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Experimental Verification of Solidification Stress Theory

by

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Objectives

- Crack-free ceramic waste form (CWF)
- Analyze Temperature Data to Analyze Cracking in CWF2
- Use Sound to Determine when Cracking Occurs
- Use Destructive Testing to Determine Where Cracking Occurs
- Verify Solidification Stress Theory
- Apply Solidification Stress Theory to explain the cracking in CWF2

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Glass/Ceramic Cylinders Crack During Formation

- Cracking (always) occurs in forming large glass or ceramic cylinders (>15 inch diameter)
- Cracking occurs in the cooldown from the formation temperature
- A crack-free formation is desired
- Crack free - more resistance to leaching

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Existence of New Stress Proposed

- Model developed here proposes a new stress - Solidification Stress
- Permanent stress set-in at high temperature - during solidification – Solidification stress
- This stress causes failure as the CWF nears room temperature.
- Thermal stress does not cause this failure

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Test for Solidification Stress

- Solidification stress is in addition to and of opposite sign of the thermal stress
- Cracking of the CWF would occur at low temperatures if solidification stress exists
- Cracking would occur at high temperatures if it does not
- Cracking would start in inner region

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Thermal Stress Model (not pre-stressed)

- Cylinder made up of concentric annuli
- All intrinsically attached in a solid cylinder
- Length determined by average temperature
- Each annulus same length as the average
- Above T_{avg} forced shorter - in compression
- Below T_{avg} forced longer so are in tension.

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Thermal Stress Model (not pre-stressed)

- Not pre-stressed means no thermal stress when all annuli are at the same temperature
- The amount that annulus i must be elongated to reach the average length, L, is

$$\Delta L_i = L\alpha(T_{avg} - T_i)$$

$$where\ \alpha = thermal\ \text{expansion}\ coefficient$$

$$T_i = Temperature\ of\ \ annulus\ i$$

$$T_{avg} = Radially\ averaged\ temperature$$

Thermal Stress Model (not pre-stressed)

- **Stress, σ_i, induced into an annulus elongated by a length, ΔL_i, is**

$$\sigma_i = \frac{E}{1-\mu}\frac{\Delta L_i}{L}$$

$$where \quad E = Young's\ Modulus$$

$$\mu = Poisson's\ ratio$$

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Thermal Stress Model (not pre-stressed)

- Combine the two equations to get Axial Stress at Midplane at any r

$$\sigma_i = \frac{E\alpha}{1-\mu}(T_{avg} - T_i)$$

- Stress at surface largest for cooling cylinder
- Circumferential surface stress same as the axial

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Pre-stresses

- A stress that exists when the temperature is uniform is pre-stress
- Pre-stress induced by solidification is set-in stress
- Total σ = set-in σ + thermal σ

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Set-in Stress Model

- CWF concentric annuli – start all liquid
- Surface lowest T; annuli solidify inwards
- Each same length when solidifying $L(T_s)$
- When second annulus solidifies, outer is shorter due to thermal contraction.
- Two intimately connected along length $L(T_0)$
- Length deficit, $L(T_s)$-$L(T_0)$, not connected

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Set-in Stress Model

- This process repeats as the annuli progressively solidify inward
- Length deficits result in dome shape on top
- Dome height larger with higher cooling rates
- Length deficit is the length not connected to the next outer annulus
- Length deficit causes set-in stress when the temperature becomes uniform

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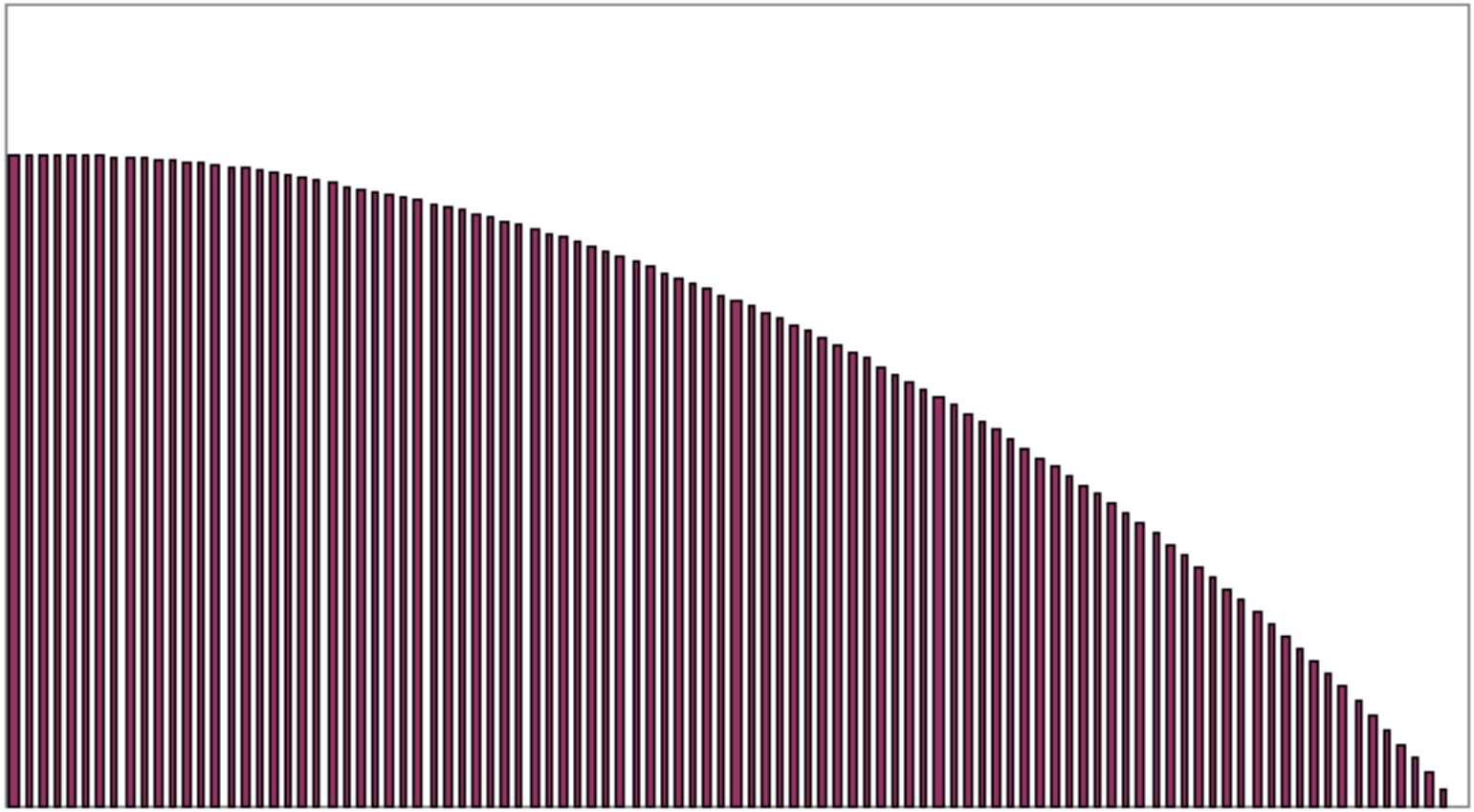
Concentric Annuli Heights



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Total Length Deficit

- Total length deficit at any r is

$$L_d = \int_{R_O}^{r} \left. \frac{dL}{dr} \right|_{s^+} dr$$

- Derivative is on outer side of solidification front, s^+

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Temperature Deficit

- Temperature deficit at annulus I

$$\Delta T_i = T_s - T_{i+1}$$

- where the annulus at r_i has just solidified,
- and T_{i+1} is the Temperature of the annulus at r_{i+1} at that time.

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Total Temperature Deficit

- Total temperature deficit at any r is

$$T_d = \int_{R_O}^{r} \left. \frac{dT}{dr} \right|_{S^+} dr$$

- Derivative is on outer side of solidification front, s^+
- Temperature deficit related to the length deficit by coefficient of thermal expansion

SET-IN Stress (Pre-stressed)

- **Temperature Deficit Stress at Midplane at any r**

$$\sigma_{\Delta i} = -\frac{E\alpha}{1-\mu}(\Delta T_{avg} - \Delta T_i)$$

- **Note the negative sign. Because deficit is the negative of the change in connective length.**

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Experiment Used to Verify

- CWF2 is a prototype vertical ceramic waste form 1-m tall, 0.5-m diameter, 400-kg, encased in stainless steel can 0.5-cm thick.
- 75% zeolite particles, 25% glass frit mixture Argon atmosphere furnace 10 day run
- Heat to melting (600 C), coalesces by over 50% to 925 C, holding at 925 C,
- Then cooling to solidification (600 C), then to room temperature.

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Measurements Available

- Stress can not be measured directly during or after formation
- CWF encased in can in furnace, so cracks cannot be observed.
- Cracking times can be recorded – correlate with temperature and calculated stress.
- Destructive examination can determine where tensile cracking occurred.

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Evidence of Cracking

- This cylinder developed many cracks during the low T portion of cooldown.
- At least 15 loud cracks were recorded over a period of 4 days. Sounds like gun shots.
- At the end of cooldown when the temperatures were below 400 C.
- Tensile Cracking pattern consistent with solidification stress theory

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Correlation of Cracking Sounds

- Surface and centerline temperatures at mid plane measured – allows calculation of midplane stress.
- Crack timing compared to calculated stress.
- Calculated total tensile stress exceeded the stress limit
- Verified cause of cracking is solidification stress, not thermal stress.

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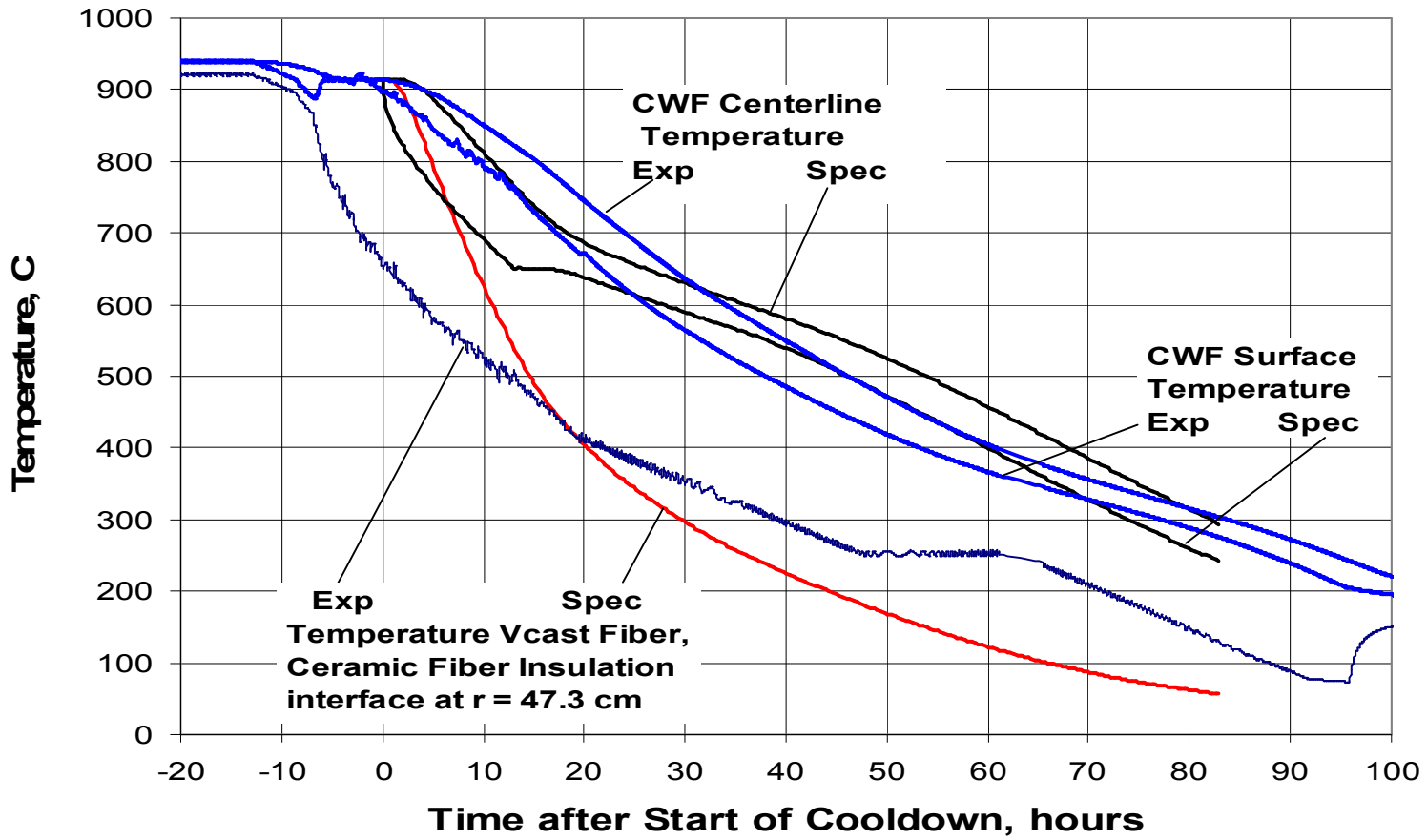
Experimental Temperatures



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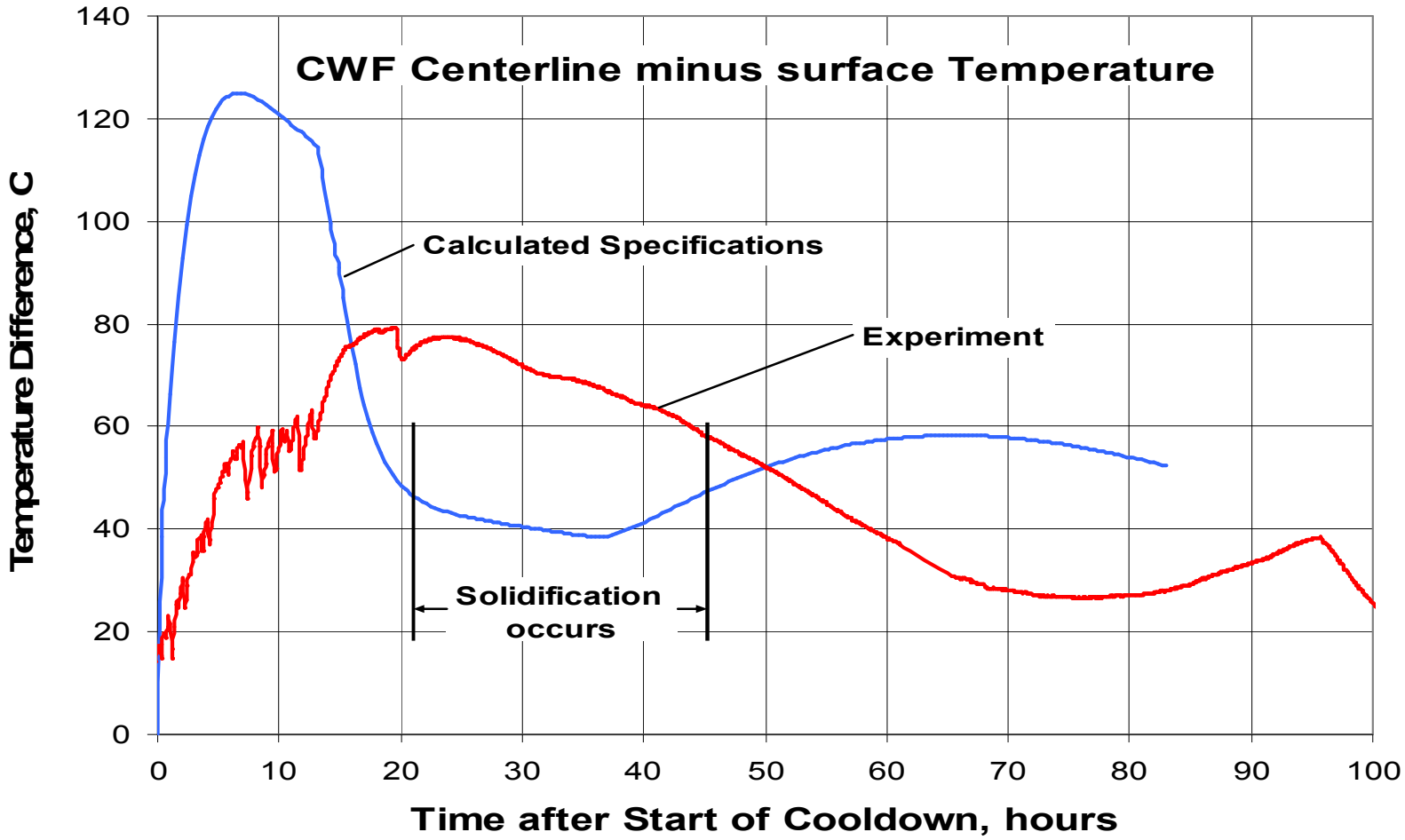
Desired vs Actual Temperature Difference



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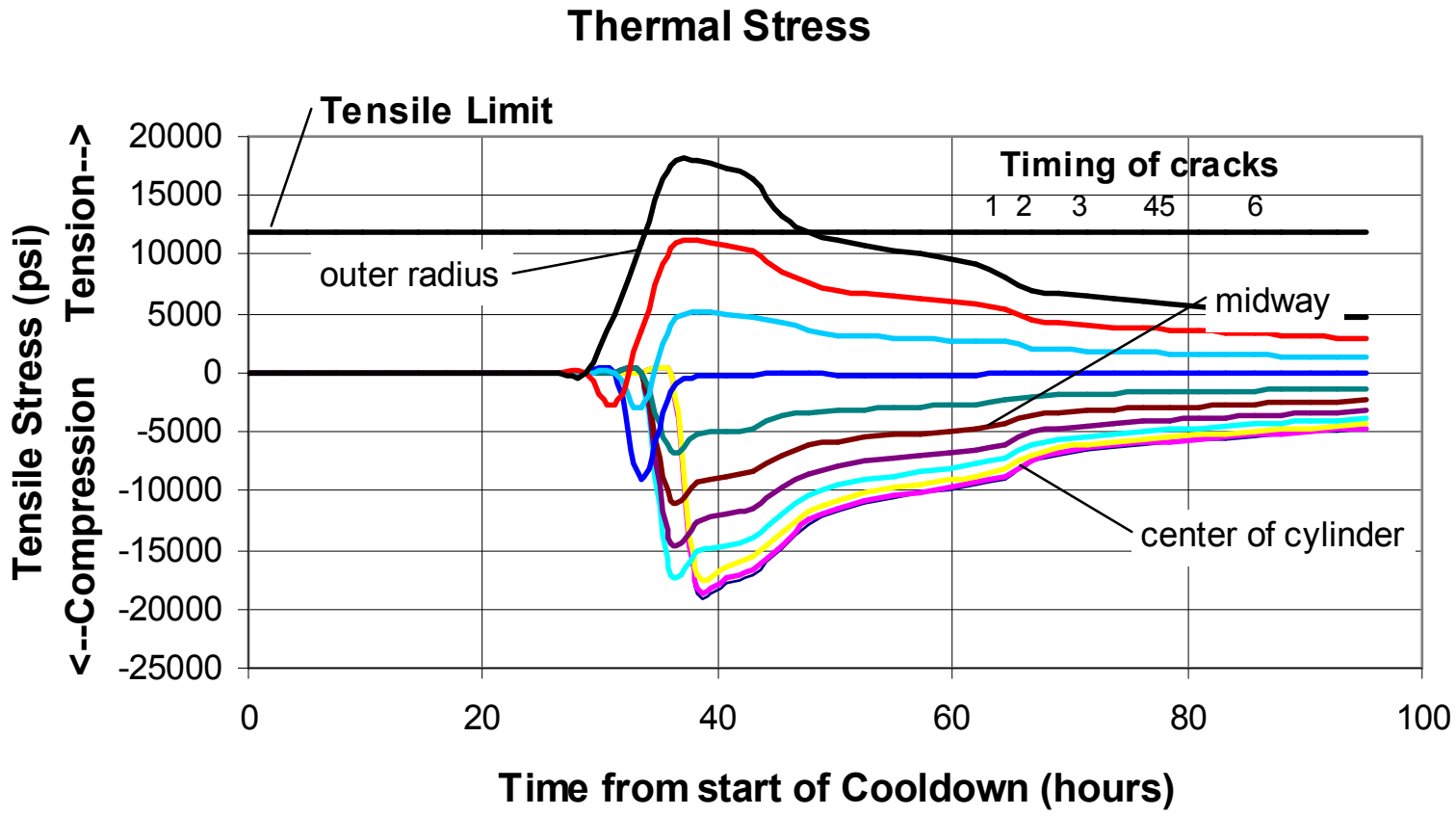
Thermal Stress Below Limit



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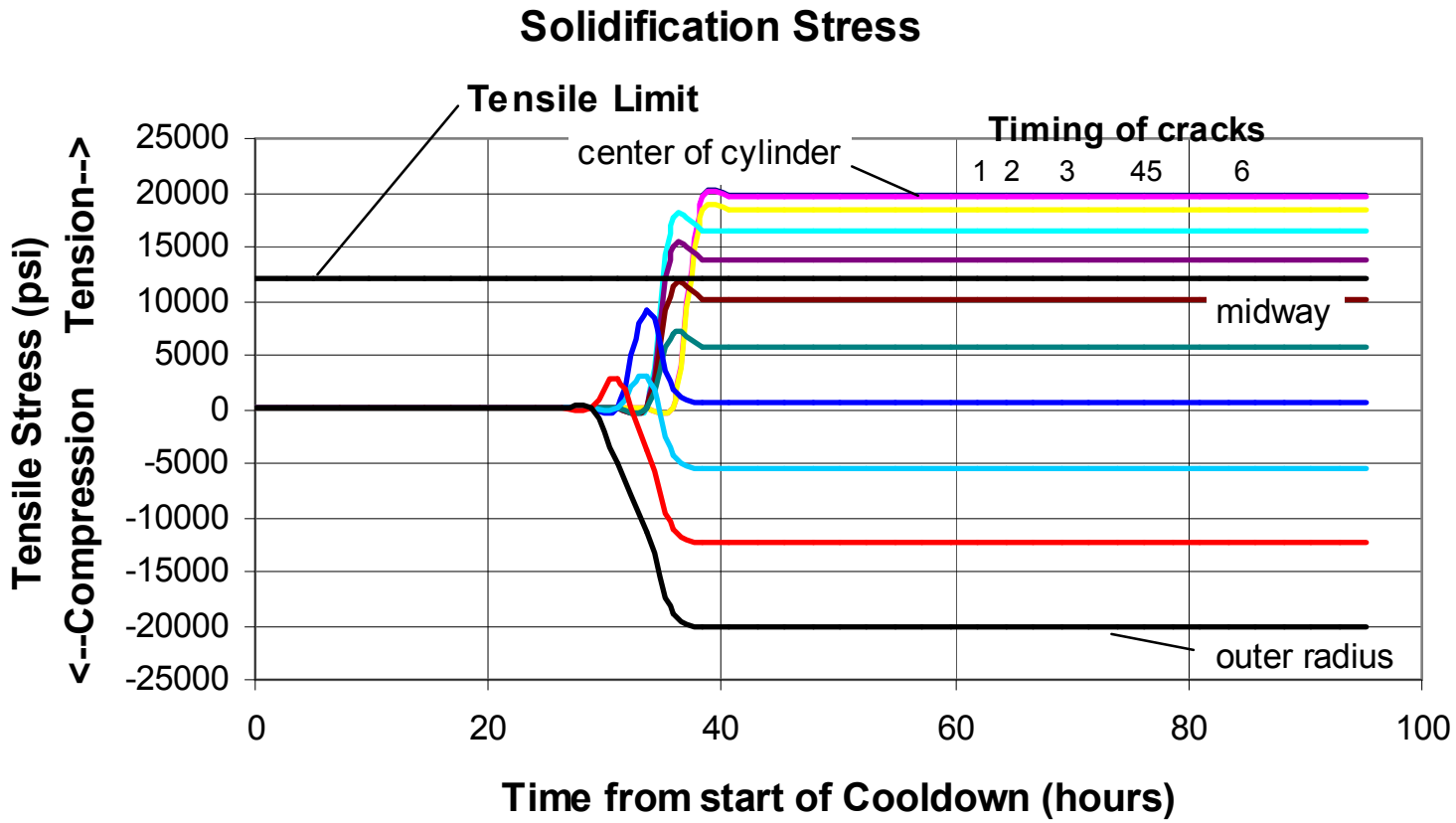
Solidification Stress Exceeds Limit



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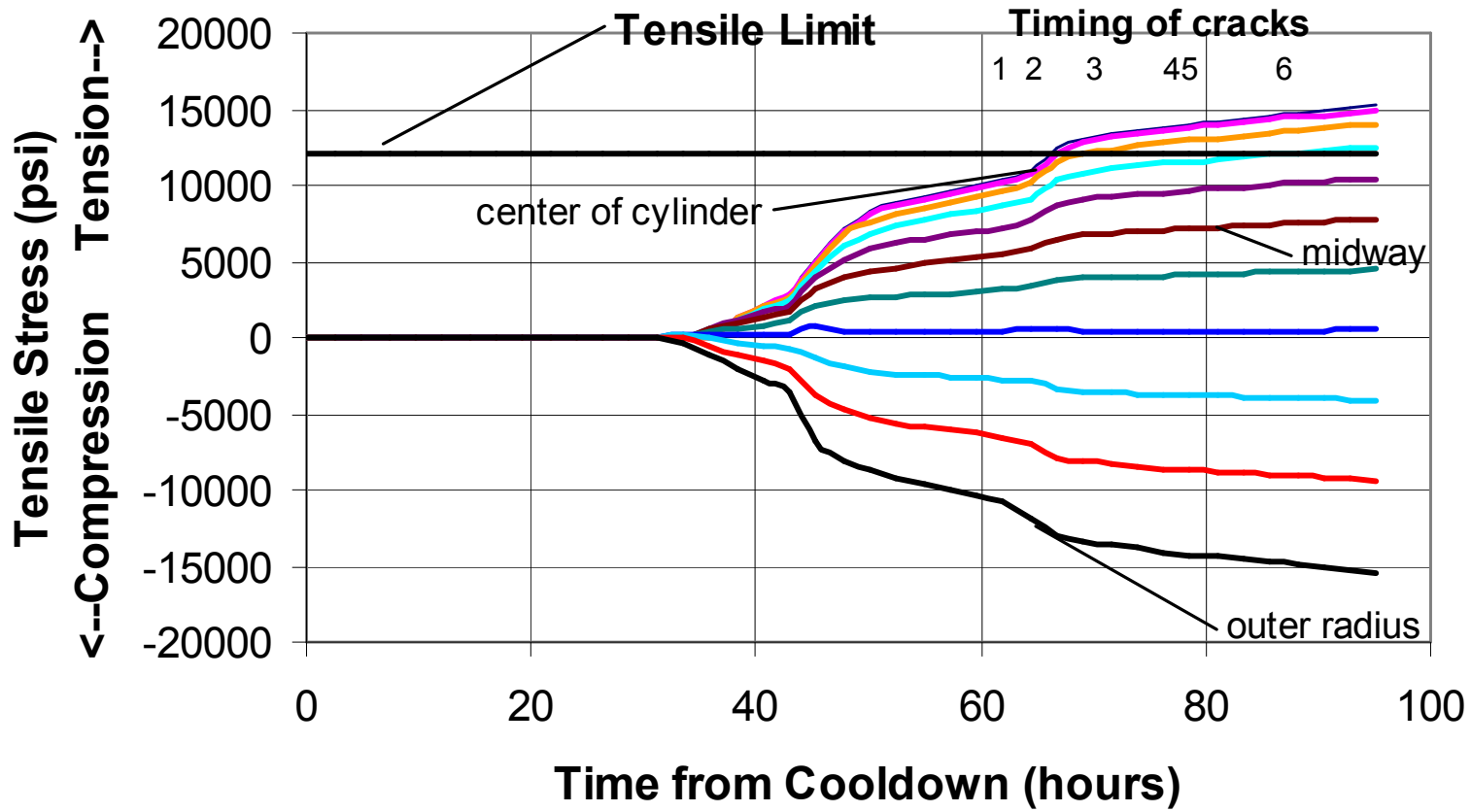
Total Stress Exceeds Limit



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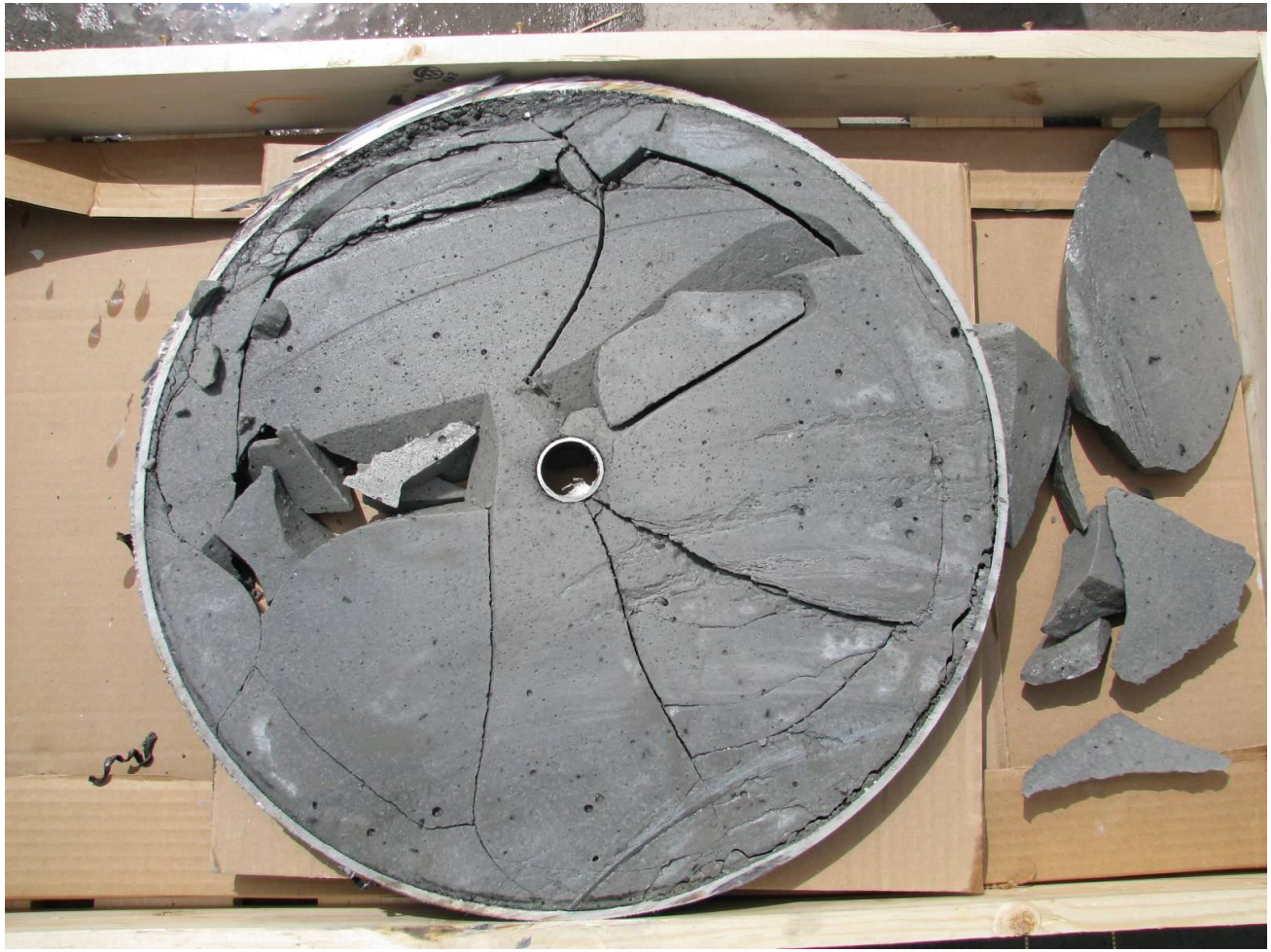
Ceramic Waste Form Damage



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Conclusions

- Cracking sounds determine when cracking occurs
- Destructive exam showed extensive cracking
- Cracking occurred during low temperature in inner region.
- Thermal stress alone predicted cracking at high temperatures in outer region.
- Solidification plus thermal stress predicts cracking at low temperatures in inner region.
- Experimental data from CWF2 verifies theory