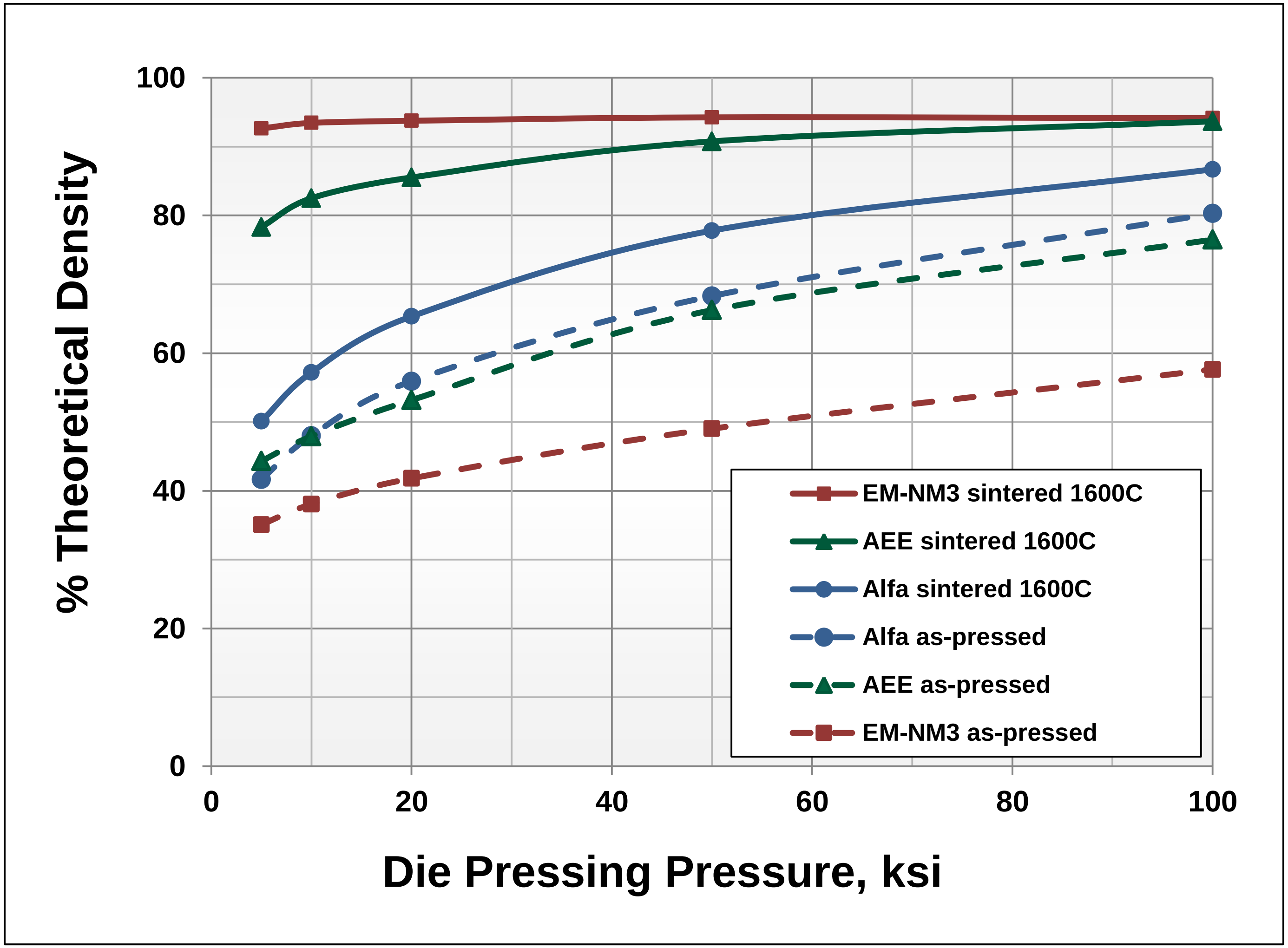


Different Lots of Mo-100 Powder Possessed Uniform Primary Particle Sizes But Varying Degrees of Agglomeration

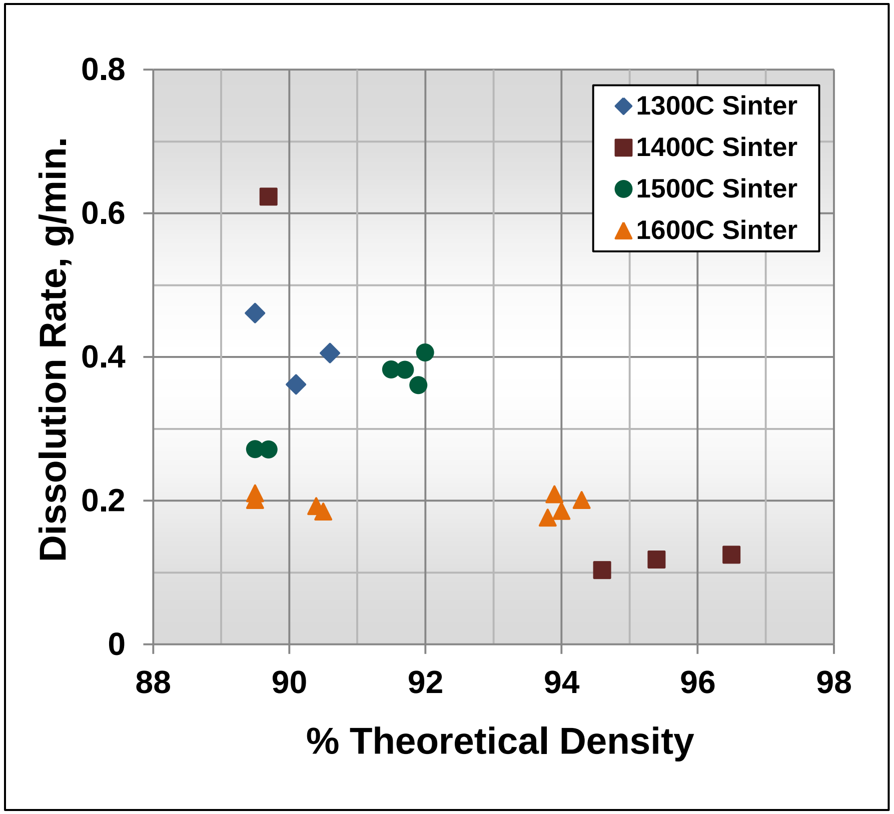
Powder Particle Characteristics Affect Compaction and Green Density

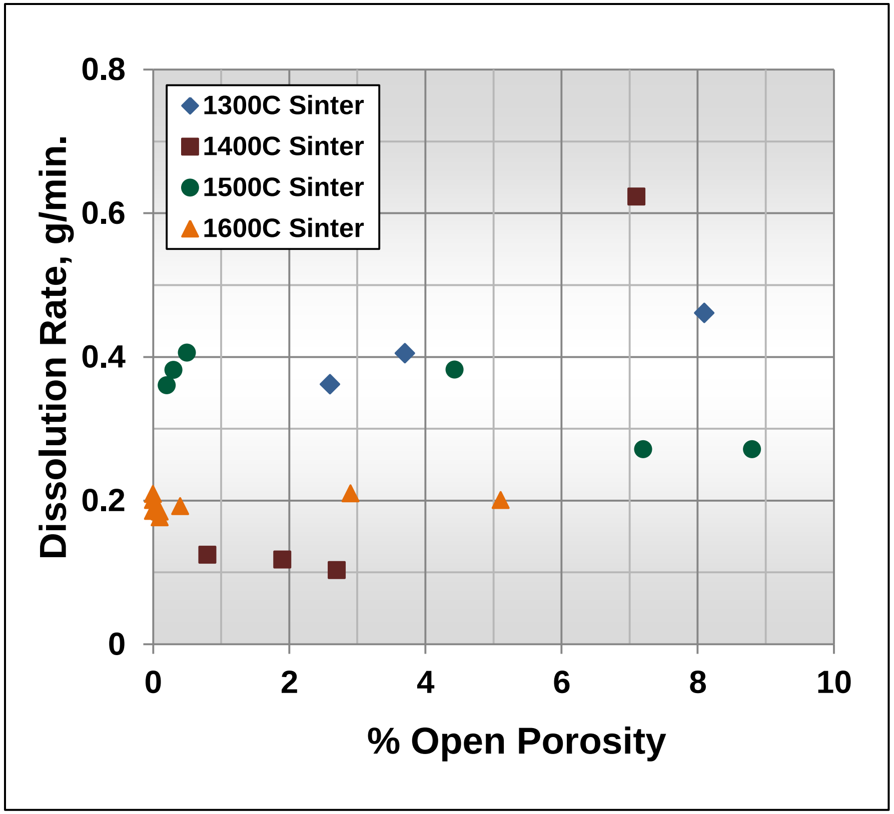


Powder Particle Characteristics Also Influence Sintering Behavior



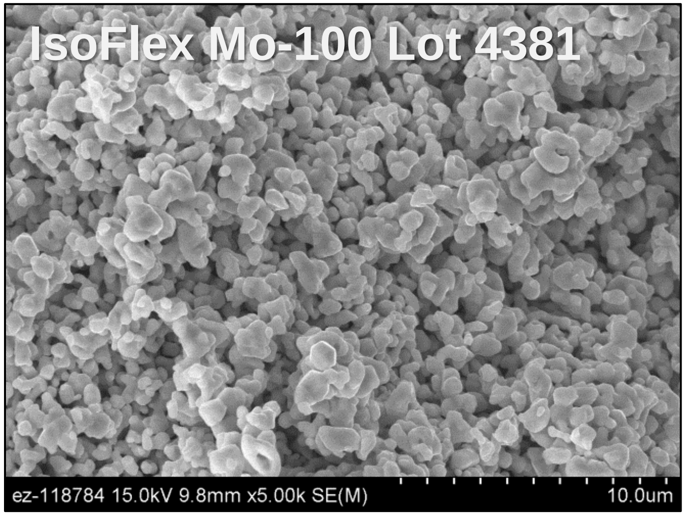
Factors That Affect Dissolution Rate Are Being Evaluated

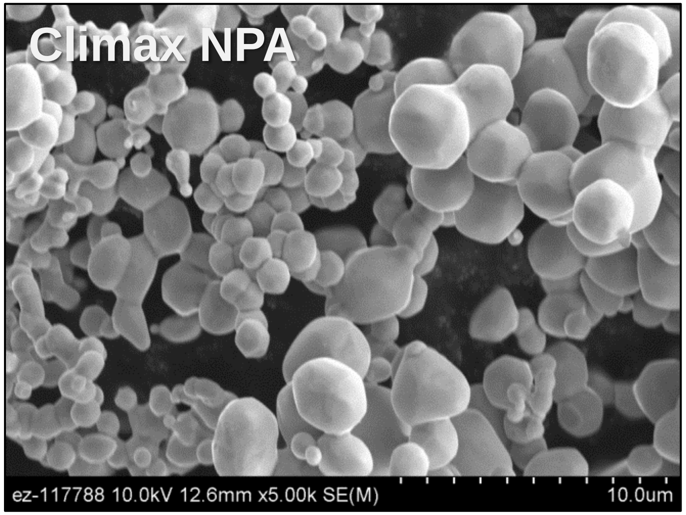


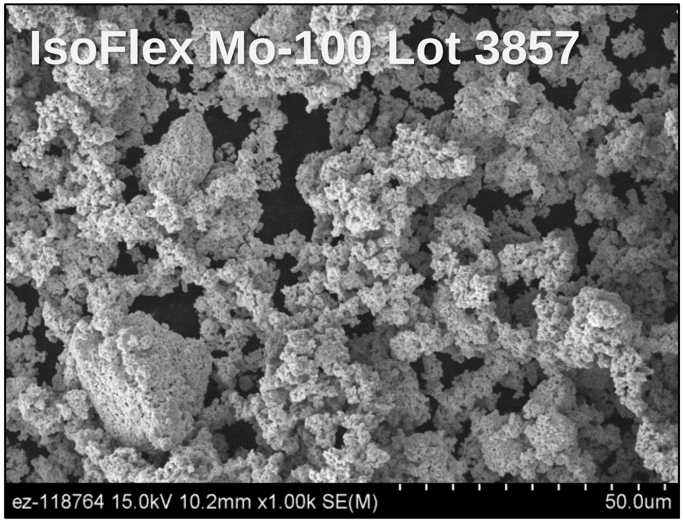


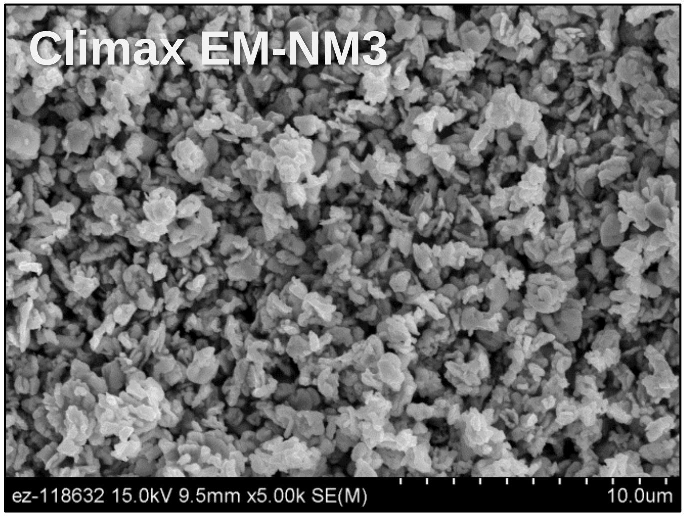
In general, samples with lower sintered densities and open porosity exhibited higher dissolution rates.

Enriched Powders Were Found to Have Characteristics In-Between Commercially-Available “Natural” Materials





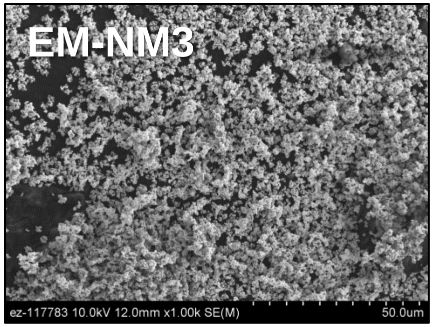


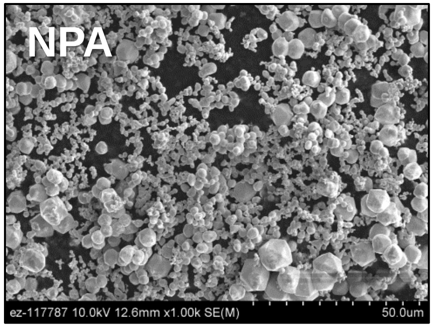


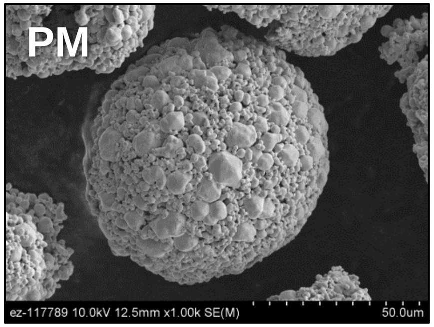
The Processing Behavior of Commercial and Recovered Powders Is Being Evaluated

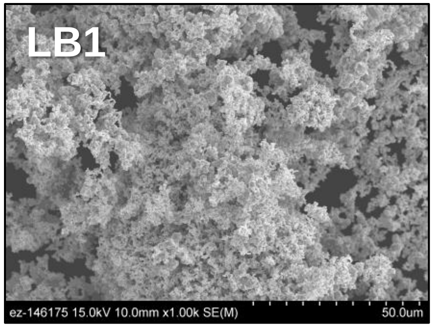
Molybdenum Supplier	Grade	Purity (% Mo)	Max. Oxygen (ppm)	Particle Size	BET (m^2/g)	Hall Flow (sec/50 g)
Climax Molybdenum	EM-NM3	99.9	1400	0.7 – 1.5 μm	2.83	No flow
Climax Molybdenum	NPA	99.95	1000	4.0 – 4.8 μm	0.45	No flow
Climax Molybdenum	PM	99.9	2000	-200/+325 mesh (spray-dried)	NM	< 45
Large-batch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

NM = not measured









Most If Not All Molybdenum Powders Can be Pressed and Sintered to 90% Density

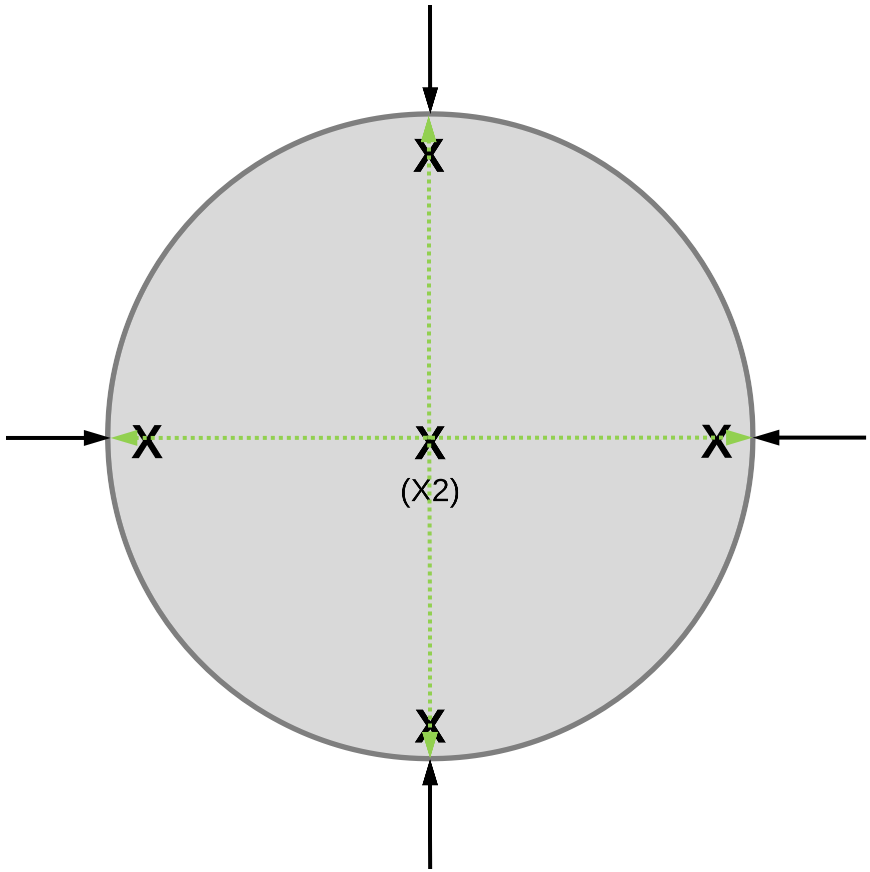
Powder	Compact Press. (ksi)	%TD (green)	Sintering (°C/h)	%TD (sintered)	Open Porosity	Diameter (mm)	Thickness (mm)	Shrinkage (%)		Cupping*
								D	t	
33 mm disks										
NPA	100	80	1500/1	90	NM	32.0	0.59	3.9	1.7	mixed
NPA	100	80	1550/1	91	NM	NM	NM	NM	NM	severe
NPA	150	85	1500/1	92.5	NM	32.5	0.56	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

* Severity of distortion, primarily cupping, was ranked using the ratio of the height of the cup minus the thickness of the disk at the peak height of the cup divided by the same thickness measurement with "**minimal**" being < 0.5, "**moderate**" between 0.5 and 1.0, and "**severe**" > 1.0.

However, the Sintered Disks Were Distorted (Cupped) and Not Uniform in Thickness

Detailed dimensional characterization was conducted to better understand the variations.

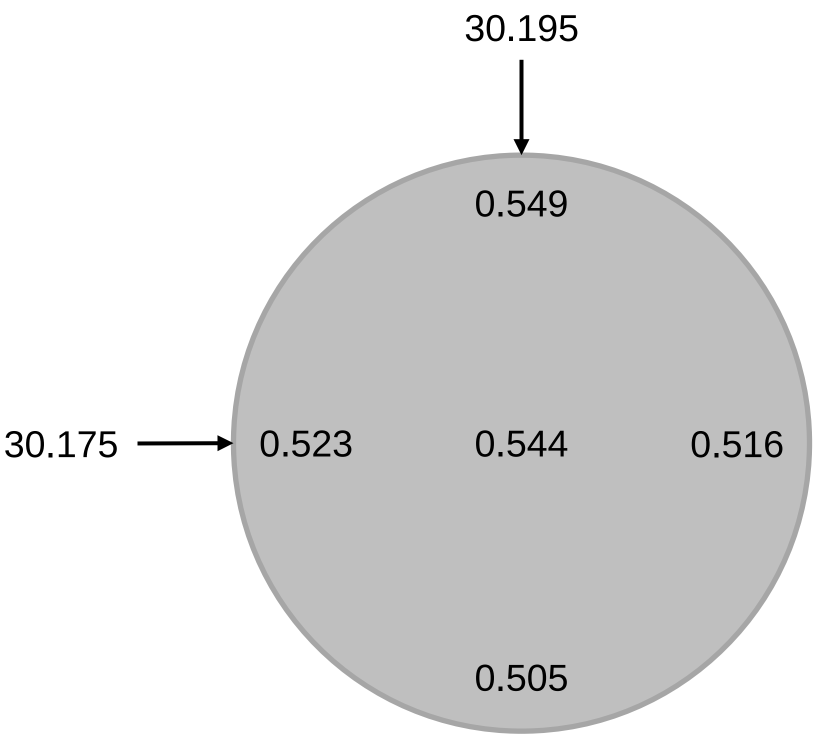
- Digital micrometer measurements at various positions
- Optical comparator measurement of diameter at various locations
- Coordinate measurement system analysis of diameter and thickness
- X-ray radiography



Climax NPA Performed Well in Earlier Trials

As-Pressed

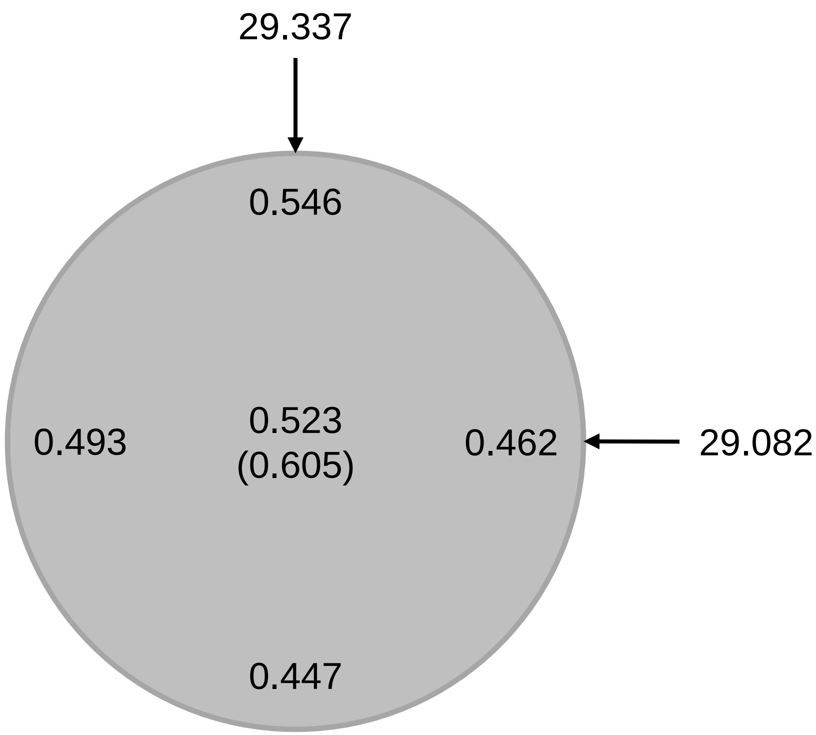
(100 ksi)



3.0314 g
78.8% TD

Sintered

(1500°C, 1 h Ar-7%H_2)

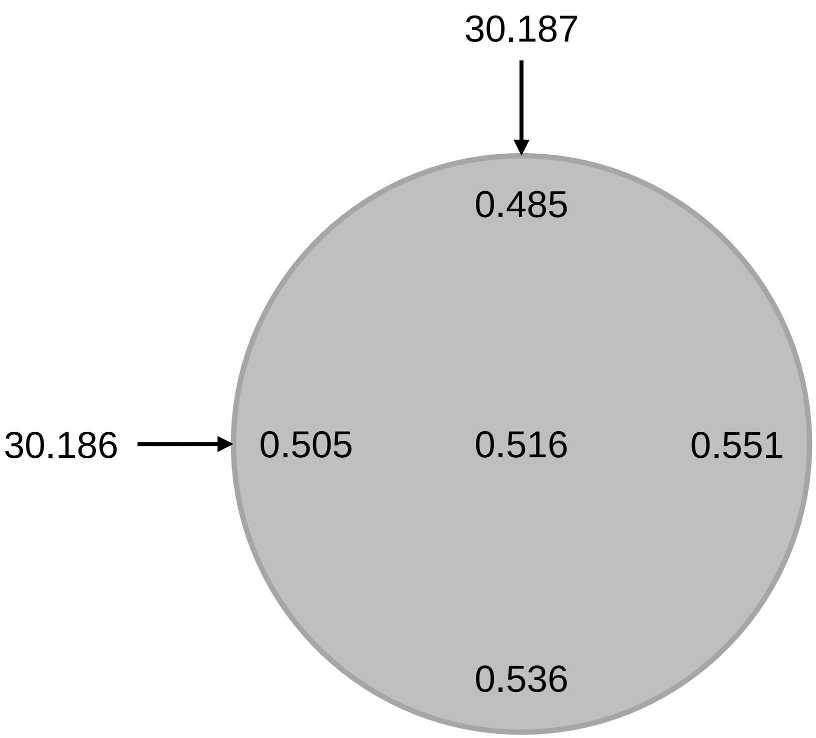


3.0256 g
89.0% TD
9.3%
open porosity

Most NPA Specimens Were Distorted and Not Uniform in Thickness

As-Pressed

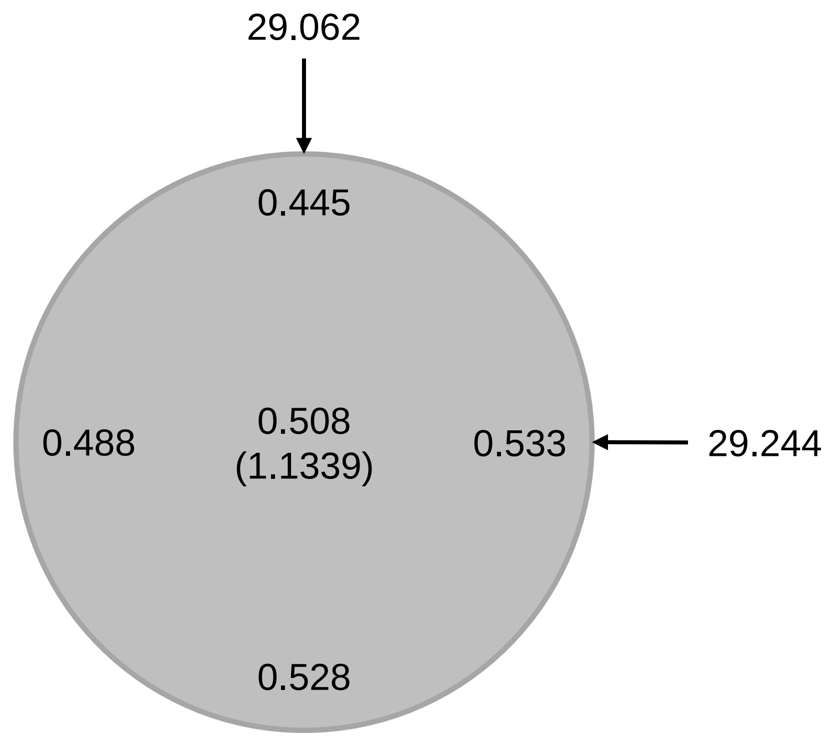
(100 ksi)



3.0261 g
79.9% TD

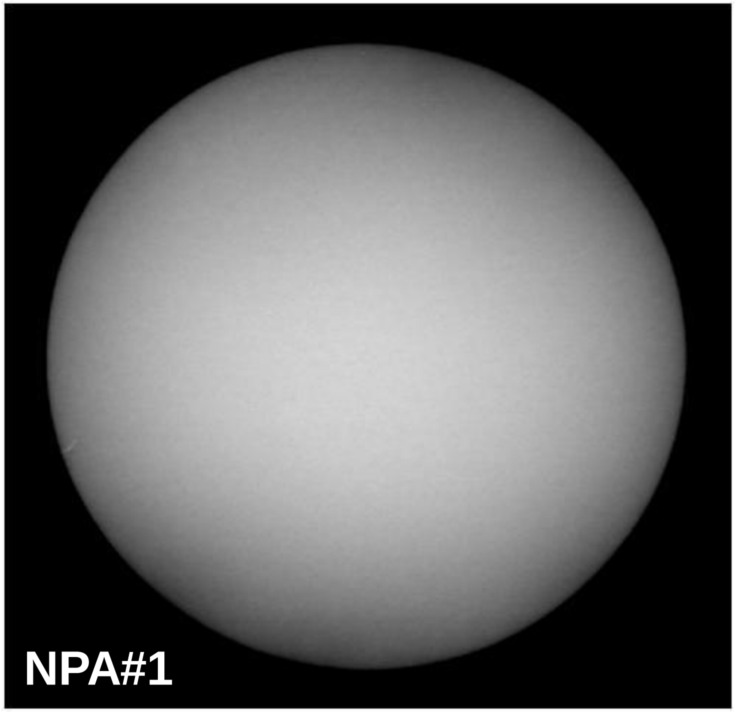
Sintered

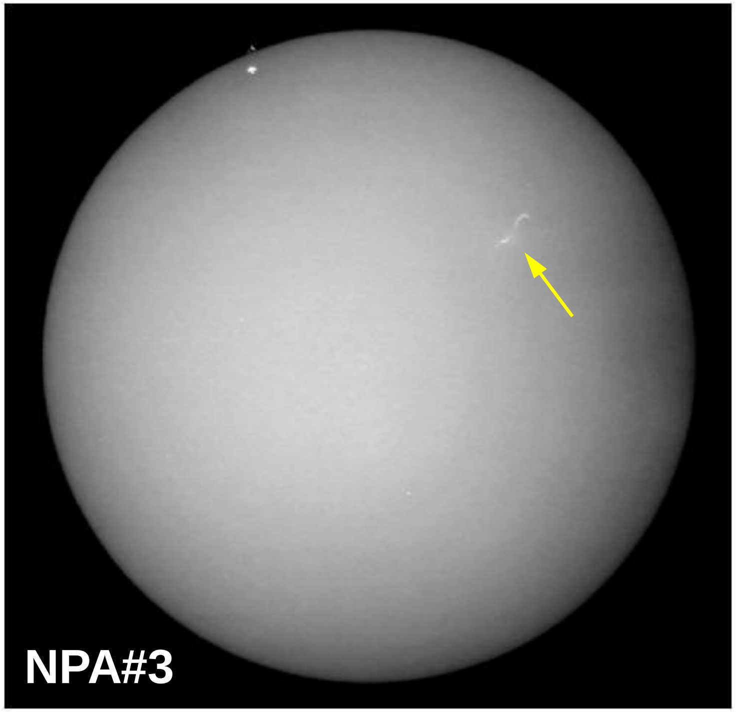
(1500°C, 1 h Ar-7%H_2)



3.0214 g
88.1% TD
10.7%
open porosity

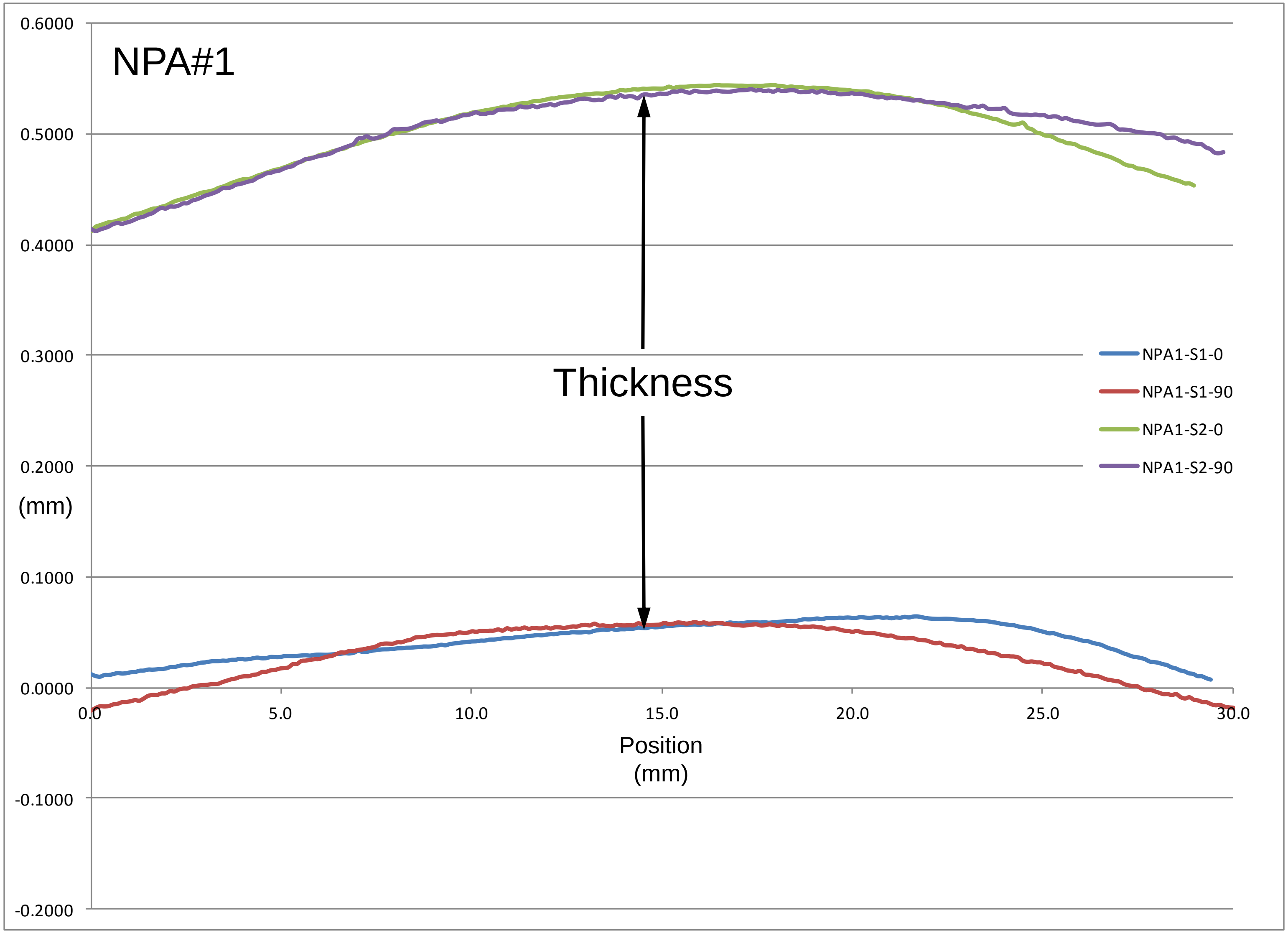
Radiographs Highlighted Density/Thickness Variations



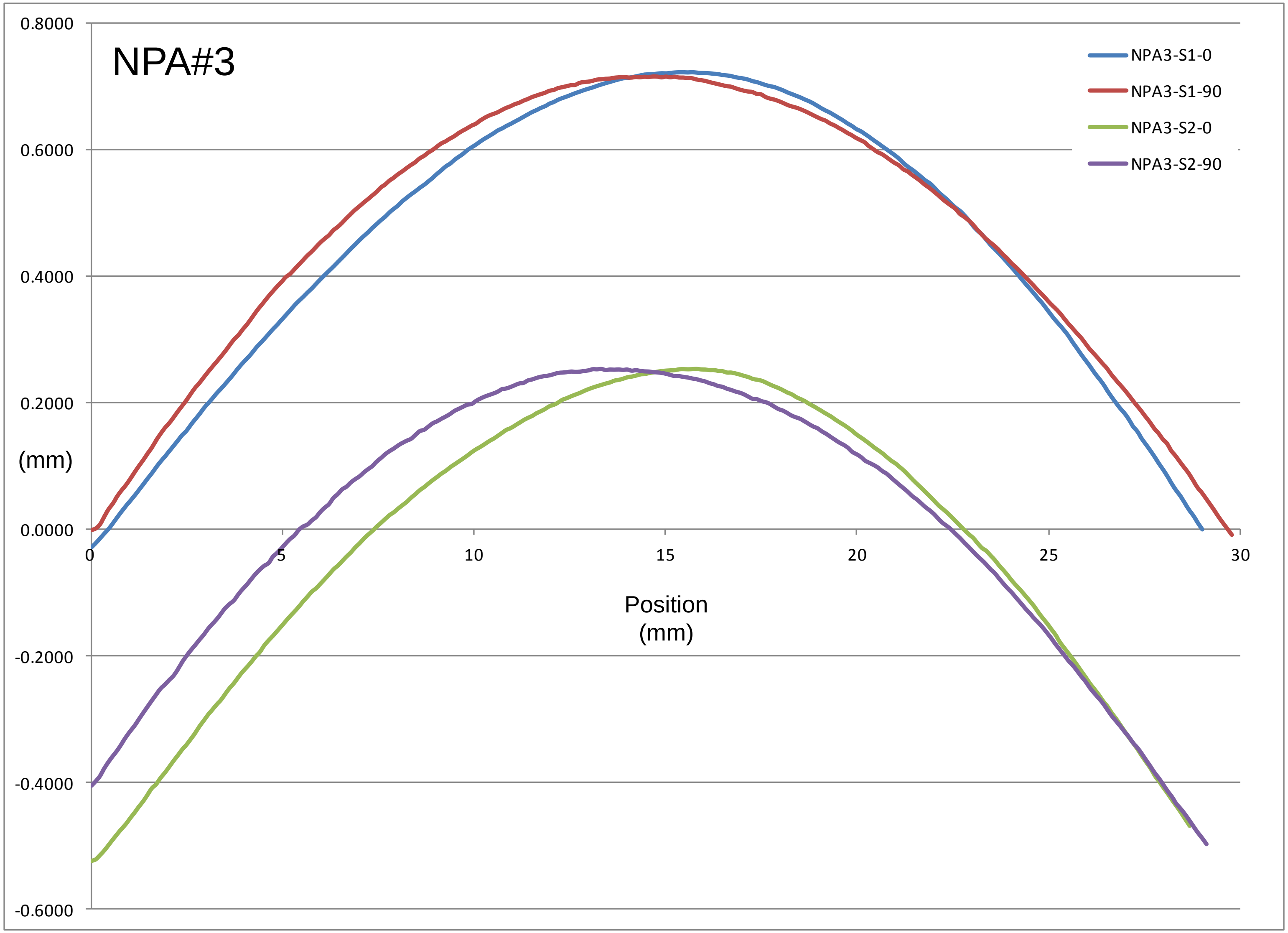


Other “flaws” were also observed.

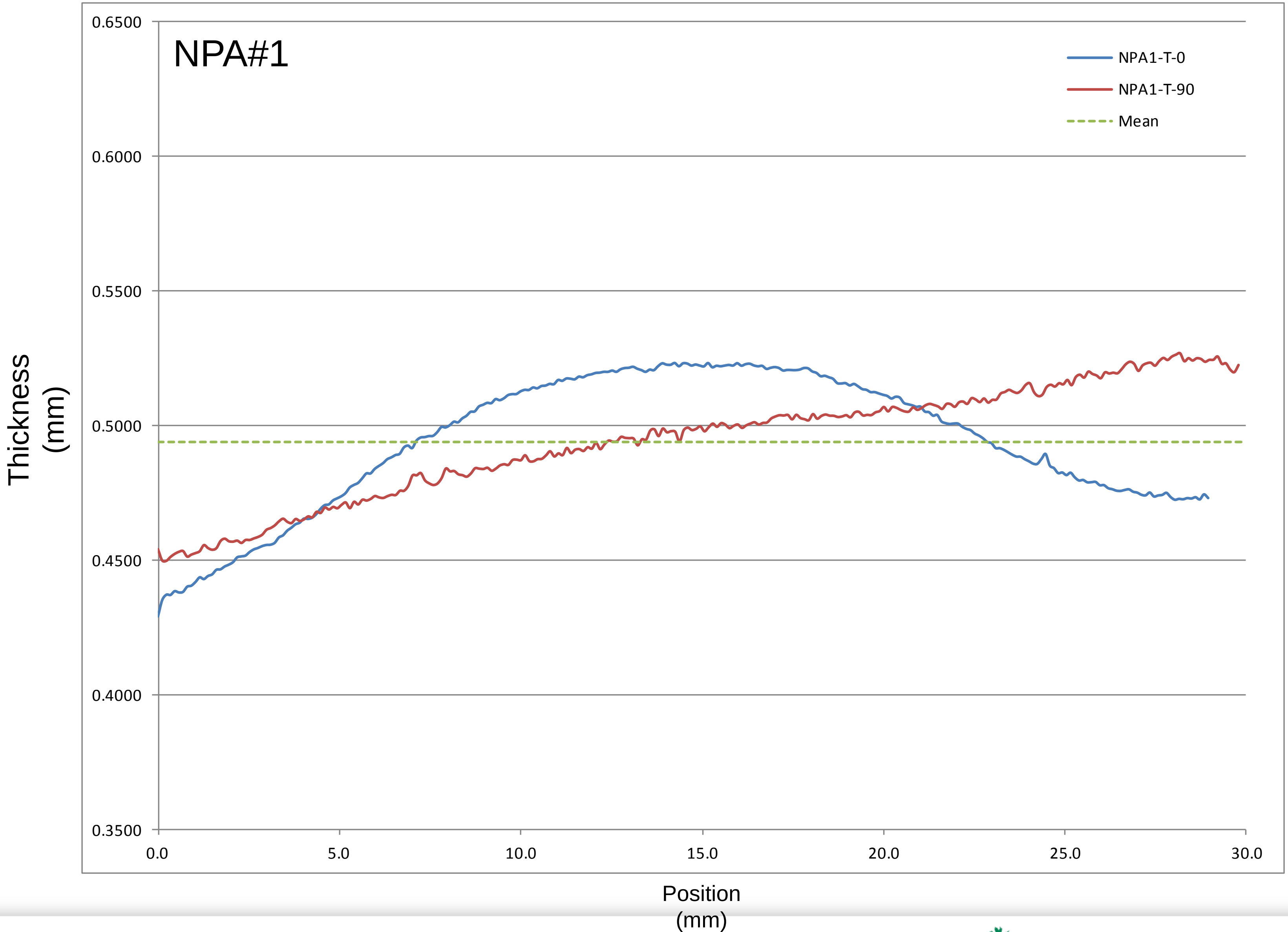
All NPA Disks Cupped During Sintering



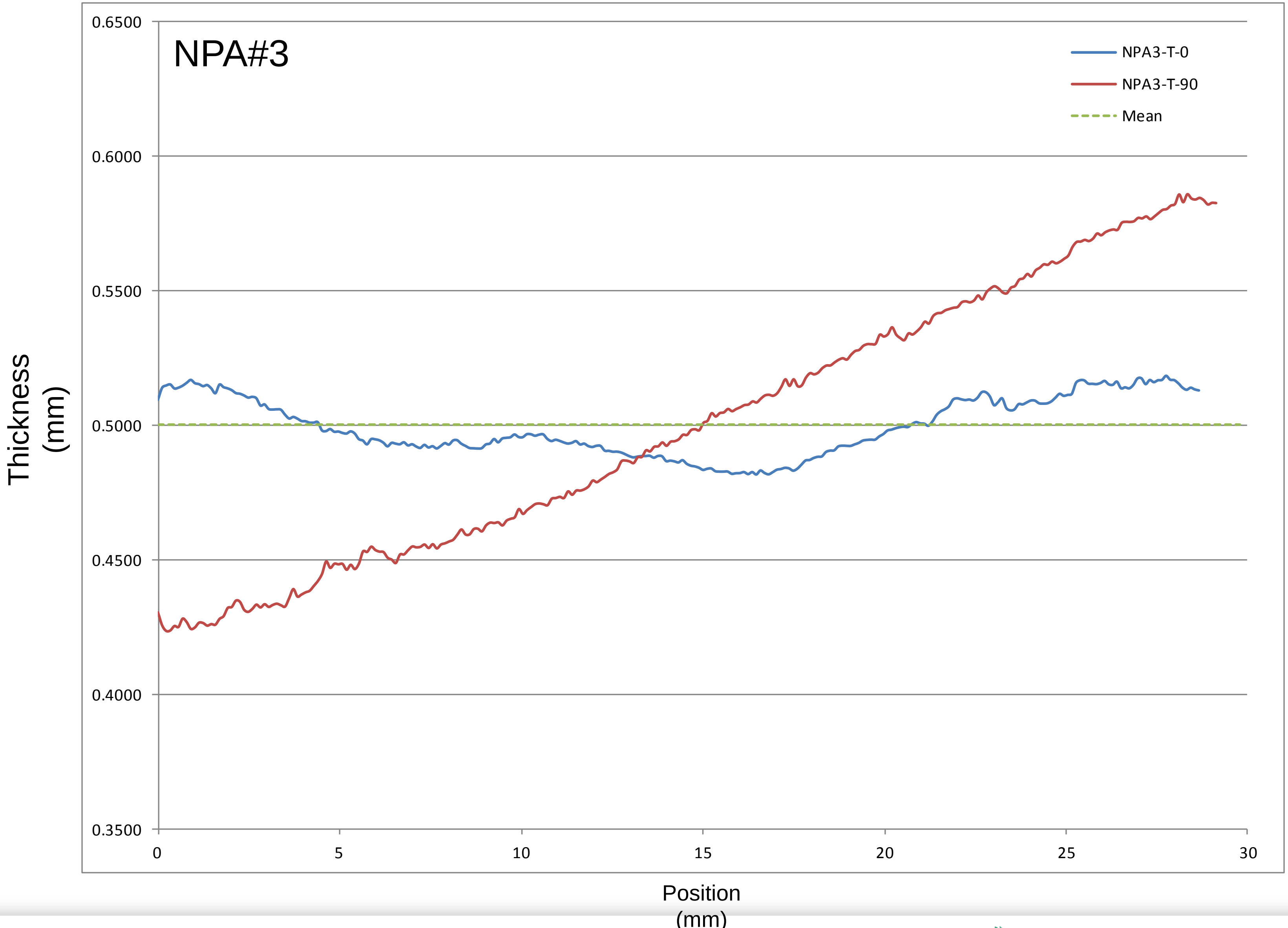
Some Disks Were Severely Cupped



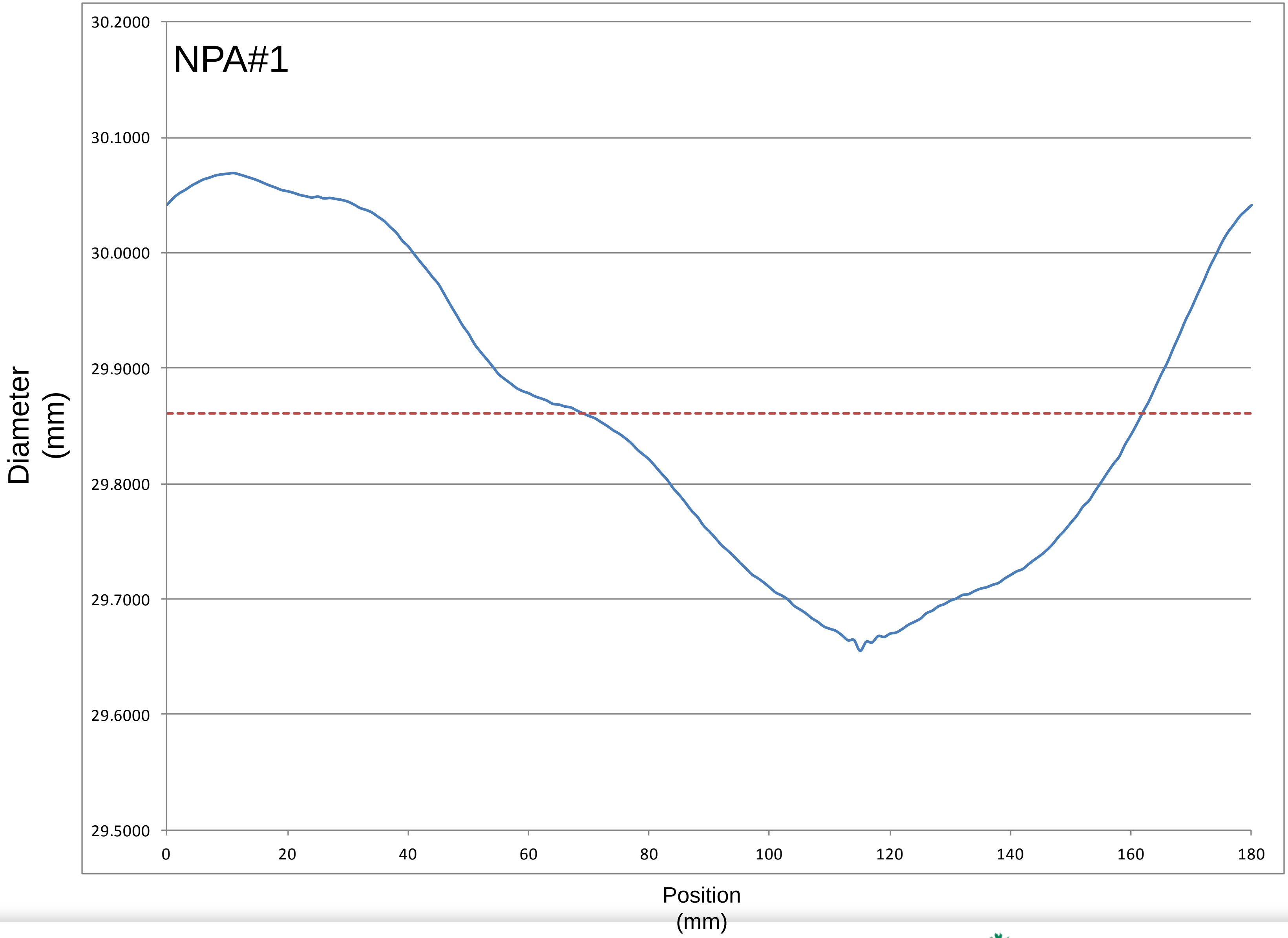
Variations in Thickness Were Also Observed



Thickness Varied Significantly For Some Disks

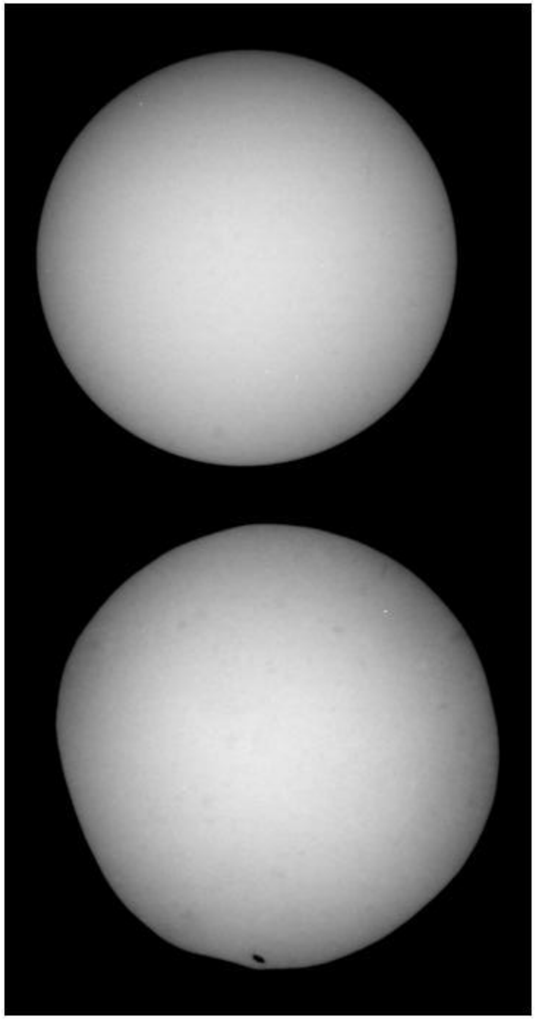


And the Disks Were Not Round

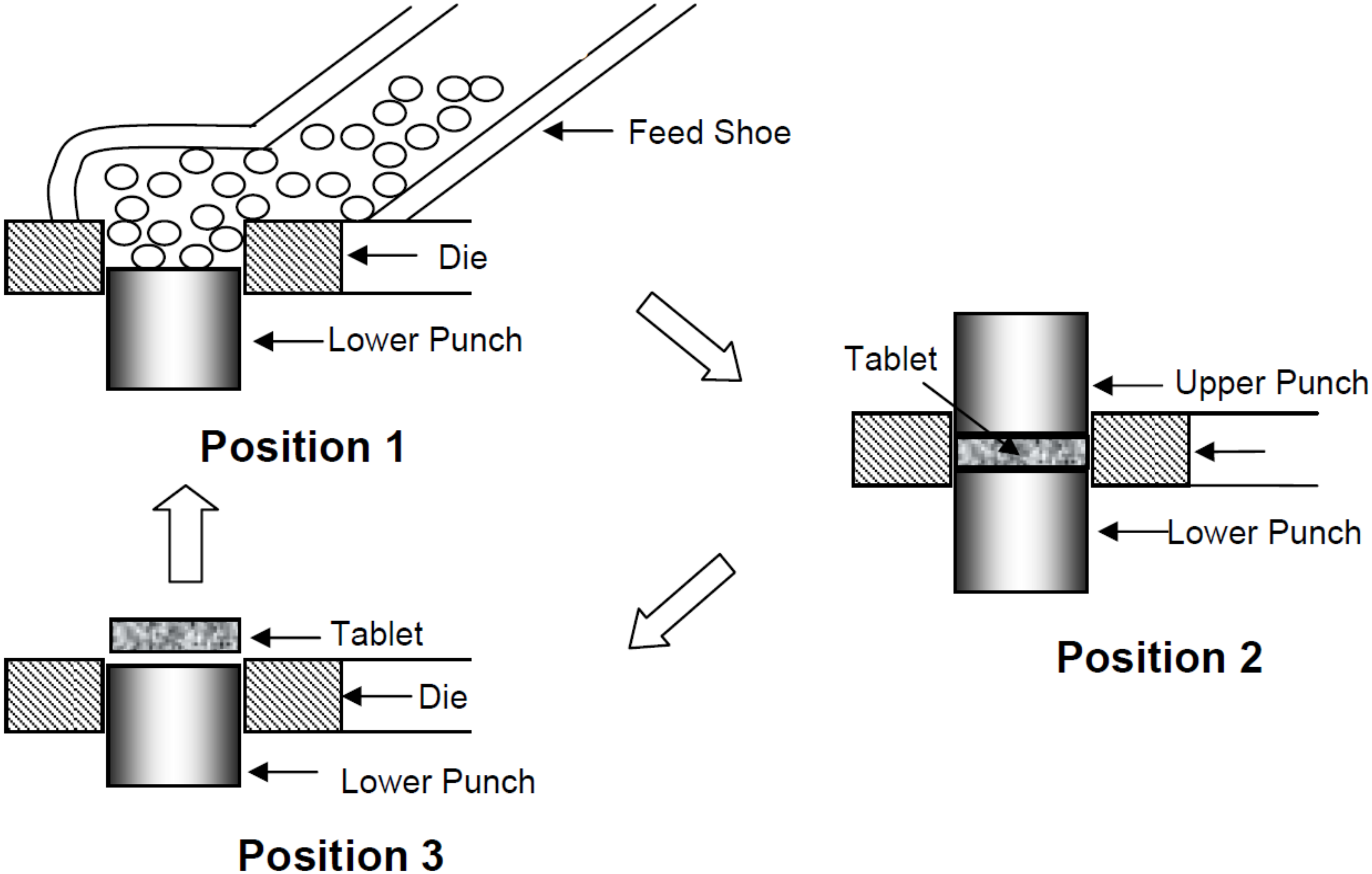


Different Approaches to Solving the Deformation Problem Were Tested

- Sintering disks lying horizontally on a bed of spherical particles.
- Restraining the disks between metal plates during sintering.
- Repressing green disks between flat plates at higher pressures.
- Repressing after sintering at room and elevated temperatures.

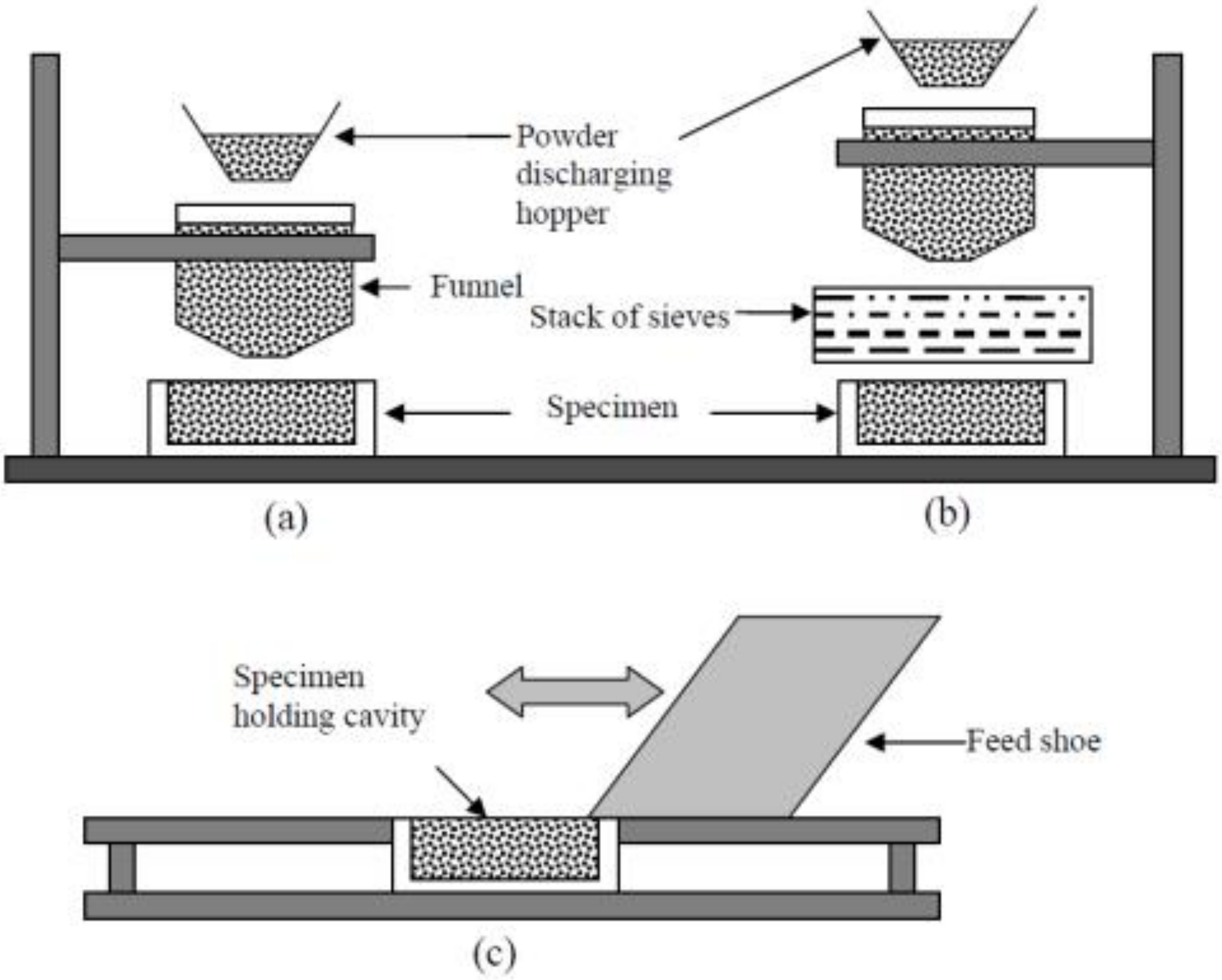


Pressing of Metal Powder is Simple

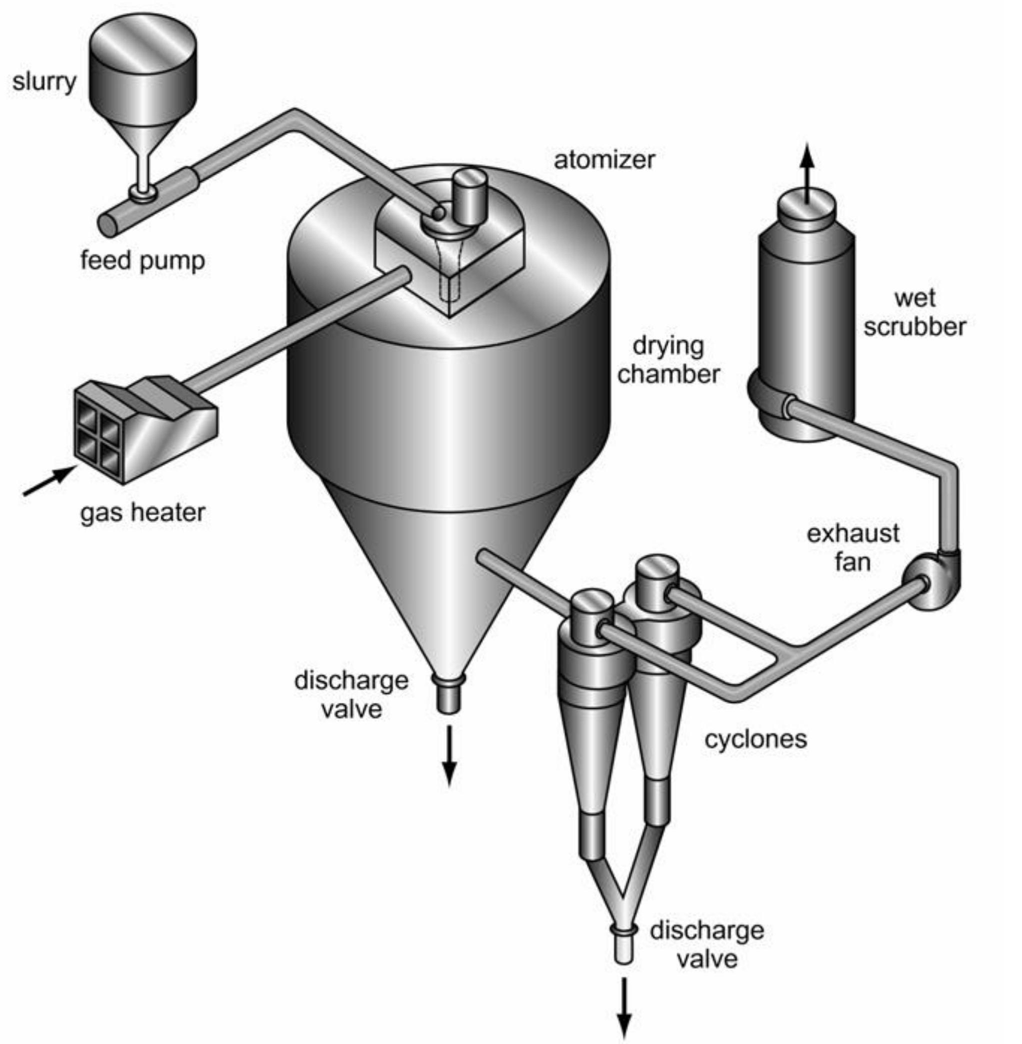


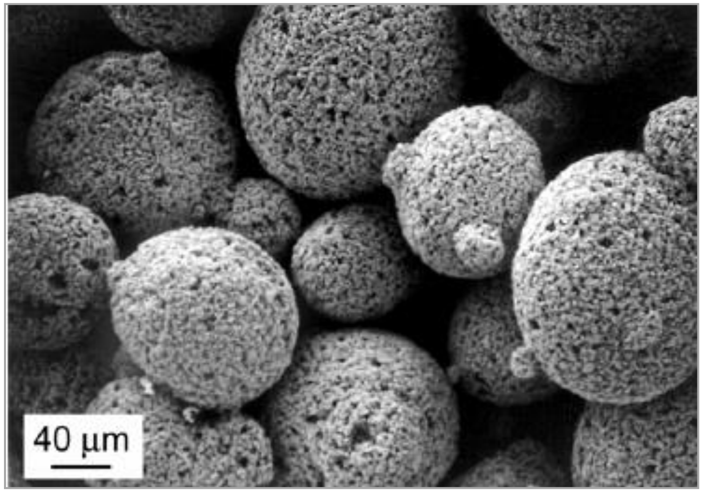
Producing Uniform, Reproducible Parts is Not.

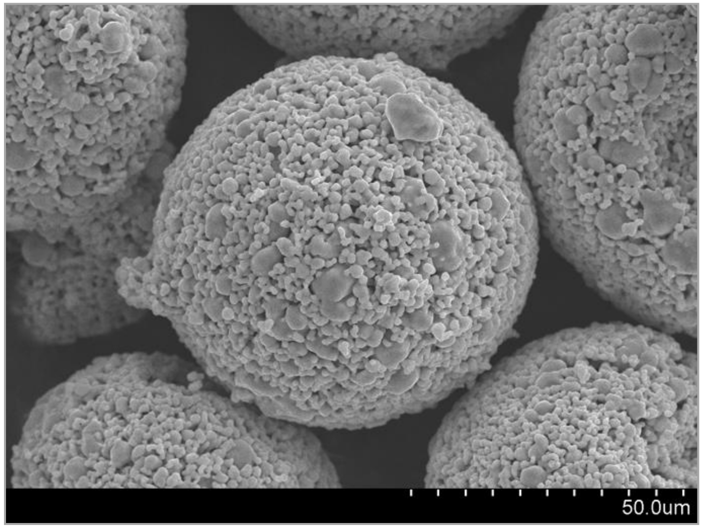
The Metal Powder Has to Flow and Uniformly Fill the Cavity of the Die



Spray-Drying of Powder Is One Approach for Controlling Flow Characteristics





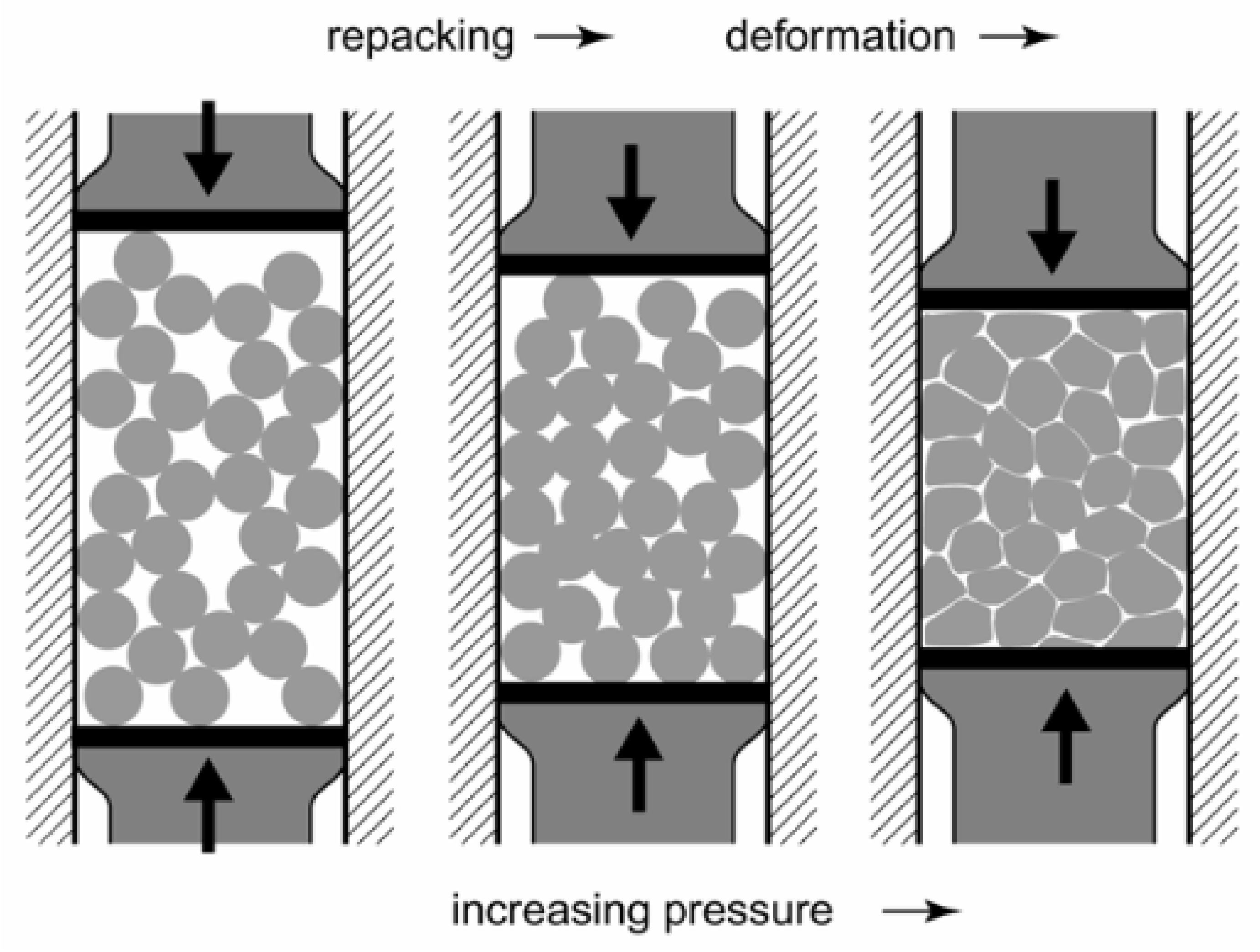


Spray-Dried Powder Flows and Presses Well Thus Produces More Uniform Parts

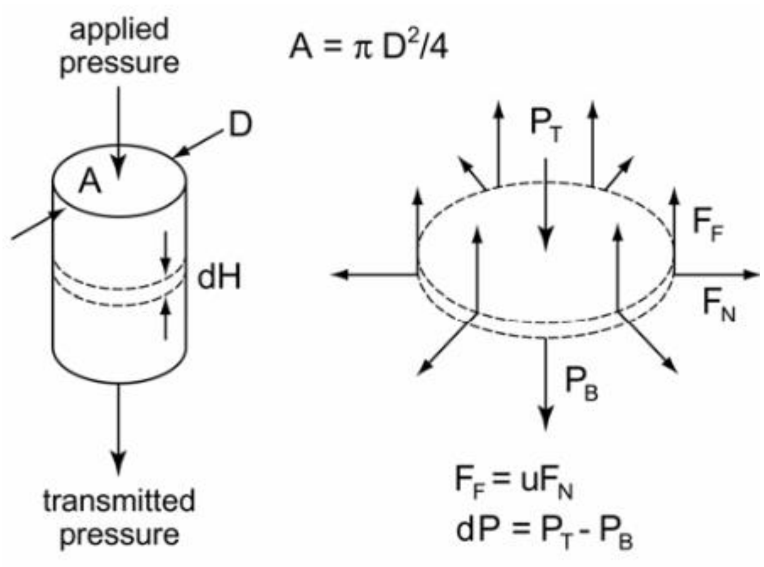
Condition	Compact Press. (ksi)	%TD (green)	Sintering (°C/h)	%TD (sintered)	Open Porosity	Diameter (mm)	Thickness (mm)	Shrinkage (%)		Cupping
								D	t	
As-Received	100	76	1500/1	85	12	29.3	0.55 ± 0.02	**3.0**	**2.6**	minimal
As-Received	100	76	1600/1	87	10	29.1	0.54 ± 0.02	**3.7**	**3.4**	minimal
As-Received	100	76	1600/4	**90**	**7**	28.8	0.54 ± 0.02	**4.7**	**5.2**	moderate
As-Received	145	79	1600/2	92	4	29.3	0.53 ± 0.02	2.9	1.1	minimal
As-Received	145	79	1600/4	93	1	29.1	0.52 ± 0.03	3.1	2.1	moderate

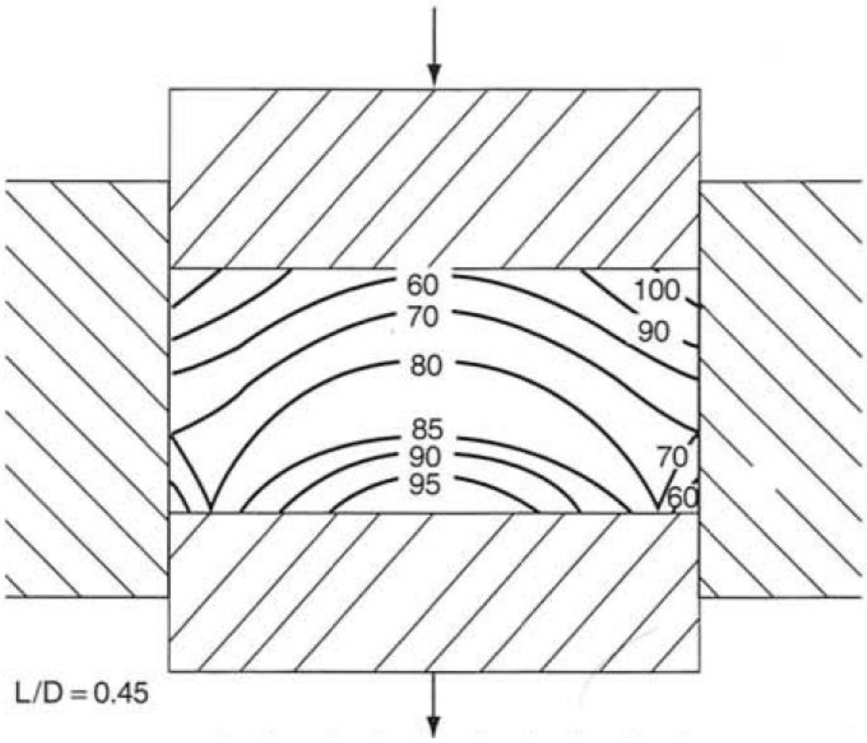
However, the disks continued to cup during sintering.

The Compaction Process Has Many Stages



Frictional Forces Associated with Tooling Surfaces and the Powder Have to be Minimized





(a) L/D = 0.45

Friction Creates Variations in Pressure Distribution Thus Differences in Density

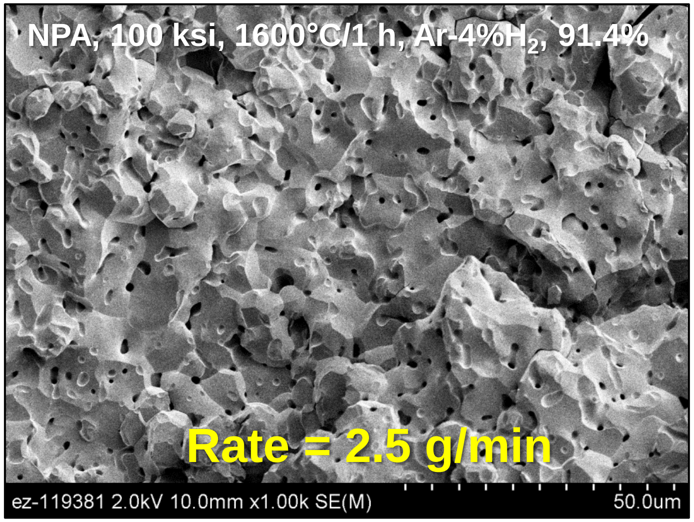
The Addition of Lubricants Improved Uniformity and Reproducibility

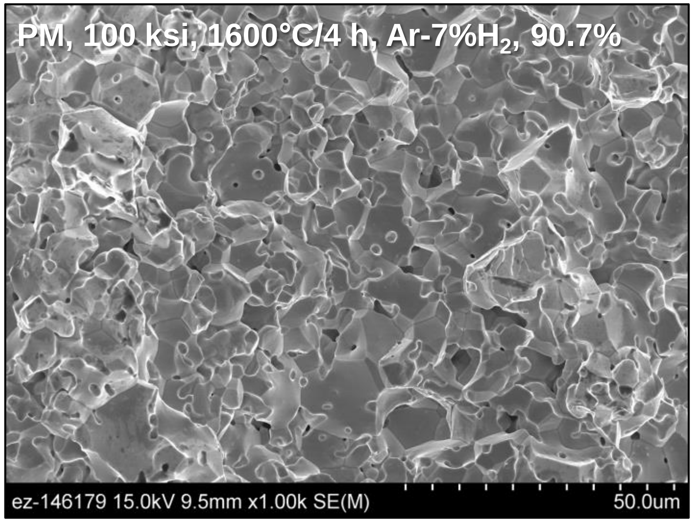
Lubricant (wt%)	Green Density (%)	Sintered Density/ Open Porosity (%)	Average Diameter (mm)	Thickness (mm)		
				Average	All Disks	Each Disk
None	76	90/7	28.8	0.54	± 0.02	± 0.04
0.25	77	90/5	28.7	0.52	± 0.005	± 0.01
0.5	76	90/8	28.7	0.53	± 0.006	± 0.01

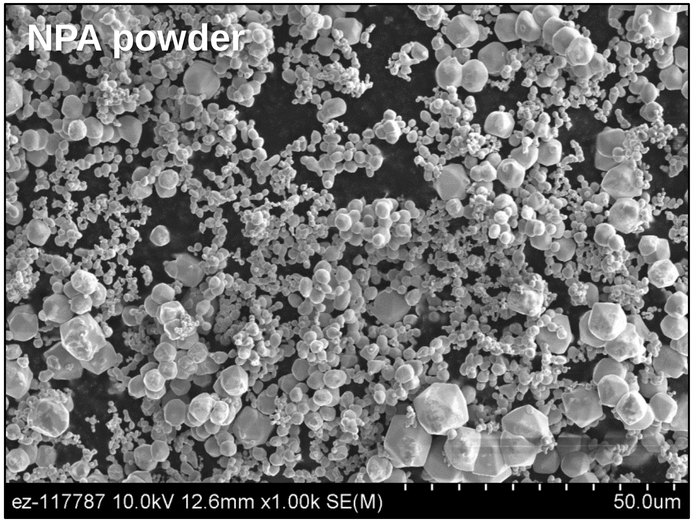
3.1 g of powder pressed at 100 ksi and sintered at 1600°C for 4 h

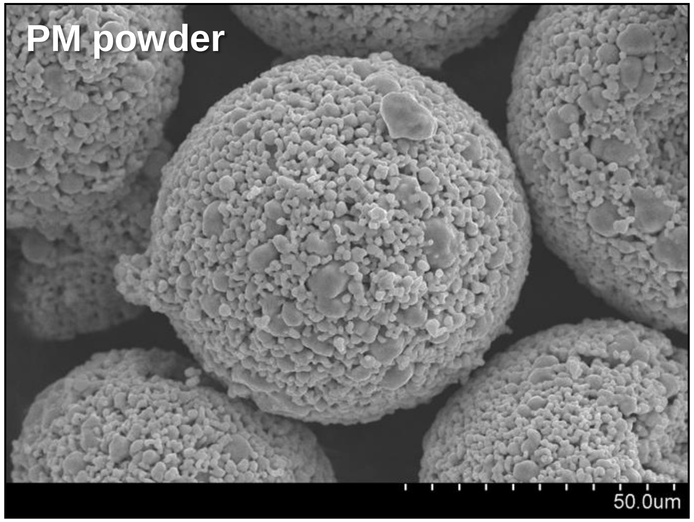
Shrinkage of ~ 5% in all directions!

Rapid Dissolution Rates for Disks Fabricated from Spray-Dried Powder Are Expected









Progress is Being Made in the Powder Metallurgy Fabrication of Target Disks for the Accelerator Production of Mo-99

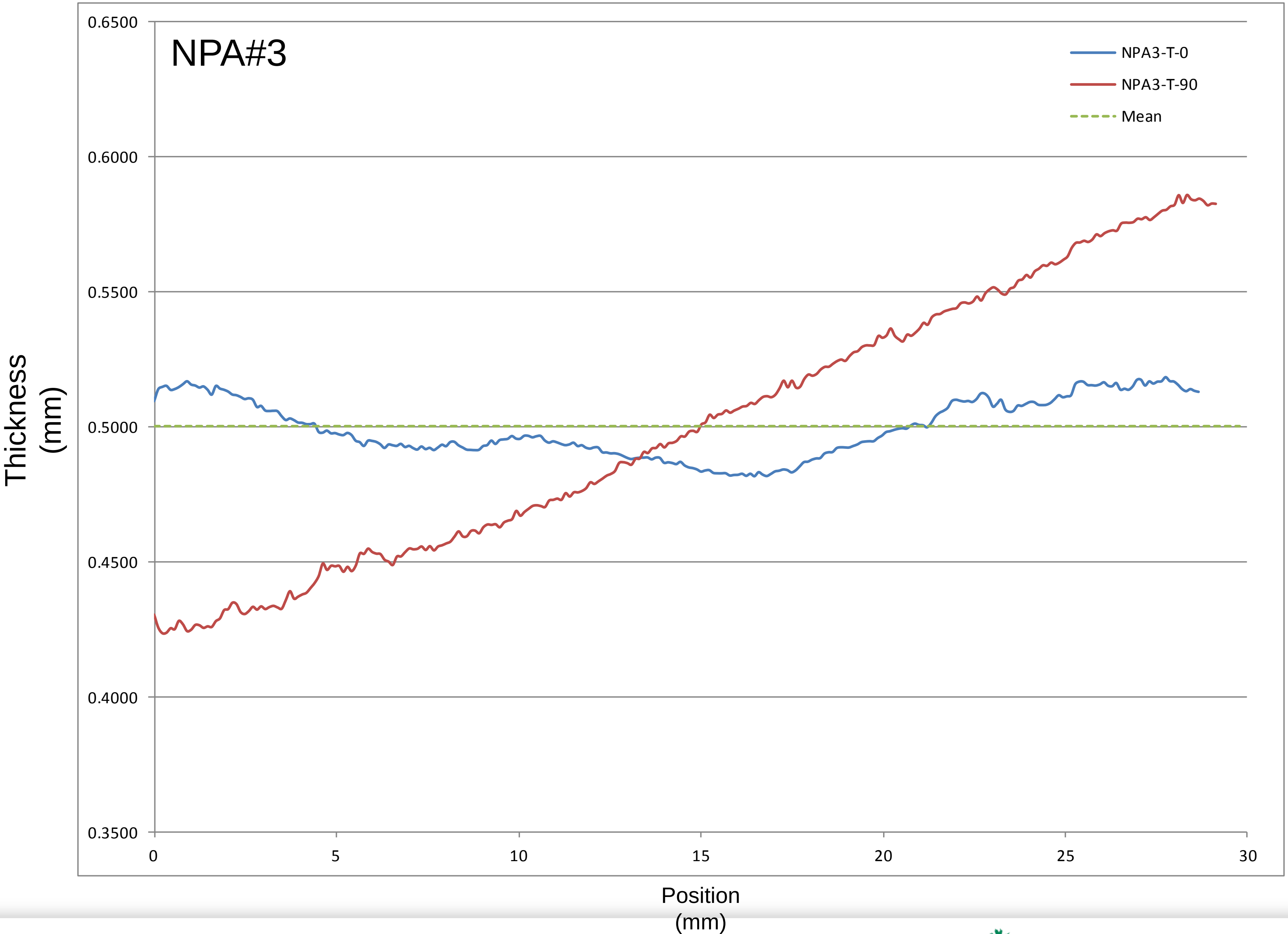
- Powder metallurgy can be used to produce accelerator target disks with minimal waste.
- All molybdenum powders can be pressed and sintered to densities greater than or equal to 90% of theoretical.
- Many factor affect the uniformity and reproducibility of thin disks fabricated from powdered metals.
- Lubricated, spray-dried powders have produced the most uniform disks to date.

Future Work

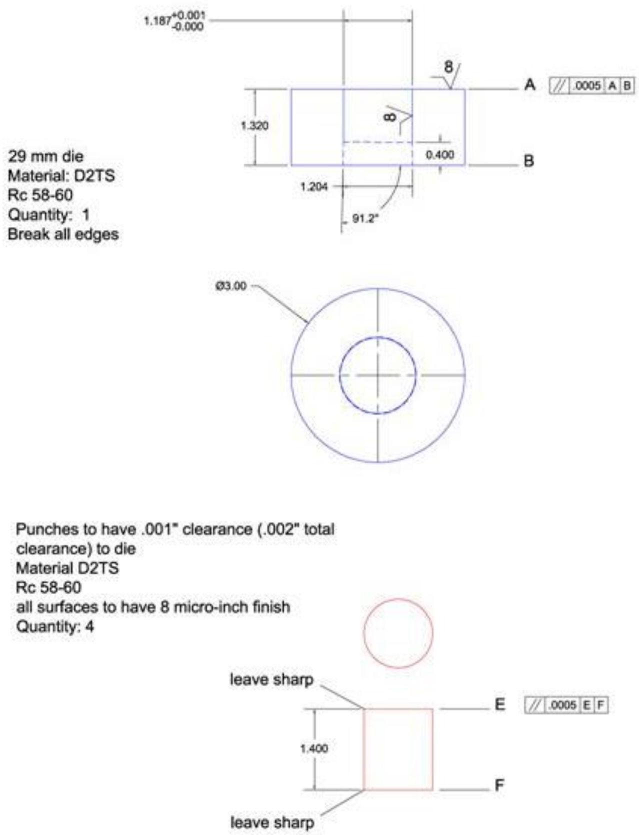
- Utilize alternate tooling designs and pressing techniques to improve uniformity and minimize distortion.
- Adjust lubricant content for optimization of compaction and sintering behavior.
- Optimize sintering schedule for complete binder removal and control of density thus open porosity.
- Evaluate dissolution rates of optimized target disks.
- Examine the thermomechanical properties of the target disks.
- Apply process to powder recovered from dissolved disks.

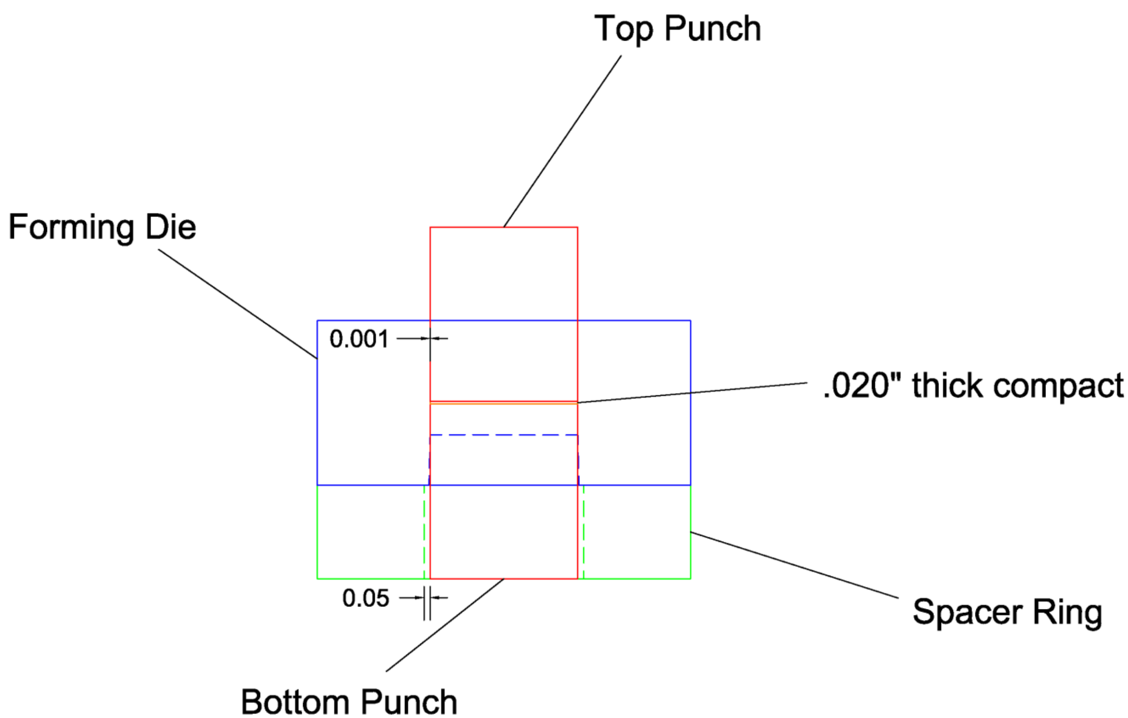
Back Up Slides

A Trend in Thickness Variation Was Observed



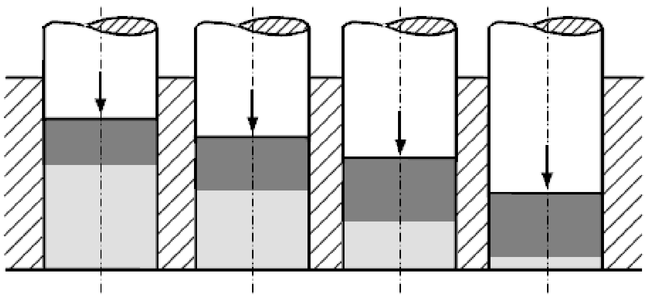
Poor Die Design and “Loose” Tolerances Can Lead to Punch Wobble



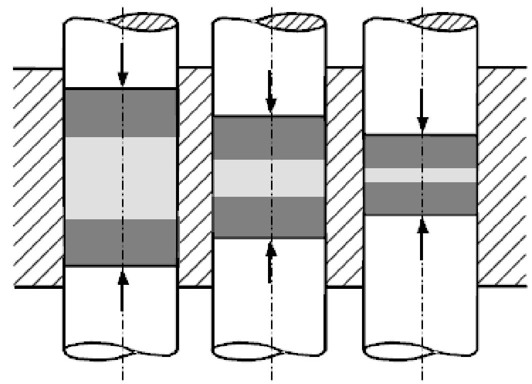


Different Pressing Techniques Can Help Reduce Pressure and Density Variations

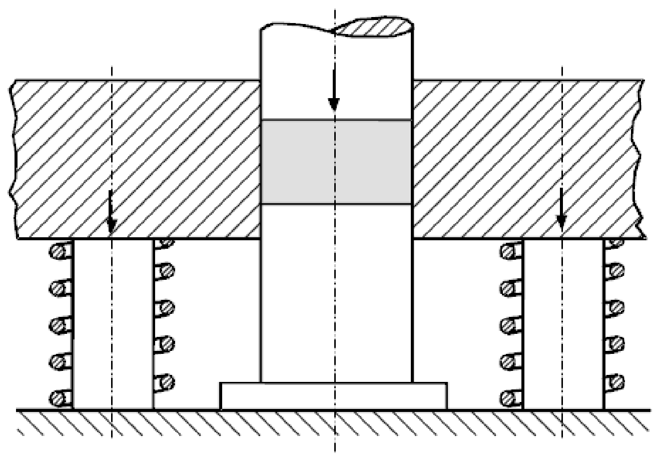
One-Sided Compacting



Two-Sided Compacting



Compacting with Floating Die



Double-Acting or Two-Sides Pressing Is an Improvement Over Single Sided

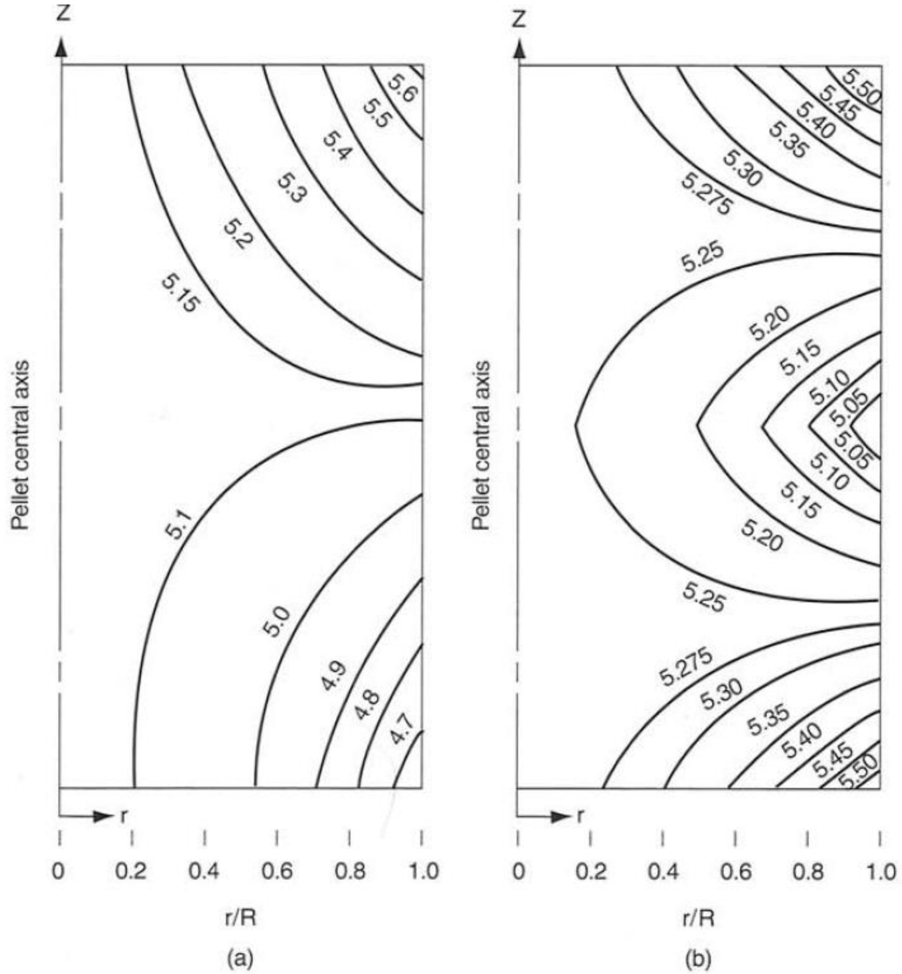


FIGURE 13.15 Decrease in pressure curves and increase in uniformity of green density by pressing compact from opposing directions with a double-acting press. (a) Single-acting press and (b) double-acting press. (From Thompson, R.A., *Am. Ceram. Soc. Bull.*, 60(2), 237–243, 1981. With permission.)