Current state of ASME code rules

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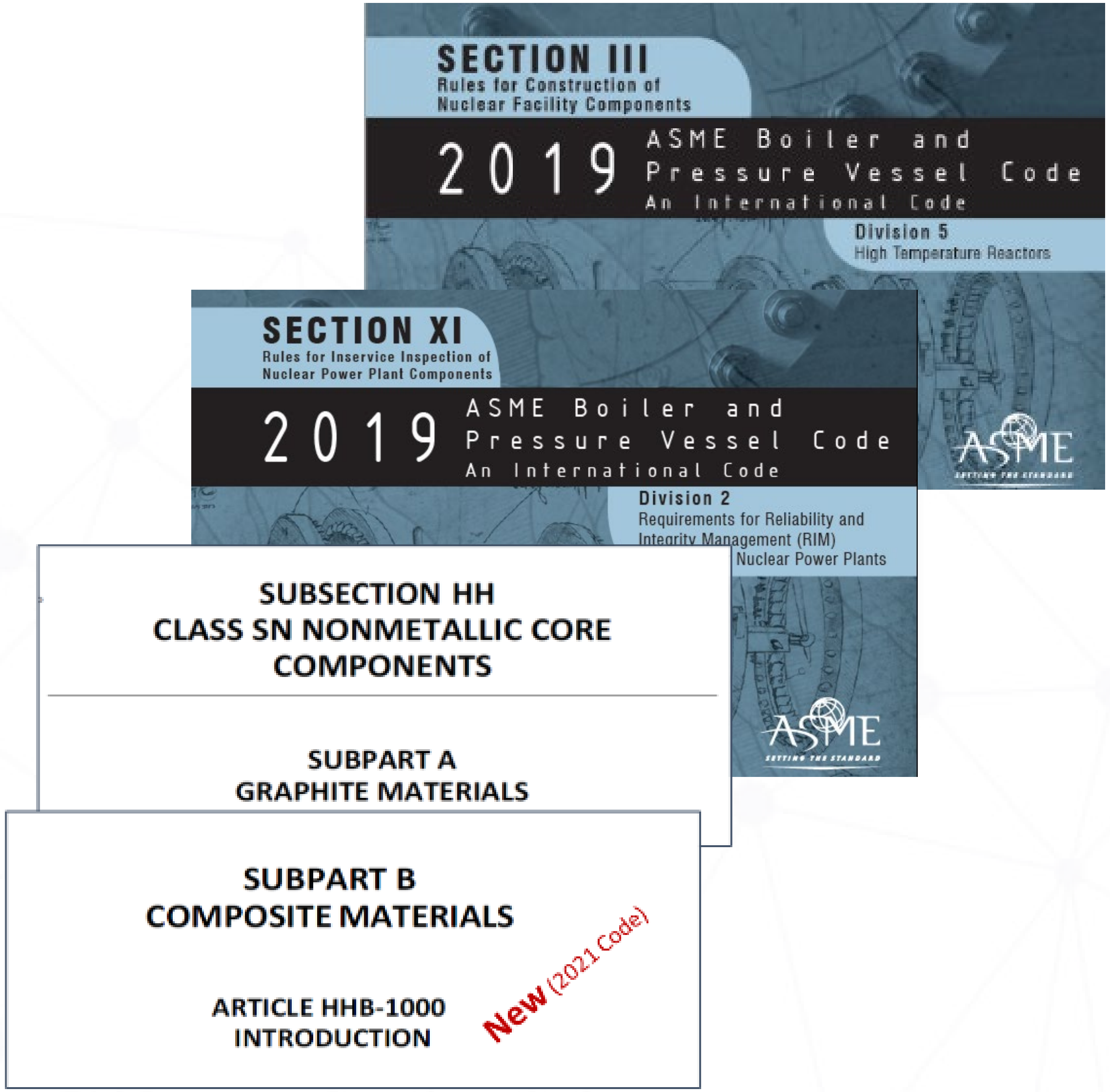
DOE-NRC Molten salt - Graphite material issues
Virtual Workshop
20-21 July 2022

ART
ADVANCED REACTOR TECHNOLOGIES

ASME Code Rules: Topics

- General review of code rules
  - What do they cover, what they don’t.
  - Probability of failure
    - *Material property parameters*
    - *Stresses and loads*
    - *Degradation on probability of failure (POF)*
- Specific Molten Salt issues of interest to the code
  - Material property changes
    - *Affect on failure in components*
  - Enhanced stresses due to intrusion
    - *Pre- and post-turnaround (crack formation) effects*
  - Degradation of material
    - *Abrasion/Erosion*
    - *Fluorination*
  - Combination effects
    - *MS + oxidation, MS + irradiation, etc.*
  - Other issues?



SECTION III
Rules for Construction of Nuclear Facility Components
2019 ASME Boiler and Pressure Vessel Code
An International Code
Division 5
High Temperature Reactors

SECTION XI
Rules for Inservice Inspection of Nuclear Power Plant Components
2019 ASME Boiler and Pressure Vessel Code
An International Code
Division 2
Requirements for Reliability and Integrity Management (RIM)
Nuclear Power Plants

SUBSECTION HH
CLASS SN NONMETALLIC CORE COMPONENTS

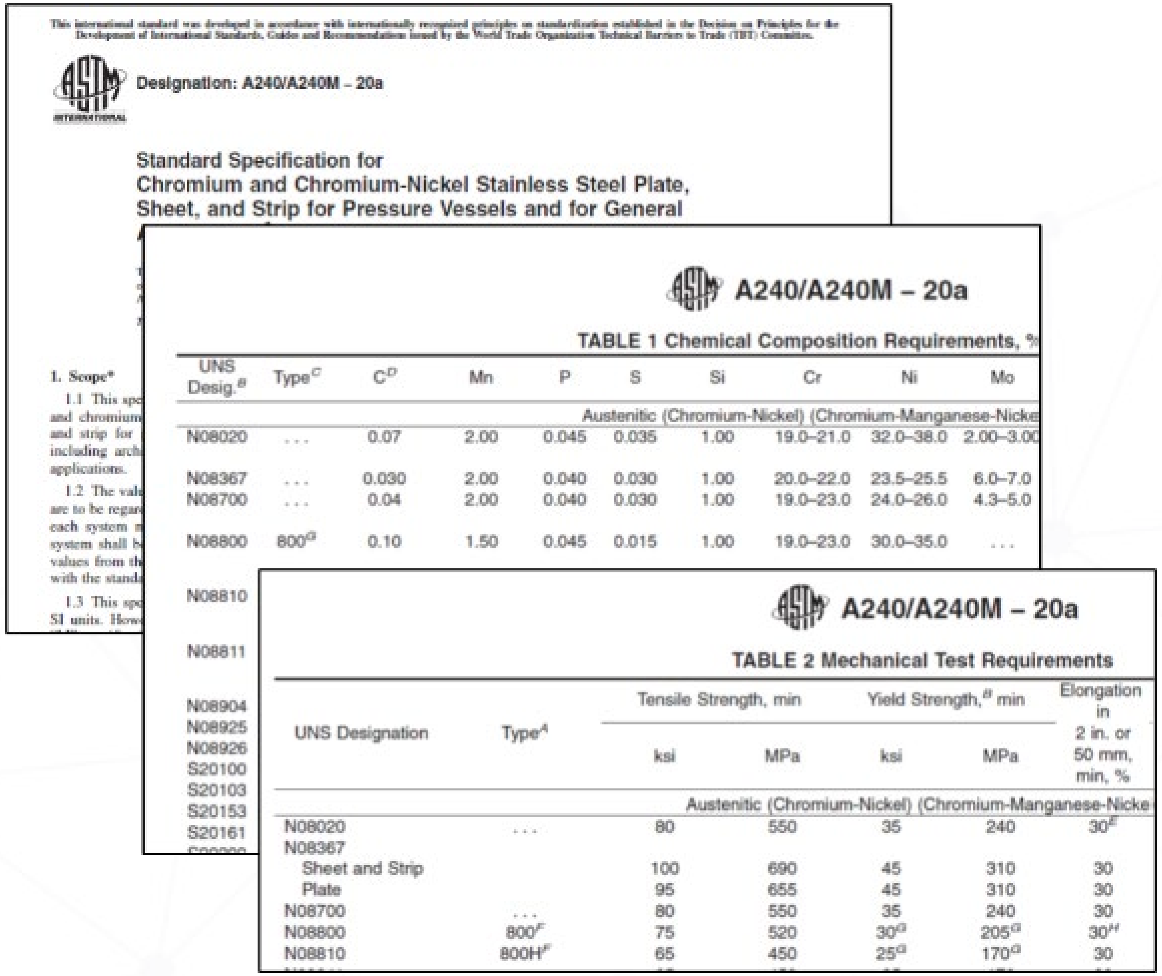
SUBPART A
GRAPHITE MATERIALS

SUBPART B
COMPOSITE MATERIALS

ARTICLE HHB-1000
INTRODUCTION

New (2021 Code)

Licensing challenge: No graphite fabrication standards



This international standard was developed in accordance with internationally recognized principles on standardization established in the Decision on Principles for the Development of International Standards, Guides and Recommendations issued by the World Trade Organization Technical Barriers to Trade (TBT) Committee.

Designation: A240/A240M – 20a

Standard Specification for
Chromium and Chromium-Nickel Stainless Steel Plate,
Sheet, and Strip for Pressure Vessels and for General

1. Scope*

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A240/A240M – 20a

TABLE 1 Chemical Composition Requirements, %

UNS Desig.[B]	Type[C]	C[D]	Mn	P	S	Si	Cr	Ni	Mo
							Austenitic (Chromium-Nickel) (Chromium-Manganese-Nicke		
N08020	. . .	0.07	2.00	0.045	0.035	1.00	19.0–21.0	32.0–38.0	2.00–3.00
N08367	. . .	0.030	2.00	0.040	0.030	1.00	20.0–22.0	23.5–25.5	6.0–7.0
N08700	. . .	0.04	2.00	0.040	0.030	1.00	19.0–23.0	24.0–26.0	4.3–5.0
N08800	800[G]	0.10	1.50	0.045	0.015	1.00	19.0–23.0	30.0–35.0	. . .
N08810									
N08811									
N08904									
N08925									
N08926									
S20100									
S20103									
S20153									
S20161									

A240/A240M – 20a

TABLE 2 Mechanical Test Requirements

UNS Designation	Type[A]	Tensile Strength, min		Yield Strength,[B] min		Elongation in 2 in. or 50 mm, min, %
		ksi	MPa	ksi	MPa	
				Austenitic (Chromium-Nickel) (Chromium-Manganese-Nicke		
N08020	. . .	80	550	35	240	30[E]
N08367						
Sheet and Strip		100	690	45	310	30
Plate		95	655	45	310	30
N08700	. . .	80	550	35	240	30
N08800	800[F]	75	520	30[G]	205[G]	30[H]
N08810	800H[F]	65	450	25[G]	170[G]	30

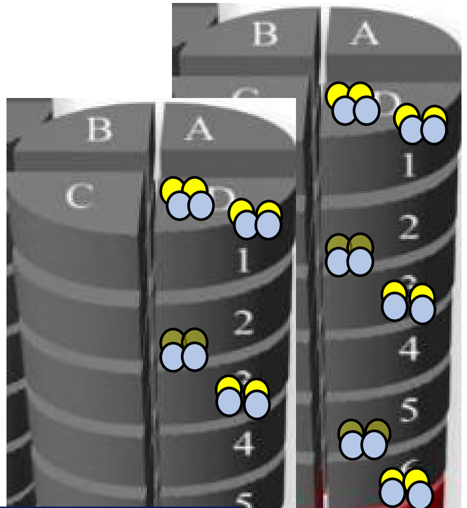
- No “Standard” nuclear graphite
  - Nothing like metals have
  - ASTM D7219 provides minimum property values (not fabrication standard)
  - This is a geologic material
- All graphite grades **are proprietary**. Only limited/general fabrication data is known.
  - Each grade has closely guarded, proprietary formulae owned by graphite suppliers
  - Unique individual change to material properties in reactor conditions
- But the good news is that all grades react similarly under nuclear core conditions
  - Specific changes are dependent upon individual grade
  - Much <u>more</u> uniform response pre-turnaround dose
  - Much <u>less</u> uniform response post-turnaround dose

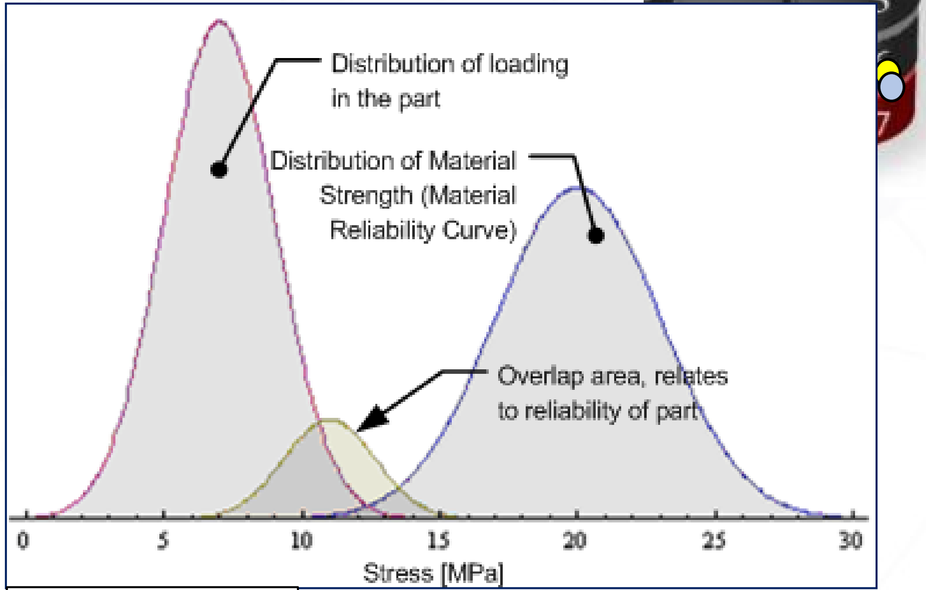
What ASME Code Rules DO cover?

Generally, the rules are pretty good and cover most of the critical areas of interest to establish a safety envelope.

- Robust un-degraded (unirradiated) construction rules
- Their weakness is in the details: How to establish and apply degradation, how to define component failure, how to calculate the probability of failure, etc.

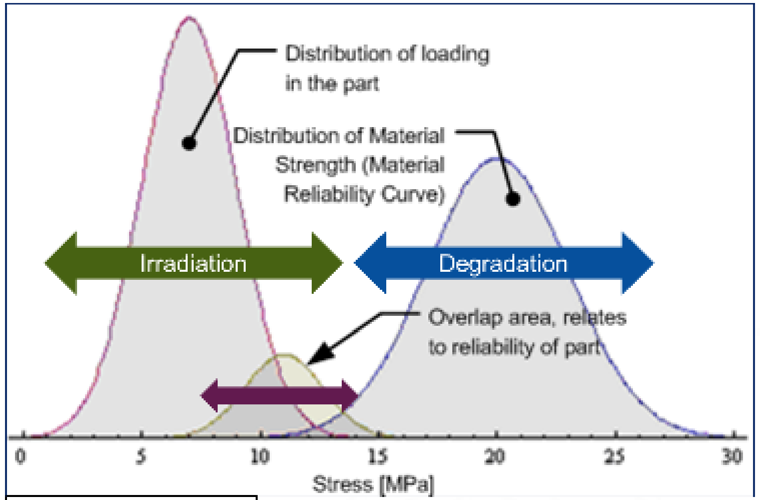
- What they **do** cover:
  - Establishes a workable probabilistic methodology
  - Establishes specific rules for probability of failure (POF)
    - *Three Assessments (Simple, Full, Test)*
  - *Establishes material properties of interest*
    - *Material Data Sheets (MDS)*
  - Establishes minimal test matrix for graphite qualification
    - *144 specimens with grain/144 against grain*
  - *Establishes <u>some</u> degradation issues*
    - *Oxidation, irradiation, combined Irr & Oxid*





From Dr. Mark Mitchell – PBMR Inc.

What ASME Code Rules DO NOT cover?

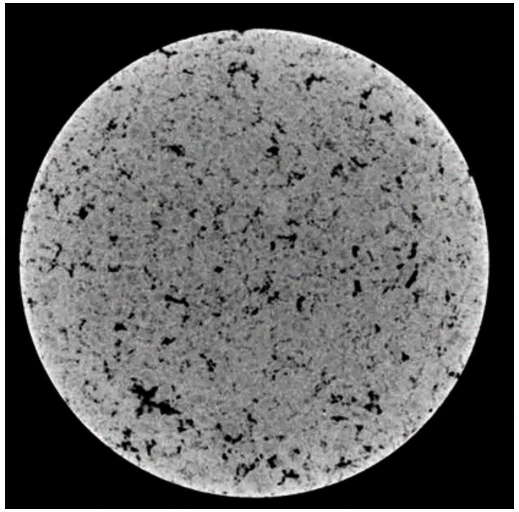


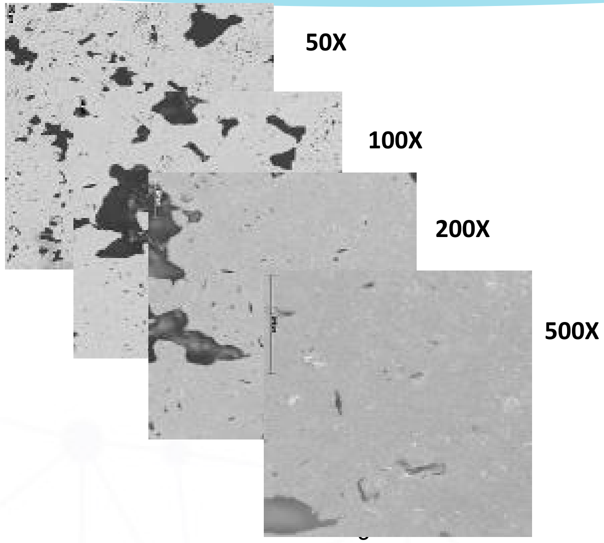
- In General, rules don't have enough <u>detail</u> on how to handle degradation
  - Section III, Div-5 are **Construction** code rules
  - But where should the degradation rules be written?
- Specifically, there are a few conspicuous areas where we are currently struggling
  - Failure and calculating failure of components
    - *Propagation of a single crack is not failure*
    - *The FEA mesh size and volume grouping methodology*
  - How to handle irradiation induced changes
    - *Before and after turnaround dose changes are critical*
      - ***Code case for each graphite grade? Or uniform behavior?***
    - *Temperature effects on irradiation changes*
  - Combined degradation effects
    - *Irradiation induced changes of oxidized material*
    - *Irradiation induced changes in molten salt environment*
  - Lack of testing standards
    - *ASME requires degraded properties but no way to get them*
  - Molten salt specific degradation issues

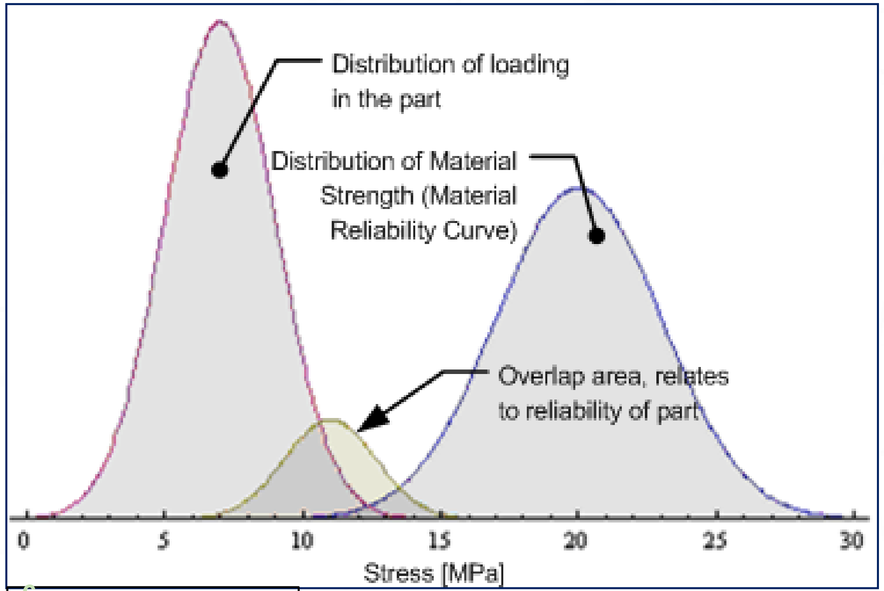
Probabilistic design

- We know nuclear graphite has significant flaws
  - *Some amount of failure (i.e., a crack) is certain*

- Therefore, core components need to be designed to accept some amount of failure.
  - *Probability of failure approach is taken*
  - *Based upon overlap of applied stresses and inherent strength of the nuclear grade used*



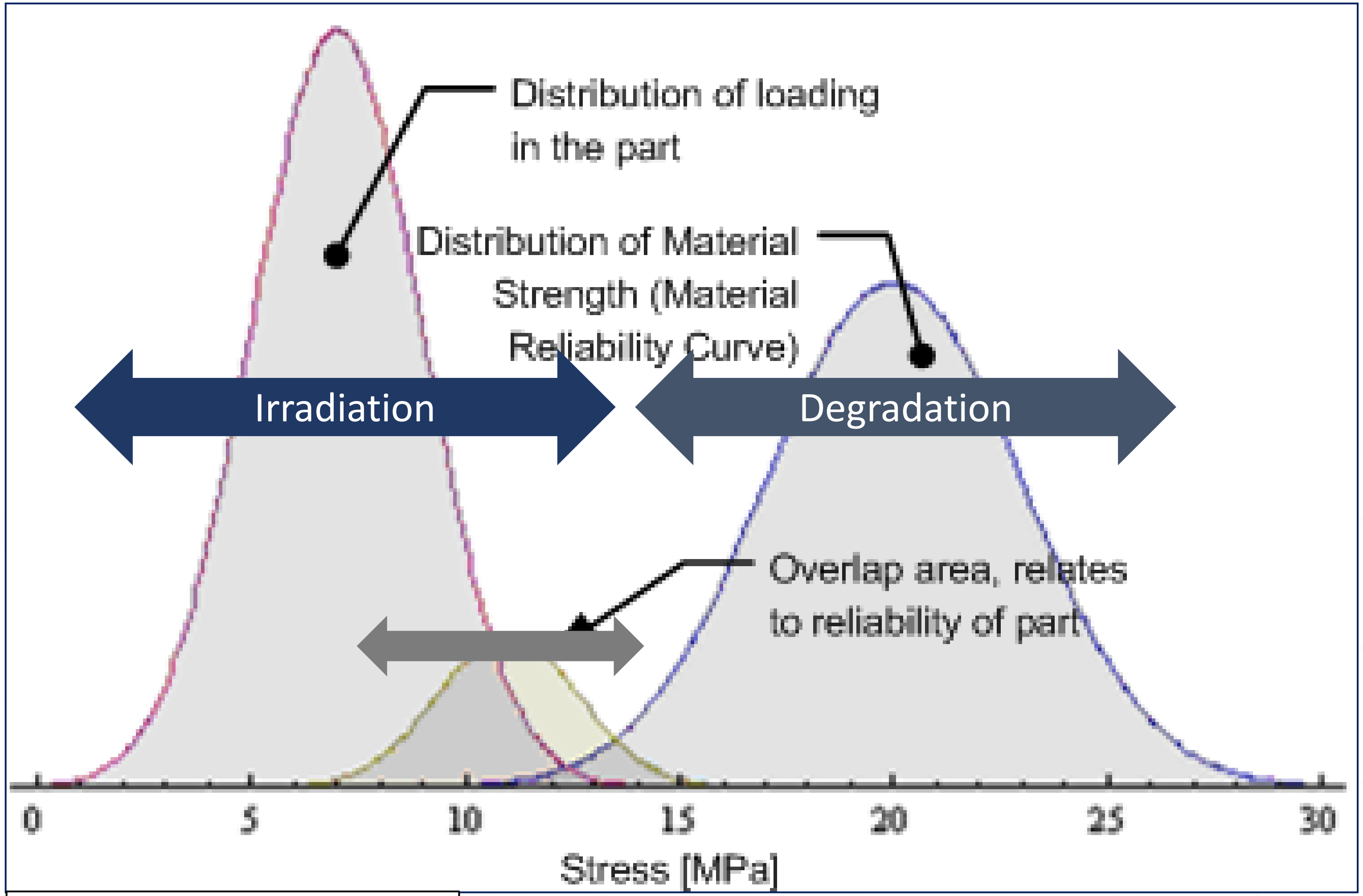




From Dr. Mark Mitchell – PBMR Inc.

Probablistic verses **deterministic** design approach
- Deterministic is generally too limiting for a brittle material
- A distribution of possible strengths in a material is needed for quasi-brittle materials (i.e., flaw size for graphite).
- Probability of failure in component based upon inherent **strength of graphite grade *and* induced stresses** during operation.

Applying degradation to POF



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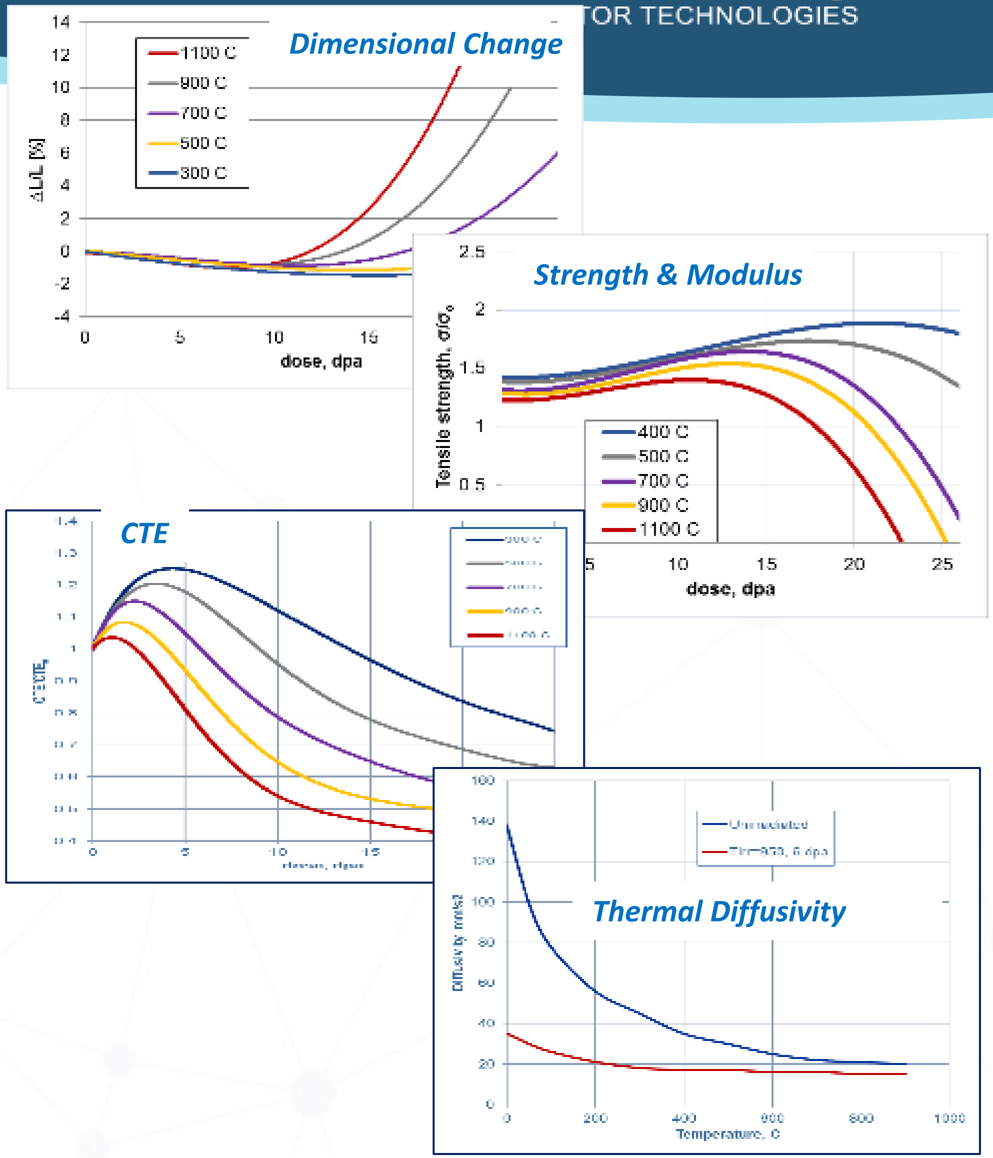
- Degradation changes the material properties
  - Irradiation strength increases
  - High temperature increases strength
  - Oxidation strength decreases
  - Molten salt strength (maybe) decreases
- Irradiation changes stress loading of the part
  - Dimensional change increases stress
  - Irradiation creep relieves stress
- Overlap will change.
  - POF will change

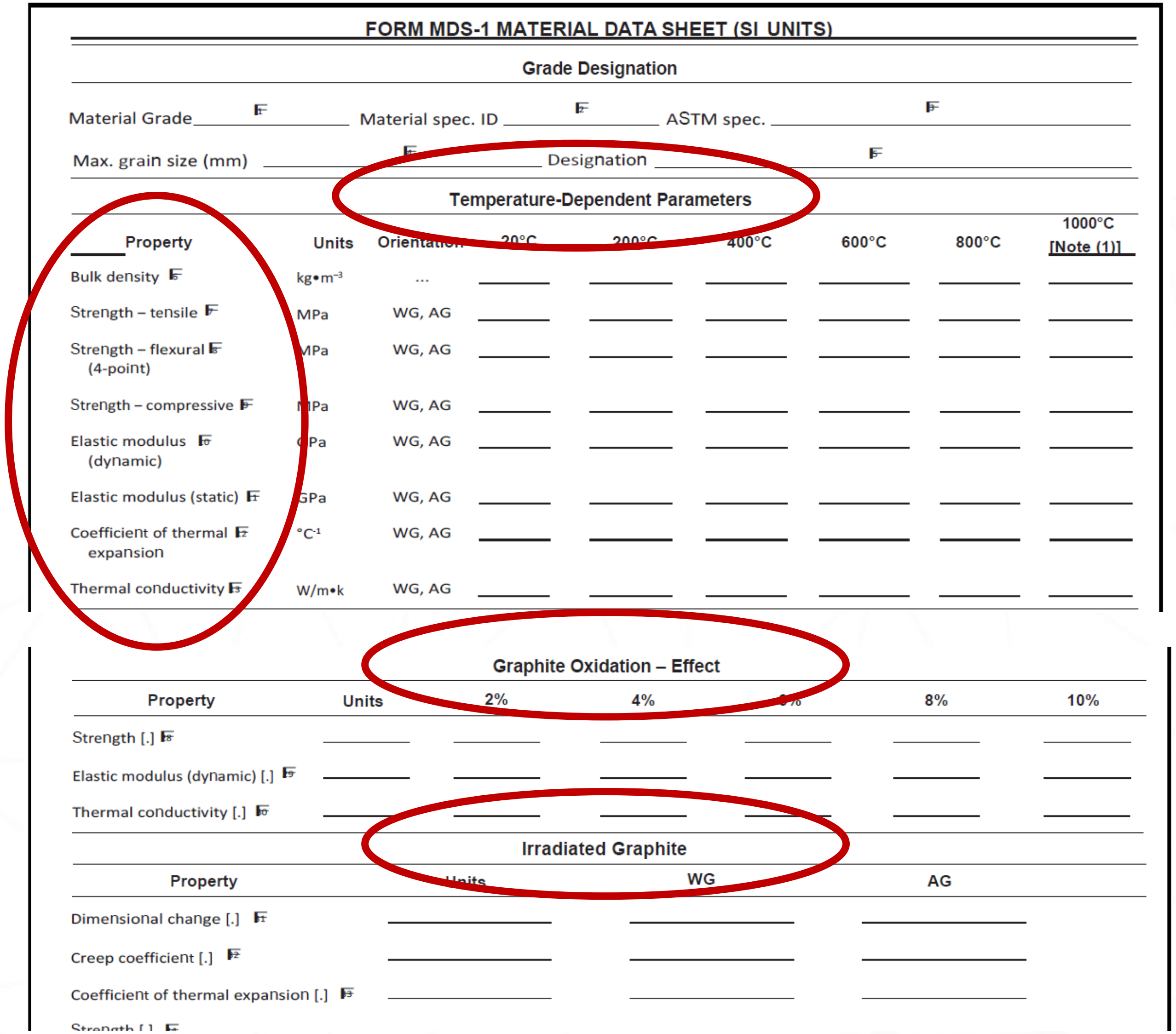
Irradiation Effects on Graphite Properties

General graphite irradiation behavior



- Significant changes occur during normal operation:
  - Dimensional change
    - *__Turnaround__ dose is key parameter*
    - *Highly temperature dependent*
  - Density
    - *Graphite gets denser with irradiation until __Turnaround__ dose*
    - *After __Turnaround__ density decreases (volumetric expansion)*
    - *Formation of microcracks (molten salt consideration)*
  - Strength and modulus
    - *Graphite gets stronger with irradiation …*
    - *Until __Turnaround__ dose is achieved. It then decreases*
  - Coefficient of thermal expansion
    - *Initial increase but then reduces before __Turnaround__*
    - *CTE is why properties are so temperature dependent*
  - Thermal conductivity
    - *Decreases almost immediately to ~30% of unirradiated values*
  - Oxidation rate
    - Increases approximately 2-3 times over unirradiated rates
- Significant changes **do not** typically occur in the following properties:
  - Neutron moderation, specific heat capacity, or emissivity

Graphite Degradation (ASME Material Data Sheets)



FORM MDS-1 MATERIAL DATA SHEET (SI UNITS)

Grade Designation

Material Grade ________ Material spec. ID ________ ASTM spec. ________

Max. grain size (mm) ________ Designation ________

Temperature-Dependent Parameters

Property	Units	Orientation	20°C	200°C	400°C	600°C	800°C	1000°C [Note (1)]
Bulk density	kg•m⁻³	...						
Strength – tensile	MPa	WG, AG						
Strength – flexural (4-point)	MPa	WG, AG						
Strength – compressive	MPa	WG, AG						
Elastic modulus (dynamic)	GPa	WG, AG						
Elastic modulus (static)	GPa	WG, AG						
Coefficient of thermal expansion	°C⁻¹	WG, AG						
Thermal conductivity	W/m•k	WG, AG						

Graphite Oxidation – Effect

Property	Units	2%	4%	6%	8%	10%
Strength [.]						
Elastic modulus (dynamic) [.]						
Thermal conductivity [.]						

Irradiated Graphite

Property	Units	WG	AG
Dimensional change [.]			
Creep coefficient [.]			
Coefficient of thermal expansion [.]			
Strength [.]			

ASME Data sheets capture most of the graphite material properties of interest:

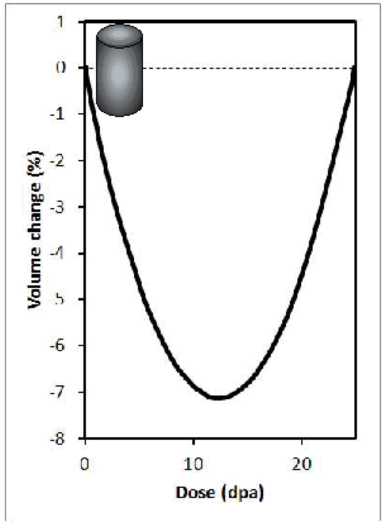
- Properties
  - *Density*
  - *Strength*
  - *Elastic modulus*
  - *CTE & Conductivity*
  - *Anisotropy*
- Temperature dependence
  - *Temperature affects everything*
- Irradiation effects
- Oxidation effects

Not covered (yet)

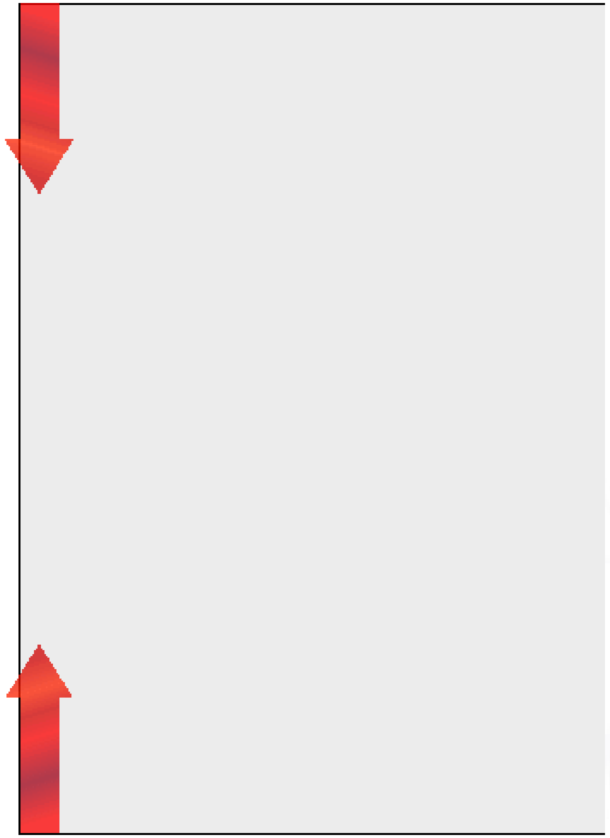
- **Molten salt issues**
- Abrasion/erosion
- Combination of degradation processes
- Details on how to use irradiation data

Induced stresses

- Fundamental material properties change with irradiation/oxidation/MS must be addressed
  - Applicant must assess stresses within component due to irradiation and thermal effects
    - *Internal stresses from dimensional change (Need creep response, too)*
    - *Turnaround dose is critical to assumptions of material response (tensile/compressive*
  - New cracks formed after turnaround



G. Haag," ***Properties of ATR-2E Graphite and Property Changes due to Fast Neutron Irradiation", Juel-4183, 2005***



- Applicant must also assess property changes to design due to irradiation, oxidation, and molten salt degradation
  - *Changes in density, strength, elastic modulus, CTE, erosion/wear, and thermal conductivity.*

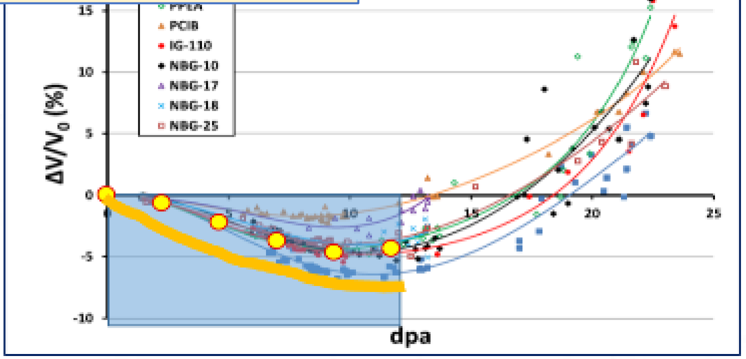
Current Nonmetallics Work Group (NWG) tasks?

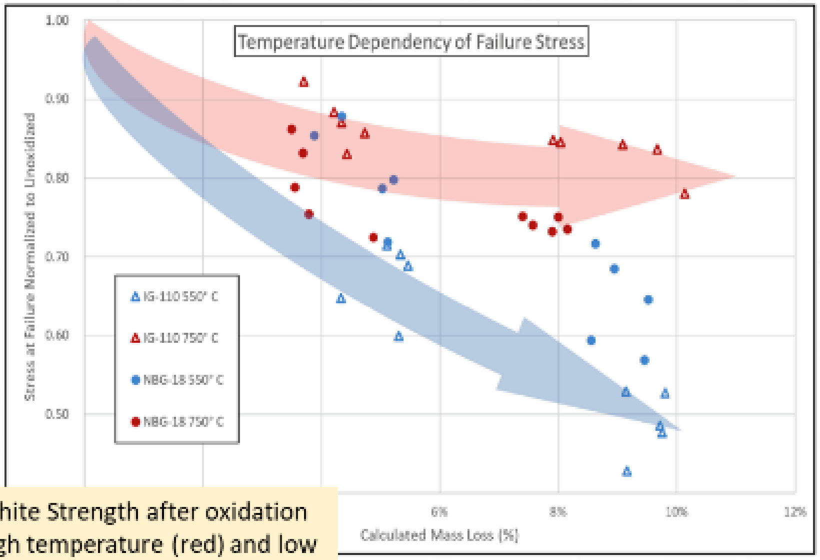
Task Groups formed within NWG:

- Failure in graphite components
    - Redefining failure other than a crack propagating
    - Review of POF assessments
        - *Underlying assumptions and why they are conservative*
- Degradation rules
    - Oxidation degradation
        - *Low temperature – maximum penetration*
        - *Component failure through oxidation*
    - Irradiation degradation
        - *Before – After turnaround induced changes*
        - *Affects on material properties, stresses, and POF*
    - Molten salt degradation issues
        - *Salt intrusion*
        - *Abrasion/Erosion issues*

Before turnaround: common response for all grades

M.C.R. Heijna, et. al, "Comparison of irradiation behaviour of HTR graphite grades", JNM, 492 (2017) 148e156





Graphite Strength after oxidation at high temperature (red) and low temperature (blue)

Cracked AGR core brick at Hunterston B power station

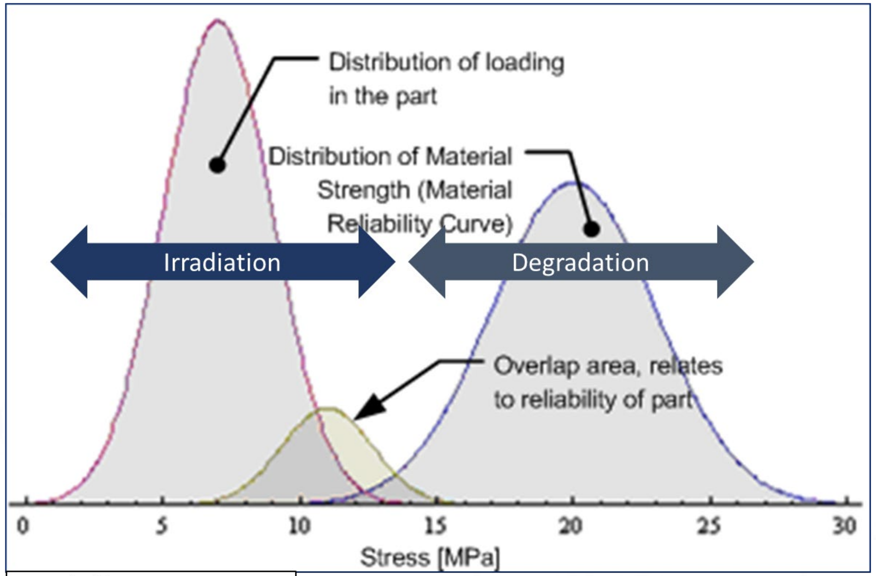
Summary of ASME issues

Induced stresses (crack propagation)

- Internal stress development
  - Irradiation induced dimensional change
  - Thermal induced changes
  - MS intrusion into microstructure
- External loads
  - Design features which produce localized loads
    - *High gas pressure*
    - *Molten salt pressure*

Changes to/in the graphite

- Material properties critical for structural integrity
  - Strength reduction
  - Thermal changes which can induce stresses
- Degradation
  - Material removal
    - *Wear, abrasion, erosion, oxidation, MS reaction*
  - Mechanisms which weaken the graphite
    - *Internal oxidation (kinetic controlled regime)*



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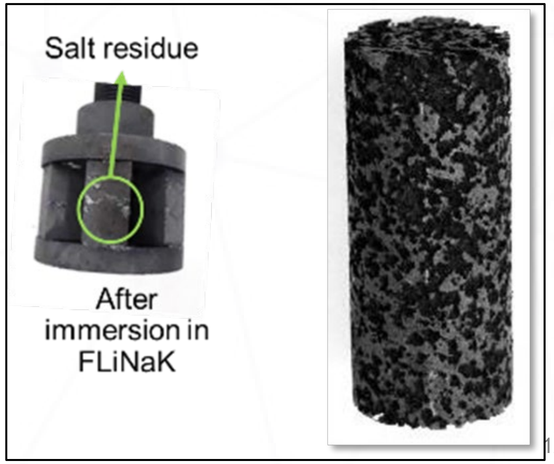
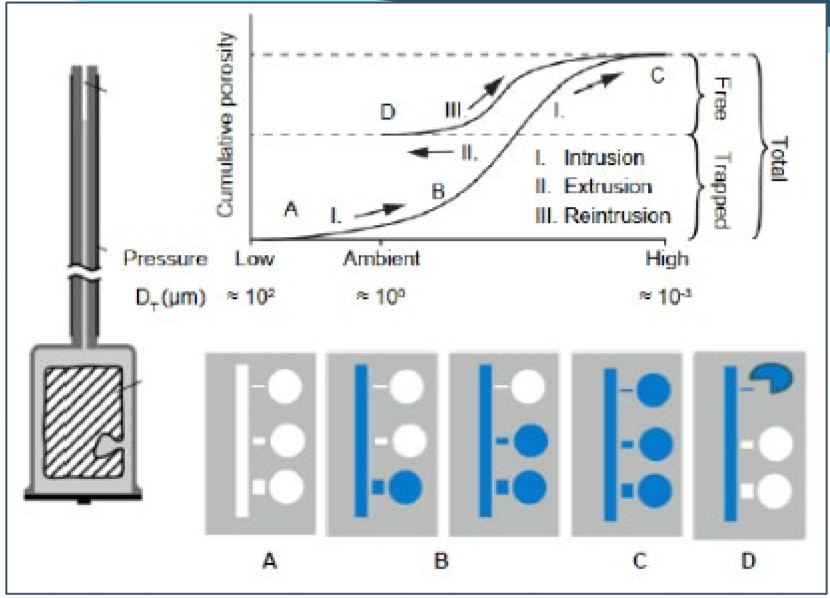


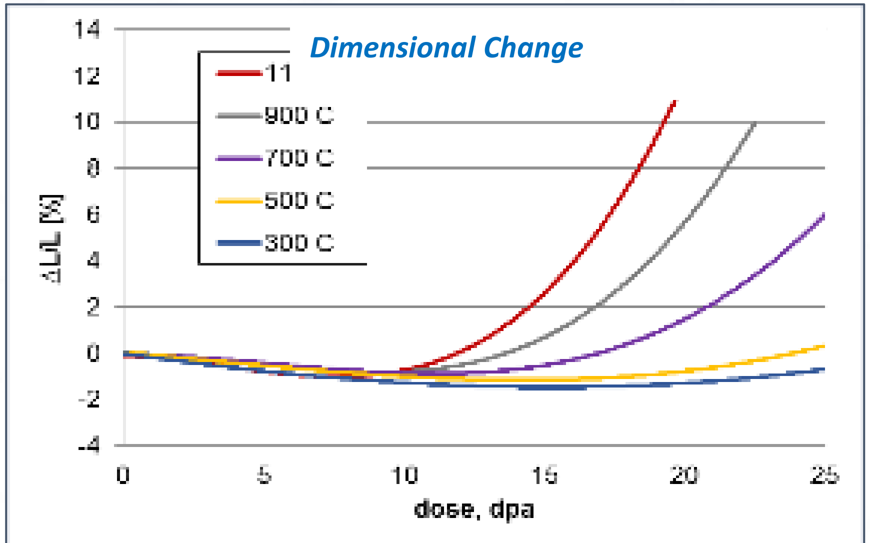
Specific molten salt issues of concern

Some issues NWG is concerned about

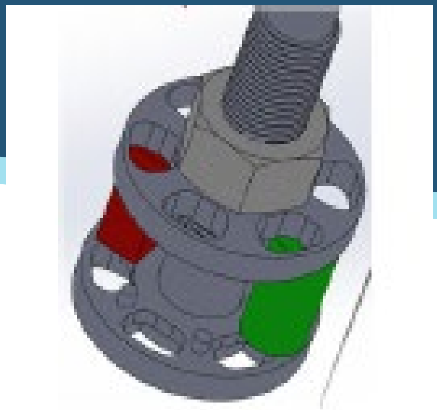
- Salt impregnation into graphite pores
  - Internal stresses in combination with irradiation
  - Physical damage, crack formation
  - “Hot spots” from fueled molten salt
- MS wear/abrasion/erosion
  - Formation of large defects/”cracks”
  - Reduction of load bearing capability (MS pressure)
- Chemical reaction (fluorine)
  - Material removal similar to wear/erosion issues
- Other non-graphite issues
  - Chemical coupling with metallic systems
  - Neutronics issues (moderation, hot spots, etc.)
  - Issues that could increase coolant blockage probability in core



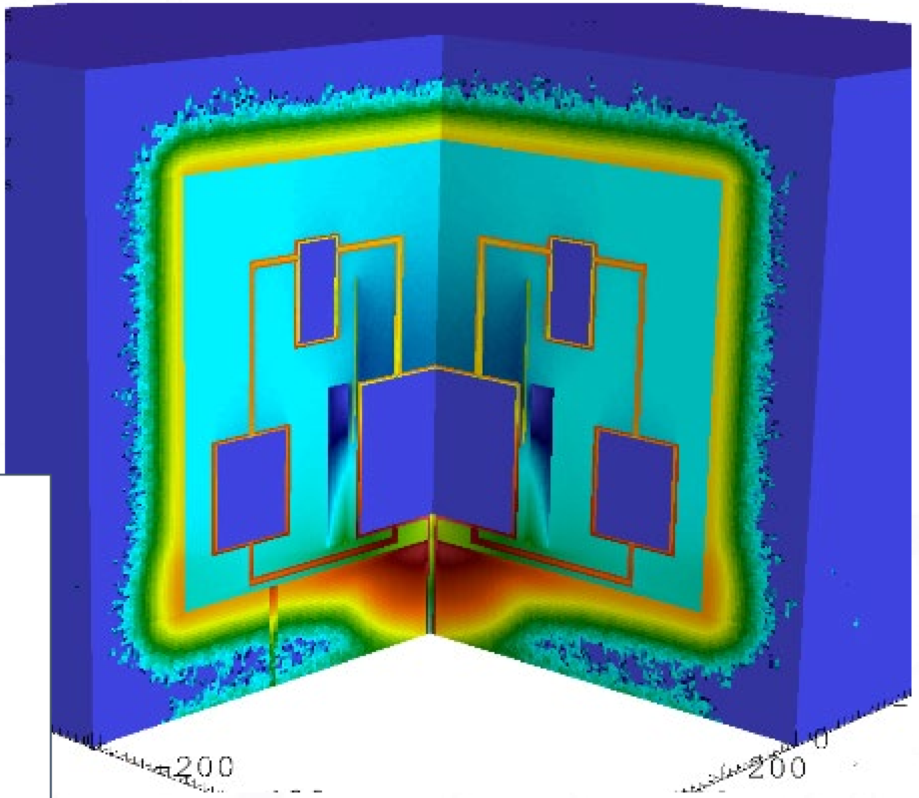
Salt residue

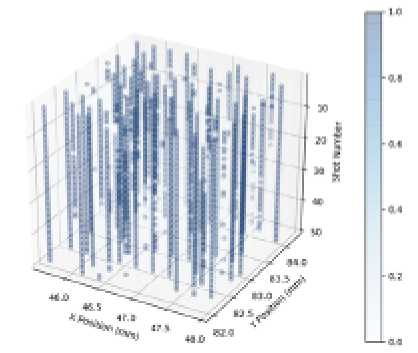


Molten Salt: What are *NOT* the issues



- Graphite – Molten Salt chemistry is considered inert
  - No chemical attack of graphite
  - No noticeable graphite attack during MSRE
- Little thermal variability within MS designs
  - Thermal transport so efficient that temperatures throughout core are evened out.
  - Small thermal induced stresses
- No combination effects
  - Currently rules don't address MS **+** oxidation **+** irradiation
- Pool type or low pressure designs
  - Minimal pressure on components
    - *Density increase at lower elevations?*
  - Coolant is relatively slow moving
  - By-pass issues are minimized





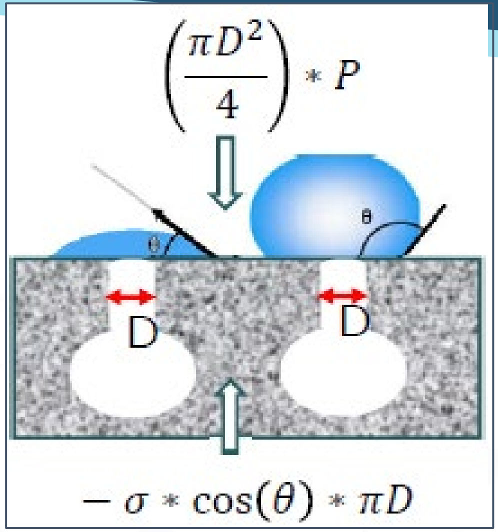
3D reconstruction of hyperspectral data showing FLINAK distribution in graphite sample exposed to molten FLINAK.

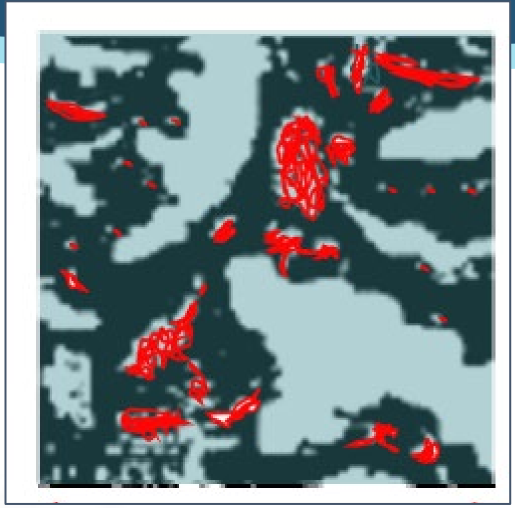
Specific issues: Salt impregnation in pores

- Molten salt diffusing into graphite pore structure
  - Nuclear graphite = 15 - 20% open porosity
  - Size of pores dependent upon grain size
    - *Pores ~ 1/3 of grain size average*

Grain Type	**Size**
Medium	< 2000 μm *(Nuclear)*
Fine	< 100 μm
Superfine	< 50 μm
Ultrafine	< 10 μm
Microfine	< 2 μm
Nanofine	< 0.1 μm

- Preliminary assessment indicated MS can penetrate any but nanofine grades
  - Nanofine grades are not really a thing
  - Nano-sized particles are difficult





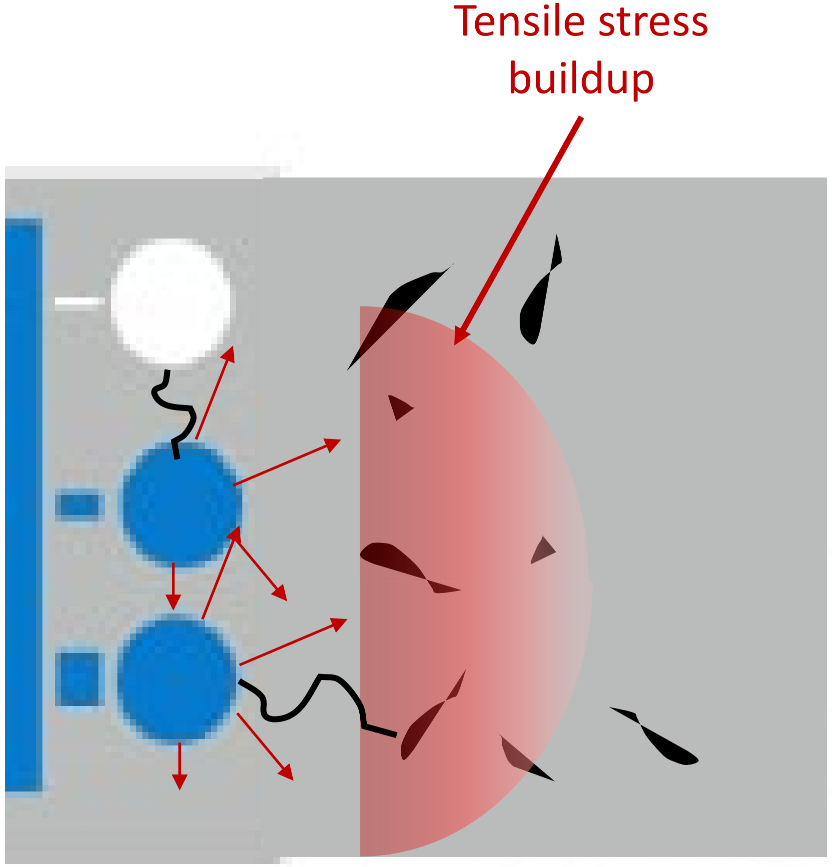
Open pores Closed pores

Graphite Grade	Grain Size, μm	Density
NBG-18	Medium < 1800	1.87
CGB (MSRE)	Medium	1.86
IG-110	Fine < 100	1.76
POCO-ZXF-5Q	Microfine < 2	1.78
POCO-AXF-50	Ultrafine < 10	1.78
POCO-TM	Ultrafine < 10	1.82
G347A	Ultrafine < 10	1.85
ETU-10	Superfine < 50	1.74

Specific issues: Salt impregnation in pores

Internal stress and crack build-up

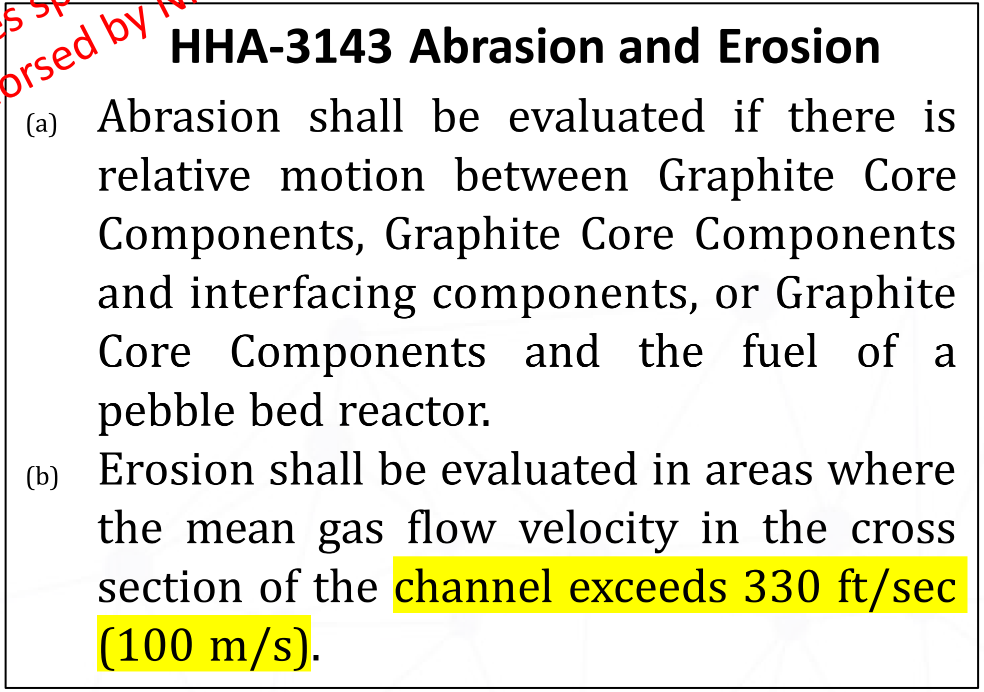
- The concern is change from liquid to solid
  - Volume change from cooling – dependent upon salt composition
  - Solid fill may prevent pore closure during irradiation induced dimensional densification
- Stress buildup
  - Interior tensile stresses build up
  - Cracks form/propagate between surface pores
    - *Spallation*
  - Cracks form/propagate into interior of graphite
    - *Interior cracking, build-up of internal strain/stress*
- Crack initiation sites already exist from existing pore structures
  - Propagation of cracks
  - Magnification of irradiation induced internal stresses
  - Induced internal stresses during cool down



Would like to know if this is a problem

Specific issues: Wear & Abrasion

Code rules specifically not endorsed by NRC



HHA-3143 Abrasion and Erosion

(a) Abrasion shall be evaluated if there is relative motion between Graphite Core Components, Graphite Core Components and interfacing components, or Graphite Core Components and the fuel of a pebble bed reactor.

(b) Erosion shall be evaluated in areas where the mean gas flow velocity in the cross section of the channel exceeds 330 ft/sec (100 m/s).

- ASME code written for gas-cooled conditions
  - Molten salt not considered in original code rules
    - ***But GCR rules are not well written, as seen***
  - Molten salts are generally higher density than graphite
    - ***Abrasion from salt is definitely possible***
    - ***Abrasion exacerbated with dust/inclusions***
  - Erosion flow limits are problematic even for gas
    - ***There is no supporting data for 100 m/s***
    - ***Not applicable to liquid MS***
  - Abrasion and erosion are design dependent
- MS Task Group working this issue

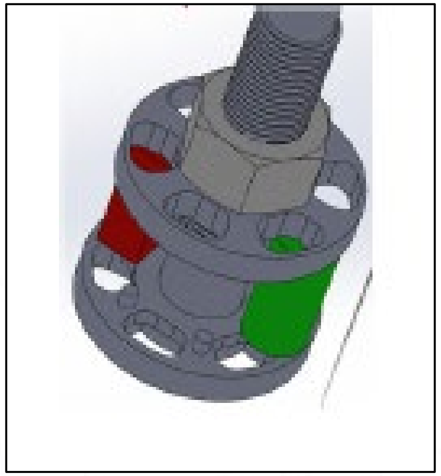
Some Graphite Grades	Density
NBG-18	1.87
CGB (MSRE)	1.86
IG-110	1.76
POCO-ZXF-5Q	1.78
POCO-AXF-50	1.78
G347A	1.85
ETU-10	1.74

Some Molten Salts	ρ (900°K)
FLiBe	> 1.9
NaFNaB	> 1.8
FLiNaK	> 2.0
FLiNaBe	> 1.9
KCl – MgCl$_2$	> 1.7
FluZirK	> 2.6

Specific issues: Chemical reaction

As noted, graphite/molten salt are all assumed inert

- If fluoride reactions are possible:
  - ***Is internal degradation possible (similar to oxidation)?***
  - ***If just the outside surface the code can handle it as material removal***
- Need a reaction rate
  - ***Temperature effects***
- Irradiation effects on reactivity?
  - ***Irradiation demonstrated to increase oxidation reactivity***
  - ***RSA sites increased***
- ASME code will tackle this similar to oxidation reaction



What ASME is concerned about

<u>Engineering issues</u>: Must affect the core safety or performance

- Salt impregnation into graphite pores
  - Must induce <u>significant</u> stresses
  - Must induce <u>significant</u> damage/cracks
  - "Hot spots" must create stresses
- Wear/abrasion/erosion
  - Most likely as engineering issue
  - Need wear rates, likelihood of cracks, and effects from temperature
- Chemical reaction
  - Must have significant damage accumulation
- "Galvanic Coupling" with metallic systems
  - Is there damage to graphite?
  - Is there significant damage to other systems?

- Salt impregnation into graphite pores
  - Don't care about 1-2 μm penetration
  - Don't care if minimal stresses created
  - "Hot spots" must be significant
- Wear/abrasion/erosion
  - Low wear rates or small (um) crack formation is not important
- Chemical reaction
  - ASME believes that graphite – MS is inert
  - Treat similar to oxidation rates
- Coupling with metallic systems
  - Needs to be significant degradation rate

We don't care about academic or material science issues

Lack of testing standards for degraded material

- A dearth of Molten Salt testing standards
  - Code rules will specify needed data but **not** specify how to get it
  - Non-MS ASTM tests have been underdevelopment since late 1990s
    - *Standardized MS tests have not even been attempted yet*
    - *Why should NRC believe data if tests have not been vetted?*
- MS testing is extremely difficult
  - How to standardize tests that very few facilities can perform?
  - How to perform in-situ testing (mechanical, thermal, etc.)?
  - How to perform elevated temperature testing?
  - How to test irradiated material in molten salt environment?
- Designer must prove their data is accurate and statistically relevant
  - Standardized tests would assist in this effort
    - *Collaborative testing (ASTM) is the best*
  - Will require significant time and large sample populations for each material property or degradation test
- How much data for each is needed?
  - ASME will need an understanding of the precision and bias for each test protocol
  - How large a sampling size is needed to determine accuracy of data
    - *Generally, for room temperature tests in air a minimum of ~30 – 50 test specimens (per test protocol) are needed for statistical accuracy.*

After immersion in FLiNaK

Sub-Sized testing

Molten Salt ASTM Testing Standard: A good first start

ADVANCED REACTOR TECHNOLOGIES

ASTM International D8091-16
"Standard Guide for Impregnation of Graphite with Molten Salt"

- Recommends a consistent procedure for controlled and reproducible impregnation of graphite with molten salts at constant temperature and pressure

$$D_0 = \frac{w_2 - w_1}{\rho\, V_{open}}$$

$$D_1 = \frac{w_2 - w_1}{\rho\, V_{total}}$$

w_1 = initial weight
w_2 = weight after impregnation
V_{open} = open pore volume
V_{total} = total pore volume
ρ = salt density at impregnation temperature

ASTM INTERNATIONAL

https://www.astm.org/

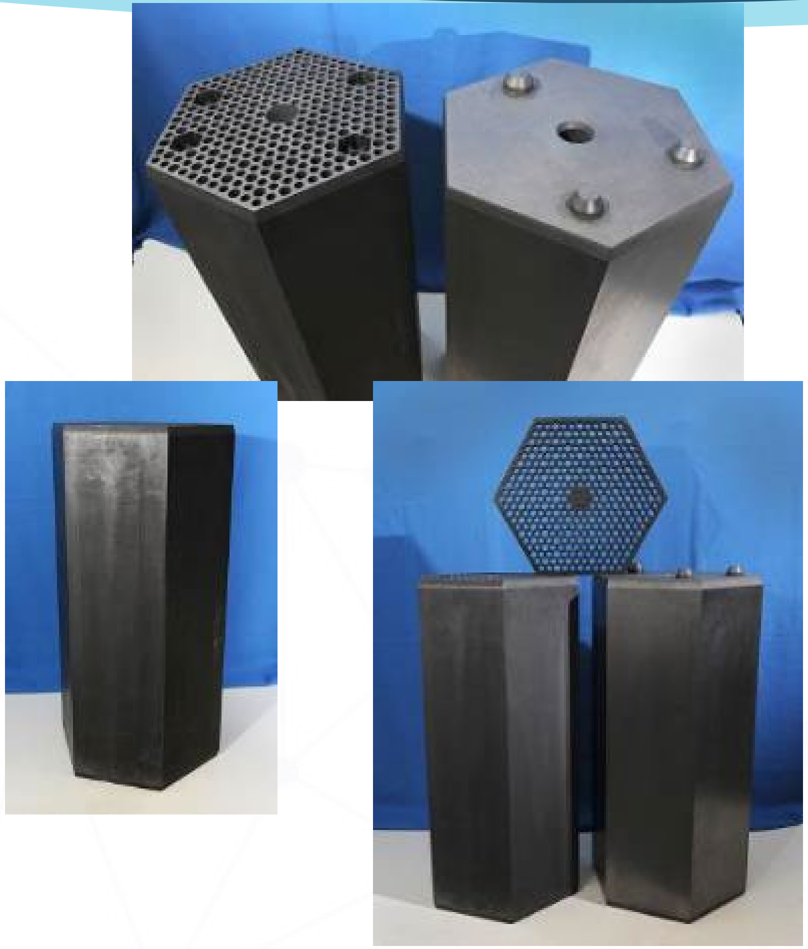
Design impact on code rules

Design functionality is critical for graphite/composite failure

- Depending upon design, component failure definition varies
  - Pore size in graphite/composite
  - Location of MS intrusion in component
    - *Where does internal stress or damage occur in component?*
  - Component wall thickness:
    - *Internal stress development, component operational life-time*
  - Lower or higher operating temperature

- Rules need to be applicable for **all** designs
  - New rules must apply to all potential designs
  - Is it applicable to all graphite-core HTR designs or just one or two designs?

- The way the code rules are organized will need generic understanding how issues will affect current code rules
  - How does it affect POF calculations?
    - ***Degradation issues***
    - ***Internal stress issues***
  - What additional material properties or degradation data are needed
    - ***Material data sheets, degradation rates, temperature effects, etc.***
  - Inspection requirements for degradation (Section XI-2)



INL

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