SAND2006-5457C

Science-Based Understanding of Ceramic Powder Compaction Through Characterization & Modeling

Kevin G. Ewsuk
Ceramic Processing and Inorganic Materials Department, 1815
Sandia National Laboratories
Albuquerque, NM 87185-1340
kgewsuk@sandia.gov

National Institute of Advanced Industrial Science and Technology (AIST)
September 13, 2006
Nagoya, Japan

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lookheed Martin company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000

Sandia National Laboratories

Acknowledgements

Sandia National Laboratories

Lupe Arguello
Denise Bencoe
Joe Cesarano
Erica Corral
Chris Diantonio
Don Ellerby
Jill Glass
Mark Grazier
Angela Gutierrez
Bonnie McKenzie
Scott Smith
Gary Zender
Dave Zeuch

University & Industry

Julie Anderson, University of New Mexico
Steven Beyea, New Mexico Resonance
Bill Blumenthal, Los Alamos National Laboratory
Julie Bremser, Los Alamos National Laboratory
Caroline Cloutier, Superior Technical Ceramics
Brian Gold, Superior Technical Ceramics
Joe Keller, Delphi Energy & Electronic Systems
Henrik Krarup, Penn State University

Sandia National Laboratories

Dry Pressing Is A Common Net-Shape Manufacturing Process

Net-Shape Manufacturing by Dry Pressing

Objective

Produce Net-Shape, Defect-Free Powder Compacts

Problems

Warping, Cracking, Capping, Laminations, Density Gradients

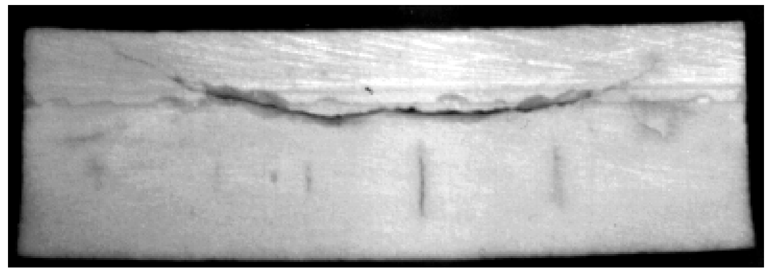
Problems Often Related to Powder Physical Characteristics & Properties

Solution

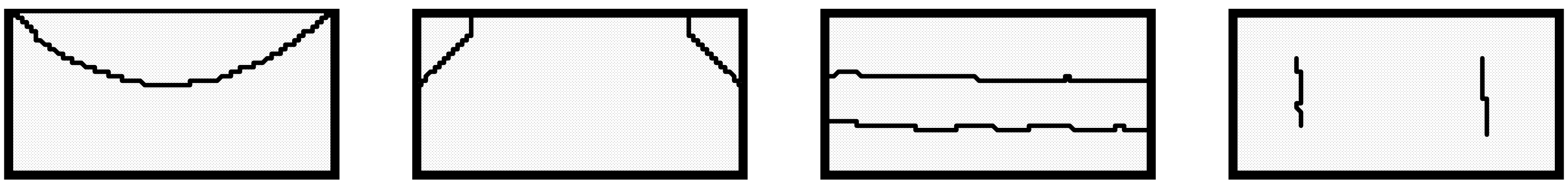
We use Characterization & Modeling to Develop
Science-Based Understanding & Control
of Ceramic Powders & Powder Compaction

Excessive Pressure, Springback, & Compaction Ratio Produce Defects

Pressed & Sintered PNZT



0.25 in

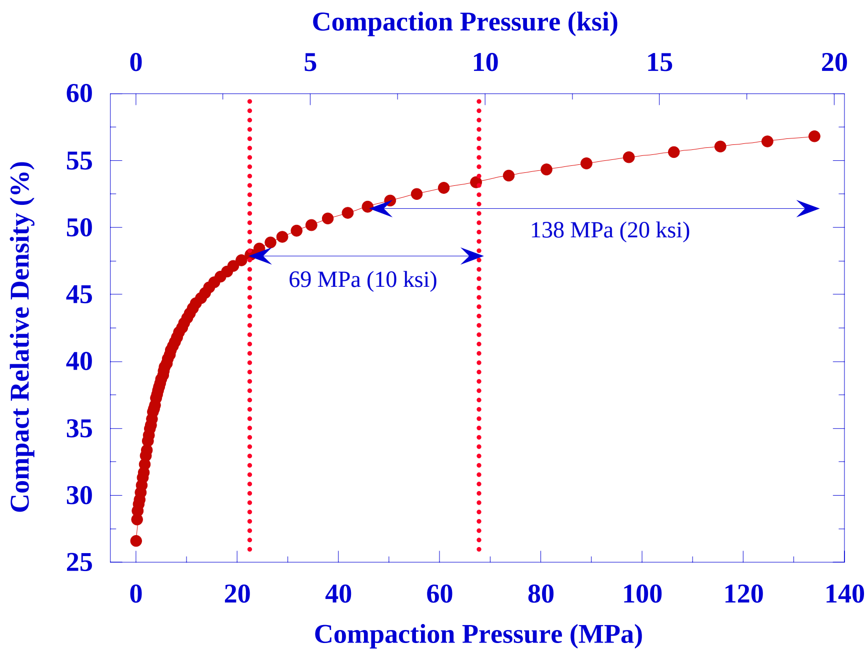


End Capping **Ring Capping** **Laminations** **Vertical Cracks**

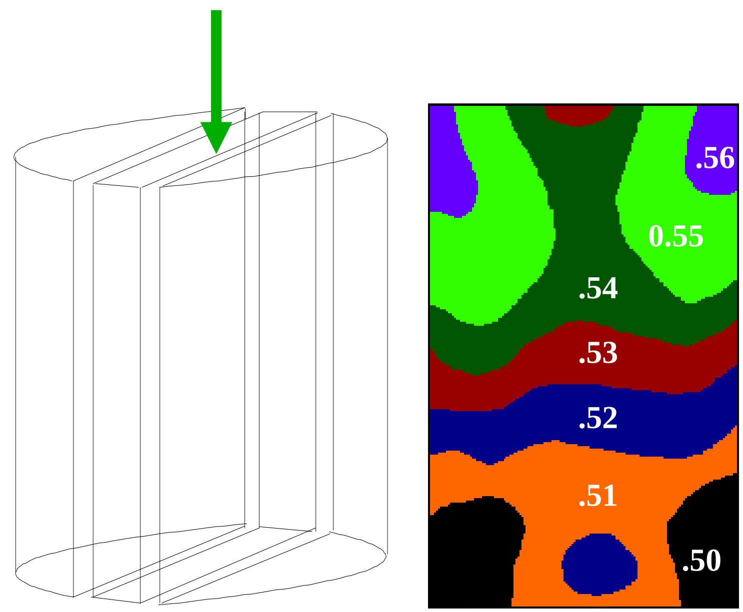
Sandia National Laboratories

Density Gradients In A Powder Compact Result From Pressure Gradients

94 wt% Alumina • 69 MPa (10 kpsi)



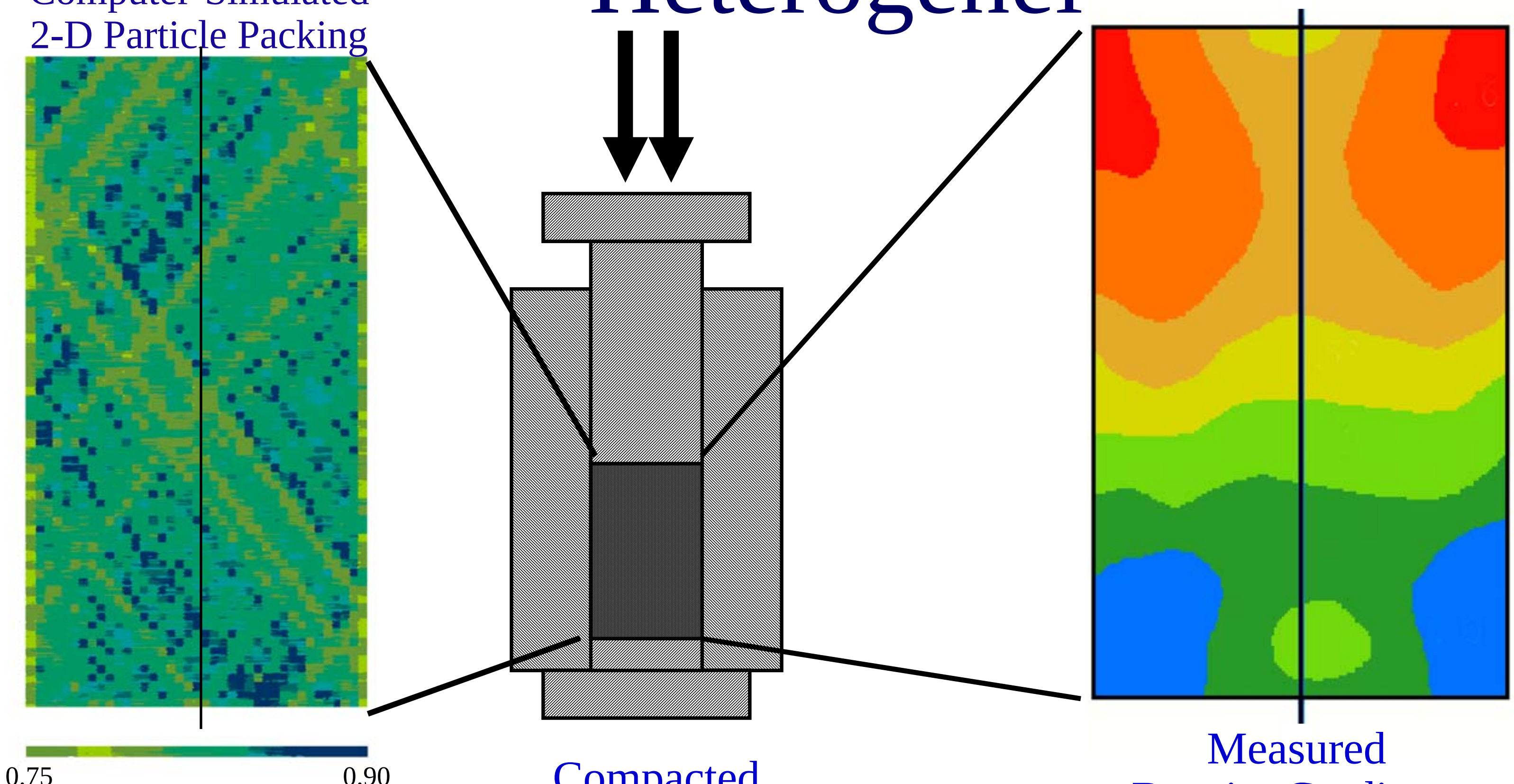
Measured Powder Compaction Curve



Measured Density Gradients
2.2 cm Diameter x 3.5 cm Tall

Variations In Compact Density Result From Packing Heterogenei

Calculated Density Gradients
Computer-Simulated
2-D Particle Packing



As-Filled
67 mm x 22.2 mm
Aspect Ratio (AR) = 3.0

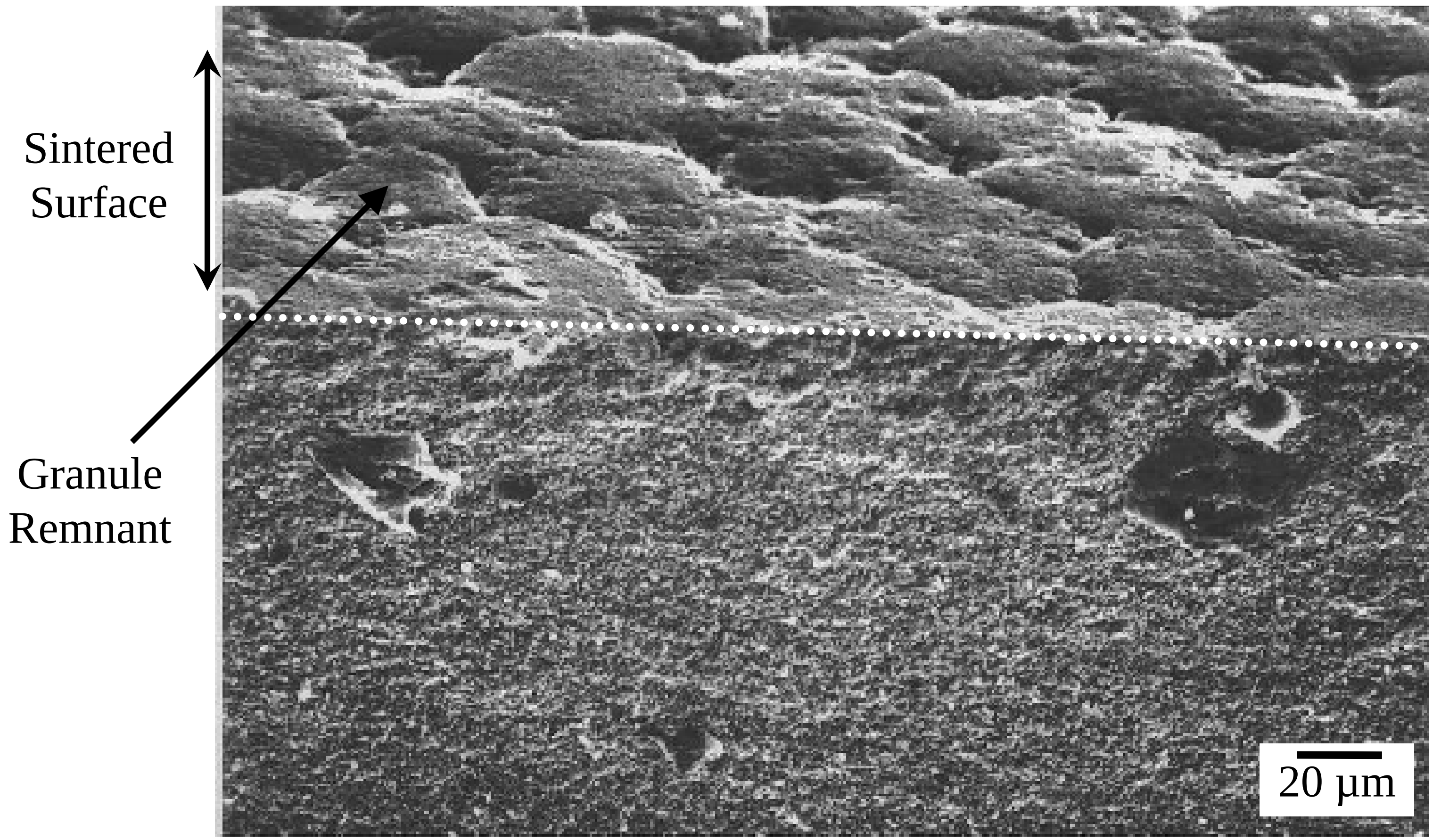
Compacted
35.1 mm x 22.2 mm
Aspect Ratio = 1.6

Measured
Density Gradients
in 94% Al_2O_3

Sandia National Laboratories

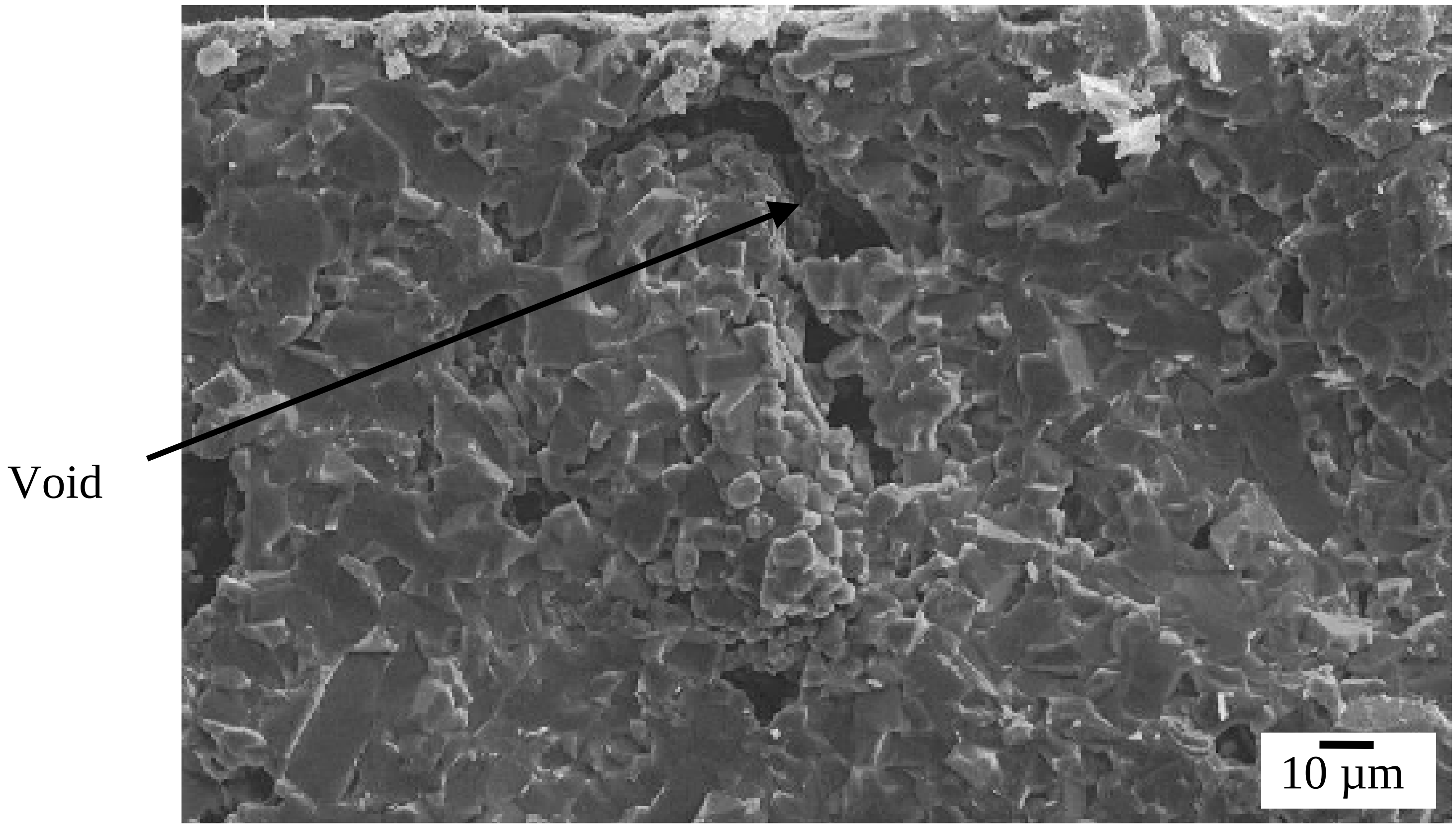
Hard Agglomerates Leave Remnants After Pressing And Sintering

Zirconia



Agglomerates/Packing Can Result In Differential Sintering & Defects

Sintered 94% $Al_2 0_3$



Powder Characteristics Influence All Three Stages Of Powder Pressing

1) Die Filling => Powder Flow & Packing

Particle/Granule Size Distribution & Shape

2) Pressing => Compaction Response & Density

Granule Density

Granule Deformation

Die Wall & Interparticle Friction

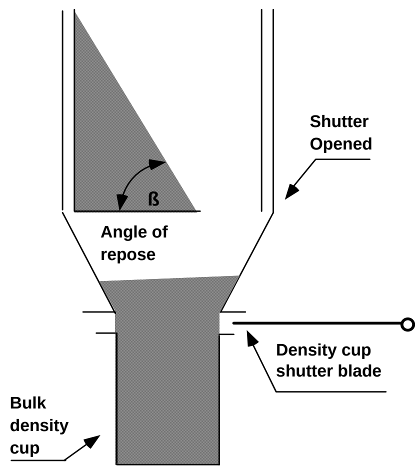
3. Ejection => Defects

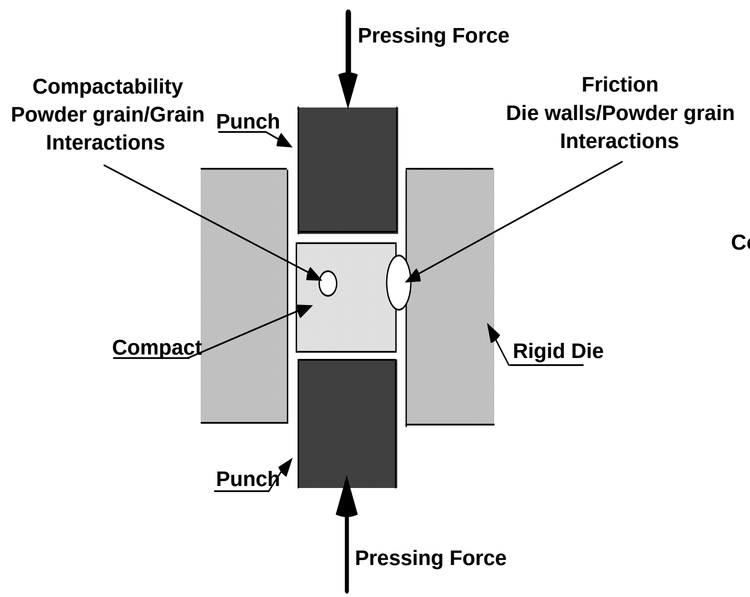
Springback

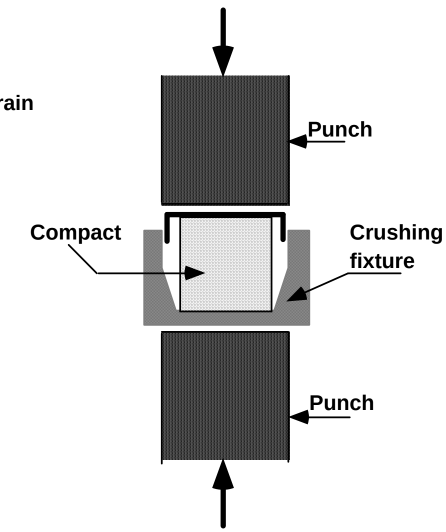
Ejection Pressure/Compact Strength

A PTC Was Used To Characterize Powder Flow, Pressing, & Ejection

KZK Powder Testing Center (PTC)







Powder Flow
<u>& Packing</u>
Angle of Repose
Bulk & Tap Density

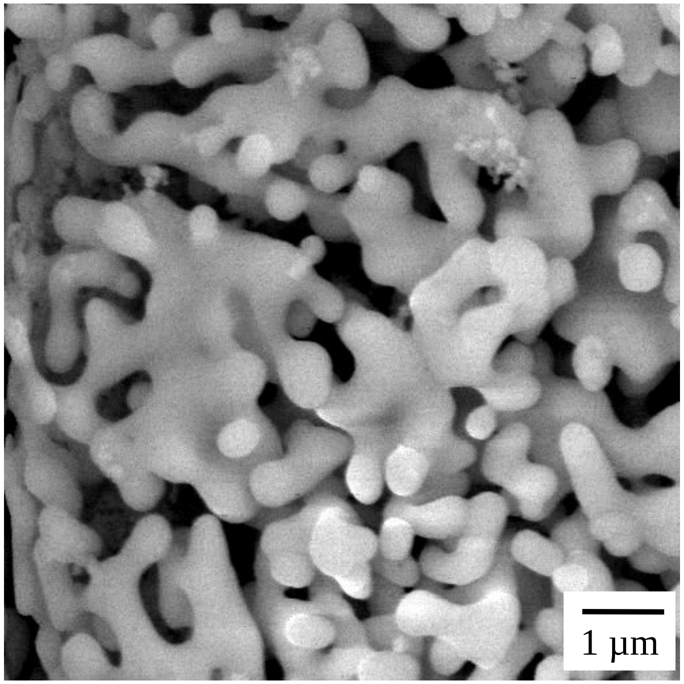
Pressing
<u>& Ejection</u>
Compaction Response
Ejection Pressure
Springback

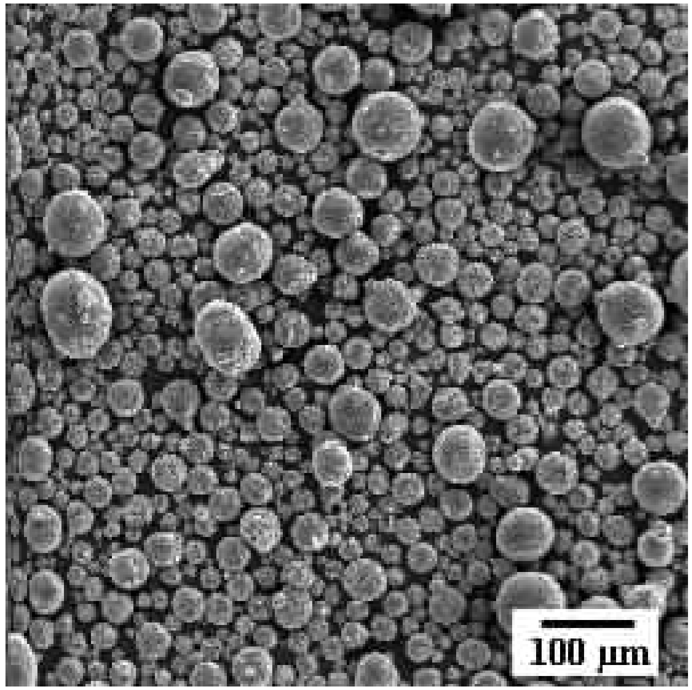
Compact
<u>Properties</u>
Compact Strength

http://kzkptc.com/

Sandia National Laboratories

Particle Size & Shape, And Granulation Affect Flow & Packing





Chemically Synthesized PZT

Angle of Repose = 83°
Bulk Density = 12.5%
Tap Density = 15.4%

Spray-Dried 94% Alumina

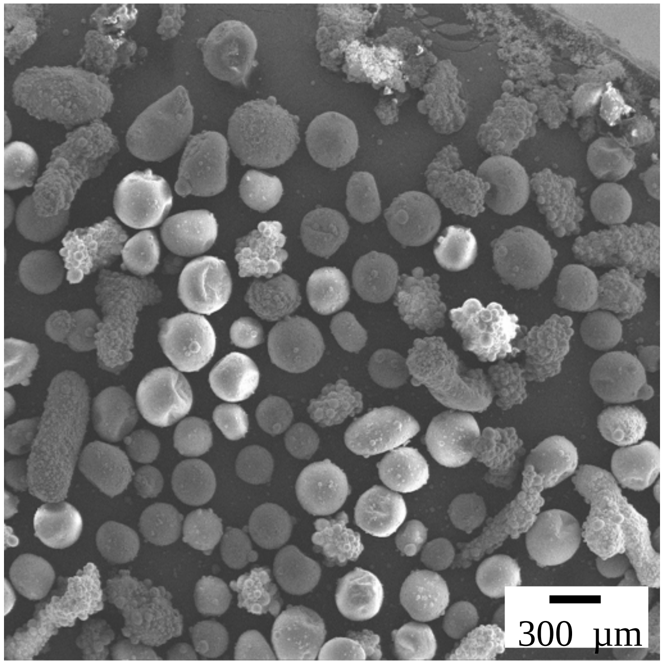
Angle of Repose = 34°
Bulk Density = 27.1%
Tap Density = 29.2%

Sandia National Laboratories

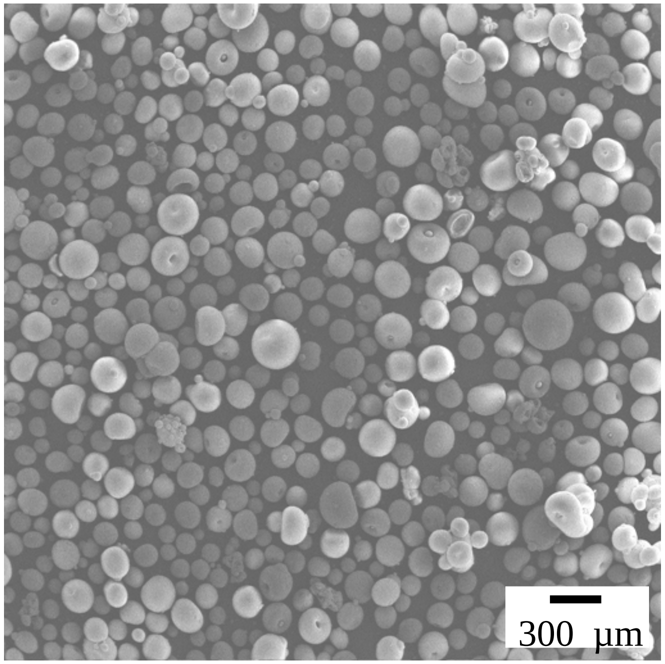
Smooth Spherical Granules Flow and Pack Better

Powder	Angle of Repose (degrees)	Bulk Density (%)	Tap Density (%)	Hausner Ratio
94% Al_2O_3	37±4	27.94±0.19	29.82±0.18	1.07±0.01
99.5% Al_2O_3	28±1	32.48±0.31	34.43±0.29	1.06±0.01

94% Al_2O_3



99.5 % Al_2O_3



113 = Agglomerate Size (µm) = 122

4.2 = Particle Size (µm) = 2.0

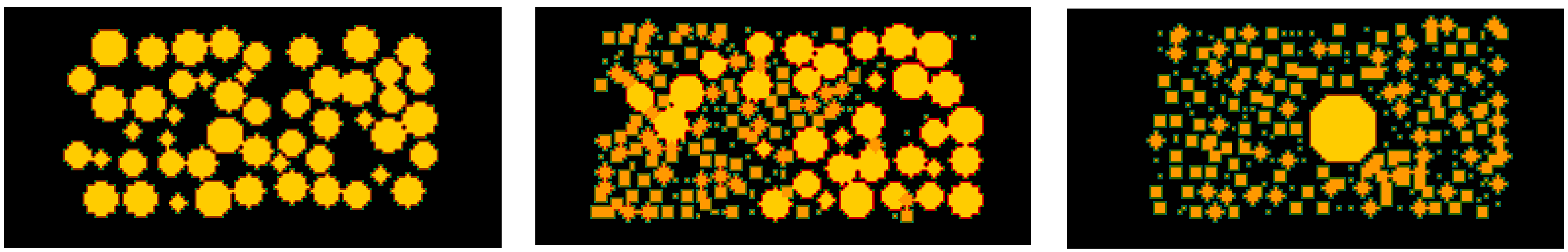
1.85 = Surface Area (m^2/g) = 3.20

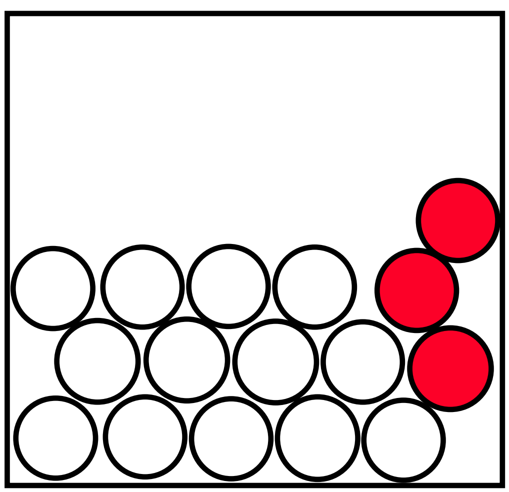
Sandia National Laboratories

Particle/Agglomerate Size And Distribution Affect Packing & Defects

Nuclear Magnetic Resonance Imaging (NMRI) of Particle Packing

Monosized | 3.2 & 6.3 mm | 3.2 & 12.7 mm

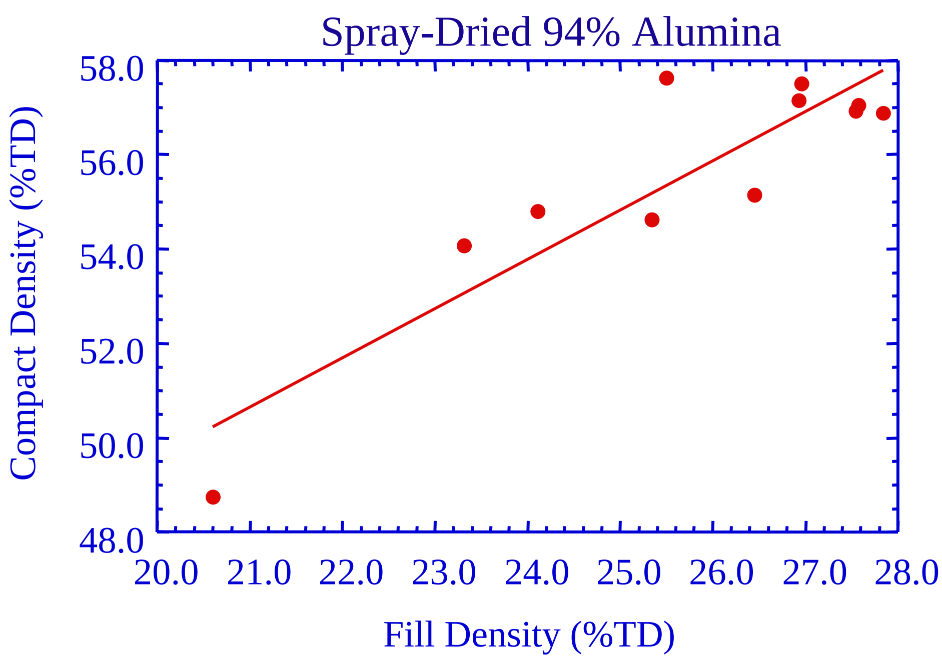




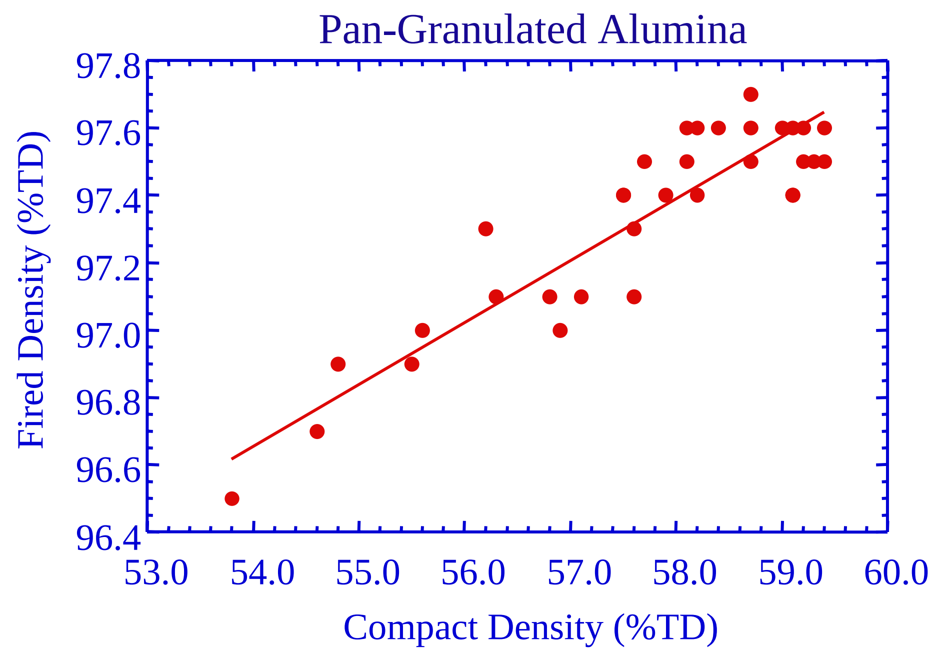
Larger particles =
larger packing defects

Packing defects are created by:
- forming die walls
- large particles
- agglomerates

Packing Defects Introduced During Die Filling Persist Throughout Processing



Compact Density Increases
With Fill Density



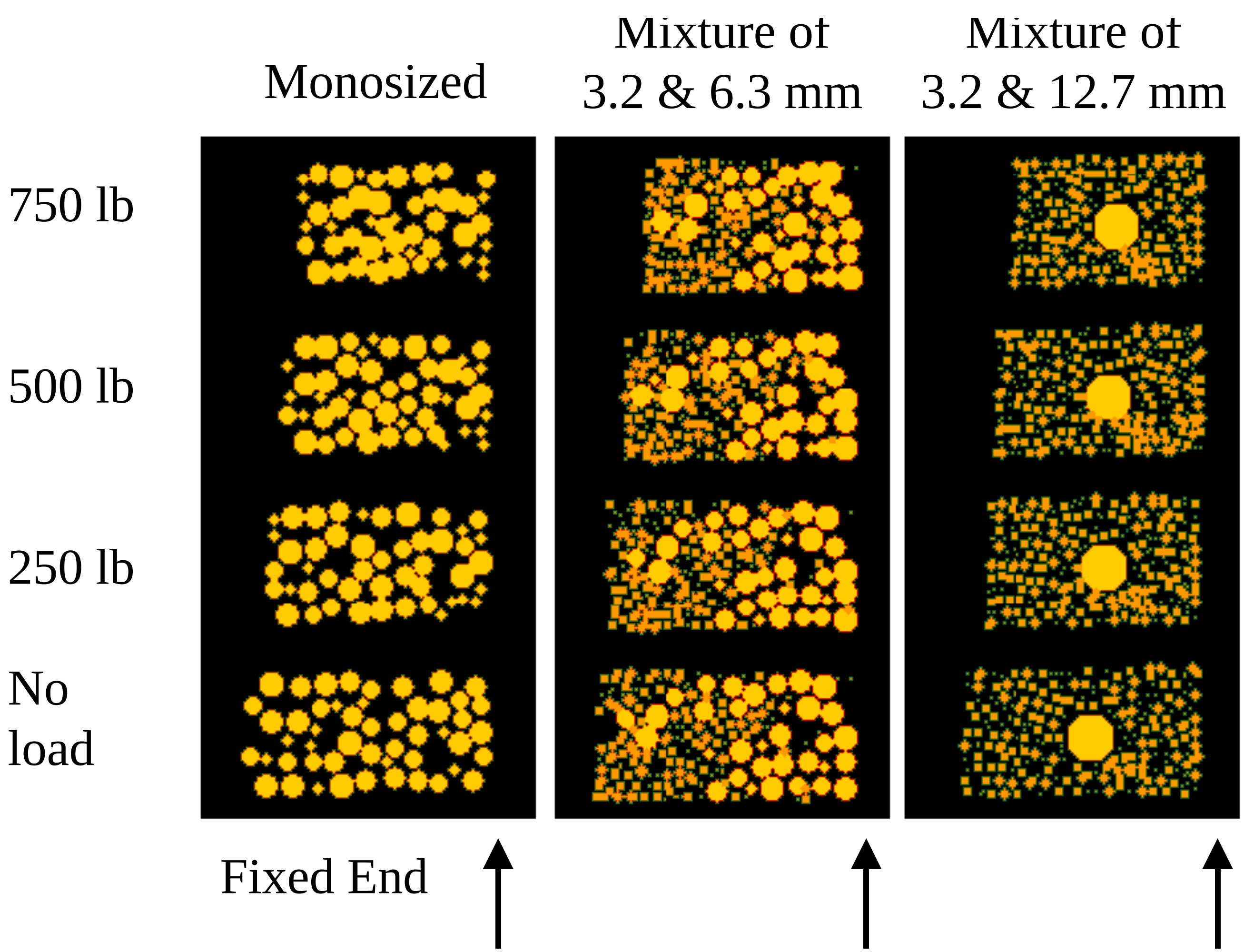
Sintered Density Increases
With Compact Density

Sandia National Laboratories

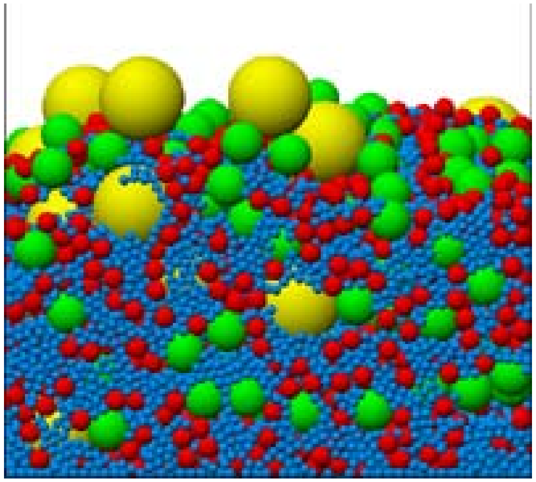
Defects Introduced In Die Filling Are Not Eliminated During Pressing

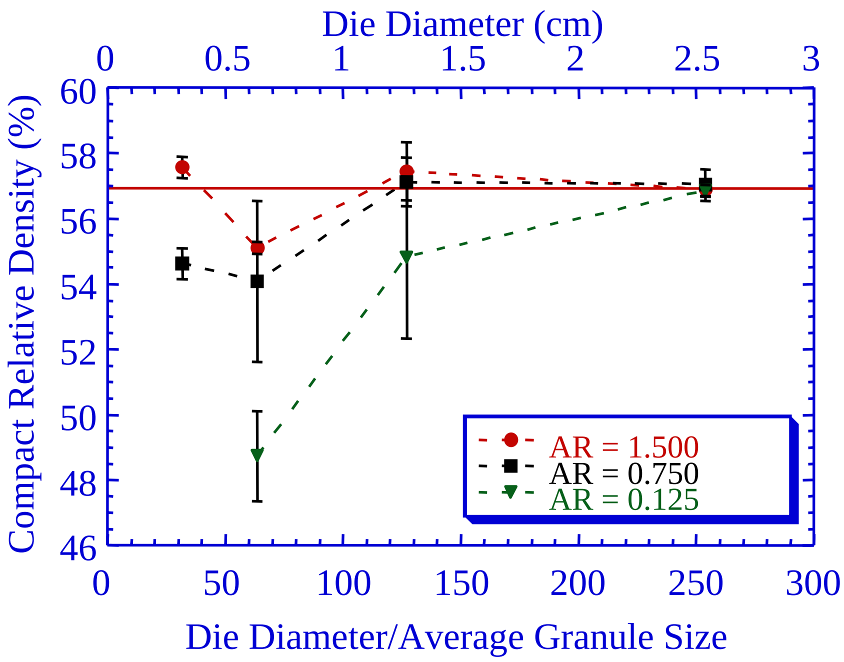
NMRI *in-situ* compaction experiments

relative particle motion < 1 particle radius



Modeling Provides Guidance On How To Minimize Packing Defects

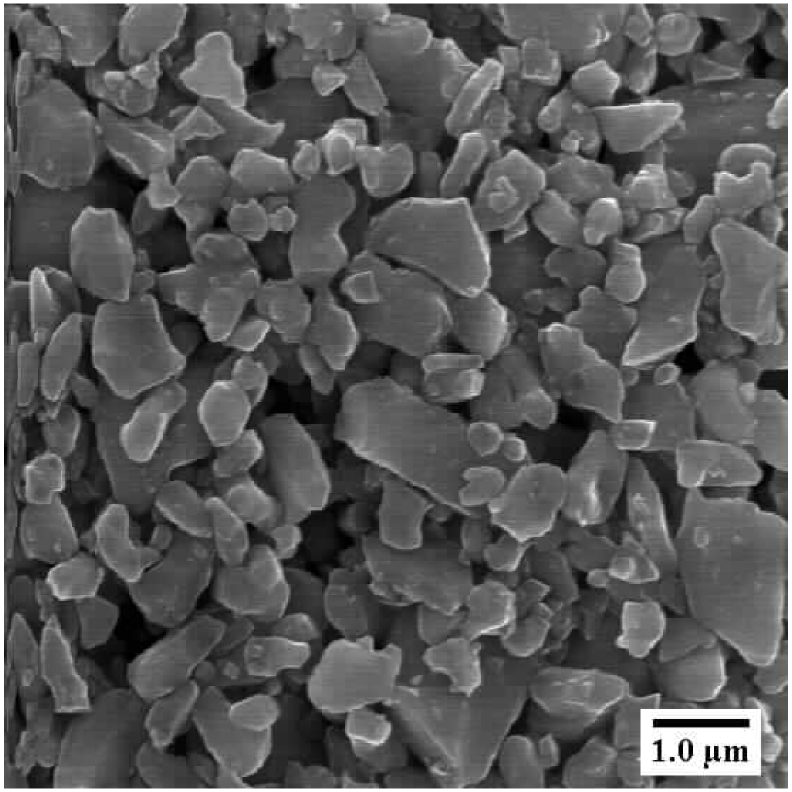




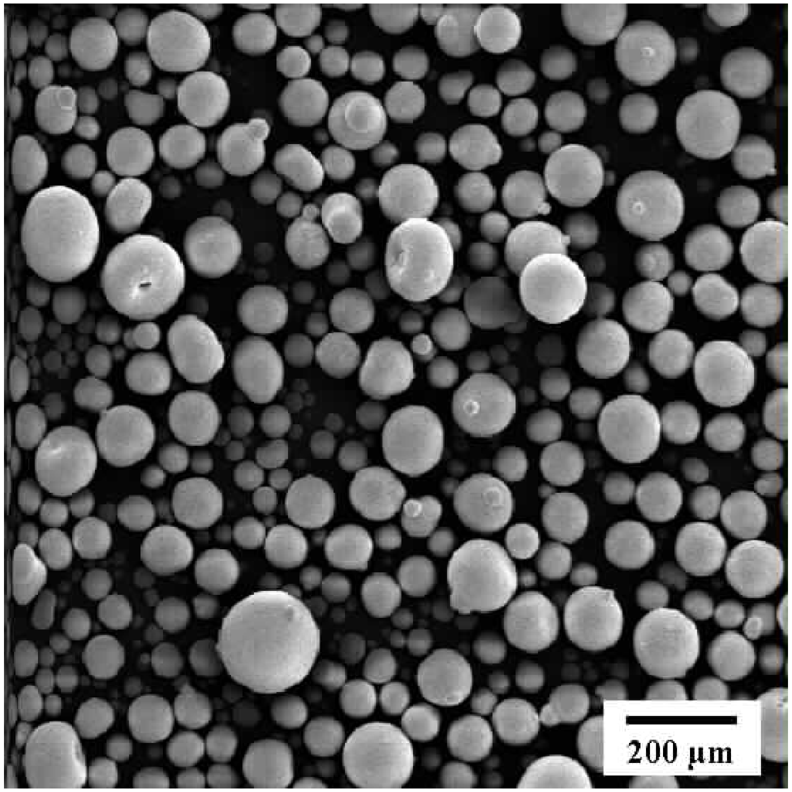
- There is a critical component size to granule size ratio of 250:1
- The ratio is important for small parts with a large surface area
- Granule sizing improve yields of small parts
- Packing models can be used to tailor granule size distributions

Sandia National Laboratories

Ceramic Powders Are Granulated To Improve Flow, Packing, & Compaction



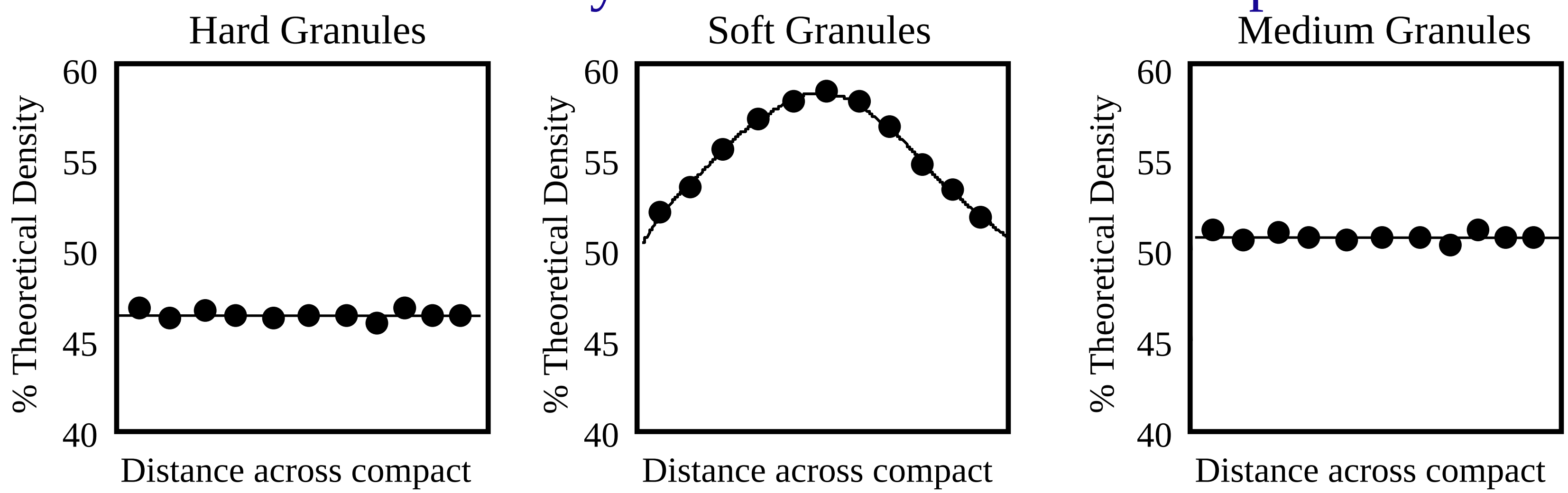
Alumina Powder

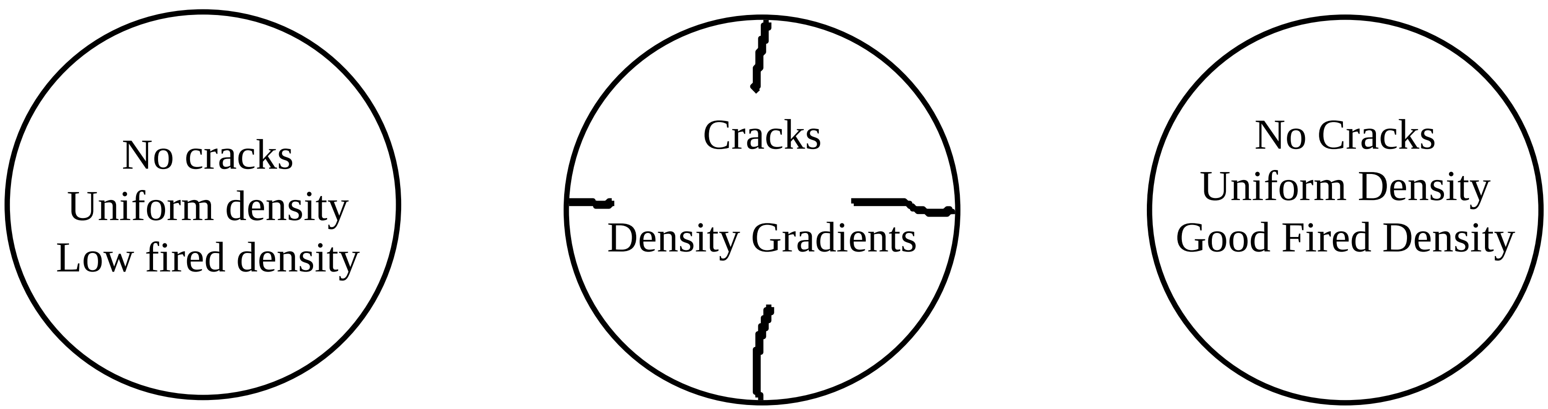


Spray-Dried
Alumina Granules

Granule Hardness Influences Flow, Packing, & Compaction

Green Density vs. Distance Across Compact

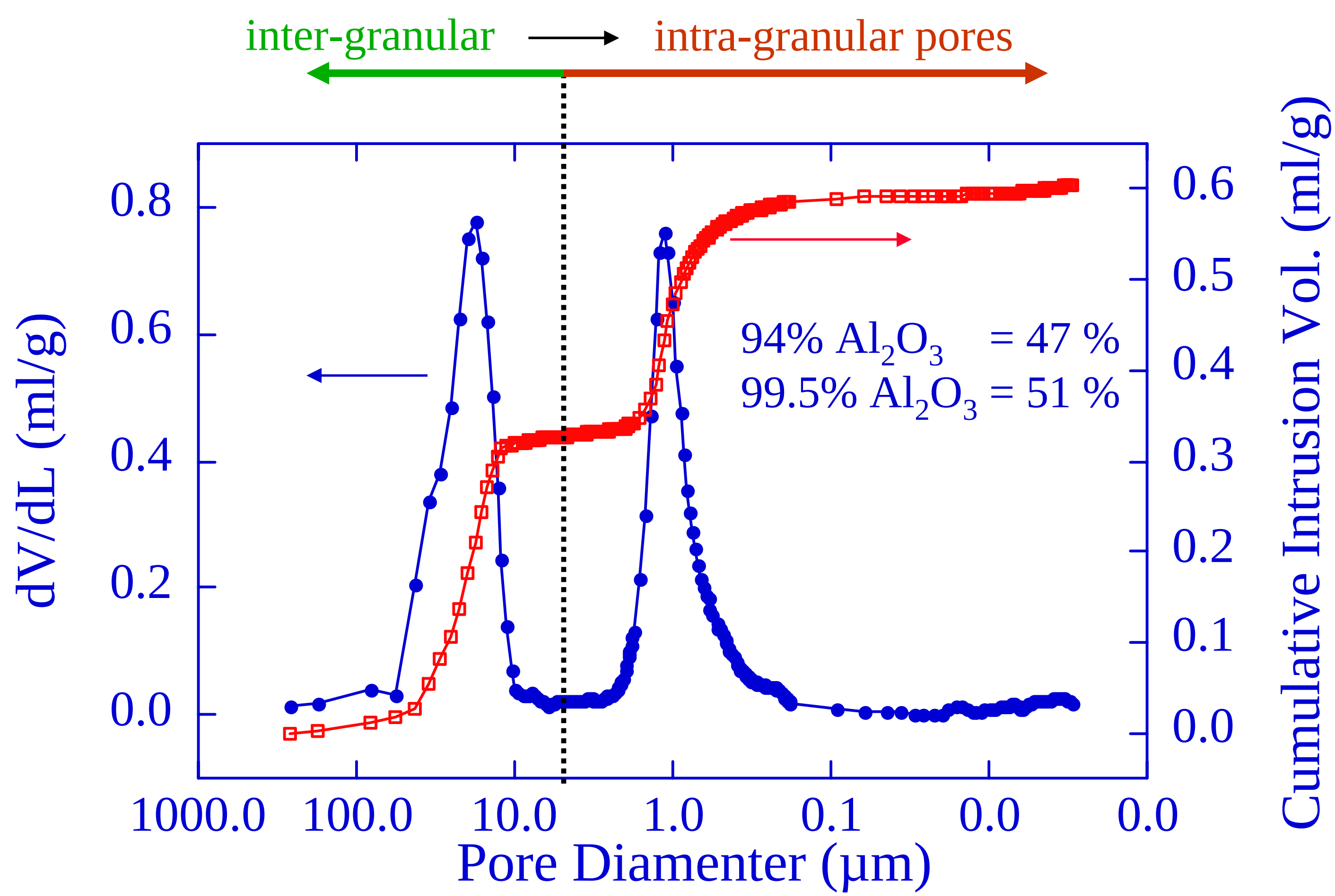




Disk Appearance After Sintering

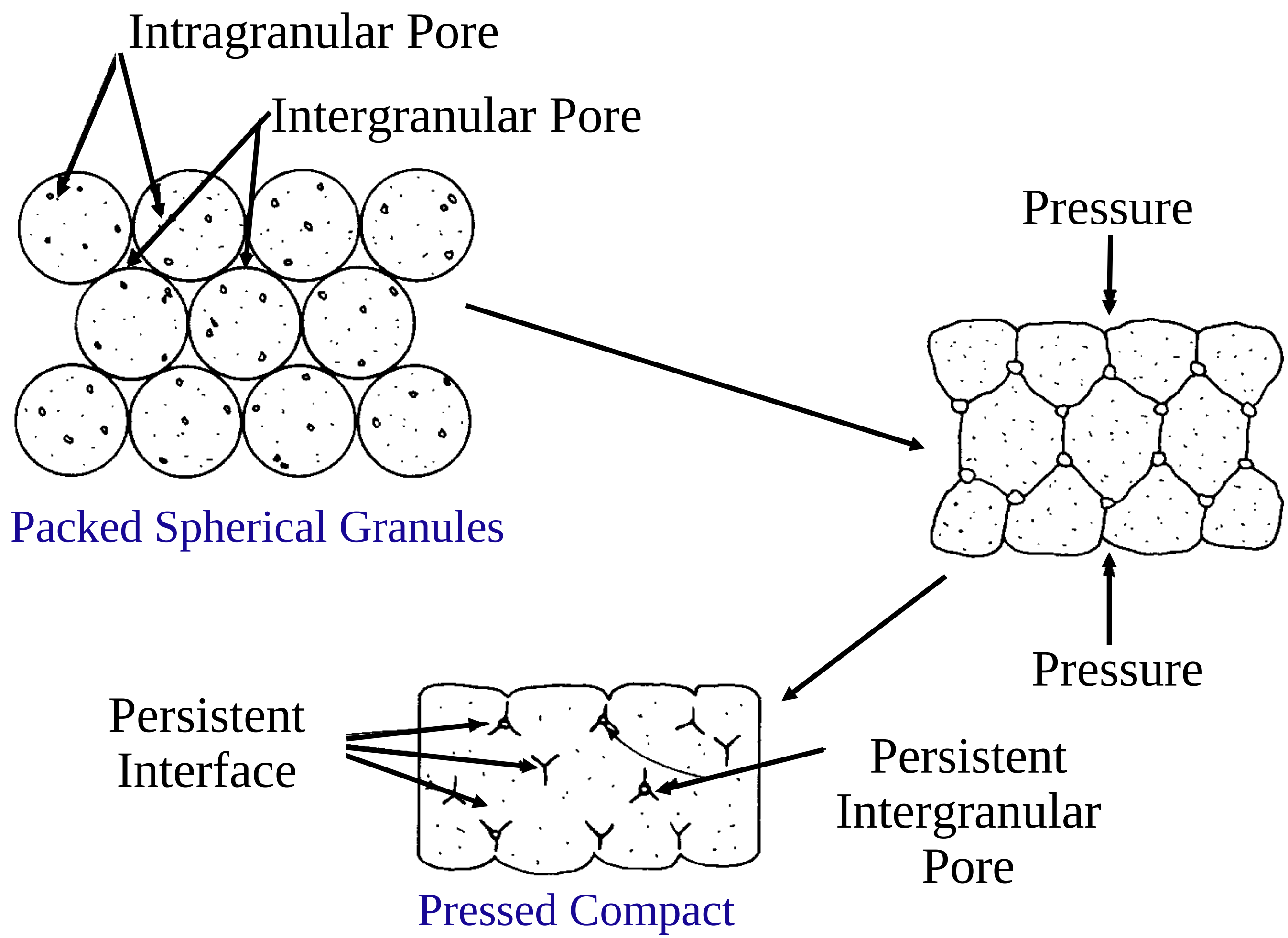
Onoda (1995)

Granule Density Can Be Measured Using Mercury Intrusion Porosimetry



A good granule density is ~ 45-50%

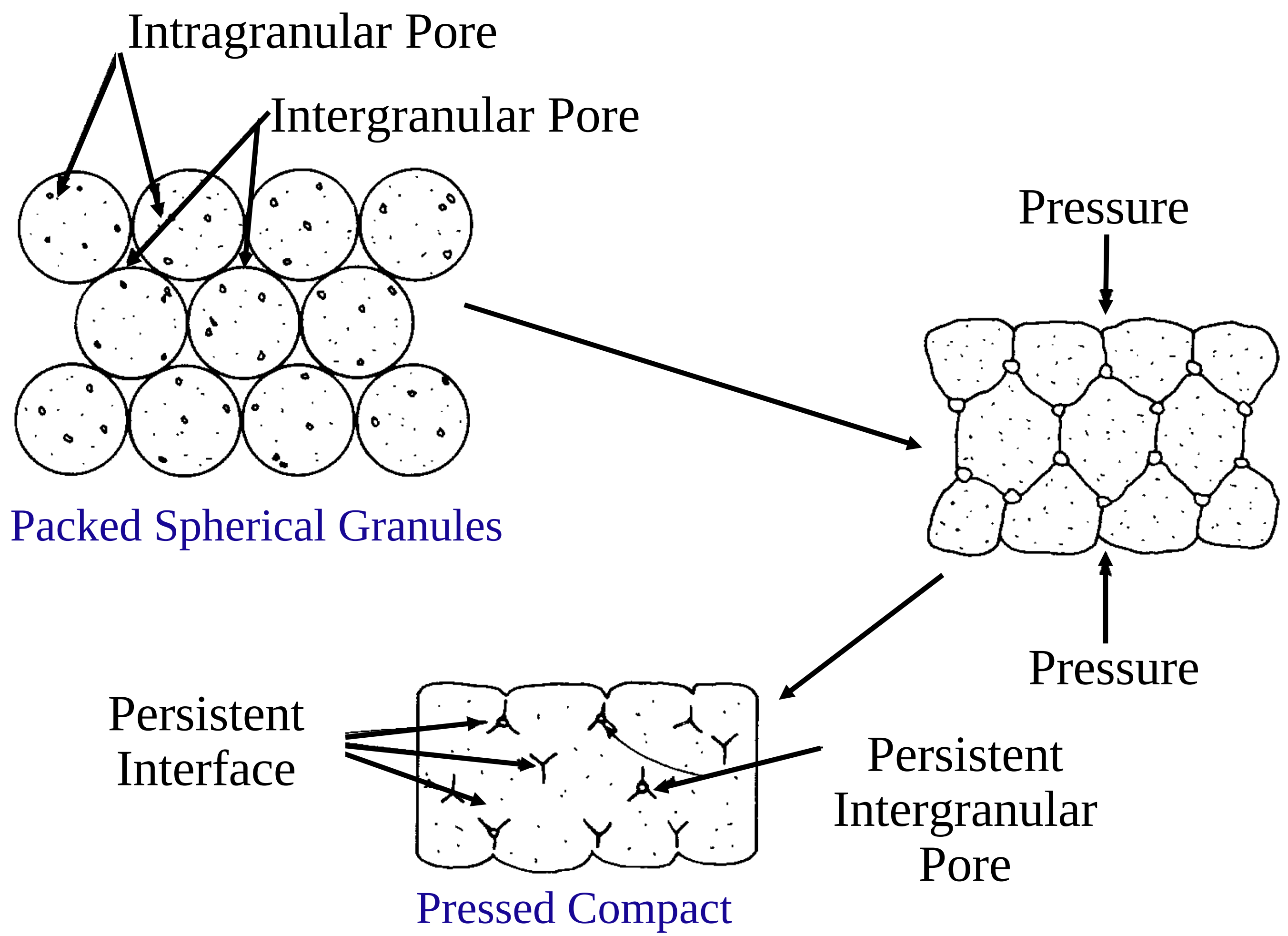
Granule Packing & Deformation Influence Compaction & Density



Reed (1988, 1995)

Sandia National Laboratories

Granule Packing & Deformation Influence Compaction & Density



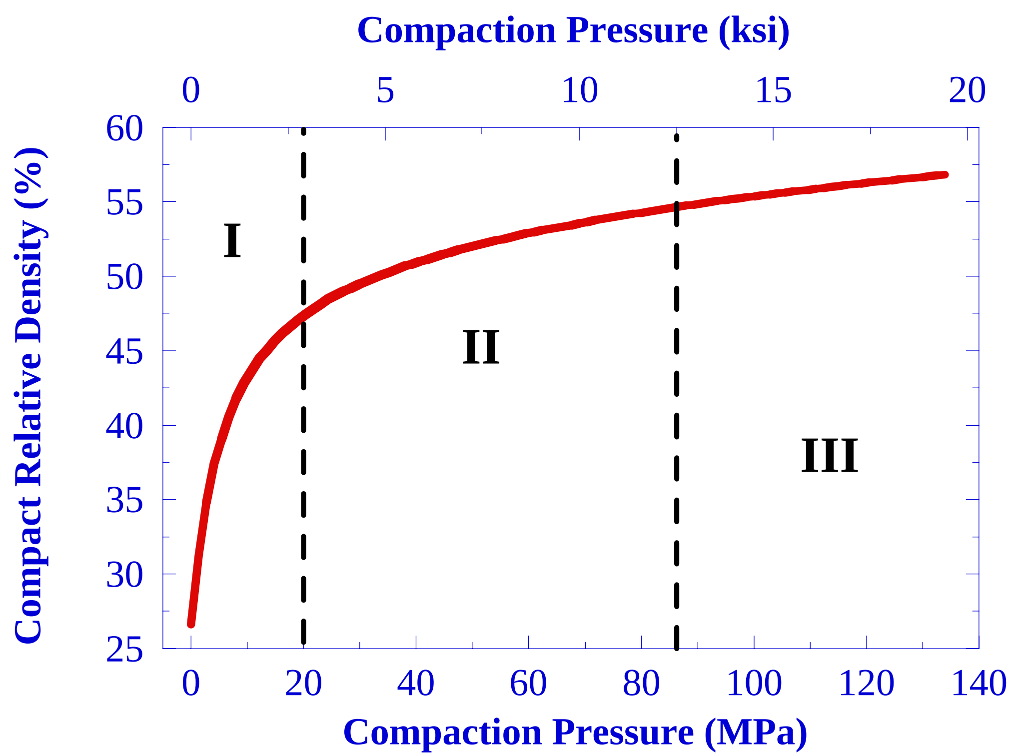
Reed (1988, 1995)

The Compaction Curve Provides A Measure Of Compaction Response

Region I
Compact density is extremely sensitive to variations in pressing pressure.

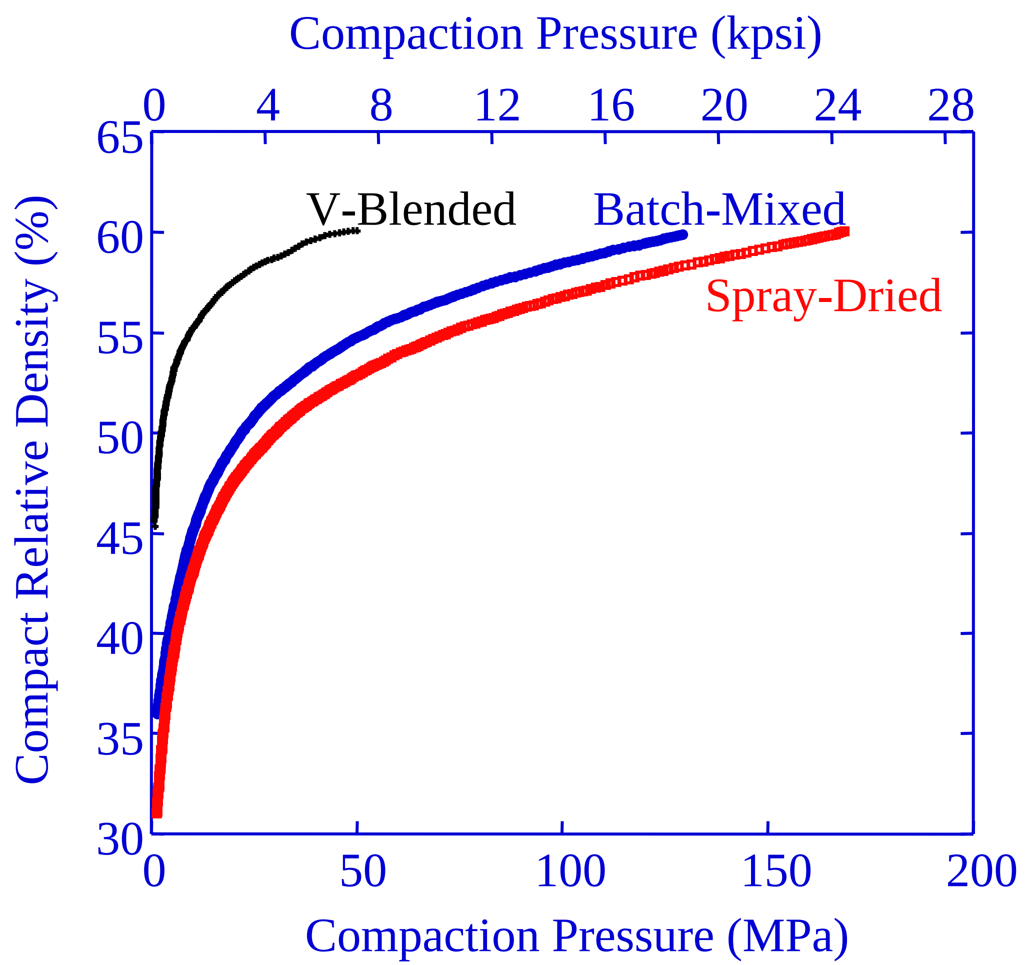
Region II
Most desirable region for manufacturing.

Region III
Compact density relatively insensitive to forming pressure.



Compaction Response Can Vary Appreciably With Granulation

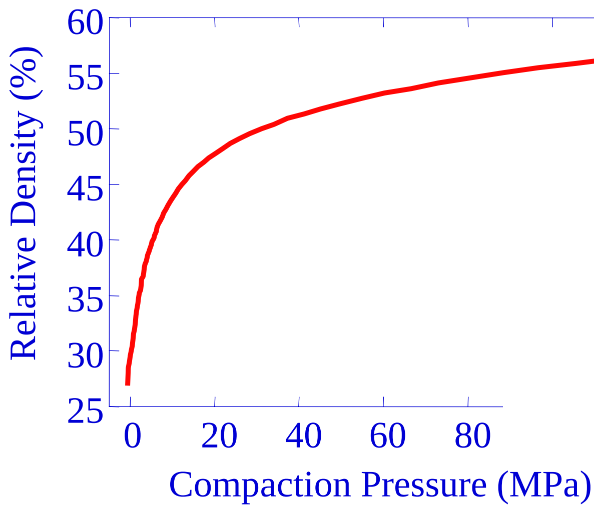
Granulated 94% Al_2O_3 Powder Compaction

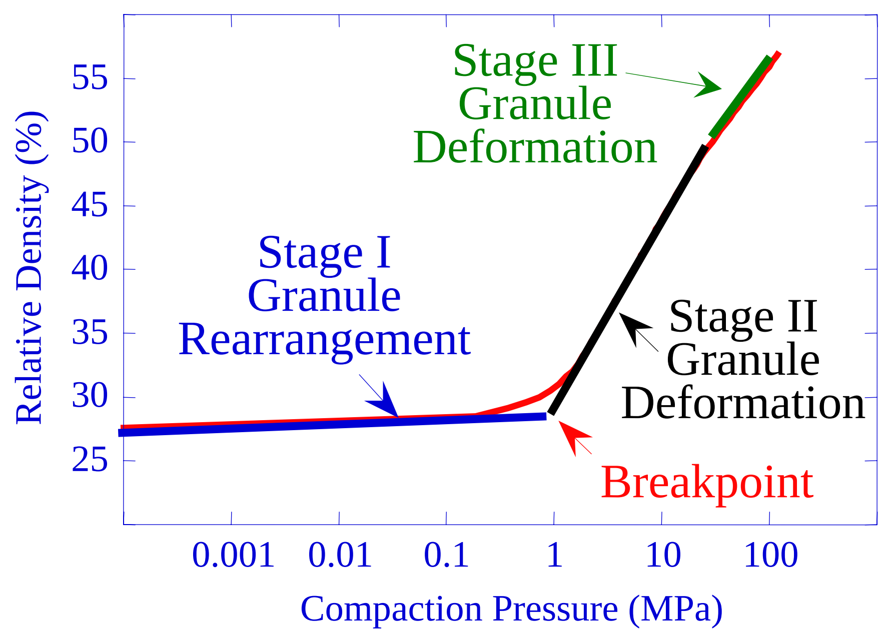


Sandia National Laboratories

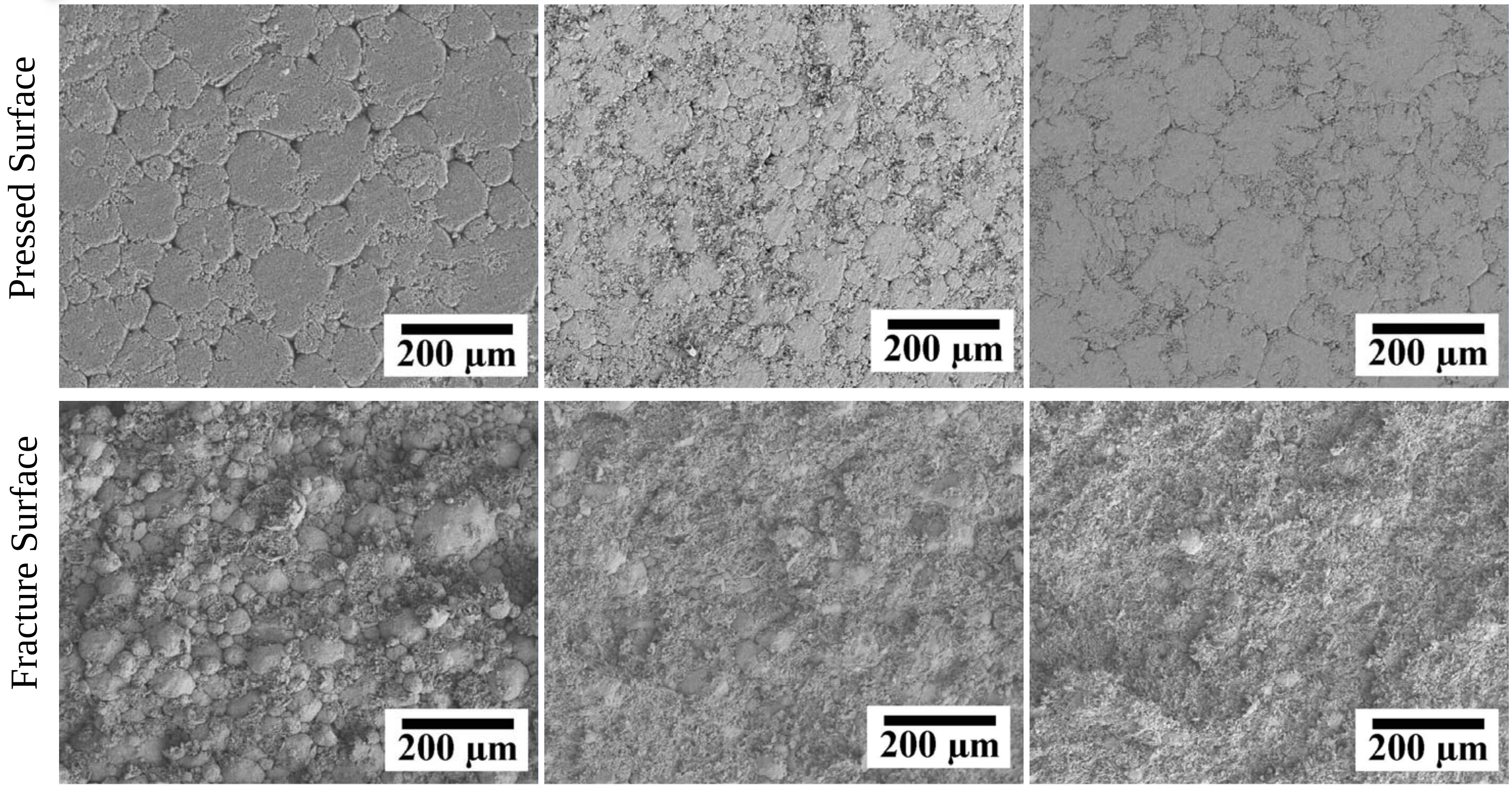
Density vs. Log Pressure Reveals Three Compaction Stages & A Breakpoint

Spray-Dried 94% Al_2O_3 Powder Compaction





Remnants of Spray-Dried Al_2O_3 Granules Disappear At ~69 MPa



17.2 MPa
2,500 psi

68.9 MPa
10,000 psi

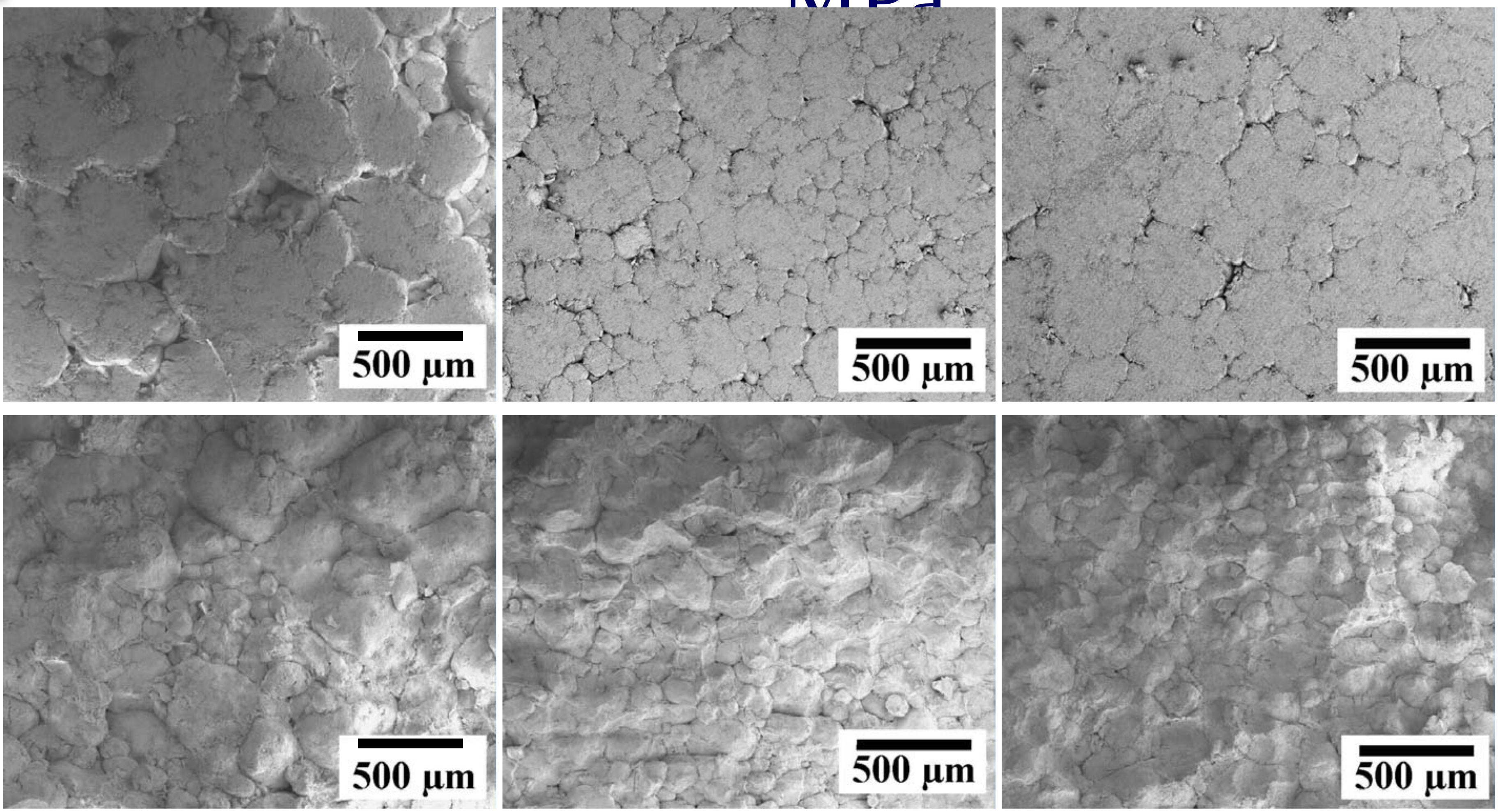
137.9 MPa
20,000 psi

Sandia National Laboratories

Remnants of Spray-Granulated Al_2O_3 Granules Persist At 137.8 MPa

Pressed Surface

Fracture Surface



17.2 MPa
2,500 psi

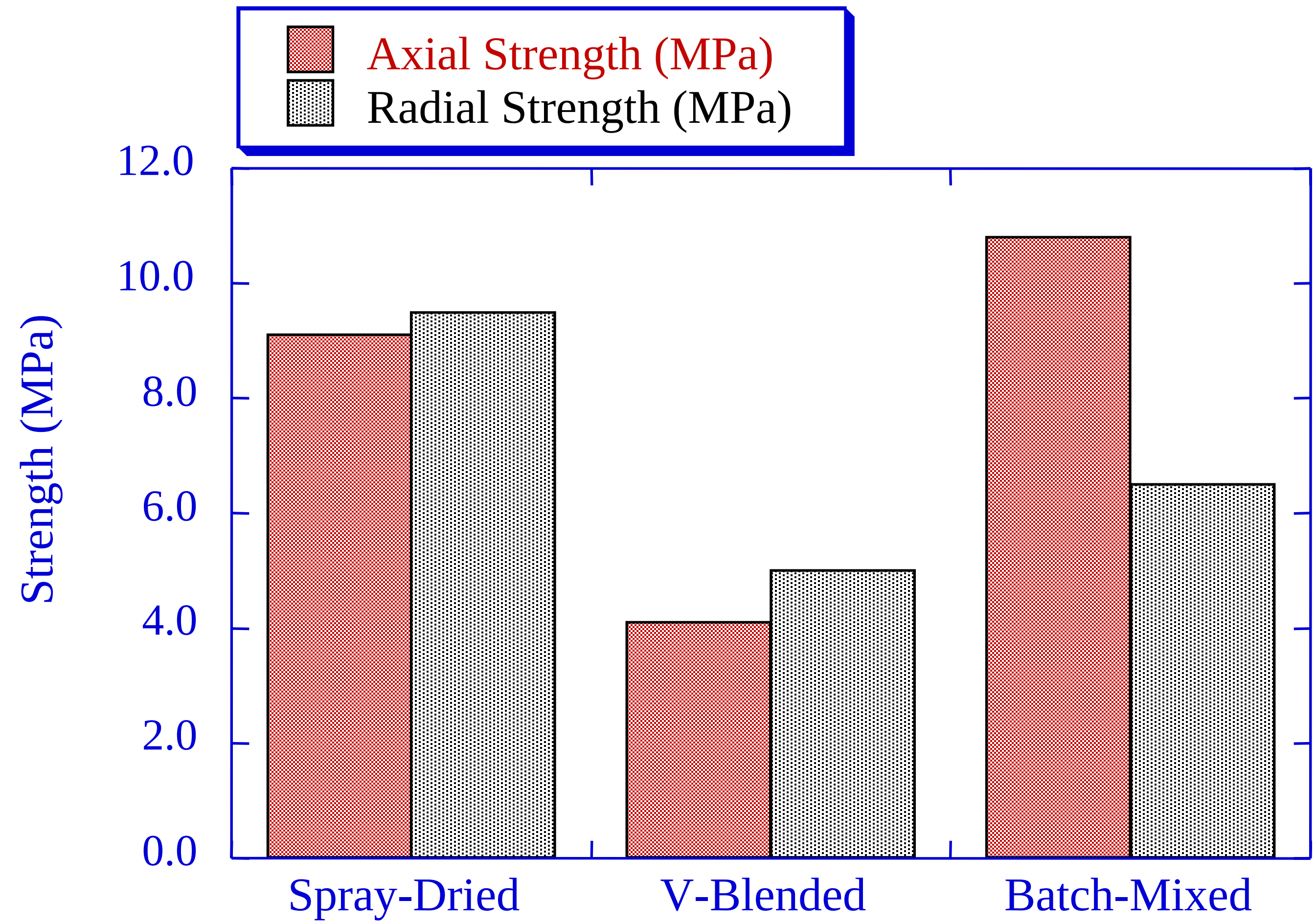
68.9 MPa
10,000 psi

137.8 MPa
20,000 psi

Sandia National Laboratories

Softer More Deformable Granules Produce Stronger Powder Compacts

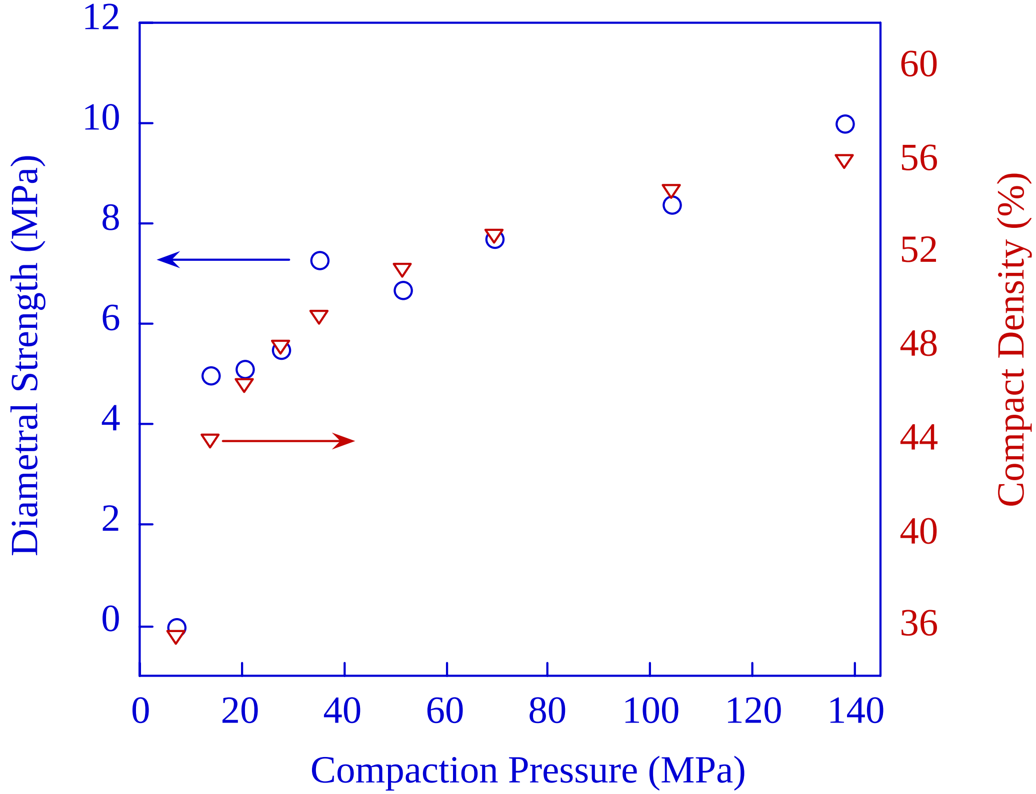
Granulated 94% Al_2O_3 Powder Compact Strength



Sandia National Laboratories

Strength Increases With Compact Density & Compaction Pressure

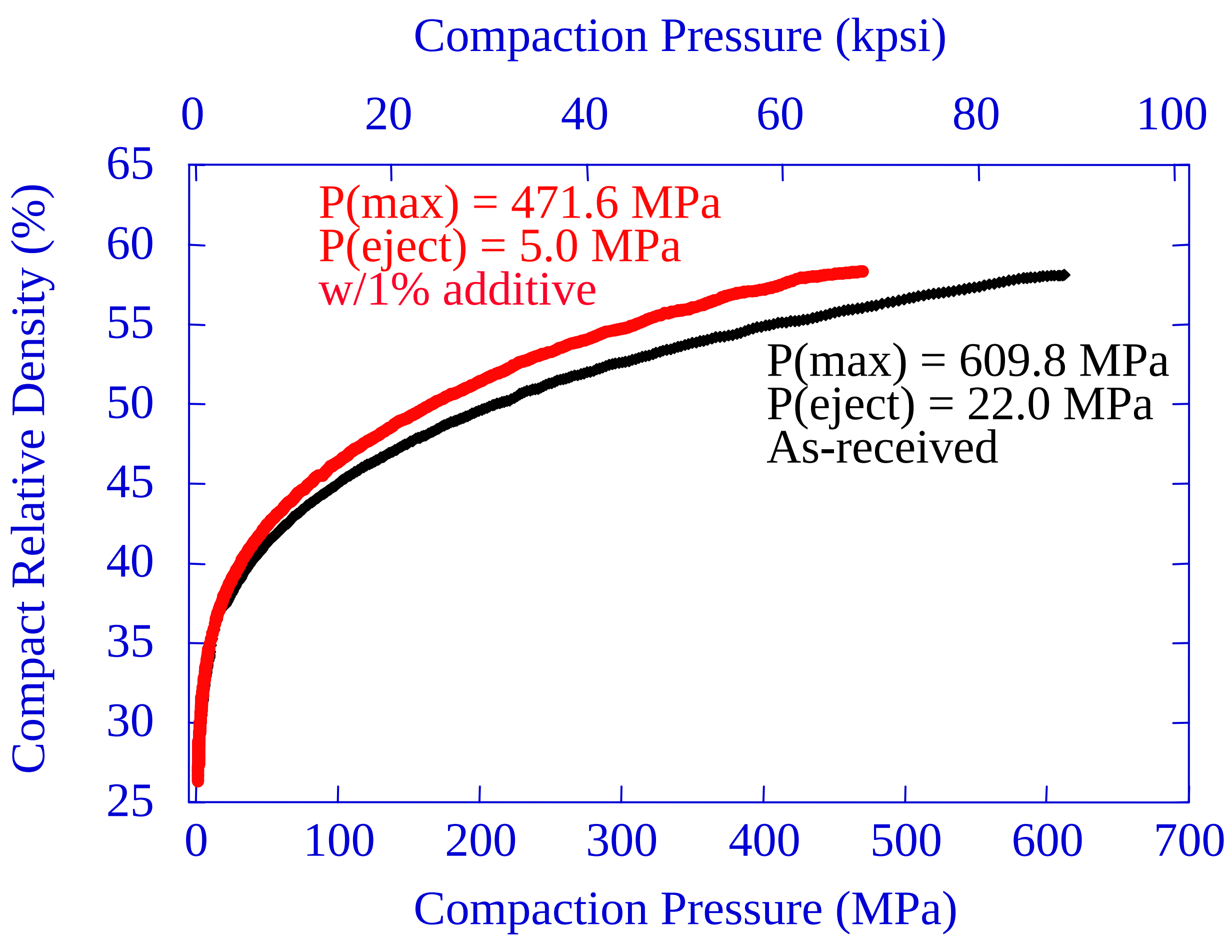
Powder	Axial Strength (MPa)	Tensile Strength (MPa)
94% Al_2O_3	1.5 ± 0.1	6.4±0.2
99.5% Al_2O_3	6.4±0.2	4.4±0.0



Sandia National Laboratories

Ejection Force & Springback Increase With Pressing Pressure

Ceramic Powder Compaction Response

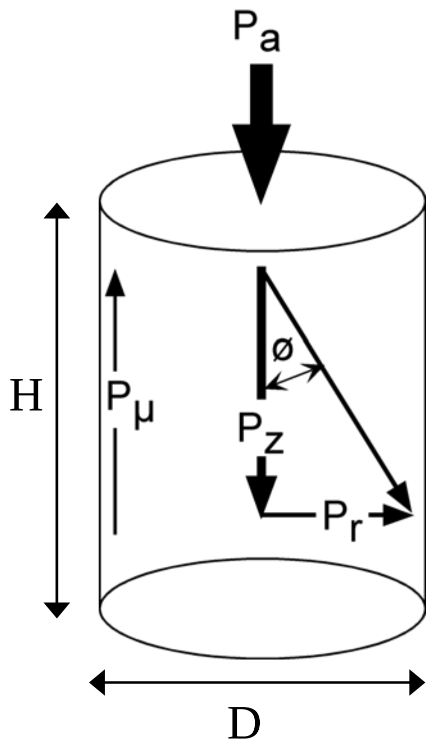


Friction Determines How Pressure Is Transmitted Through The Compact

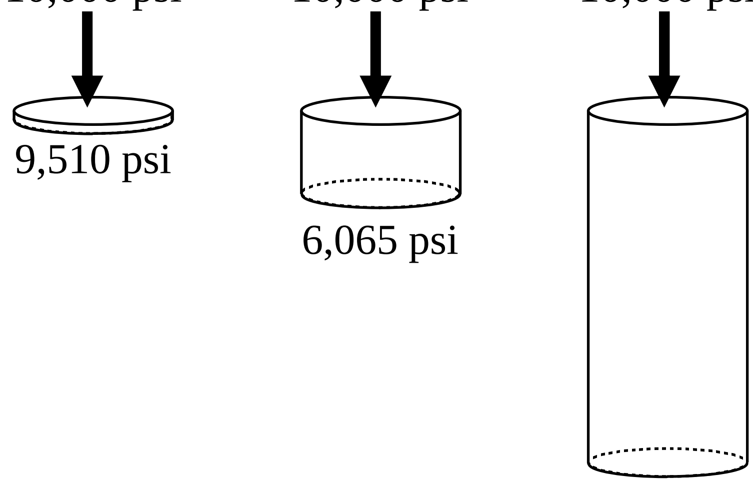
$$P_z = P_a \exp(-4/D \cdot z \cdot \mu \cdot \alpha)$$

$$\mu \cdot \alpha = \ln (P_z/P_a) / (-2/r \cdot z)$$

$$\alpha = \tan ø$$



H/D =	0.1	1	4
P_a =	10,000 psi	10,000 psi	10,000 psi
P_H =	9,510 psi	6,065 psi	1,350 psi

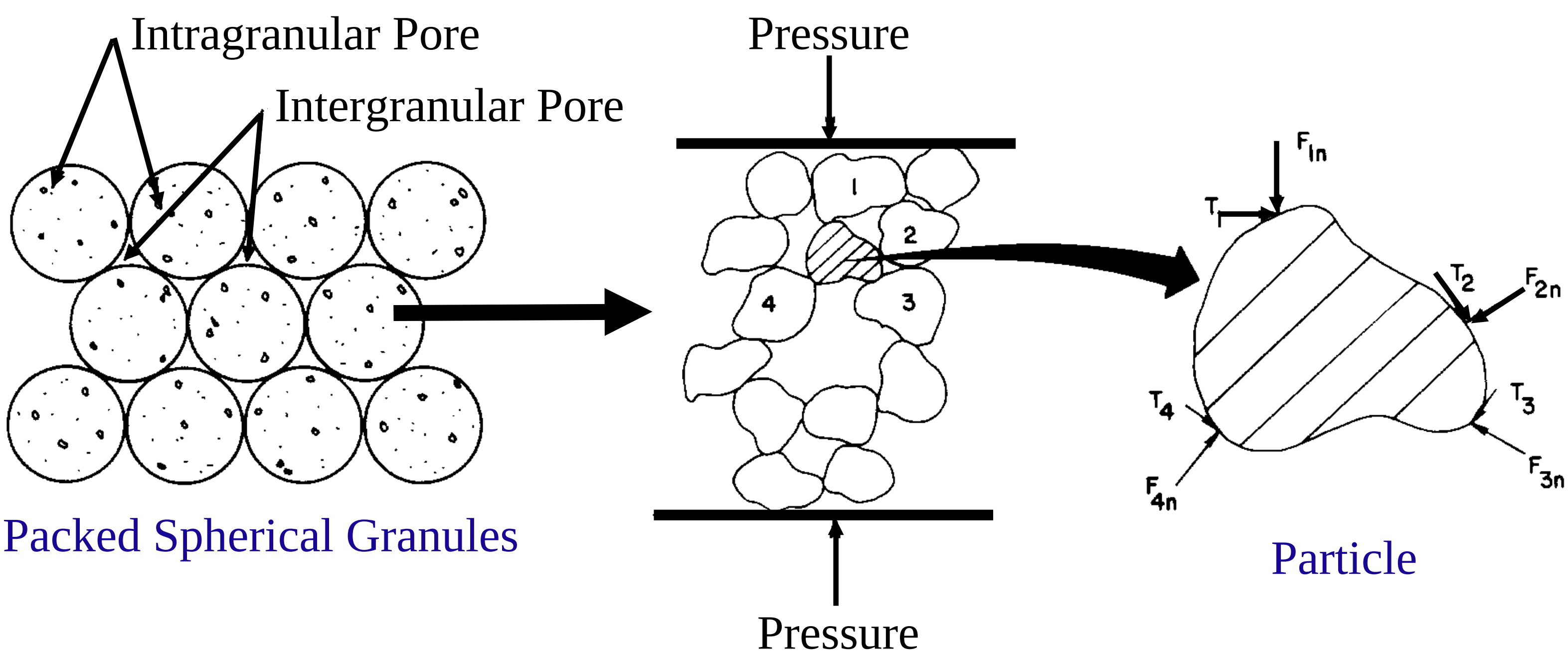


Transmitted pressure calculated for different H/D values (μ = 0.25 and α = 0.5)

Janssen (1895)

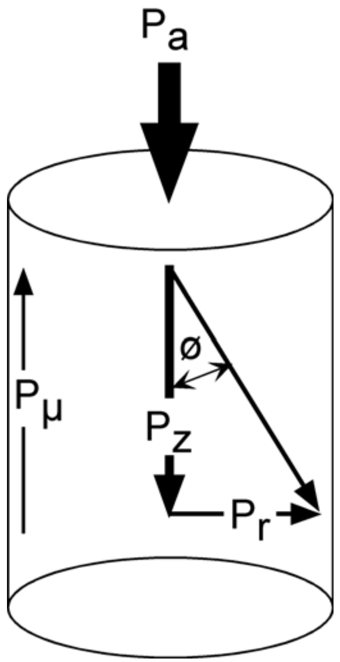
Sandia National Laboratories

Particle Size & Shape Influence (Powder) Internal Friction



Reed & Runk (1976)

The Applied & Transmitted Pressure Provide Information on μ & α



$P_r = \alpha \bullet P_z$

$P_\mu = \mu \bullet \alpha \bullet P_z$

Jansen's Analysis

$P_z = P_a \exp(-2/r \bullet z \bullet \mu \bullet \alpha)$

$\mu \bullet \alpha = \ln (P_z/P_a) / (-2/r \bullet z)$

$\alpha = \tan ø$

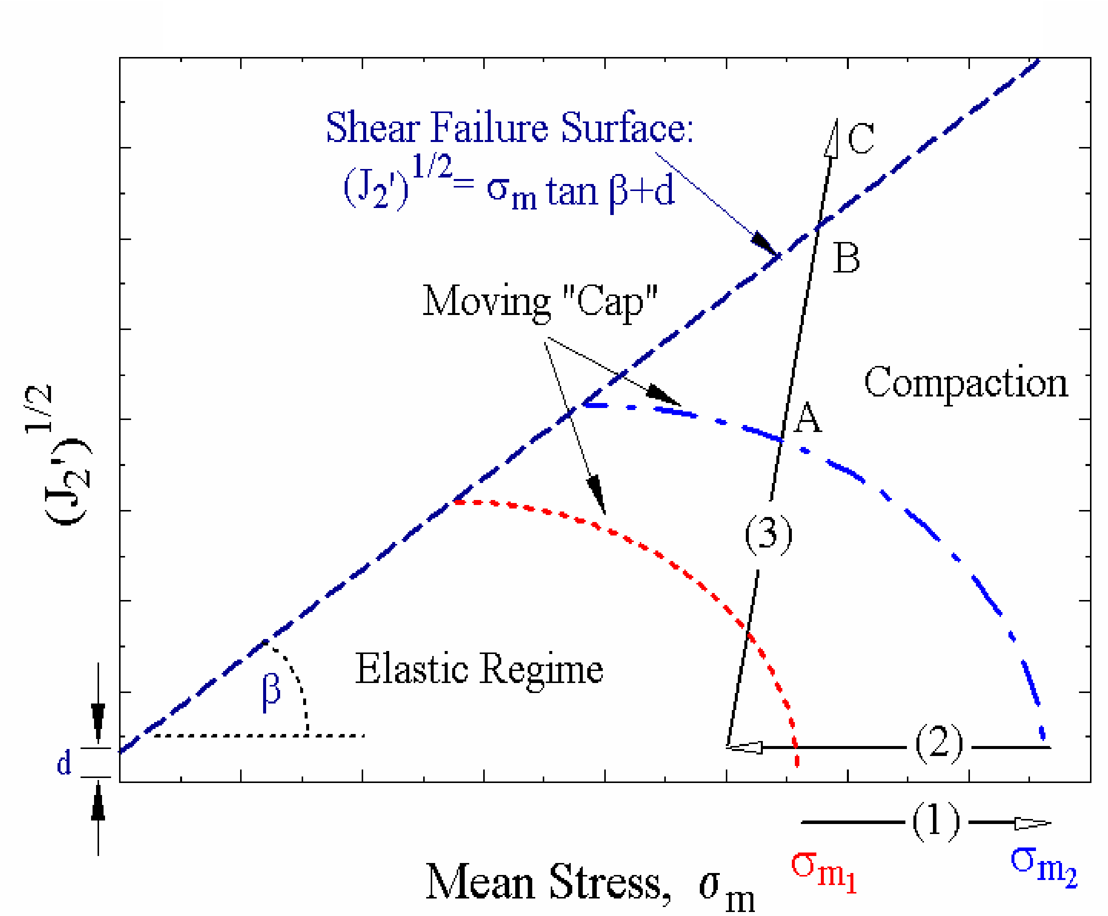
The PTC Slide Coefficient, η

$\ln \eta = 2r/z \bullet \ln (P_z/P_a)$

$\ln \eta / -4 = \mu \bullet \alpha$

μ ~ die wall friction α ~ powder friction

Ceramic Powder Compaction Is Described By A Cap-Plasticity Model

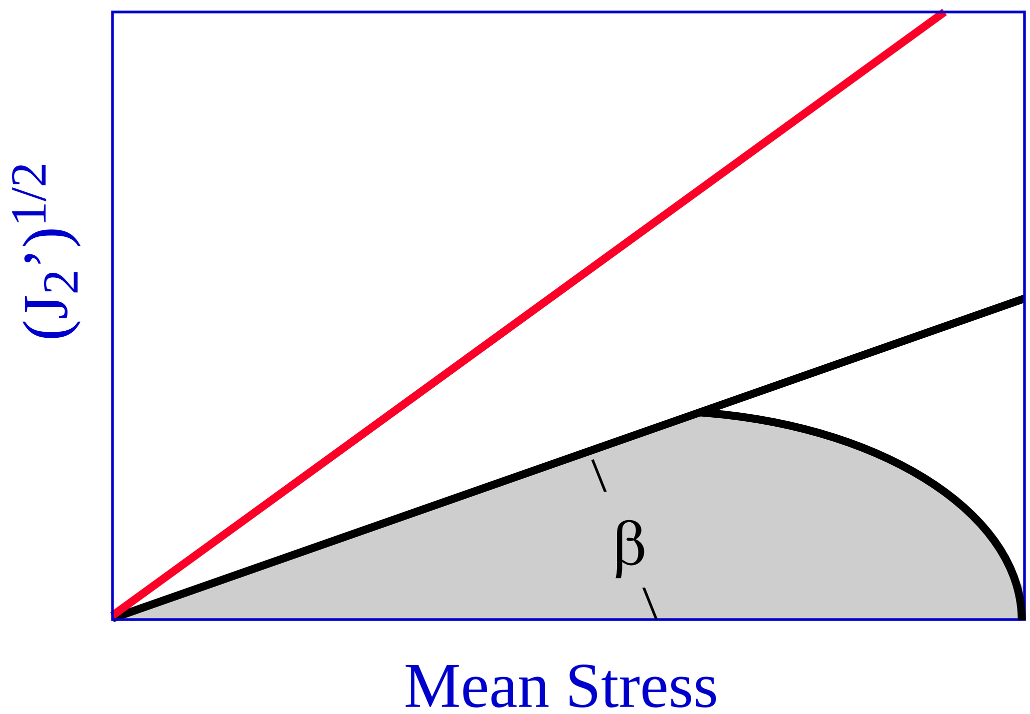


Model Features

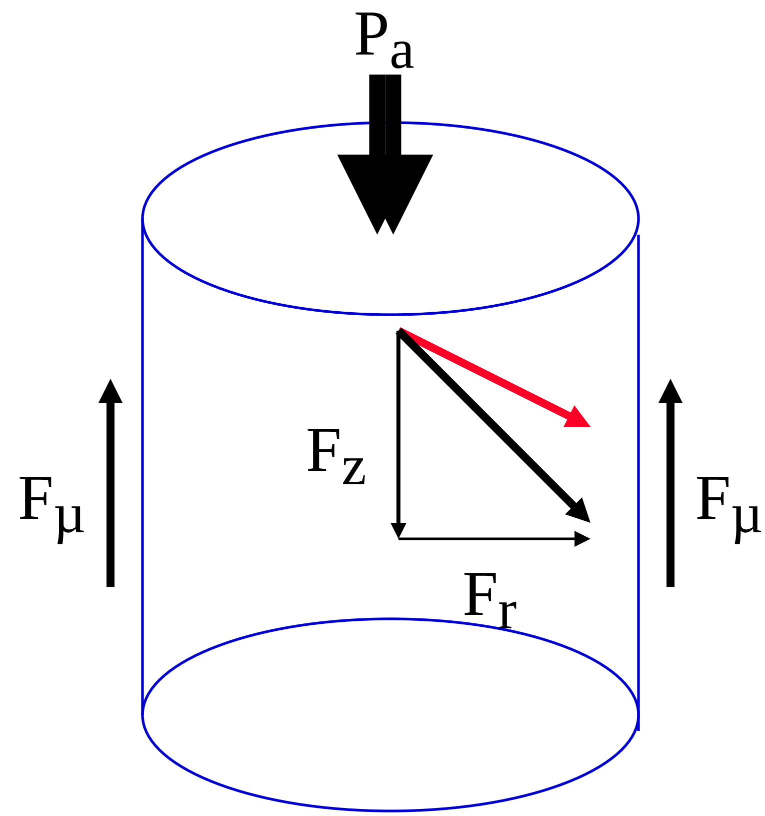
- Stationary shear failure surface
  - cohesion, d
  - angle of internal friction, β.
- Elliptical Cap that moves
- An elastic regime circumscribed by the two foregoing surfaces

Sandia National Laboratories

Powders Having A Larger Friction Angle β Are More Difficult To Press

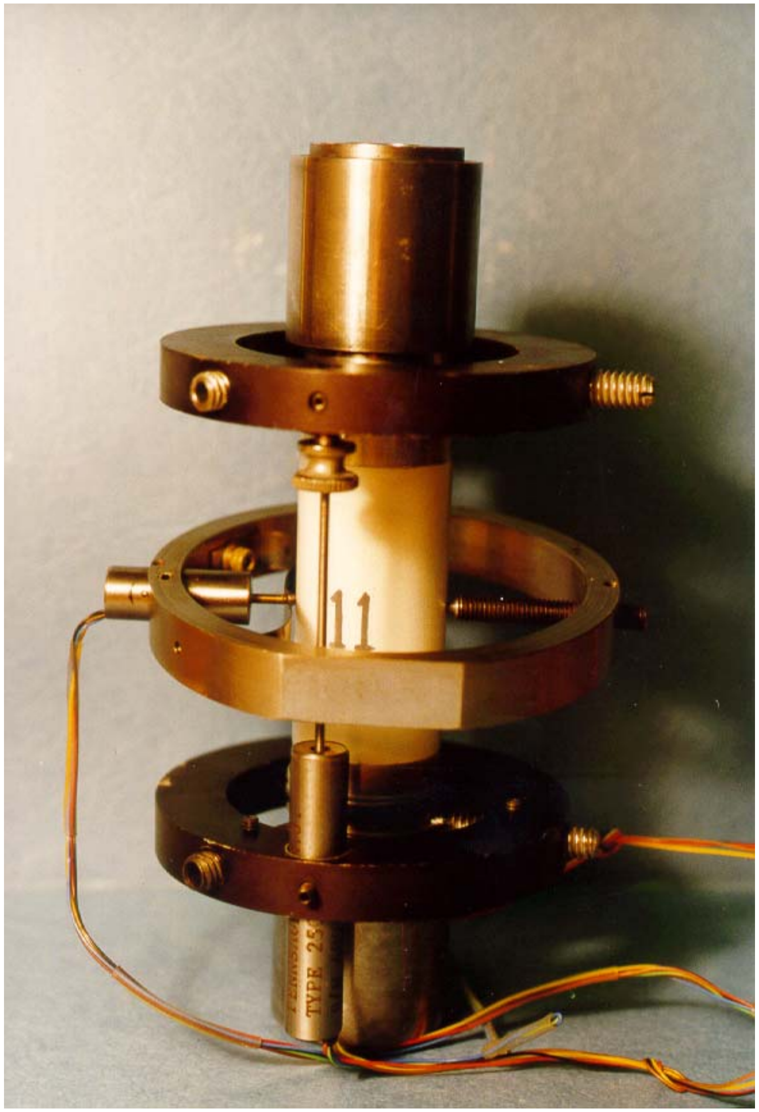


Cap Model



Jansen Model

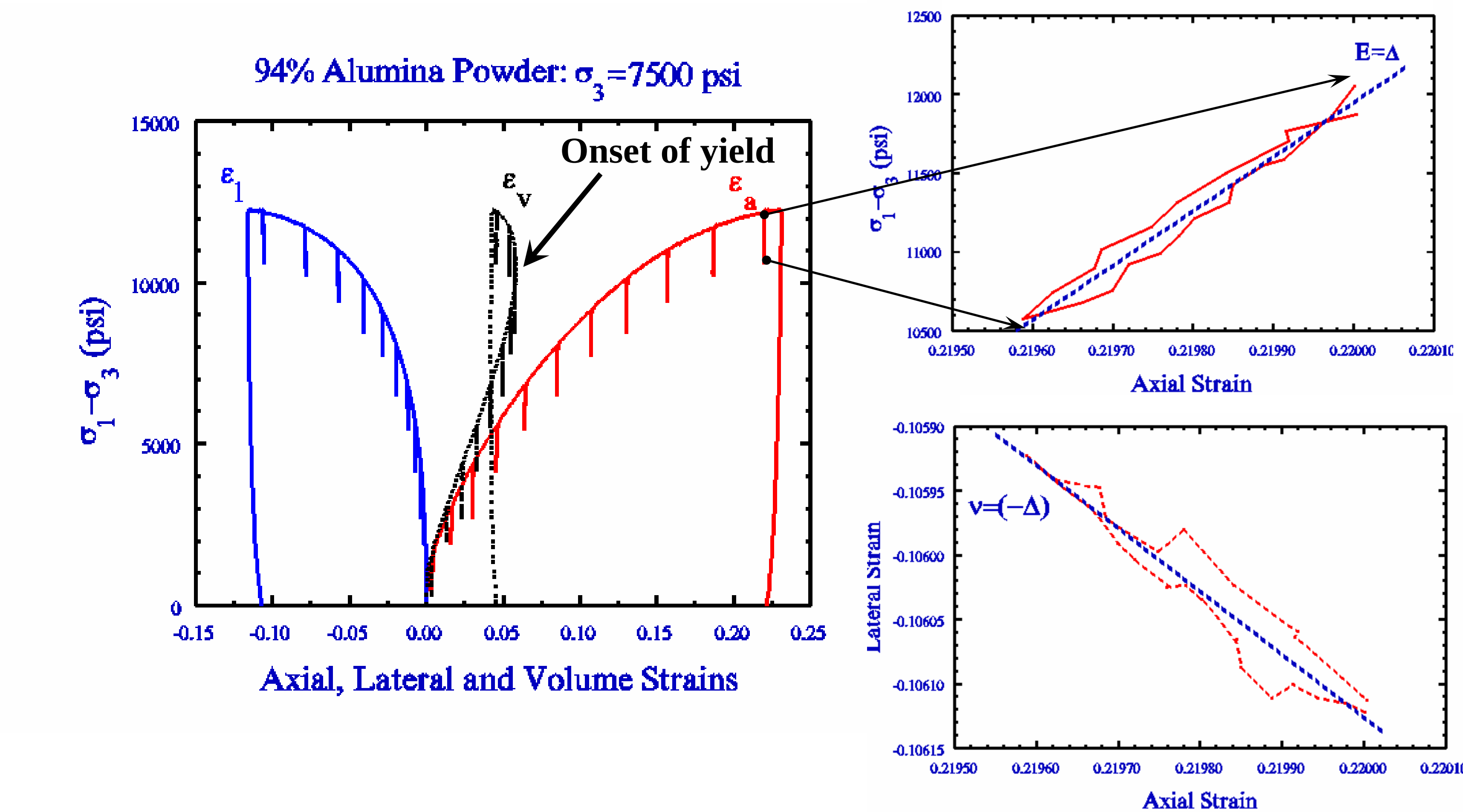
Powders Were Characterized In Hydrostatic & Triaxial Compression



Triaxial Compression Tests

- 2.54 cm dia. by 5.08 cm tall compacts
- Pressurized to target pressure (i.e., the hydrostatic forming pressure)
  - Monitored sample strains
- Hydrostatic pressure looped at target pressure
  - Determined K
- Loaded axially (i.e., deviatoric stress)
- Completed periodic, axial unload/reload loops
  - Determined E, ν
  - Calculated G from K, E, & ν

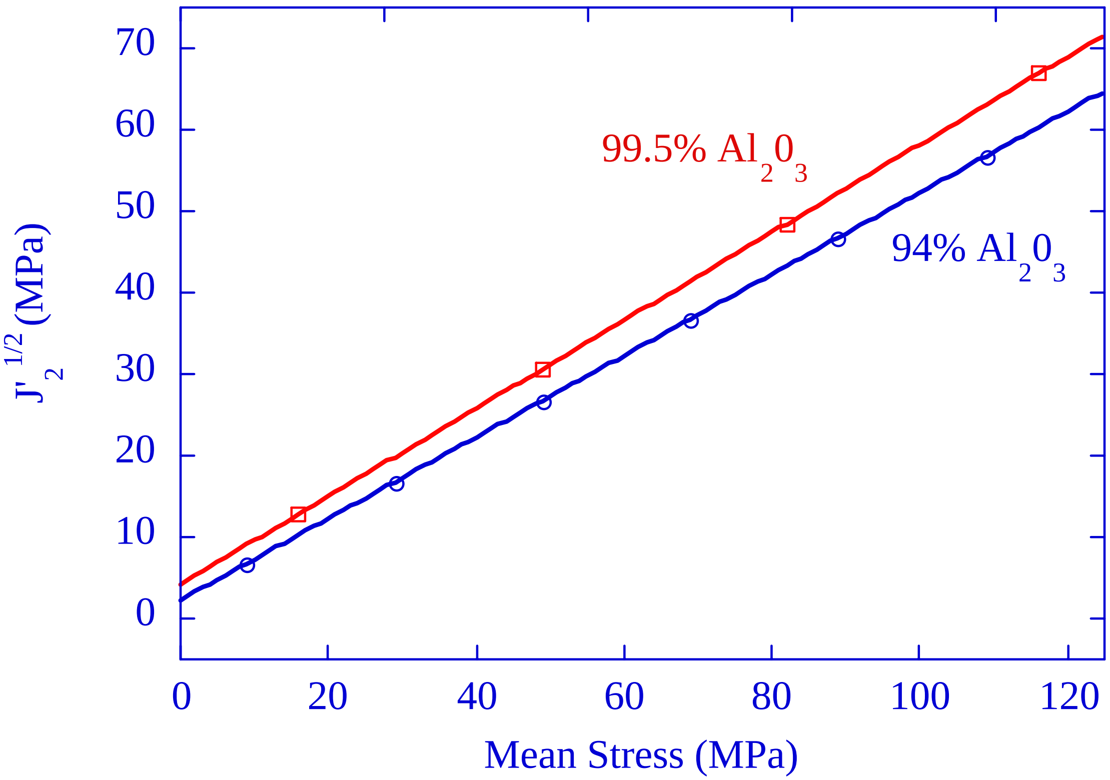
Mechanical Properties Were Determined From Stresses & Strains



Sandia National Laboratories

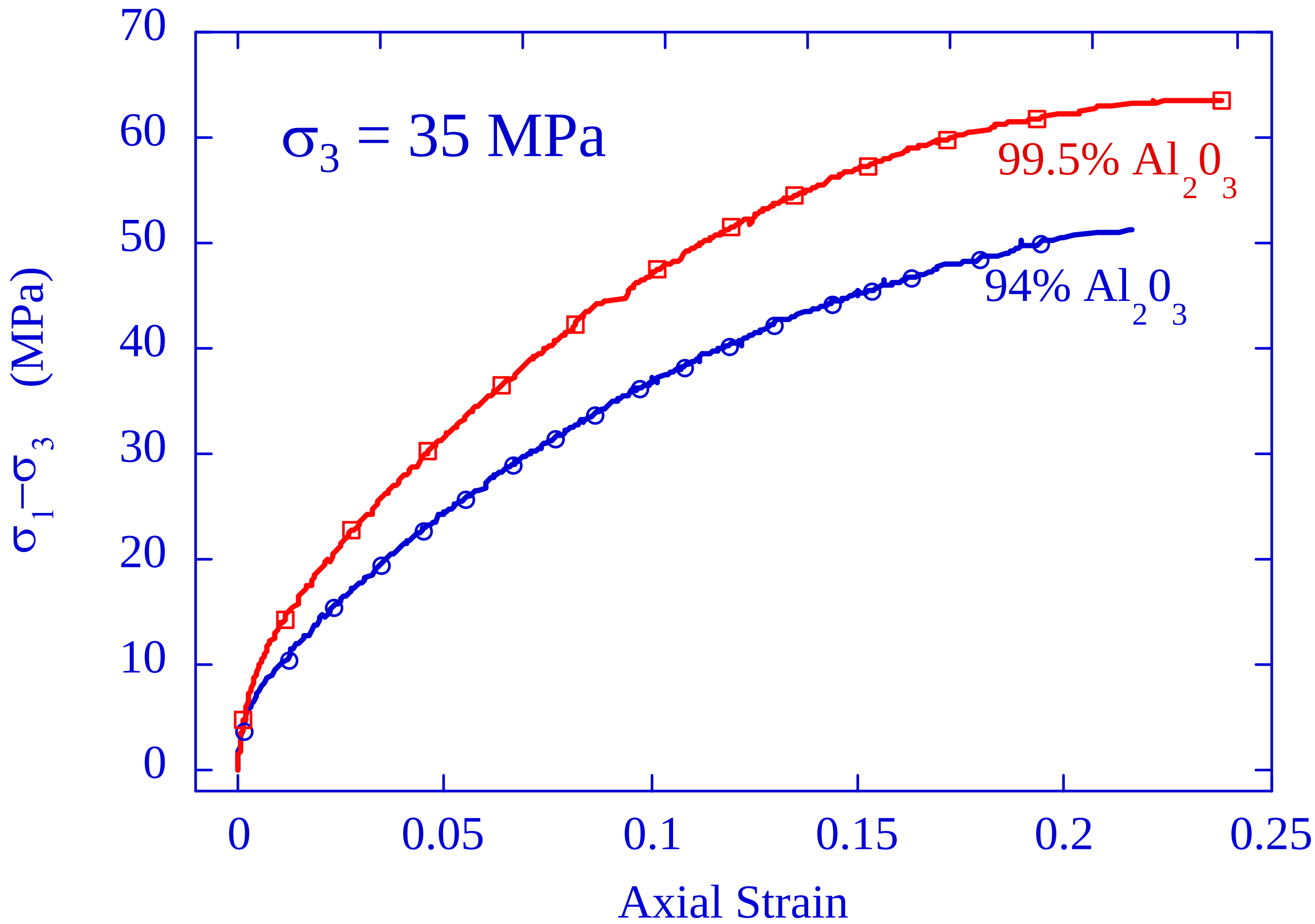
More Pressable 94% Al_2O_3 Has A Smaller Angle of Internal Friction, β

Powder	Cohesion (MPa)	Angle of internal friction (degrees)	Tan β
94% Al_2O_3	2.3	26.6	0.50
99.5% Al_2O_3	4.2	28.4	0.54



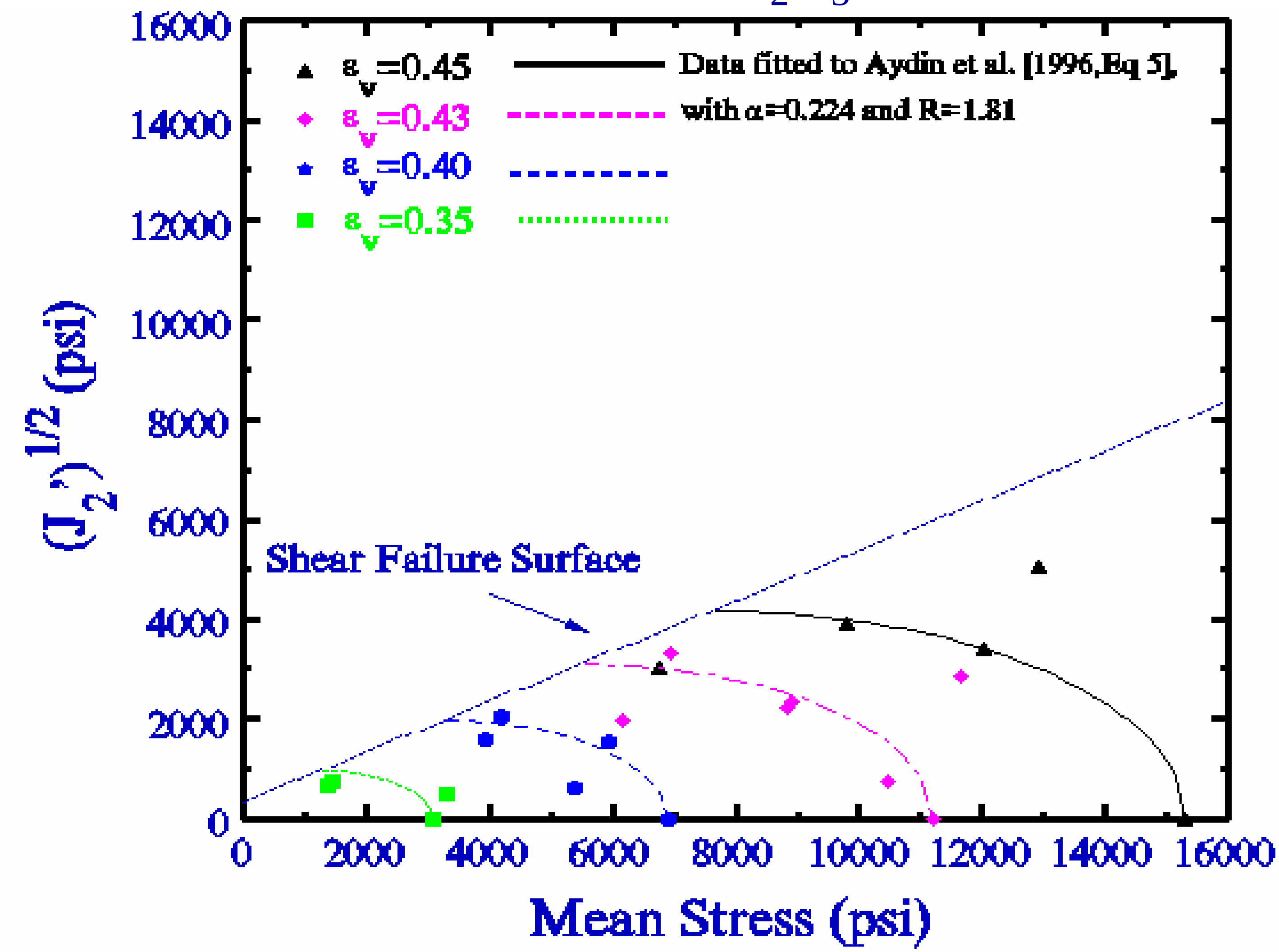
Sandia National Laboratories

Harder To Press 99.5% Al_2O_3 Is Stronger In Triaxial Compression



Drucker-Prager Parameters Were Determined To Support Modeling

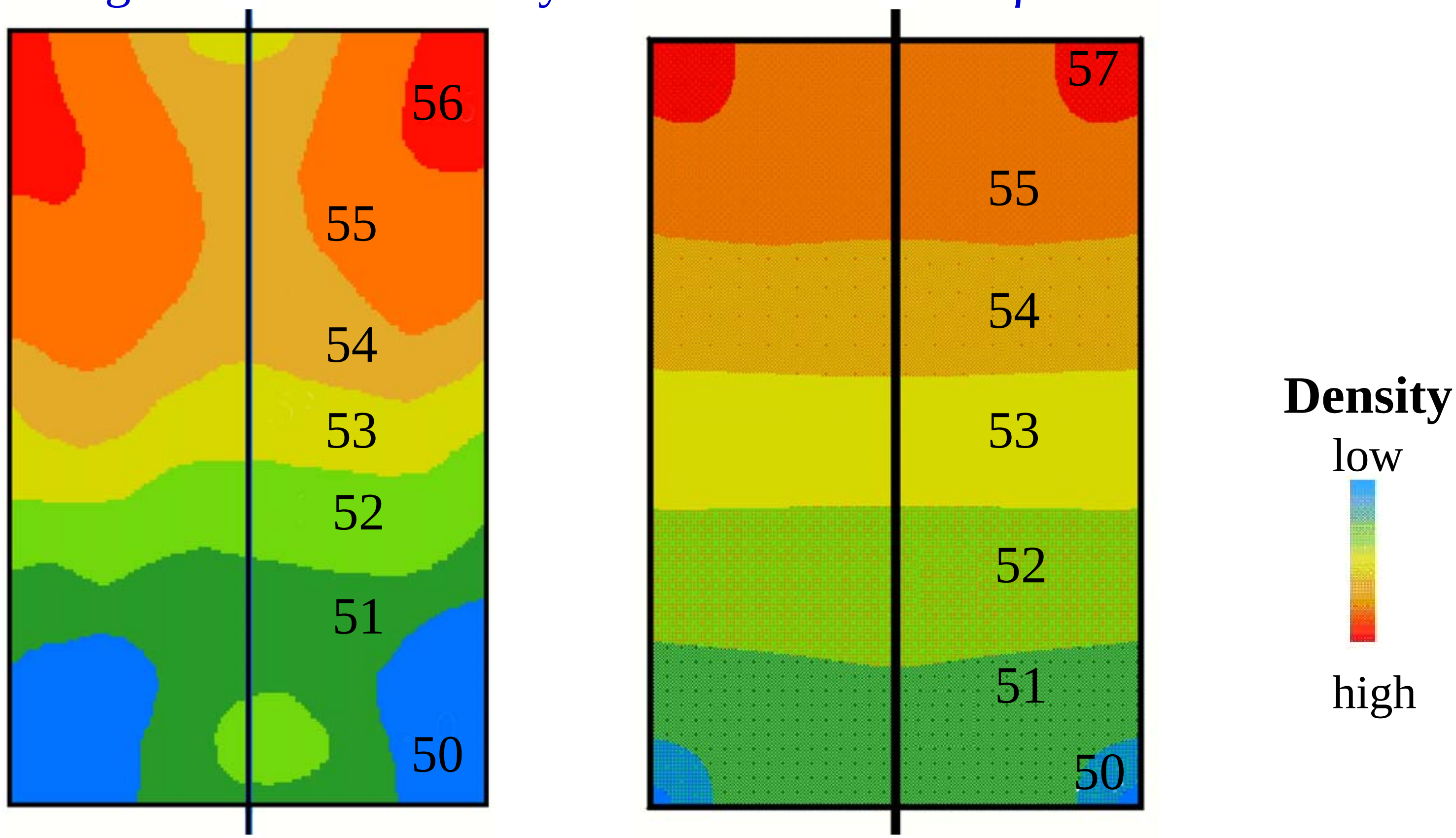
94% Al_2O_3



Model Simulations Show Excellent Agreement With Experimental Results

94% Alumina

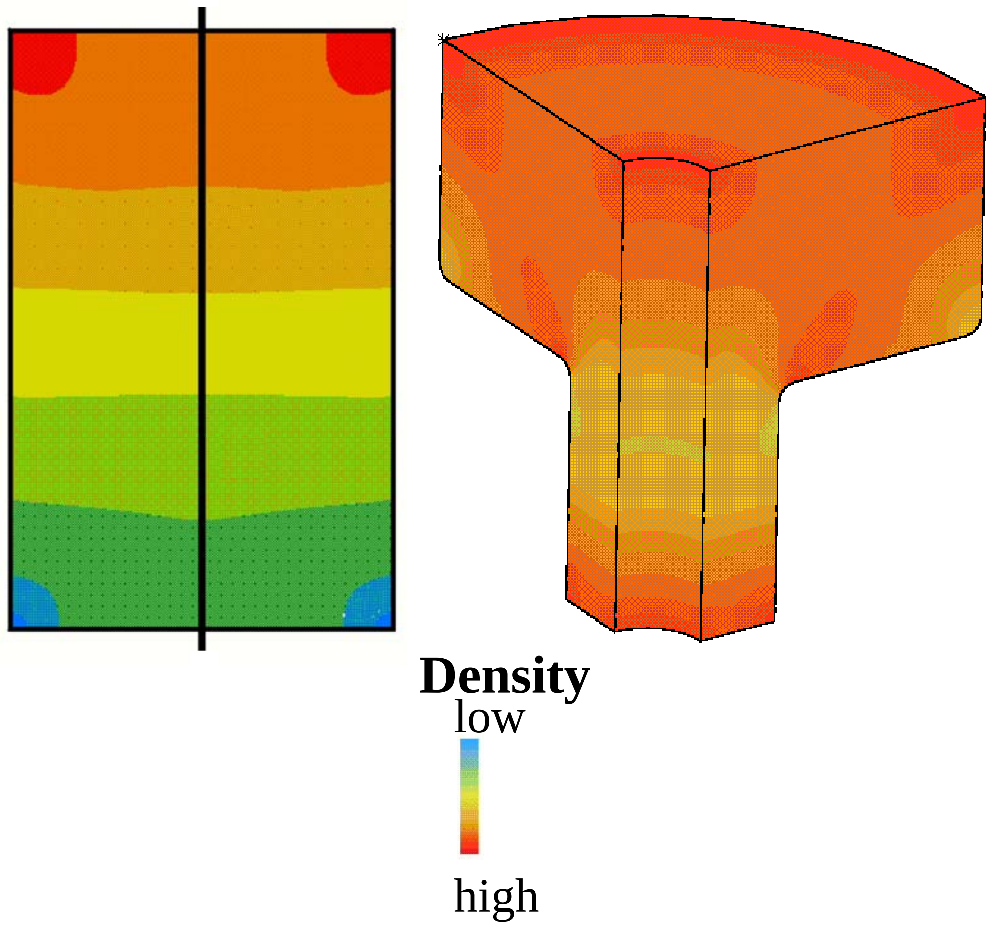
Single-Action Uniaxially Pressed at 69 MPa • μ = 0.2



Ultrasound Measured Density Gradients

FE Model Predicted Density Gradients

The Simulation (CPU) Time Also Provides A Measure Of Pressability



FE Compaction Simulations

<u>Cylindrical Compact</u>

- 2.2 cm Dia x 6.7 compacted to 3.5 cm

• Single-Action, Top-Down Pressing

- 94% alumina - 747 CPU s - 1.0 X

- 99.5% alumina - 9919 CPU s - 13.3X

<u>Bushing Compact</u>

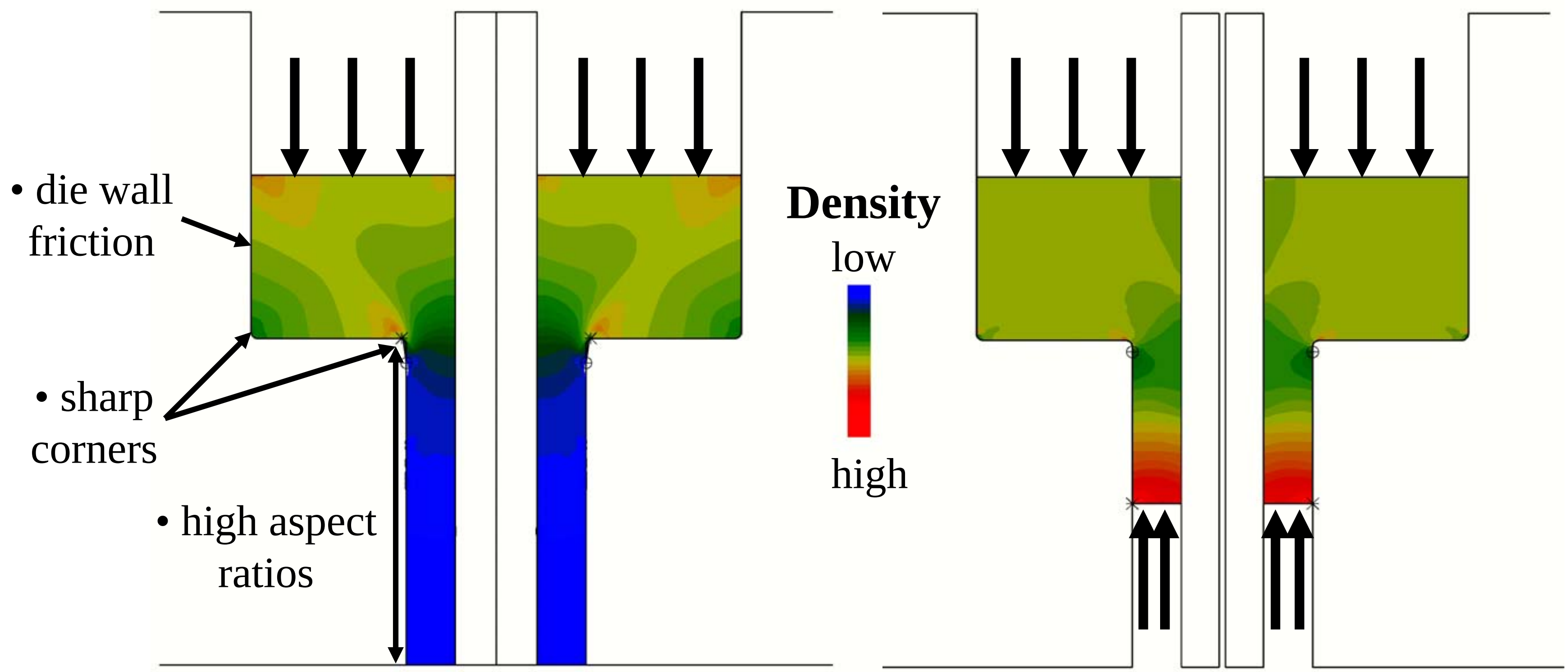
• Balanced, Dual-Action Pressing

- 94% alumina - 1297 CPU s - 1.7X

- 99.5% alumina - 154,200 CPU s - 206X

Modeling & Simulation Can Guide Die Design & Pressing

Cross sections of 3D Finite Element Method (FEM) compaction model simulations predict the density gradients in a complex component geometry

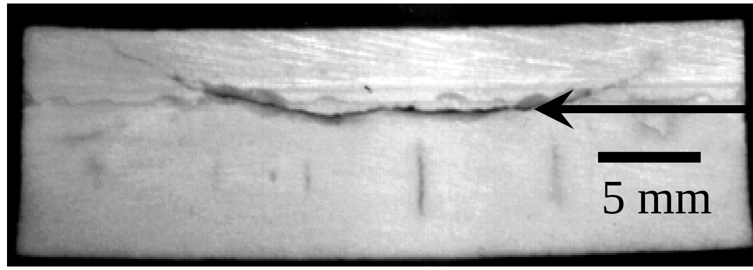


Single-Action Pressing

Dual-Action Pressing

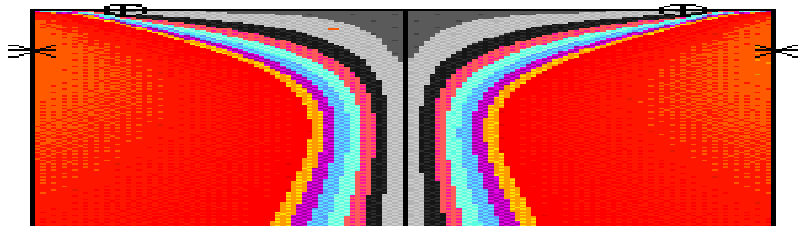
Defects Are Produced If The Stress Path Intersects The Shear Failure Surface

Pressed & Sintered
Lead Zirconate Titanate

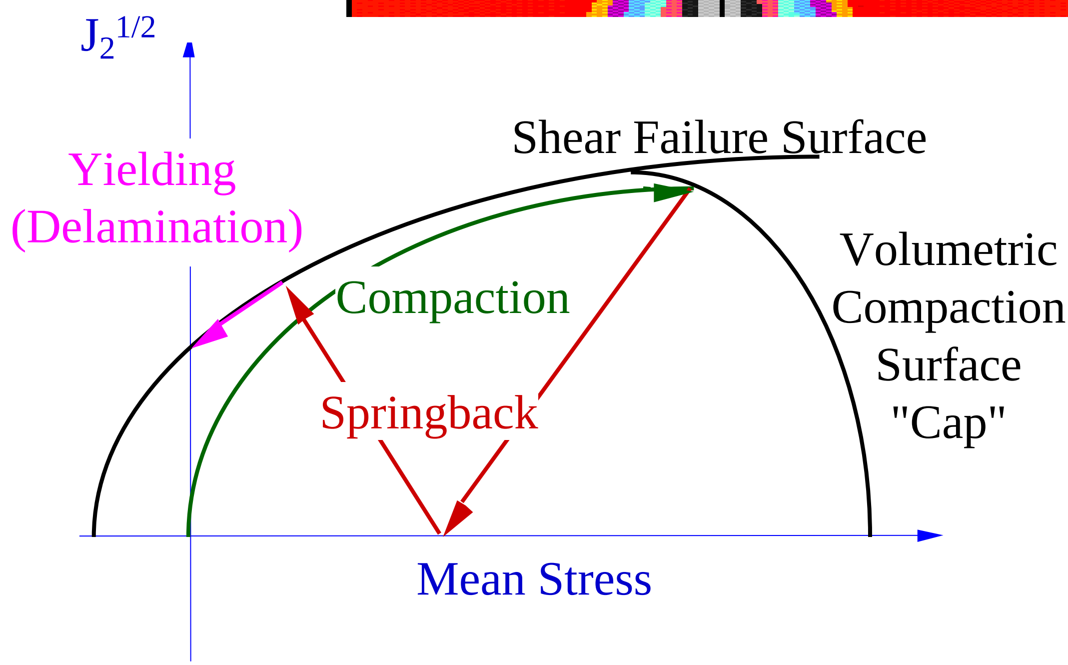


End Cap
Defect

FEM Simulation

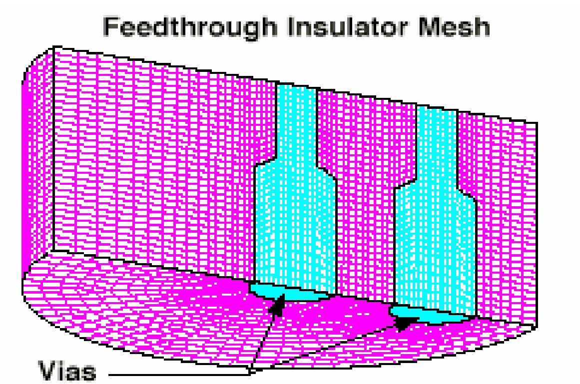


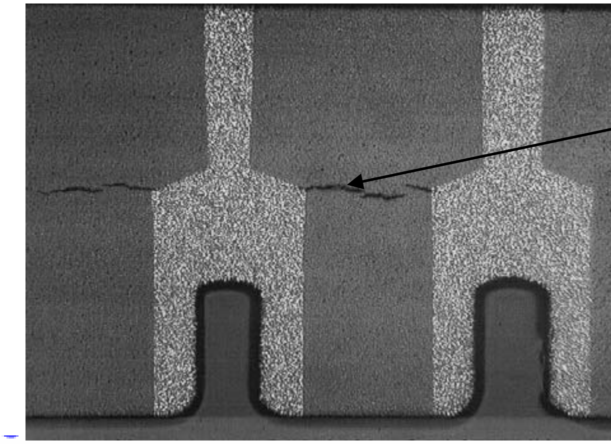
Shear Stress Map
-45 psi
1000 psi



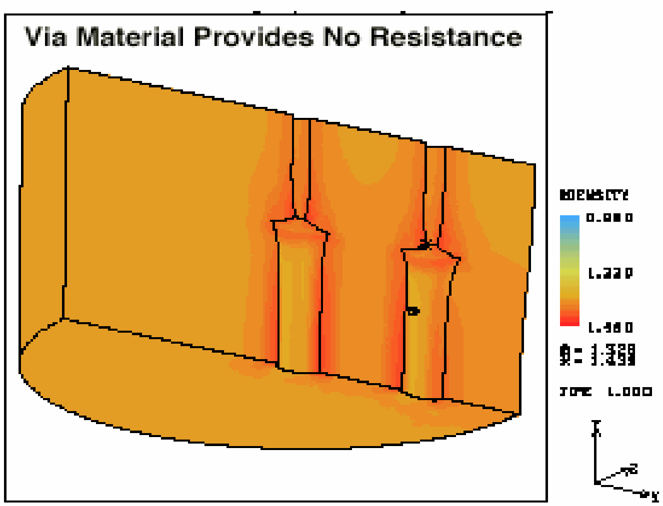
Sandia National Laboratories

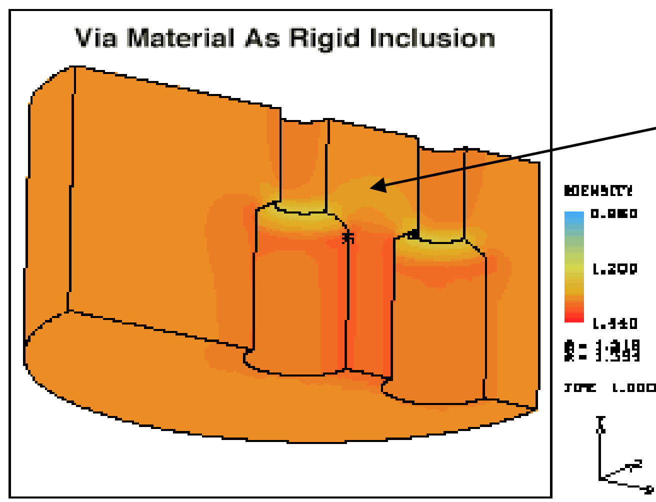
Compaction Simulations Have Linked Defects To Processing





Crack Between Cermet Vias

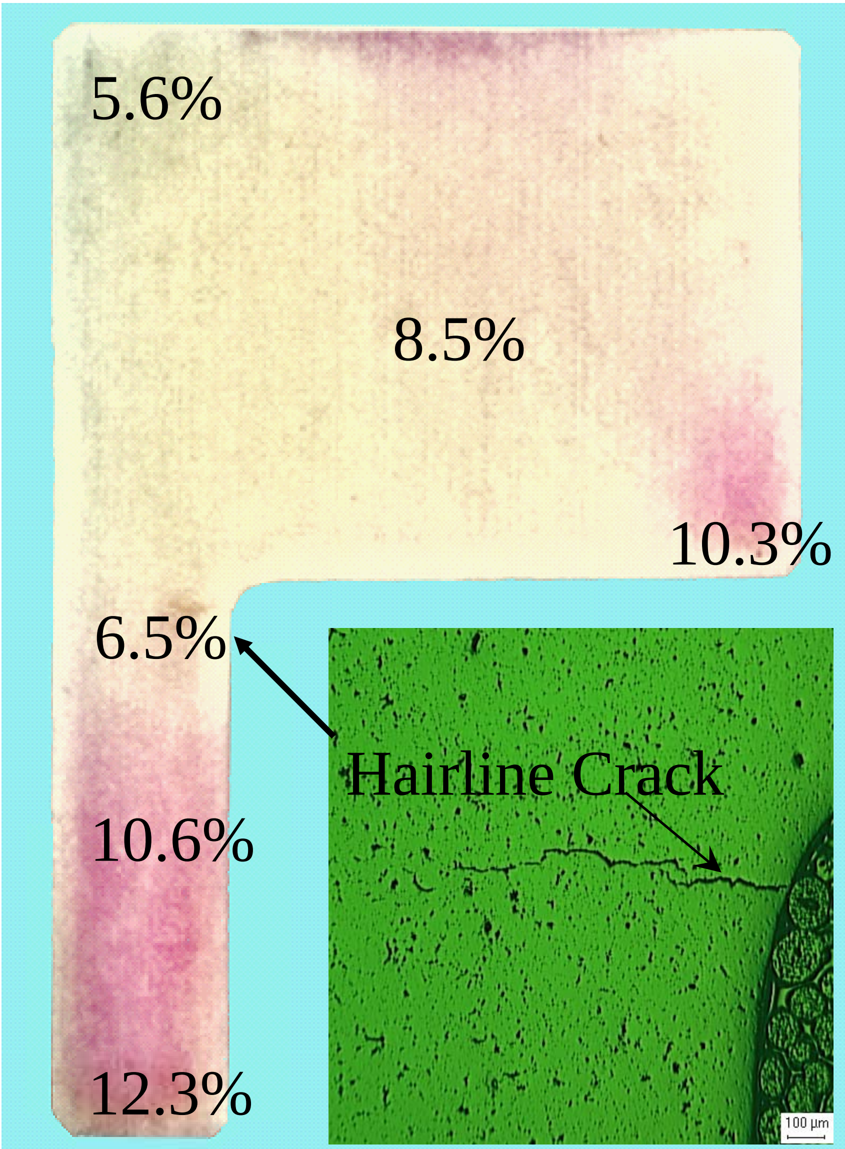




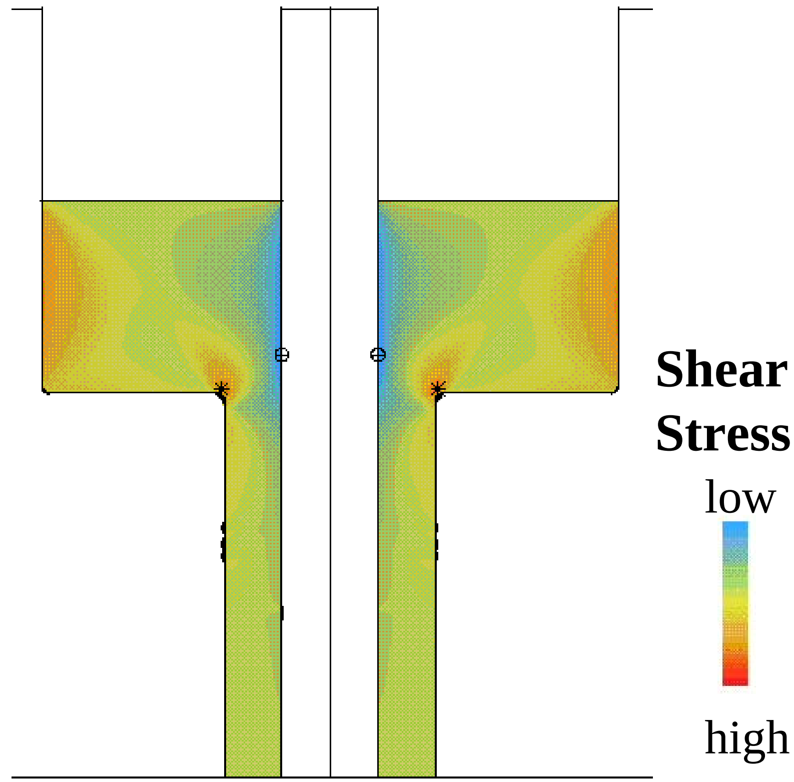
Low Density Between Cermet Vias

Sandia National Laboratories

Simulations Predict Stress And Density Gradients That Result In Cracks

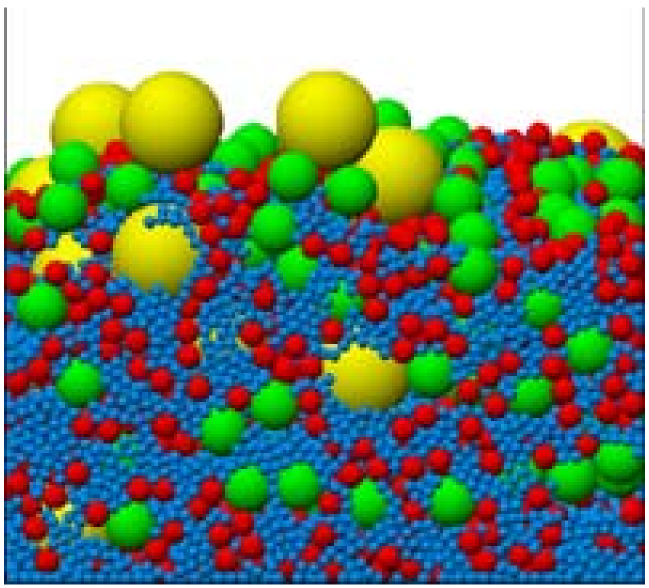


94% Al_2O_3
Dual-Action,
Uniaxial Pressing



Sandia National Laboratories

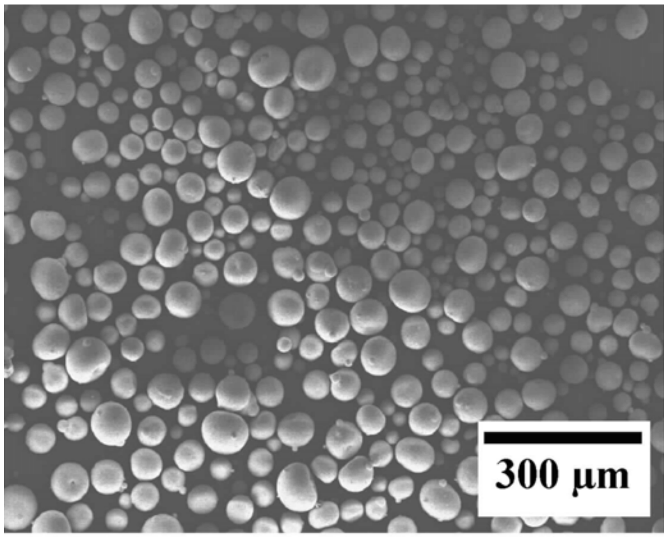
Defects Persist Throughout Processing

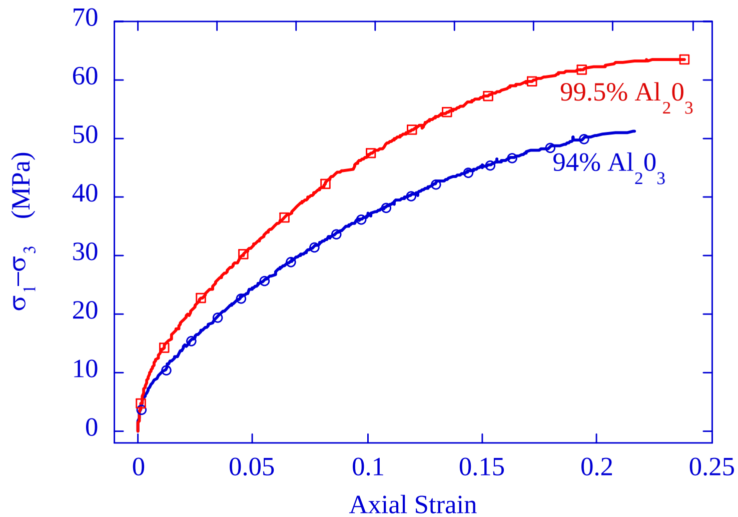


- Defects introduced during die filling persist through compaction & Sintering
  - control stickiness to optimize packing
  - minimize bulk & tap density difference
- Particle packing models can be used to anticipate & avoid packing defects
  - 250:1 critical die size to granule size
  - critical ratio is important for small parts
  - granule sizing can improve yields
  - use models to tailor size distributions

Sandia National Laboratories

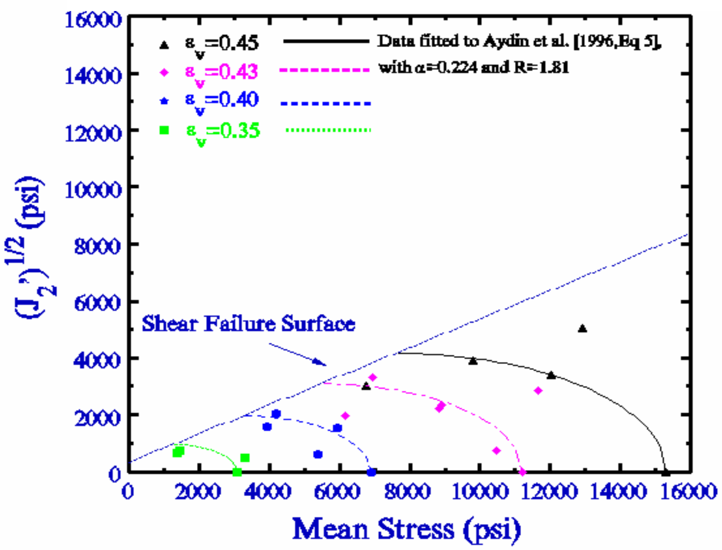
Powder Pressibility Is Related To Powder Characteristics/Properties

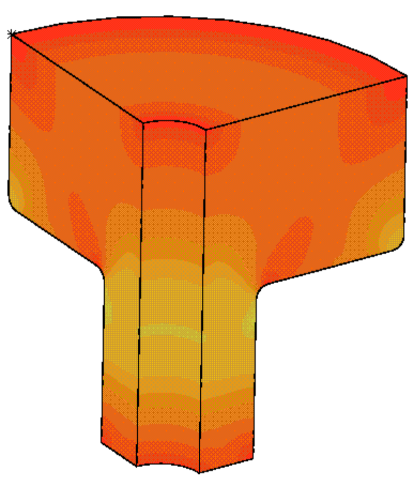




- Granule size, shape, & density affect powder flow, packing, & compaction
  - size ~ 50-400 µm
  - spherical
  - angle of repose < 40°
  - granule density ~ 45-50%
- A combination of techniques is required to assess compaction response
  - compaction response curve
  - green compact strength
  - green compact microstructure
- Powder pressibility can be assessed from compaction response and from powder mechanical properties
  - $\mu \cdot \alpha = \ln (P_z/P_a) / (-2/r \cdot z)$
  - angle of internal friction, β
  - strength in triaxial compression

Compaction Response Can Be Characterized, Predicted, & Controlled





- Ceramic Powder Compaction is Described by a Cap Plasticity Model
- FE Modeling Accurately Predicts Density Gradients From Pressing
- Modeling has been used to Establish Guidelines to Improve Pressing
- Improved Understanding and Control of Materials and Processing Will Result in More Reproducible Manufacturing

Sandia National Laboratories