Laser ultrasonic measurements of grain size during processing of metals and alloys.

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Real time microstructure control

- Complementary tool to control metallurgical processes
- Estimate optimum process parameters for novel metal and alloys
Thermo-mechanical processing lab
Real time sensing at high temperature

15 mm
Principle of the technique

**Broadband ultrasound pulse (2 to 30 MHz)**

Generation and detection laser pulses

Ultrasound pulse

Thermocouple

FEM simulation

Up to 50 waveforms measured per second

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Measured ultrasonic parameters

- Properties of ultrasound compressional waves
- Time of arrival of echoes -> Velocity $V$
- Amplitude of echoes -> Attenuation $\alpha(f)$

**Filtered signal**

\[
V = \frac{2(e + \epsilon)}{\tau} \\
\alpha(f) = \frac{20}{2e} \log \left( \frac{A_{echo(i)}}{A_{echo(j)}} \right)
\]
Velocity of ultrasonic wave

Rotated Elastic Tensor

\[ C_{ijkl} = \int c'_{ijkl} f(odf) \]

\[ T_{ik}(\vec{n}) = C_{ijkl} \vec{n}_j \vec{n}_l \]

\[ V = \sqrt{\sum \frac{K(odf)}{\rho}} \]

What can be investigated?
Phase transformation
Second phase/Precipitation
Recrystallization

EBSD to **Velocity map** (mm/µs)

**Velocity Distribution**

**EBSD to Velocity map (mm/µs)***

**Velocity Distribution**

**Pure Titanium**

**Fraction of Orientation**

**Pressure wave velocity (mm/µs)**

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In polycrystalline metals, scattering depends on ultrasonic wavelength

- **Rayleigh Region**
  \[ \alpha(D, \lambda) = C_r D^3 \lambda^{-4}, \lambda \gg D \]

- **Stochastic Region**
  \[ \alpha(D, \lambda) = C_s D \lambda^{-2}, \lambda \approx D \]

- **Diffusion Region**
  \[ \alpha(D, \lambda) = C_d / D, \lambda \ll D \]
How to estimate the grain size?

1) Reference sample $D_0$
2) **ONE ECHO METHOD**

Isolate only grain scattering

Measurement precision < 10 %

$$\alpha(f) = a + b f^n$$

Frequency dependant grain size parameter

$$b = C(T)[D_i^{n-1}(t) - D_0^{n-1}(t_0)] f^n$$

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Application to austenite in steel

1) Reference fine grain sample at room temperature

\[ \alpha(f) = a + bf^3 \]

2) Calibration developed at Timken (S.E. Kruger et al., Iron Steel Technol, (2005), 2(10),25
Application to hot rolling processes

✓ Grain size measurement after hot-deformation in Mo-TRIP steel

✓ Strain = 0.2 and 0.4
Austenite grain refinement

- Larger grain refinement at higher deformation strain

![Graph showing grain size vs. time with T_{def} = 900°C and 25 µm scale bars](image)
Nickel based super alloys

- Control the grain growth + dissolution of second phase particles prior to forging
- Starting structure has 20 µm polygonal grain
- + 2 to 3 % of delta phase precipitates
Stage of heterogeneous grain growth

- Local Nb microsegregations affect the stability of the second phase leading to heterogeneous grain growth (Fraction of large and small grains)
Metallographic analysis

- Evaluation of the mean grain size $\text{EQAD} = \sqrt{\frac{4\bar{A}}{\pi}}$
- Maximum 1% largest grain diameter

<table>
<thead>
<tr>
<th>Time (s)</th>
<th>$\text{EQAD}$(μm)</th>
<th>$D_{\text{MAX}}$(μm)</th>
<th>$\frac{D_{\text{MAX}}}{\text{EQAD}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>15</td>
<td>56</td>
<td>3.7</td>
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<tr>
<td>30</td>
<td>18</td>
<td>120</td>
<td>6.7</td>
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<tr>
<td>75</td>
<td>19</td>
<td>139</td>
<td>7.3</td>
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<tr>
<td>175</td>
<td>33</td>
<td>139</td>
<td>4.2</td>
</tr>
<tr>
<td>480</td>
<td>36</td>
<td>155</td>
<td>4.3</td>
</tr>
<tr>
<td>900</td>
<td>42</td>
<td>172</td>
<td>4.1</td>
</tr>
</tbody>
</table>

Cumulative Volume Fraction

Reduced grain size ($D/\text{EQAD}$)

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Correlation at 1050°C

• Evolution of the scattering parameter \( b \) with the relative change in mean grain size.

• Direct measurement of the coefficient \( C^* \)

\[
\sqrt{|1000 \cdot b(t_i, D_i)|} = C^* \sqrt{D_i^2(t_i) - D_0^2(t_0)}
\]

Linear regression coefficient \( C^* = 0.022 \)
Grain grow tests

- Insight into the grain growth behavior.
- Different grain growth stages
  
  1) Zener \[ \frac{dD}{dt} = K \left( \frac{1}{D} - P_0 \right) \]
  
  2) Rapid grain growth
  
  3) Parabolic \[ D^2 - D_{init}^2 = Kt \]
Criteria for abnormal grain growth

- Normalization procedure
- Time at the onset of abnormal grain growth

<table>
<thead>
<tr>
<th>$D_{init}$ (μm)</th>
<th>$K$ (μm$^2$.s$^{-1}$)</th>
<th>$P_0$ (μm$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>24</td>
<td>1.6</td>
<td>0.0574</td>
</tr>
</tbody>
</table>
Cobalt super alloys

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Empirical correlation

\[ \alpha(f) = a + bf^3 \]

\[ \sqrt{|1000b|} = \Gamma(T)\delta \left( \sqrt{|D_i^2 - D_0^2|} \right)^{1-\epsilon} \]

Grain size dependence

Temperature dependence
Grain growth model

\[ D^m - D_{\text{init}}^m = \Phi(T)t \]

Effective mobility

\[ \Phi(T) = \lambda_1 \exp\left(-\frac{\lambda_2}{kT}\right) \]
Closer look at the attenuation spectrum

- These empirical approaches require adjusting the effective bandwidth
- Because they do not account for multiple regimes of scattering

\[ \alpha(f) = a + bf^3 \]
Computer generated grain structure

- Finite element simulation of wave propagation on polycrystalline materials
- Centroidal voronoi tessellation: all cells have 6 faces but the final structure is not ordered
Material properties

- Single crystal stiffness tensor
- FCC iron at 1423 K (Zarestky et al., 1987, Phys.Rev. B 35(9), pp.4500)
- Single crystal elastic constant:
  \[ c_{11} = 154 \text{ GPa} \]
  \[ c_{12} = 122 \text{ GPa} \]
  \[ c_{44} = 77 \text{ GPa} \]
- Zener Anisotropy factor
  \[ c_{44}/c' = 4.8 \]
- Crystallographic orientation

![Velocity Graph](attachment:image.png)
FEM simulation of ultrasound propagation

- Displacement field for austenite (D = 300μm)
Results: Attenuation spectrum

- $D = 30 \, \mu m$
- $D = 70 \, \mu m$
- $D = 200 \, \mu m$
- $D = 300 \, \mu m$
- $D = 100 \, \mu m$
- $D = 400 \, \mu m$
Validation using austenite calibration

- By selecting appropriate frequency range, the austenite calibration provide satisfying agreement with FEM generated attenuation spectrum

![Graph 1](image1.png)

![Graph 2](image2.png)
Validation with scattering theory

Normalized attenuation

Normalized frequency

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New approach for grain size measurements

- Evaluation of grain size accounting for multiple regimes of scattering
- Using FEM simulated attenuation and/or scattering theory to predict grain size.
Quantitative tool to validate LUMet results

Example: Attenuation spectrum measured in austenite at high temperature.

Least square approach on FEM data provide quantitative estimate of the mean grain size.
Conclusions

• Ultrasonic attenuation can be sensitive to the self similarity of grain size distribution.

• FEM are integrated to simulate the wave propagation in anisotropic aggregate.

• Although in 2D (plain strain), it gives quantitative results.

• Empirical methodology (single scattering regime) have limitations in coarse grained structure.
### Reference (isotropic) material

- Random ODF (Volume Fraction of orientation V)
- Weighted average on elastic tensor (T)

\[
\langle T \rangle^{\text{Reuss}} = \left[ \sum_{m=1}^{M} V_m T^{-1}(g_m^c) \right]^{-1}.
\]

\[
\langle T \rangle^{\text{Voigt}} = \sum_{m=1}^{M} V_m T(g_m^c).
\]

<table>
<thead>
<tr>
<th>Stiffness tensor (Unit: GPa)</th>
<th>Reuss (isoStress)</th>
<th>Hill (Average)</th>
<th>Voight (isoStrain)</th>
</tr>
</thead>
<tbody>
<tr>
<td>X (RD)</td>
<td>173.6 112.0 112.4</td>
<td>2.1 1.1 -1.0</td>
<td>188.3 104.6 105.1</td>
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<tr>
<td>Y (TD)</td>
<td>112.0 173.9 112.2</td>
<td>-1.0 -2.1 -1.0</td>
<td>104.6 188.5 104.9</td>
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<tr>
<td>Z (ND)</td>
<td>112.4 112.2 173.4</td>
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<tr>
<th>Wave Velocity (mm/us)</th>
<th>P-Wave</th>
<th>S1-Wave</th>
<th>S2-Wave</th>
<th>P-Wave</th>
<th>S1-Wave</th>
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<tr>
<td>X (RD)</td>
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<td>2.0735</td>
<td>1.9493</td>
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<tr>
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<td>Z (ND)</td>
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<td>1.9542</td>
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<td>2.4053</td>
<td>2.2821</td>
<td>5.1625</td>
<td>2.6948</td>
<td>2.5685</td>
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Selection of appropriate averaging

- Velocity in the small grain size sample should be close to satisfy the isotropic condition.

\[ D = 30 \text{ um} \Rightarrow V = 5.1035 \pm 0.005 \]

\[ D = 100 \text{ um} \Rightarrow V = 5.134 \pm 0.048 \]

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