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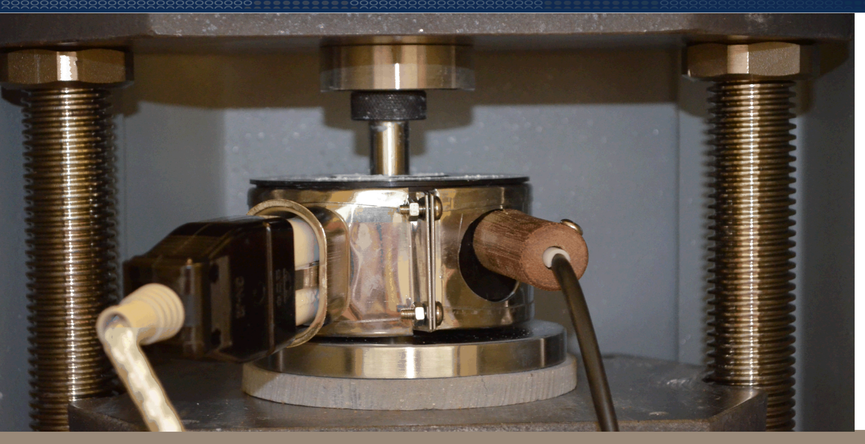

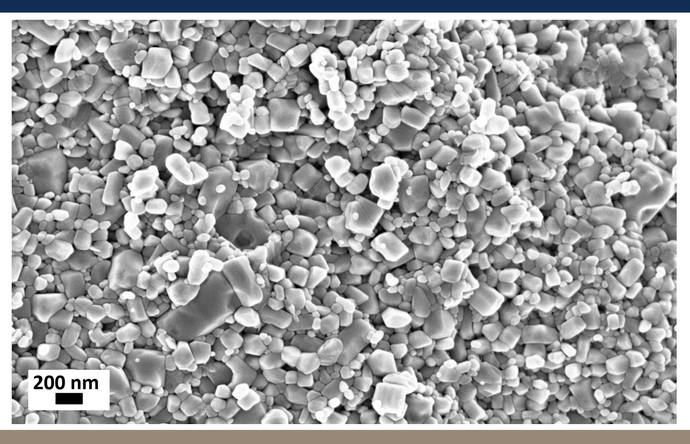

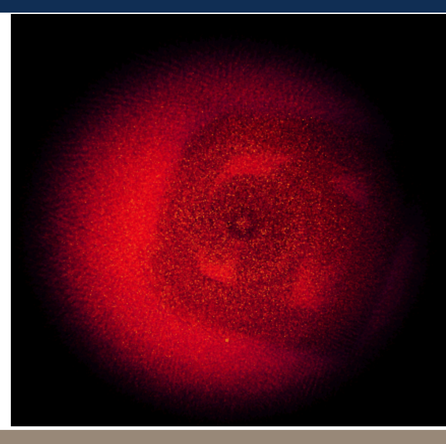

Cold Sintering Process

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Abstract

Ceramic pellets formed through the cold sintering process, which utilizes high pressure (300 MPa) and low temperature (100°C) compared to conventional sintering, were found to have comparable densities to traditionally sintered ceramics (~95%). This process allows for close control of grain size for applications such as varistors. Additionally, polymers were successfully integrated into ceramic pellets. Ceramic pellets containing a 3D printed object (ABS- acrylonitrile butadiene styrene) were successfully sintered to high density, and 28 vol.% PTFE*ZnO composites were successfully produced via cold sintering.

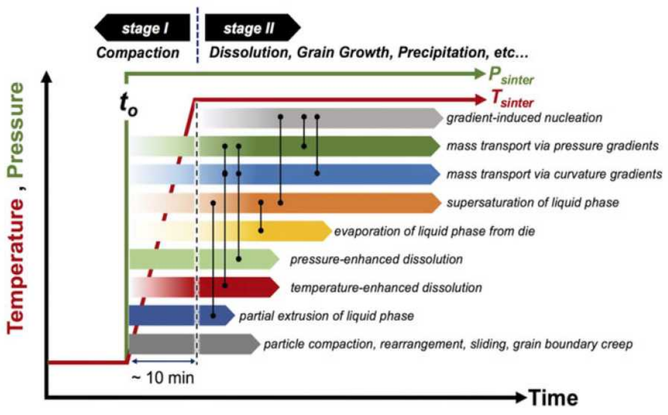
Introduction

- Cold sintering process (CSP):
  - Low temperatures (100°C)
  - High pressures (300 Mpa)
  - Low vol% liquid media

Binary compounds	Ternary compounds	Quaternary compounds	Quinary compounds
MoO_3 ($P2_1/c$)	Li_2CO_3 ($C2/c$)	$LiFePO_4$ ($Pnma$)	$LiAl_{0.5}Ge_{1.5}(PO_4)_3$
WO_3 ($P2_1/n$)	$CsSO_4$	$LiCoPO_4$ ($Pnma$)	$Li_{0.5x}Bi_{1-0.5x}Mo_xV_{1-x}O_4$
V_2O_3 ($R\bar{3}c$)	Li_2MoO_4 ($R\bar{3}$)	KH_2PO_4 ($Fdd2$)	…
V_2O_5 ($Pmmn$)	$Na_2Mo_2O_7$ ($Cmca$)	$Ca_5(PO_4)_3(OH)$ ($P6_3/m$)	…
ZnO ($P6_3mc$)	$K_2Mo_2O_7$ ($P\bar{1}$)	$(LiBi)_{0.5}MoO_4$	…
Bi_2O_3 ($P2_1/c$)	$ZnMoO_4$ ($P\bar{1}$)	…	…
CsBr ($Pm\bar{3}m$)	$Gd_2(MoO_4)_3$ ($Pba2$)	…	…
MgO ($Pm\bar{3}m$)	Li_2WO_4 ($R\bar{3}$)	…	…
PbTe ($Pm\bar{3}m$)	Na_2WO_4 ($Fd\bar{3}m$)	…	…
Bi_2Te_3 ($R\bar{3}m$)	$LiVO_3$ ($C2/c$)	…	…
NaCl ($Fm\bar{3}m$)	$BiVO_4$ ($C2/c$)	…	…
ZnTe ($F\bar{4}3m$)	$AgVO_3$ ($C2/c$)	…	…
AgI ($P6_3mc$)	Na_2ZrO_3 ($C2/c$)	…	…
CuCl ($F\bar{4}3m$)	$BaTiO_3$ ($P4mm$)	…	…
ZrF_4 ($C2/c$)	$NaNO_2$ ($Im2m$)	…	…
ZrO_2 ($P2_1/c$)	$Mg_2P_2O_7$ ($P2_1/c$)	…	…
…	$BaMoO_4$ ($I4_1/a$)	…	…
…	Cs_2WO_4 ($Pnma$)	…	…
…	$Na_xCo_2O_4$	…	…
…	$Ca_3Co_4O_9$ ($C2/m$)	…	…
…	KPO_3 ($Pa\bar{3}$)	…	…

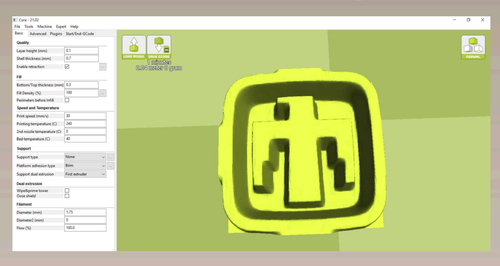
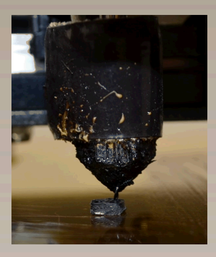
List of materials sintered to date using CSP [1].

- Mechanisms:
  - Particle rearrangement
  - Pressure- and temperature-assisted dissolution and precipitation



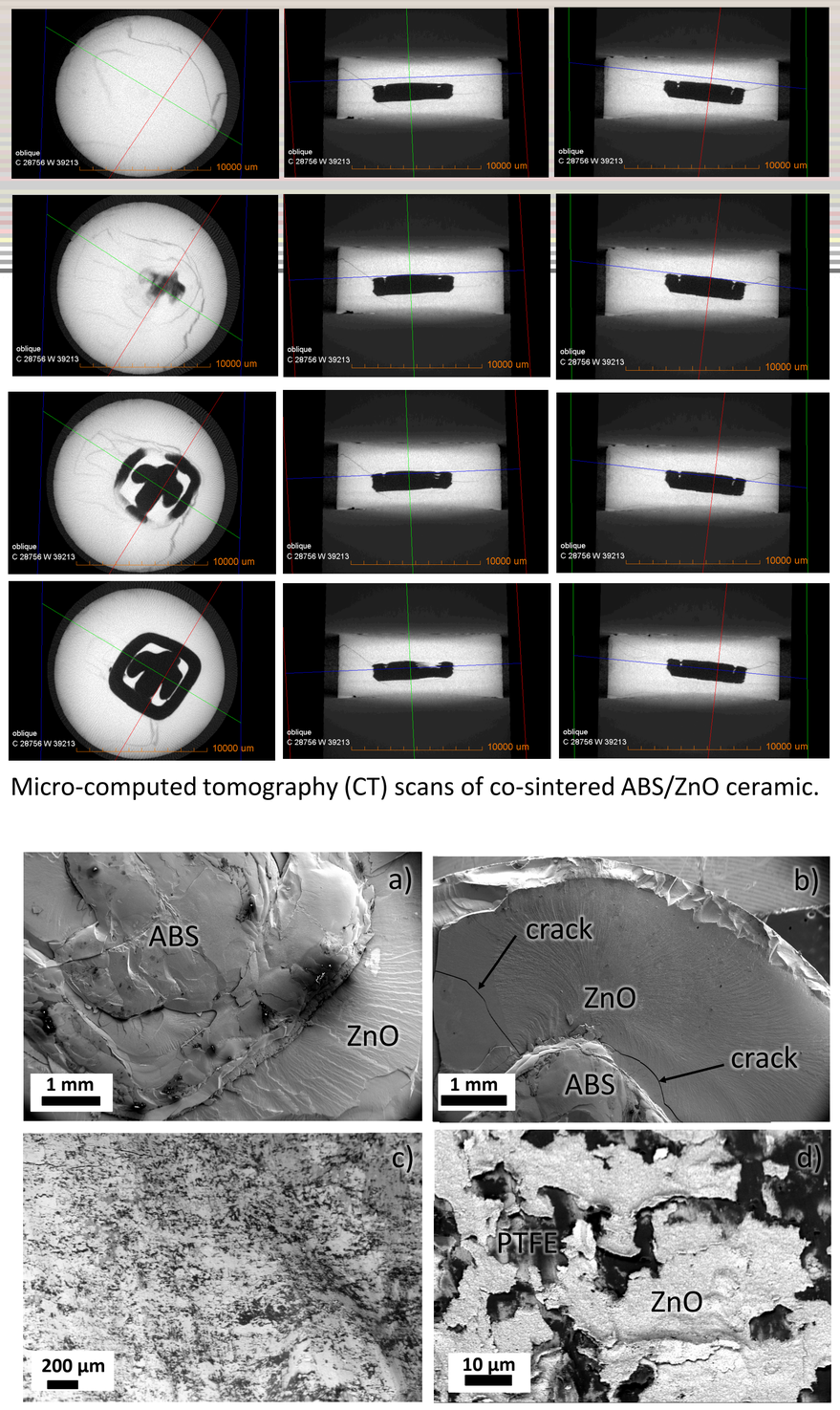
Densification stages and mechanisms of cold sintering [1].

Co-sintered Polymer-Ceramics

126°C, 387 MPa, CH_3COOH

3D printed ABS Thunderbird in cold sintered ZnO
Sintered at 126°C, 387 MPa using acetic acid.

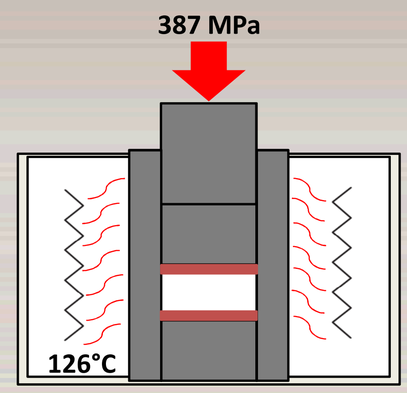


Micro-computed tomography (CT) scans of co-sintered ABS/ZnO ceramic.

SEM images of cold sintered pellets: a) and b) 3D printed ABS cold sintered in ZnO, c) and d) PTFE/ ZnO cold sintered composite.

Methods

Processing Parameters	
Powder	ZnO
Liquid	Acetic Acid (1M, 10 wt%)
Pressure	387 MPa
Temperature	126°C
Hold Time	½ hour
Density	95%



Schematic of the cold sintering process.

Results & Discussion

- 95% theoretical densities were achieved
- Cracking of ram/ceramic interface
  - Mitigated with Kapton tape
- Polymer/ceramic composites successfully sintered at 126°C
- Cracking of ceramic when integrating 3D printed objects
  - Friction against die walls
  - CTE mismatch between polymer and ceramic
  - Yielding of polymer due to applied pressure

Conclusion

- Cold sintering enables a new class of composite materials with disparate thermal budgets
- Cold sintering is a disruptive processing technology
- Varies from >10,000 years of evolutionary ceramic processing

Future Work

- Grain size control to understand the effects of grain boundary on electrical properties.
- Crack origination and mitigation in pellets with 3D parts.
- Evaluate cold sintered composite properties

First cold sintered ceramics produced at Sandia: Successfully demonstrated a way to integrate polymers into ceramics

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[1] Maria, J., Kang, X., Floyd, R., Dickey, E., Guo, H., Guo, J., . . . Randall, C. (2017). Cold sintering: Current status and prospects. *Journal of Materials Research,* 1-14. doi:10.1557/jmr.2017.262
[2] J. Guo, H. Guo, A. L. Baker, M. T. Lanagan, E. R. Kupp, G. L. Messing, C. A. Randall, *Angew. Chem. Int. Ed.* 2016, 55, 11457.