SAND2018-8960C

Additive Manufacturing of Alumina Components by Extrusion of *in-situ* UV-cured Pastes

Lok-kun Tsui,[1] Erin Maines,[2] Lindsey Evans,[2] David Keicher,[3] Judith Lavin[2]

[1] Center for Micro-Engineered Materials, University of New Mexico

[2] Advanced Materials Laboratory, Sandia National Laboratories

[3] Integrated Deposition Solutions Inc.

UNM CENTER *for* MICRO-ENGINEERED MATERIALS

Sandia National Laboratories

 Revised 2018-08-09

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

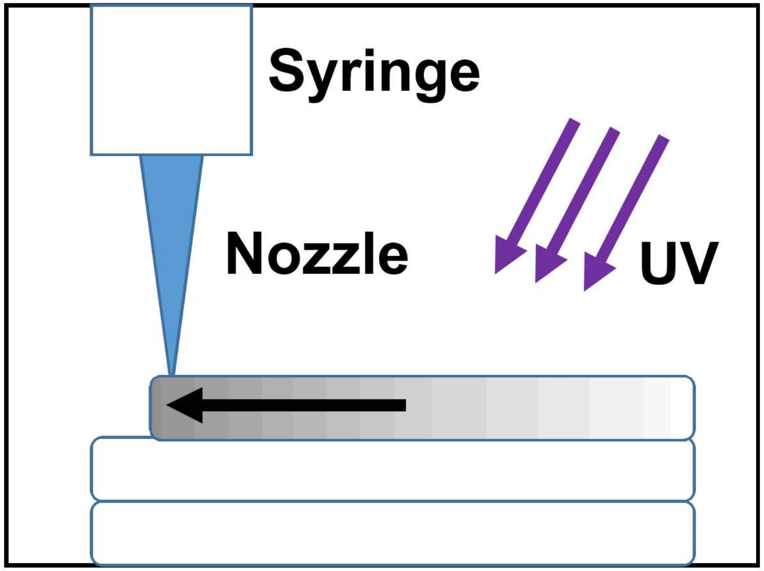
THE 29TH ANNUAL INTERNATIONAL SOLID FREEFORM FABRICATION SYMPOSIUM – AN ADDITIVE MANUFACTURING CONFERENCE – 2018

Overview

- Motivation: Additive manufacturing of ceramics by extrusion of UV-curable pastes.
- Printer Hardware Overview
- Process Overview: Paste formulation, printing, and sintering.
- Studies varying particle size, solids loading, and sintering conditions on morphology and density of components.
- Conclusions and Future Work

Motivation and Objectives

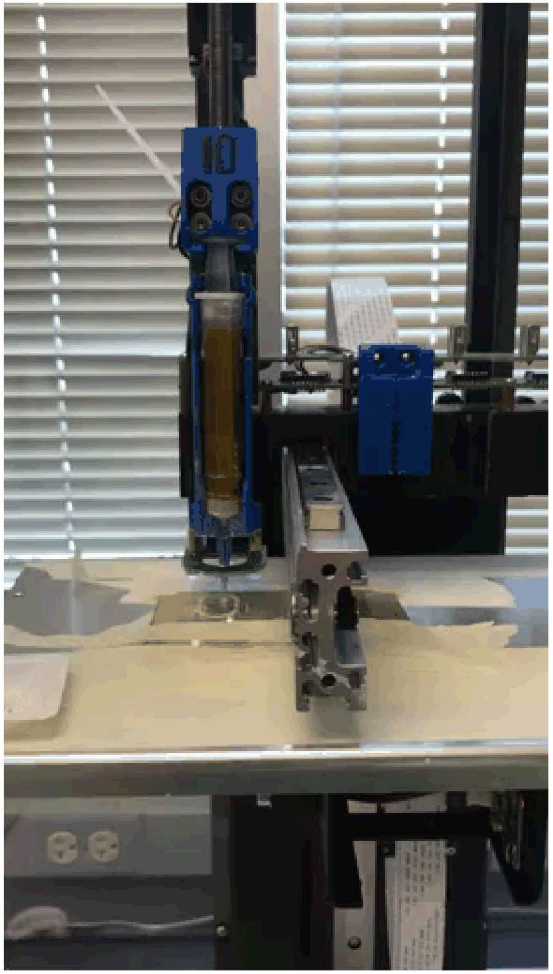
- Additive manufacturing enables the low cost, rapid prototyping of ceramic parts at small scales.
- Applications: Biomedical parts, electronics components and packaging, structural ceramics, electrochemical gas sensors.[1,2]
- Extrusion printing: UV-curable paste is extruded from a syringe and solidified with UV light.
- Relatively low equipment cost, small quantity (10s of grams) of material needed, can be done in at ambient temperature.
- Optimize for high density parts and understand rheology of pastes and sintering behavior of components.



[1] N. Travitzky, et al. Adv. Eng. Mater. 16 (2014) 729–754.
[2] L.-K. Tsui, et al. Prog. Addit. Manuf. (2018).

Motivation and Objectives

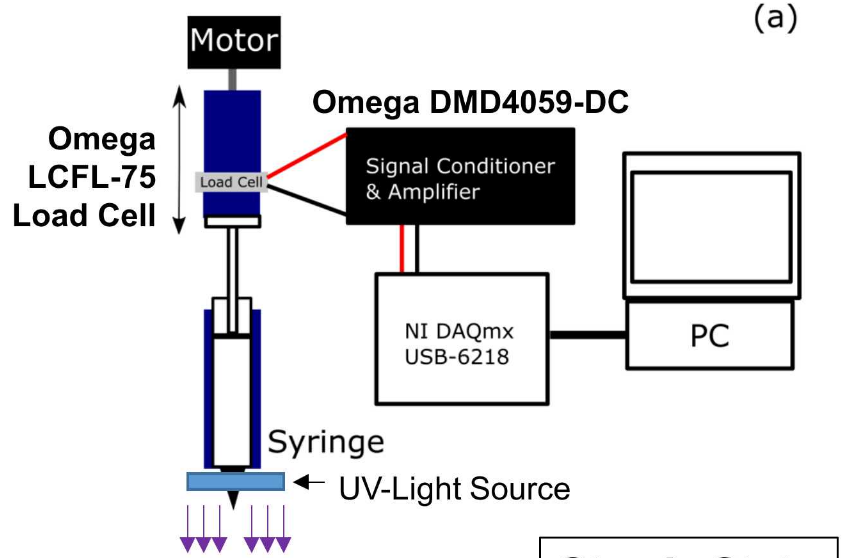
- Additive manufacturing enables the low cost, rapid prototyping of ceramic parts at small scales.
- Applications: Biomedical parts, electronics components and packaging, structural ceramics, electrochemical gas sensors.[1,2]
- Extrusion printing: UV-curable paste is extruded from a syringe and solidified with UV light.
- Relatively low equipment cost, small quantity (10s of grams) of material needed, can be done in at ambient temperature.
- Optimize for high density parts and understand rheology of pastes and sintering behavior of components.



[1] N. Travitzky, et al. Adv. Eng. Mater. 16 (2014) 729–754.
[2] L.-K. Tsui, et al. Prog. Addit. Manuf. (2018).

Hardware Overview

- Hyrel System 30M printer – Supports several material types including pastes, gels, and plastic filaments.
- CSD-5 – 5mL Syringe head with 365 nm UV LEDs.
- Modification of print head with a load cell to support force sensing.



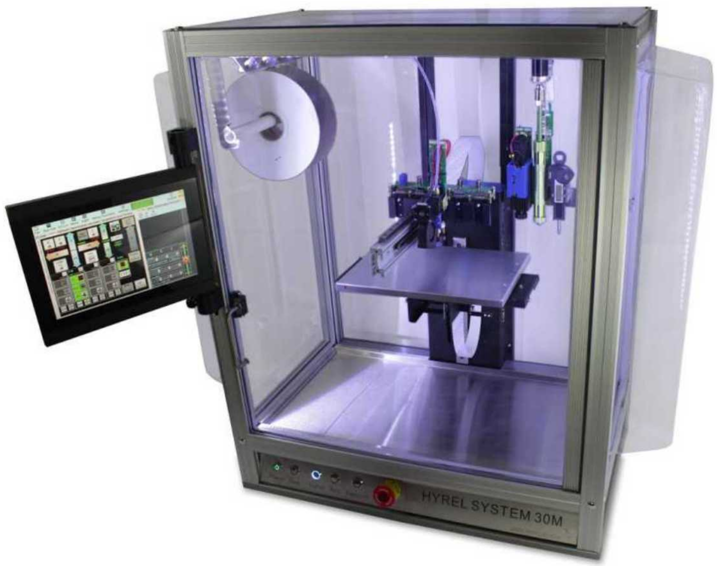
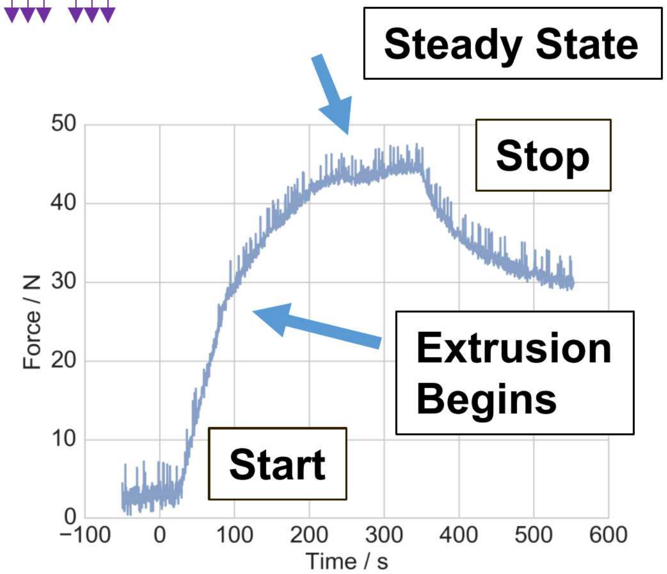


Image:http://www.hyrel3d.com/core-suystems/system-30m/



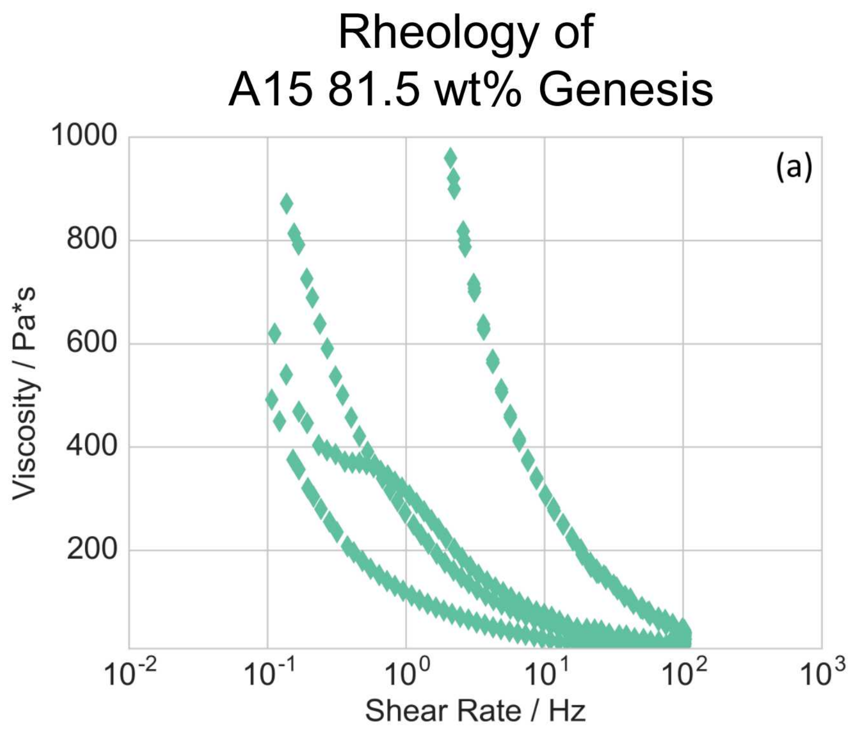
Paste Composition and Formulation

Materials

- Alumina powder:
  - Almatis A15: 0.5 – 4 μm (Bimodal)
  - Almatis A16: 0.5 μm
- Lubricant: Oleic Acid
- Dispersant: Hypermer KD1 (A16 Only)
- UV-Curable Resin: Tethon3D Genesis

Processing and Formulation

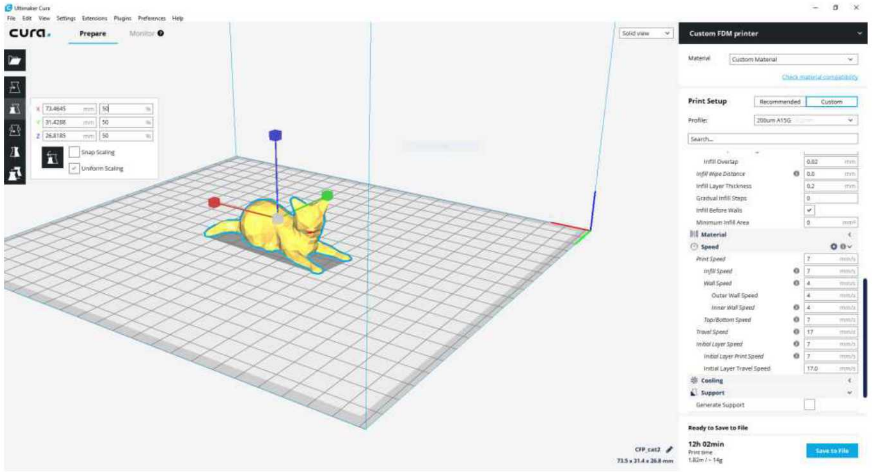
- Powders milled with lubricant and dispersant, dried and sieved.
- Mass loading varied for A16, 81.5 wt% used for A15
- Mixed with planetary vacuum mixing at 2000RPM
- Loaded into a syringe and centrifuged to remove air bubbles.
- Rheology measured to show that pastes exhibit shear thinning behavior



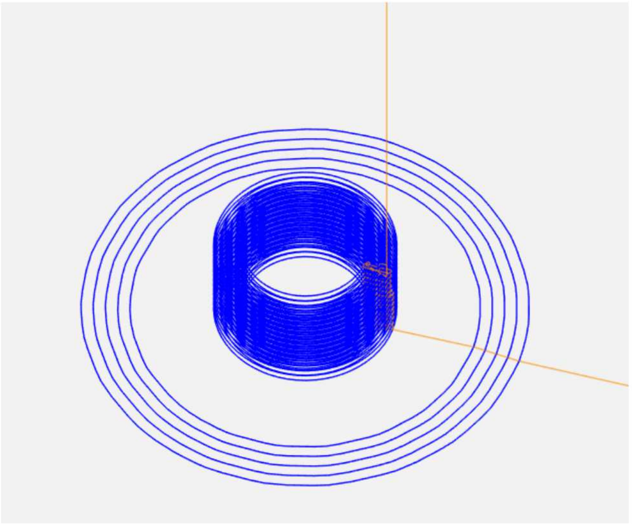
Printing Procedure

- Toolpath generation performed using Cura: FOSS slicing software designed for plastic 3D printing.
- Nozzle: black opaque tip with orifice diameter 635 μm.
- Substrate: Glass slide
- Printing achievable at 200 μm layer thickness with print speed of 5 mm/s and 70-90% infill.

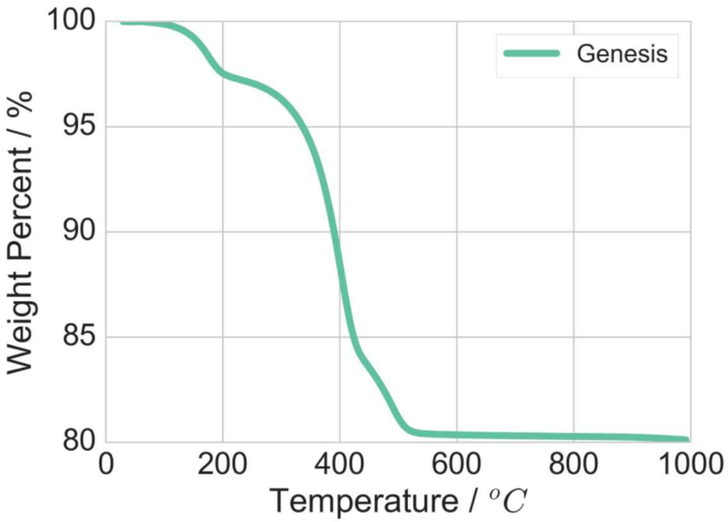
Cura Software



Example Ring Toolpath



Burnout and Sintering



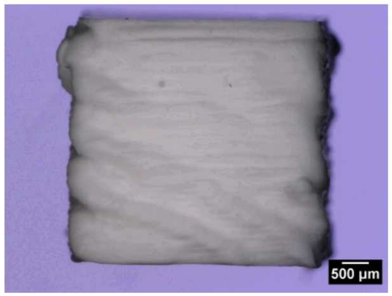
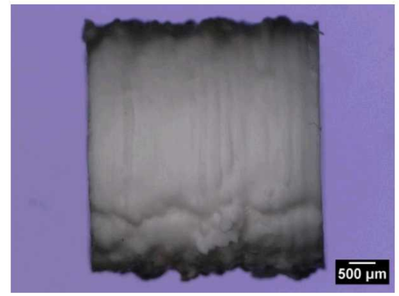
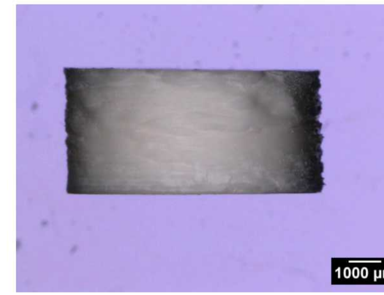
TGA of 81.5 wt% A15 in Genesis

Temperature (°C)	Ramp Rate (°C/min)	Hold Time (Hours)
Burnout (Air atmosphere)		
225	0.5	0.5
625	0.5	1.0
1000	1.0	1.0
25	-3.0	0.0
Sintering (Air atmosphere)		
1625	3.0	2.0
25	-5.0	0.0

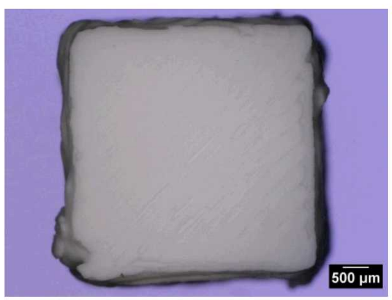
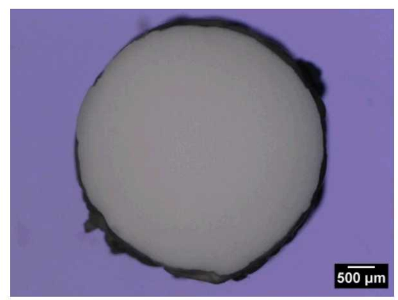
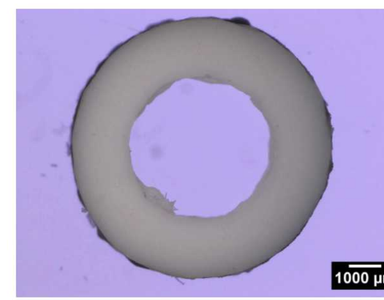
Sample Components (After Sintering)

A15 Test Structures

A16 Test Structure

	7.5 mm Cube	**7.5 mm cylinder**	**8 mm ring**
Side View			

Higher roughness observed along axis perpendicular to bed.

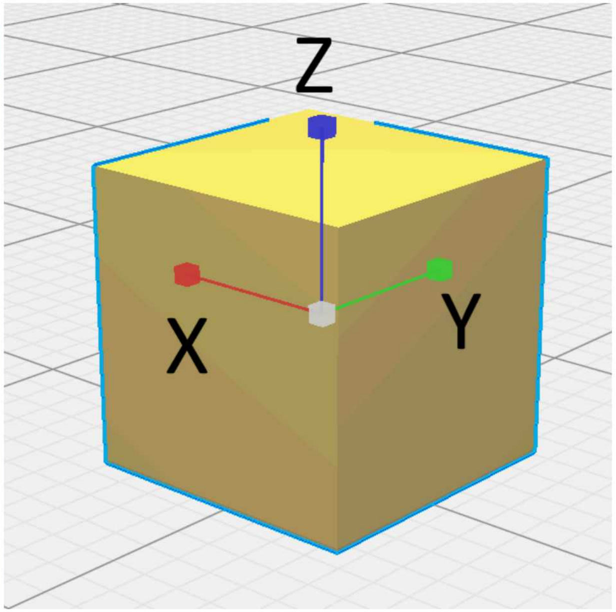
Bottom View			

Bottom surface smooth. Top surface smoothed by polishing.

A15 Components: Precision, Shrinkage, Density

7.5 x 7.5 x 7.5 mm Cube

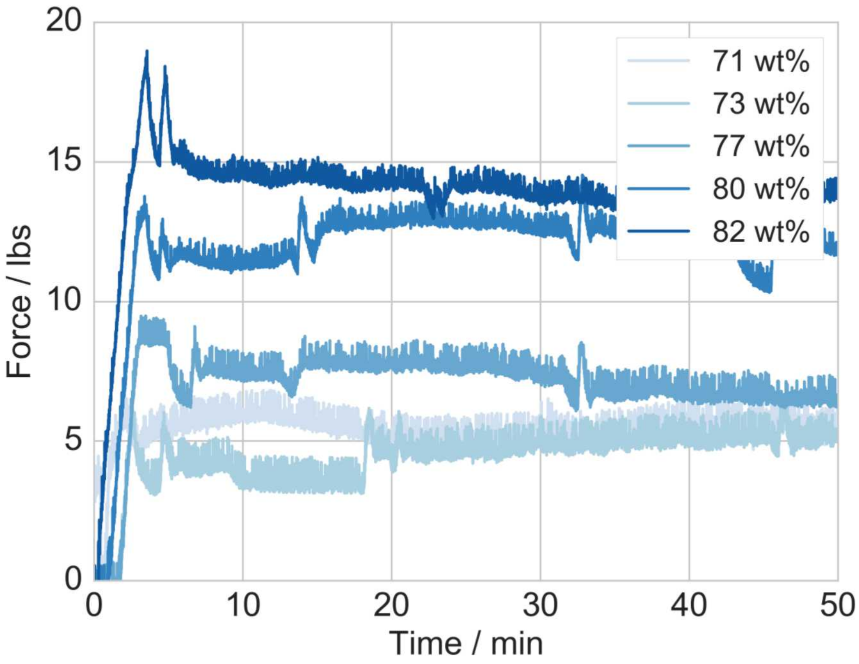
Parameter Name	Nominal	As Deposited	Final Sintered
Length (X)	7.5 mm	7.50 mm (±0.09 mm)	6.37 mm (±0.03 mm)
Width (Y)	7.5 mm	7.54 mm (±0.15 mm)	6.35 mm (±0.19 mm)
Height (Z)	7.5 mm	7.51 mm (±0.08 mm)	6.10 mm (±0.08 mm)
Aspect Ratio	1.0	0.999 (±0.01)	0.959 (±0.01)
Length Shrinkage (X)			15%
Height Shrinkage (Z)			19%
Volumetric Shrinkage			42%



- Greater shrinkage observed in Z due to more voids.
- Final density measured by Archimedes technique: maximum of 91.5% of theoretical density with all A15 components after sintering at 1625 ^{o}C.

Improving density of printed components with smaller particle size alumina

- A15 contains particles on the order of 4 μm.
- Using 0.5 μm A16 should improve the driving force for sintering due to higher surface area.
- Loading varied between 71 – 82 wt%.
- Sintering temperature for 80 wt% varied between 1400 – 1665 °C.
- Determine rheological properties of the pastes using force sensing.



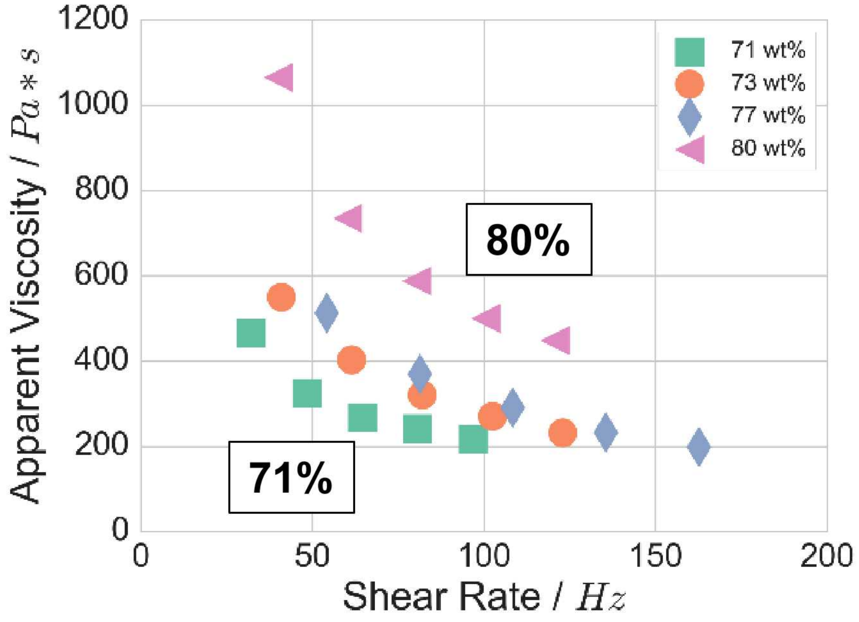
Extrusion force during deposition of A16 from 71-82 wt% loading

THE 29TH ANNUAL INTERNATIONAL SOLID FREEFORM FABRICATION SYMPOSIUM – AN ADDITIVE MANUFACTURING CONFERENCE – 2018

Rheological Measurements from Extrusion Force

- Cao et al. [1] developed a model to calculate apparent viscosity through a nozzle from applied force.
- Maximum force is recorded as a function of dispensing rate varied from 0.5 to 1.7 μL/s.
- Viscosity increases with increasing powder loading.
- All pastes exhibit shear thinning behavior.

Apparent Viscosity of A16 Pastes

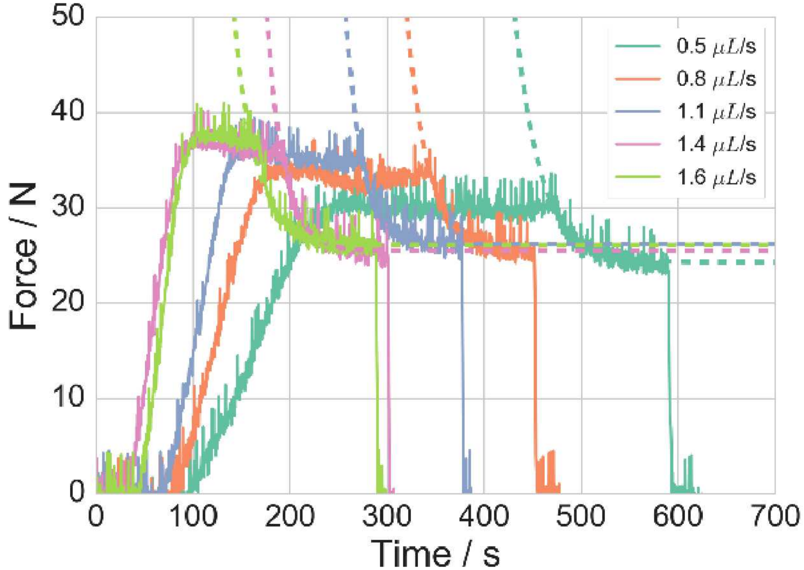


[1]: K. Cao, Y. Liu, C. Tucker, M. Baumann, G. Grit, S. Lakso, ACS Comb. Sci. 16 (2014) 198–204.

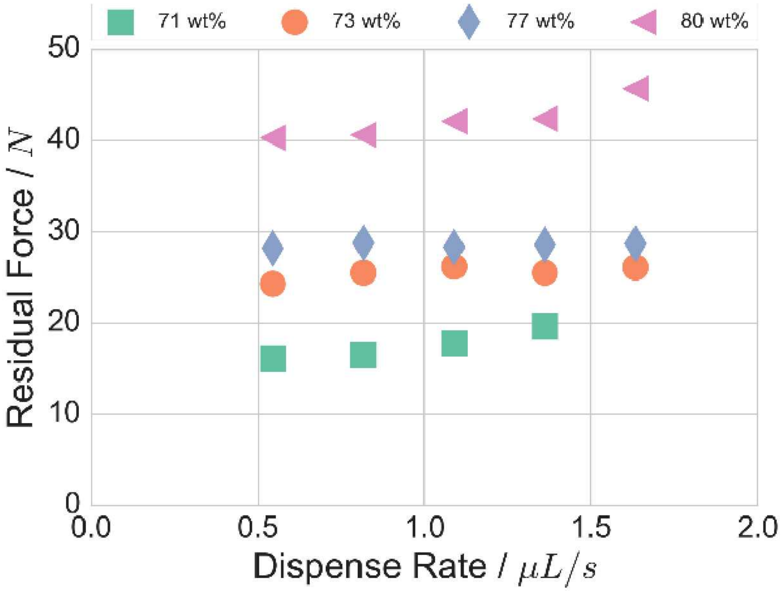
THE 29TH ANNUAL INTERNATIONAL SOLID FREEFORM FABRICATION SYMPOSIUM – AN ADDITIVE MANUFACTURING CONFERENCE – 2018

Measurement of residual force after dispensing.

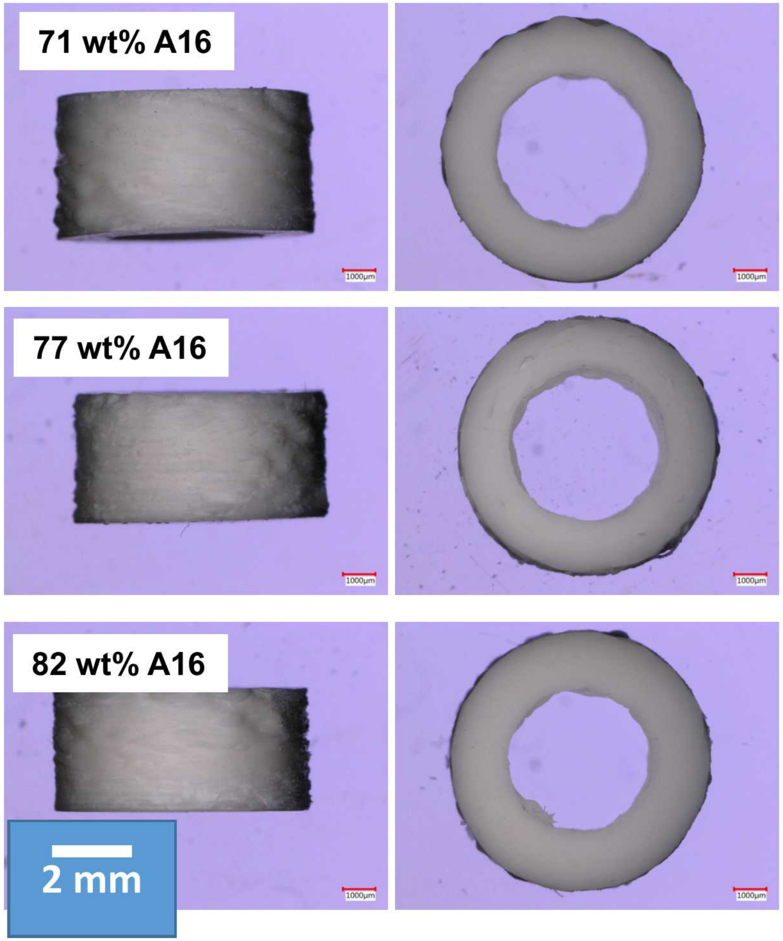
- After dispensing stops, material continues to flow until a steady-state force is reached.
- This represents the minimum force needed to extrude material.
- Residual force determined by fitting exponential decay to force transient.
- Residual force is independent of initial dispense rate.

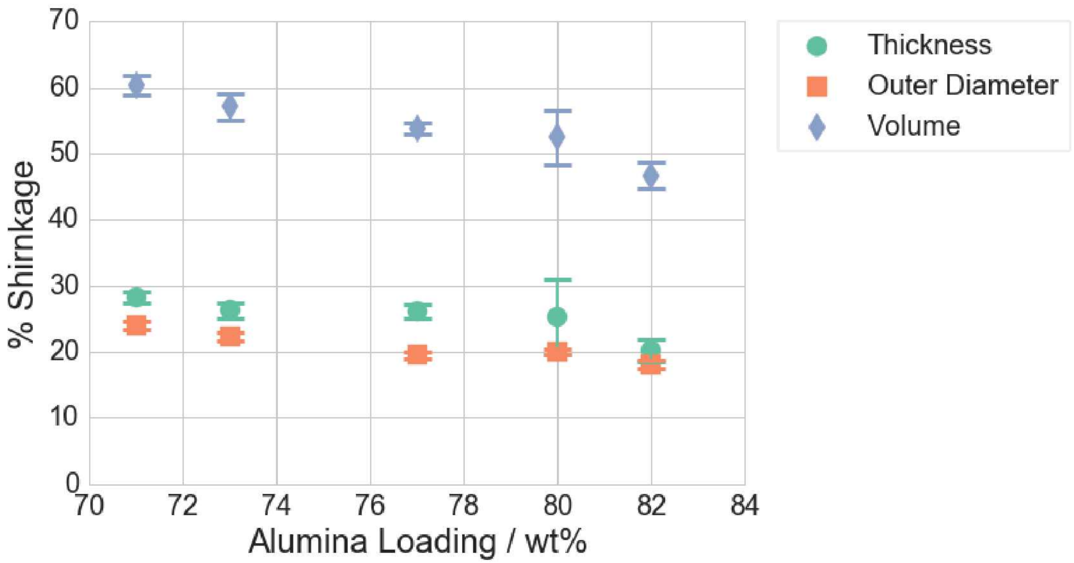


73 wt% A16 Paste



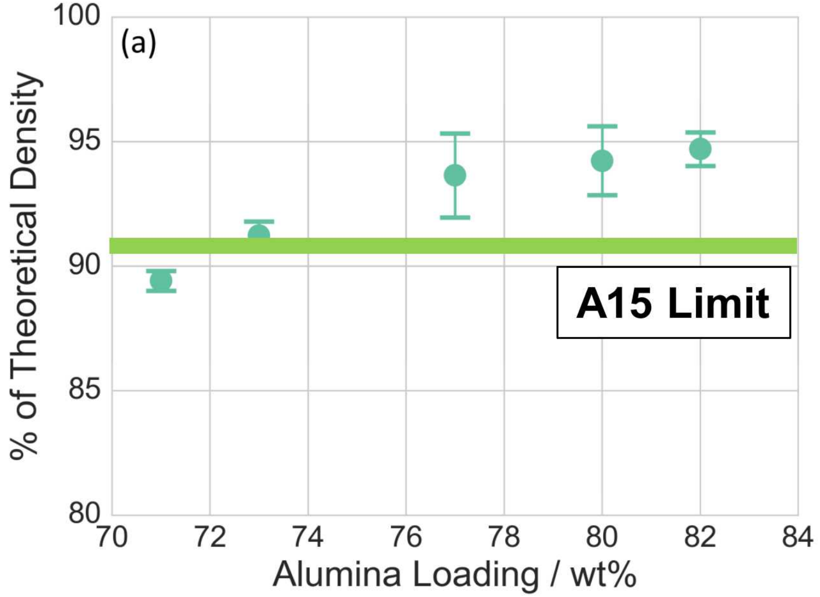
Morphology and Shrinkage of Parts Printed with A16

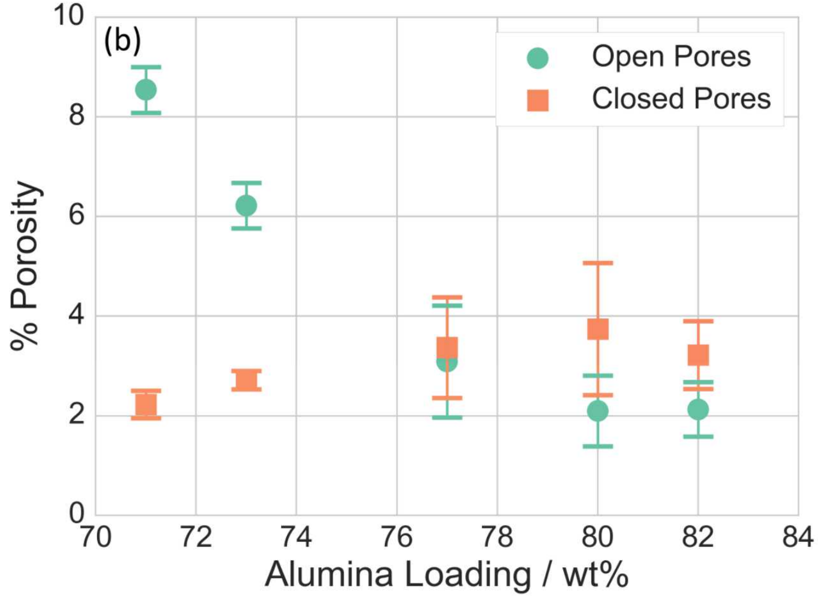




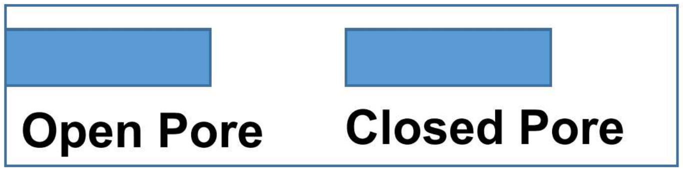
- Samples printed to compensate for expected shrinkage and same sizes achieved.
- Shrinkage decreases linearly as a function of loading.
- Shrinkage in thickness always greater than shrinkage in OD.

Influence of A16 alumina loading on density



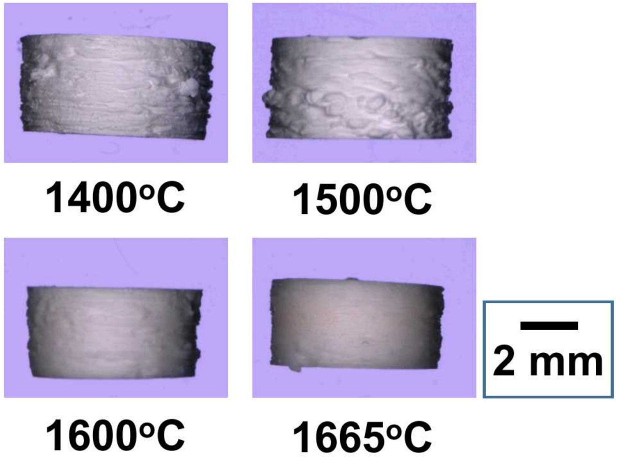


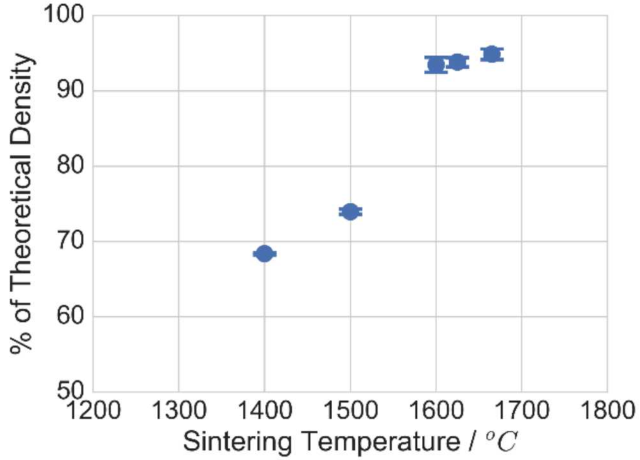
- Particle loading varied between 71-82 wt%.
- Density increases from 88 – 95% of theoretical density of alumina, exceeding limitation of A15.
- Fraction of open pores (cracks, exterior voids) decreases with loading while fraction of closed pores (trapped voids) increased.

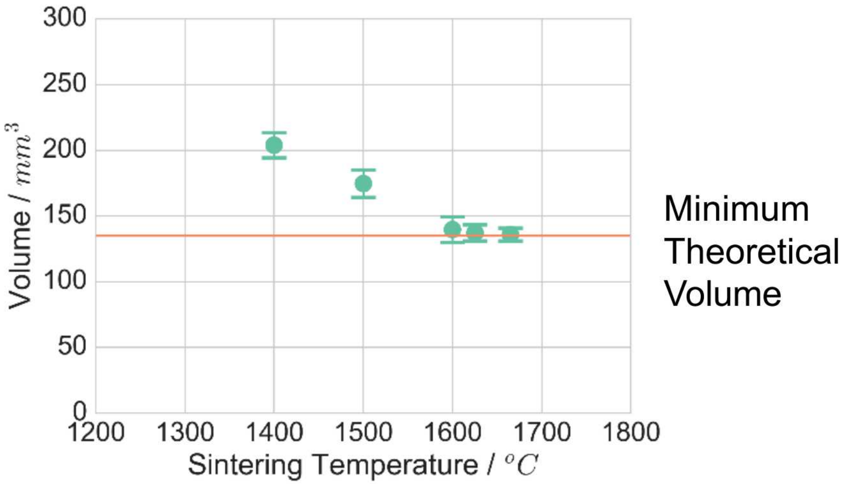


Influence of sintering temperature on final density of A16 parts

- 2 hour sintering hold at temperatures from 1400-1665 °C using 80 wt% A16 paste
- Decrease in qualitative exterior roughness after sintering at 1600 °C.
- Diminishing returns in density with temperature above 1600 °C.
- Convergence to minimum theoretical volume implies remaining deficiency associated with microscopic defects.







THE 29TH ANNUAL INTERNATIONAL SOLID FREEFORM FABRICATION SYMPOSIUM – AN ADDITIVE MANUFACTURING CONFERENCE – 2018

X-ray Computed Tomography

- XCT provides non-destructive imaging of the interior structure of additively manufactured samples.
- Sample sintered at 1625 °C with A16 loading of 82 wt%.

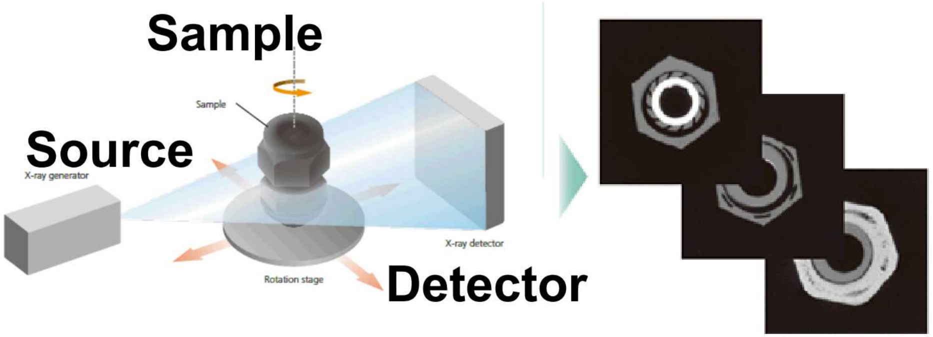
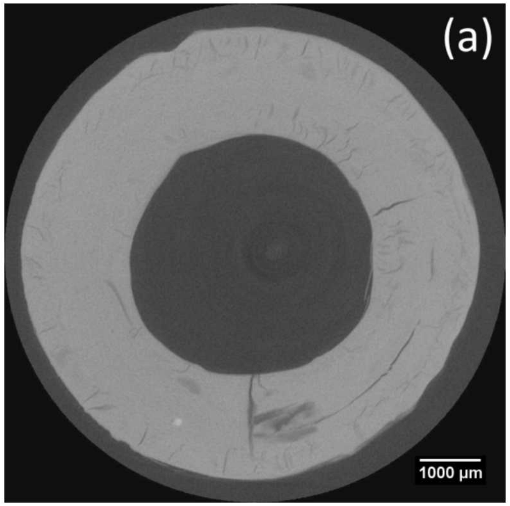
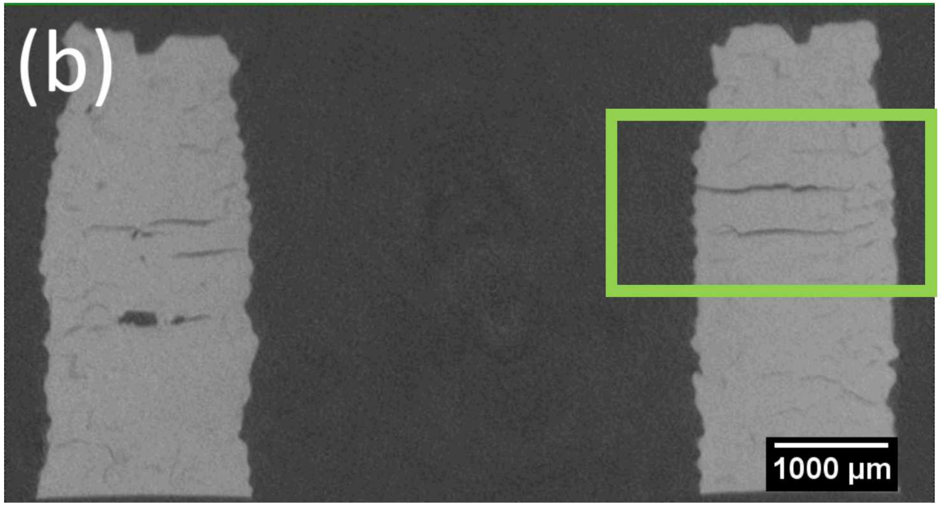


IMAGE: https://www.shimadzu.com/an/ndi/ct/smx90ct.html



Top View

Small cracks observed near edges.



Side View

Defects associated with inter-layer interfaces.

THE 29TH ANNUAL INTERNATIONAL SOLID FREEFORM FABRICATION SYMPOSIUM – AN ADDITIVE MANUFACTURING CONFERENCE – 2018

Conclusions

- Demonstrated the capability to print simple geometric shapes with densities of 94.5%.
- Applied force sensing to study rheological properties of pastes: apparent viscosity, residual forces.
- Non-destructively identified defects using x-ray CT.

Future Work

- Continue to optimize deposition parameters to minimize void formation inside our components.
- Demonstrate capability to print more complex structures and larger structures beyond simple geometric shapes.
- Demonstrate capabilities for other ceramics beyond alumina. (e.g. YSZ)

Acknowledgments

- **Nelson Bell (Sandia Nat'l Labs) –** Assistance in obtaining and analyzing rheology data.
- **Joseph A. Romero (Sandia Nat'l Labs) –** Assistance in XCT measurements.
- Funding:

Sandia National Laboratories

LDRD Program.

Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc. for the US Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Extra Slides

Rheological Measurements from Extrusion Force

Maximum force is recorded as a function of dispensing rate varied from 0.5 to 1.7 μL/s.

$$\tau_R = \frac{P_{Max} R}{2L}$$

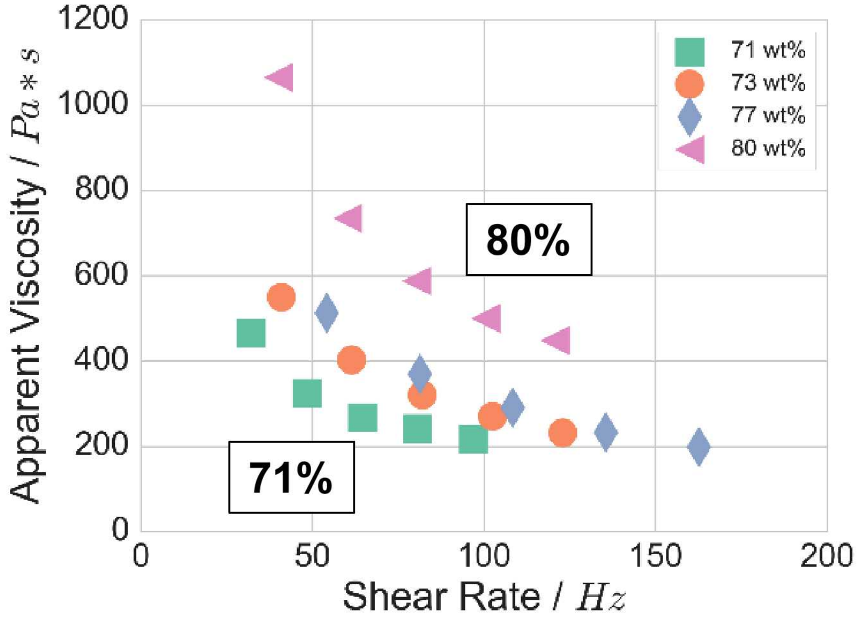
τ_R is the wall stress. P_{max} is the maximum pressure observed, R is the radius of orifice, and L is length of material dispensed.

$$\gamma_R^* = \frac{Q}{\pi R^3}\left[3 + \frac{d\,ln\left(\frac{Q}{\pi R^3}\right)}{d\ln \tau_R}\right]$$

$\gamma*$ is shear rate, Q is dispense rate, and derivative obtained from the $\ln(\tau_R)$ vs. $\ln(Q/\pi R^3)$ plot.

$$\eta = \frac{\tau_R}{\gamma_R^*}$$ Apparent Viscosity

Apparent Viscosity of A16 Pastes



Adapted from: K. Cao, Y. Liu, C. Tucker, M. Baumann, G. Grit, S. Lakso, ACS Comb. Sci. 16 (2014) 198–204.

PSD from Almatis