Consolidation of Advanced Powders by Severe Plastic Deformation

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**Consolidation of Advanced Powders by Severe Plastic Deformation**

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See also ADM001672., The original document contains color images.
Collaborators

- I. Anderson, DOE Ames Laboratory and Iowa State University, (Powder fabrication)
- R. Barber, Texas A&M University, (ECAE processing)
- J.T. Im, Texas A&M University, (Amorphous materials characterizations)
- H.J. Maier, University of Paderborn, (Transmission electron microscopy)
- J. Robertson, Texas A&M University, (Amorphous materials characterizations)
“Consolidation of Advanced Powders by Severe Plastic Deformation”

Talk Outline

1. Description of ECAE
2. Materials
   - Bulk Nanostructured Cu
   - Bulk Amorphous Zr-based Alloy
3. Experimental Results
4. Lessons Learned
5. Questions Remain
ECAE Concept

**Conditions**

1. Inlet and outlet channels have nearly the same dimensions
2. Channel intersection is abrupt
3. Lubrication and other means are used to reduce friction

**Results**

1. Simple shear occurs
2. Effective strain is $(2/\sqrt{3}) \cot \psi$ or 1.16 for $\psi=45^\circ$
3. Effective strain for multiple (N) extrusions is 1.16 N for $\psi=45^\circ$
4. Strain is relatively uniform
Consolidation of powder by ECAE

**Can/Powder Description**
- Inert Can Material
- 0.75 x 0.75 x 3.5 inch
- 0.50∅ x 1.5 inch Long Cavity
- Loose Powder with ~0.35 Void Fraction
- Vacuum Bake/Outgas
- e-beam Weld Seal
- Instrumented with Thermocouples

**Deformation Conditions**
- 90° Die Angle
- Isothermal Tool
- Constant Punch Speed
- Hydrostatic Pressure
- Simple Shear Uniformly Deforms Can and Encapsulate
- Heat of Deformation
- Collect Measurements
  - Load-Stroke
  - Time-Temperature

**Extruded Billet Characteristics**
- Near Full Density
- Shorter Billet (Cavity Length Decreases by ~1/3)
- Cavity Geometry Changes Shape (Depends on Number of Passes and Route)
Potential Benefits of Powder Consolidation by ECAE

- Small heated cross-section relative to conventional area reduction extrusion (better heat transfer conditions)
- Large product cross-sections may be possible (conservation of cross-section during extrusion)
- High length/diameter ratio product may be possible
- Combined compaction and shear
- Consolidation to near full density after a single extrusion
- Consolidation to full density at lower temperature than needed for HIPing
- Lower punch loads than for area reduction extrusion
# ECAE Route Descriptions

<table>
<thead>
<tr>
<th>Route name</th>
<th>Min. # of passes</th>
<th>Billet rotations about the extrusion axis</th>
<th>Material Yield*</th>
<th>Effect on microstructure</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>1</td>
<td>0° 0° 0° etc.</td>
<td>0.58</td>
<td>elongation (lamellar)</td>
</tr>
<tr>
<td>B (Bₐ)</td>
<td>2</td>
<td>+90° -90° +90° etc.</td>
<td>0.67</td>
<td>elongation (filamentary)</td>
</tr>
<tr>
<td>C</td>
<td>2</td>
<td>180° 180° 180° etc.</td>
<td>0.83</td>
<td>back/forth shearing</td>
</tr>
<tr>
<td>C' (Bₜ)</td>
<td>4</td>
<td>+90° +90° +90° etc.</td>
<td>0.67</td>
<td>back/forth cross-shearing</td>
</tr>
<tr>
<td>E</td>
<td>4</td>
<td>180° 90° 180° etc.</td>
<td>0.78</td>
<td>back/forth cross-shearing</td>
</tr>
</tbody>
</table>

* Theoretical yield of fully deformed material after N=4 in billet with length/width ratio of 6
### Theoretical Change in Particle Surface Area for Different ECAE Routes

<table>
<thead>
<tr>
<th>Route Name</th>
<th>Percent increase in cubic element surface area for different numbers of passes (N values)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
</tr>
<tr>
<td>A</td>
<td>0</td>
</tr>
<tr>
<td>B</td>
<td>0</td>
</tr>
<tr>
<td>C</td>
<td>0</td>
</tr>
</tbody>
</table>

**Flow Plane of CuAg Composite**

- **N=0**
- **N=1 (Route A)**
- **N=2 (Route A)**
- **N=4 (Route A)**
- **N=2 (Route C)**
ECAE Tool Characteristics

- Billet Cross Section: ARO 19x19 mm, TEXAS 25x25 mm
- Billet Length: ARO 200 mm, TEXAS 150 mm
- Max Isothermal Temp.: ARO 500°C, TEXAS 300°C
- Rapid Billet Ejection: Yes, Yes
## ECAE Consolidation Processing Conditions

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Cu Nanopowder</th>
<th>Amorphous Zr-based</th>
</tr>
</thead>
<tbody>
<tr>
<td>Encapsulation Material</td>
<td>Ni</td>
<td>Ni</td>
</tr>
<tr>
<td>Open or Closed Can</td>
<td>Vacuum</td>
<td>Vacuum</td>
</tr>
<tr>
<td>Extrusion Temp (°C)</td>
<td>23</td>
<td>400-440</td>
</tr>
<tr>
<td>Punch Speed (mm/s)</td>
<td>2.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Max. Temp Rise (°C)</td>
<td>10</td>
<td>10-20</td>
</tr>
<tr>
<td>Max. Punch Load (kip/kN)</td>
<td>~190/~850</td>
<td>~80/~360</td>
</tr>
<tr>
<td>Est. Hydrostatic Pressure (ksi/MPa)</td>
<td>~160/~1100</td>
<td>~70/~480</td>
</tr>
</tbody>
</table>
Cu Nanopowder Project Motivation

- Difficult to achieve grain sizes less than 100 nm using SPD techniques starting from coarse grain structures.
- Could consolidation of nanoparticles to full density be a method to obtain bulk samples?
- Investigate deformation and mechanical properties in bulk nanocrystalline materials.
- Conflicting results on fatigue response of UFG materials. Do the SPD microstructures really deteriorate the fatigue properties? Is it possible to improve ductility?

Note: Problems in nanoparticle consolidation: residual porosity, dynamic recrystallization, abnormal grain growth, bimodal porosity distribution, not much mechanical property data: only hardness measurements.
Initial Cu Powders

Electroexploded Nanopowder
\(O_2 \approx 0.1\) wt% (FNAA)

Agglomerates

Average size 67 nm (X-Ray analysis)

Micropowder (DOE Ames)
99.99 wt% Cu, -325 mesh
Ave. Grain Size: 4.2 microns (X-Ray analysis)

Average size 130 nm (X-Ray analysis)
## Extrusion and Testing Conditions

<table>
<thead>
<tr>
<th>ECAE Route</th>
<th>Can Material</th>
<th>Extrusion Speed</th>
<th>Powder Size</th>
</tr>
</thead>
<tbody>
<tr>
<td>1A</td>
<td>Copper</td>
<td>0.1&quot;/sec</td>
<td>- 325 mesh</td>
</tr>
<tr>
<td>2A</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2B</td>
<td>Copper</td>
<td>0.1&quot;/sec</td>
<td>130 nm</td>
</tr>
<tr>
<td>2C</td>
<td>Nickel</td>
<td>0.1&quot;/sec</td>
<td></td>
</tr>
<tr>
<td>4C</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2B</td>
<td>Nickel</td>
<td>0.1&quot;/sec</td>
<td>3.50 mm</td>
</tr>
<tr>
<td>2C</td>
<td></td>
<td></td>
<td>3.50 mm</td>
</tr>
<tr>
<td>4E</td>
<td>Annealed Bulk Copper</td>
<td>75 µm</td>
<td></td>
</tr>
</tbody>
</table>

Thickness: 1.00 mm

9 cm

Thickness: 4.00 mm

8.00 mm

4.00 mm

3.50 mm
Tension Experiments
ECAE Processed Bulk Samples

Cu 101
Tension Experiments at RT

As Received Material
ECAE at RT, 1A
ECAE at RT, 2C
ECAE at RT, 4C
ECAE at RT, 2C + 100 C for 2 hrs.

Stress, MPa
Strain

0.00 0.05 0.10 0.15 0.20 0.25 0.30
0 50 100 150 200 250 300 350 400 450 500

Strain
Microcrystalline Powder Consolidate Tension Experiments

Microcrystalline Cu powder
Processed with ECAE
Tension @ RT (Along Extrusion Direction)

- Route A, 1 pass
- Route A, 2 pass
- Route B, 2 pass
- Route C, 2 pass
- Route C, 4 pass
- Annealed Cu (75 µm grain size)
Microcrystalline Powder Consolidate Compression Experiments

Microcrystalline Cu powder processed with ECAE Compression @ RT

- Route A, 1 pass, normal to flow plane
- Route A, 2 pass, normal to flow plane
- Route B, 2 pass, normal to flow plane
- Route C, 2 pass, normal to flow plane
- Route C, 4 pass, normal to flow plane
Microstructural Evolution of Microcrystalline Powder Consolidate

2A
both high and low dislocation density areas

2B
~ 200 nm dislocation free subgrains

2C
very high dislocation density, not well-developed subgrains

4C
very high dislocation density, well-developed subgrains
Nanocrystalline Powder Consolidate Tension Experiments

Nanocrystalline Cu powder (150 nm) Processed with ECAE Tension @ RT Tension axis = Extrusion Direction

Route B, 2 Passes
Route E, 4 Passes
Annelaed Bulk Copper (75 µm)
Nanocrystalline Powder Consolidate Compression Experiments

Nanocrystalline Cu powder (150 nm)
Processed with ECAE Compression @ RT
Compression axis = Extrusion Direction
Microstructural Evolution of Nanocrystalline Powder Consolidate

Initial Powder

2B

2C
Fracture Surface of Nanocrystalline Powder Consolidate
# Grain Size vs. Strength Relationship

<table>
<thead>
<tr>
<th>ECAE Route</th>
<th>Powder Size</th>
<th>Grain Size</th>
<th>Tension (Extrusion Direction)</th>
<th>Compression</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>X-Ray</td>
<td>TEM</td>
<td>$E$ (GPa)</td>
</tr>
<tr>
<td>1A</td>
<td>- 325 mesh (4.2 µm from X-Ray)</td>
<td>315 nm</td>
<td>200 – 300 nm (some grains &gt;500 nm)</td>
<td>115</td>
</tr>
<tr>
<td>2A</td>
<td>300 nm</td>
<td>200 – 300 nm (some grains &lt; 100 nm)</td>
<td>108</td>
<td>433</td>
</tr>
<tr>
<td>2C</td>
<td>250 nm</td>
<td>200 - 300 nm</td>
<td>114</td>
<td>-</td>
</tr>
<tr>
<td>4C</td>
<td>260 nm</td>
<td>250 nm</td>
<td>115</td>
<td>418</td>
</tr>
<tr>
<td>2B</td>
<td>130 nm (from X-Ray, about 100 nm from TEM)</td>
<td>110 nm</td>
<td>70 - 100 nm</td>
<td>104</td>
</tr>
<tr>
<td>2C</td>
<td>140 nm</td>
<td>~200 nm and 50 - 80 nm</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>4E</td>
<td>-</td>
<td>~250 nm and 40 – 80 nm</td>
<td>92</td>
<td>516</td>
</tr>
<tr>
<td>ECAE processed bulk Cu (1A)</td>
<td>-</td>
<td>~200nm- (&gt;1 µm in the elongated direction)</td>
<td>120</td>
<td>287</td>
</tr>
<tr>
<td>ECAE processed bulk Cu (2C)</td>
<td>-</td>
<td>200-500nm</td>
<td>116</td>
<td>310</td>
</tr>
<tr>
<td>ECAE processed bulk Cu (4C)</td>
<td>-</td>
<td>200-500nm</td>
<td>125</td>
<td>346</td>
</tr>
<tr>
<td>Annealed Bulk Copper</td>
<td>-</td>
<td>75 µm</td>
<td>120</td>
<td>51</td>
</tr>
</tbody>
</table>

FD = Flow Direction, ED = Extrusion Direction
Conclusions

- Successful consolidation of microcrystalline copper particles to full density. Route 2B resulted in the best results.

- Nanoparticles were consolidated with relative success. Tensile strength of 550 MPa and Compressive strength of 780 MPa were achieved. Low tensile ductility is attributed to the inter-agglomerate debonding which might be due to moisture or an oxide layer.

- ECAE appears to be a viable approach to obtain bulk nanocrystalline (<100 nm) materials for structural applications.
Interest in production of bulk amorphous metal for structural applications.

Approach:
- ECAE consolidation of gas-atomized Zr-based amorphous metal powder into bulk amorphous metal. Vitreloy 106a is chosen because of a large $T_x - T_g$.
- Composition: $(\text{Zr}_{58.5}\text{Nb}_{2.8}\text{Cu}_{15.6}\text{Ni}_{12.8}\text{Al}_{10.3})$

Objectives:
- No crystallization
- Good particle-to-particle bonding
- Tensile strength comparable to cast counterpart
- Consolidate dimensions greater than casting
Initial Amorphous Zr-based Powders

- **Powder characteristics**
  - $\text{Zr}_{58.5}\text{Nb}_{2.8}\text{Cu}_{15.6}\text{Ni}_{12.8}\text{Al}_{10.3}$
  - Gas atomized at AMES-MPC
  - $38 \mu\text{m} < \text{Diameter} < 150 \mu\text{m}$
  - Batch 1: $\sim 1280$ ppmw oxygen ($0.57\text{ at }\%$) and $\sim 266$ carbon ppmw in dia. $< 75 \mu\text{m}$
  - Batch 2: $\sim 780$ ppmw oxygen
  - Amorphous character

As-Received Powder

Batch 1: $T_g = 398 \degree\text{C}; \quad T_x = 460 \degree\text{C}$

$\Delta T = 62 \degree\text{C}$

Batch 2: $T_g = 403 \degree\text{C}; \quad T_x = 480 \degree\text{C}$

$\Delta T = 77 \degree\text{C}$
# Extrusions Conditions

<table>
<thead>
<tr>
<th>Billet ID</th>
<th>Cu050</th>
<th>Cu051</th>
<th>Cu052</th>
<th>Cu053</th>
<th>Cu054</th>
<th>Cu058</th>
<th>Ni024</th>
<th>Ni023</th>
<th>Ni029</th>
<th>Ni029</th>
<th>Ni041</th>
</tr>
</thead>
<tbody>
<tr>
<td>Extrusion Route</td>
<td>1A</td>
<td>1A</td>
<td>1A</td>
<td>1A</td>
<td>1A</td>
<td>1A</td>
<td>1A</td>
<td>1A</td>
<td>1A</td>
<td>2B</td>
<td>2C</td>
</tr>
<tr>
<td>T(_{\text{die}}) (°C)</td>
<td>440</td>
<td>420</td>
<td>420</td>
<td>420</td>
<td>400</td>
<td>430</td>
<td>410</td>
<td>430</td>
<td>410</td>
<td>420</td>
<td>410/420</td>
</tr>
<tr>
<td>T(_{\text{maximum}}) (°C)</td>
<td>459</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
<td>420</td>
<td>451</td>
<td>415</td>
<td>433</td>
<td>421</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>Punch Speed (mm/s)</td>
<td>6</td>
<td>1</td>
<td>6</td>
<td>12</td>
<td>6</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Time above T(_{g}) (s)</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
<td>148</td>
<td>195</td>
<td>382</td>
<td>231</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>Time above T(_{\text{tool}}) (s)</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
<td>23</td>
<td>77</td>
<td>44</td>
<td>147</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>Microhardness (HV(_{500}))</td>
<td>520 ± 35</td>
<td>493 ± 25</td>
<td>475 ± 15</td>
<td>480 ± 20</td>
<td>480 ± 33</td>
<td>484 ± 15</td>
<td>488 ± 4</td>
<td>490 ± 5</td>
<td>NA</td>
<td>493 ± 5</td>
<td>497 ± 4</td>
</tr>
</tbody>
</table>

Batch 1 (1280 ppmw O\(_2\)) (HV\(_{500}\) = 467 ± 35)  
Batch 2 (780 ppmw O\(_2\)) (HV\(_{500}\) = NA)

- Nickel cans contain V106a with 780 ppmw oxygen (T\(_{g}\) = 403°C - T\(_{x}\)=480 °C)

- Copper cans contain V106a with 1280 ppmw oxygen (T\(_{g}\) = 398°C - T\(_{x}\)=460 °C)

## Controlled Variables:

- Temperature  
- Strain Rate  
- Extrusion Rate  
- Oxygen Content  
- Hydrostatic Pressure
Temperature versus time for billets Cu058 and Ni023 showing the sample time above Tg and the rise in temperature due to material deformation as it passes through the shear zone. Both billets were extruded with the die at 430°C and with a punch speed of 6 mm/s and 0.5 mm/s for Cu058 and Ni023, respectively. Note that the horizontal axis is only a time scale and not an indicator of the amount of time into the processing.
Thermal History of V106a Powder
Consolidations on TTT Diagram

- 20°C/min - 1280 ppmw O
- 20°C/min - 780 ppmw O
- 40°C/min - 1280 ppmw O

Tx Batch 1
Tx Batch 2

Tg
DSC Curves for V106a Powder and Consolidates

- Combined effects of oxygen content, temperature, strain rate, and can material
Fine Interparticle Cracking

Ni029
410/420 °C
Route 2B
0.5 mm/s
Effect of Oxygen Content

1280 ppmw

Cu058
430 °C
Route 1A
6 mm/s

Ni023
430 °C
Route 1A
0.5 mm/s

780 ppmw

Cu052
420 °C
Route 1A
6 mm/s

Ni029
410/420 °C
Route 2B
0.5 mm/s
Compressive Response of Consolidated V106a Powder

Compressive Fracture Surfaces

430 °C, 6 mm/sec

420 °C, 6 mm/sec
Conclusions

- Full consolidation with one ECAE pass at temperatures of $T_g$ and higher without significant crystallization.

- Fine inter-particle cracks are sometimes present in the consolidate.

- Higher oxygen content level restricts t-T space for consolidation, decreases ductility, decreases $\Delta T$, promotes crystallization and inhibits interparticle bonding due to surface oxides.

- Zr-based amorphous metal powders with a substantial TTT opportunity window can be consolidated in the supercooled liquid region by ECAE.
Lessons Learned

Problems Identified

1. It is difficult to achieve grain sizes $\leq 100\text{nm}$ using ECAE when starting from coarse grain structures.

2. **Residual porosity** (from initial powder agglomerates) may be a problem for nanoparticle consolidation and mechanical properties.

3. **Particle surface contamination** (Oxygen, water, etc.) is highly detrimental to effective consolidation.

4. Severe plastic deformation may result in **local heating** and dynamic recrystallization (for crystalline phases) or crystallization (for amorphous phases).

5. Brittle material (whether the precursor is particulate or bulk) is difficult to ECAE process without cracking.
Lessons Learned

Encouraging Results

1. Cu nanopowder can be effectively consolidated by ECAE: One pass gives nearly full density; two passes improves mechanical properties.

2. ECAE consolidated Cu nanopowder has higher tensile and compression strengths than does wrought Cu given severe plastic deformation.

3. Amorphous metal powder, with a substantial supercooled liquid region, can be consolidated to nearly full density by one pass ECAE without significant crystallization.
Questions Remain

1. Are the properties of ECAE processed powder consolidates isotropic?

2. How do HIPing, area reduction extrusion and ECAE compare with respect to effectiveness (level of material properties in consolidate and ease of processing) of powder consolidation?

3. Can the ECAE process for powder consolidation be scaled up for the production of high efficiency structural components?

4. Can amorphous metal powder be consolidated below $T_g$?