Ceramic Material Solutions for Nuclear Waste Disposal Canisters

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High Temperature Integrity
Mechanical Integrity for Energy Systems

EMPA
Materials Science & Technology

Ceramic solutions for nuclear waste disposal canisters

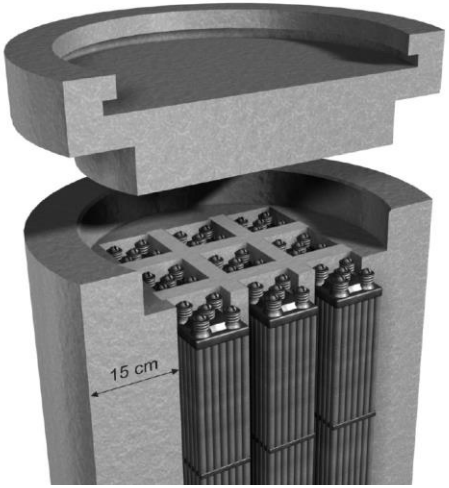
ENSI presentation (5.Nov-2015)

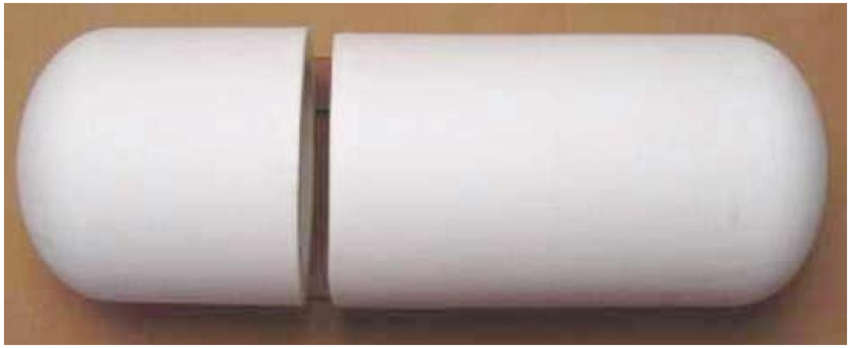
- Background and Introduction
- Mechanical integrity
- Susceptibility to environmental damage and impact on geological barrier
- Large product-form fabrication
- Container sealing solutions
- Coatings
- Concluding remarks

Ceramic solutions for nuclear waste disposal canisters

NAGRA canister review: Canister dimensions

- Disposal of spent fuel (SF)
  - *5m long x 760mm ID, with t ≤ ~150mm (depending on material solution)*
  - *Alternative configurations possible, but length is fixed*
- Disposal of vitrified high level waste (HLW)
  - *3m(1.5m) long x 450mm ID, with t ≥ 50mm (depending on material solution)*
  - *HLW cylinders are typically 1.34m long x 430mm diam.*





Ceramic solutions for nuclear waste disposal canisters

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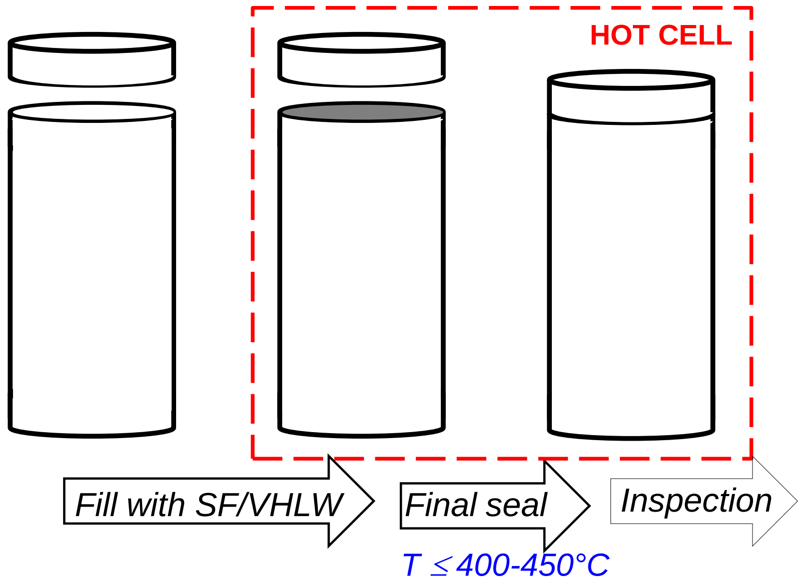
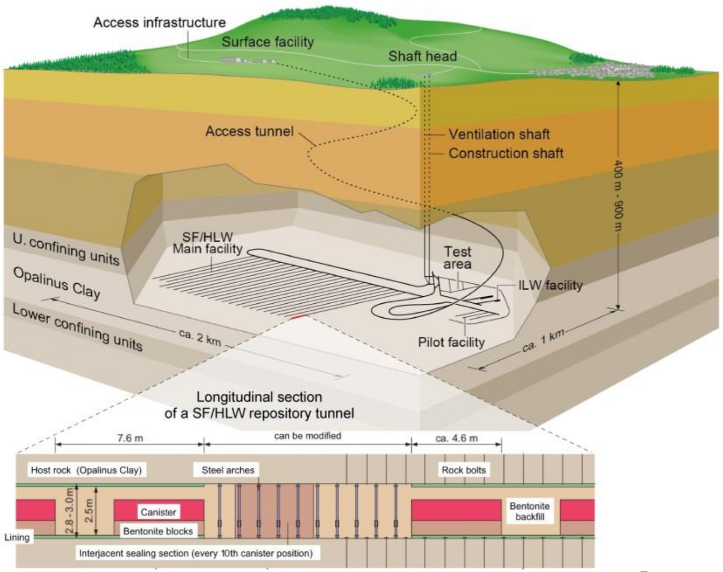
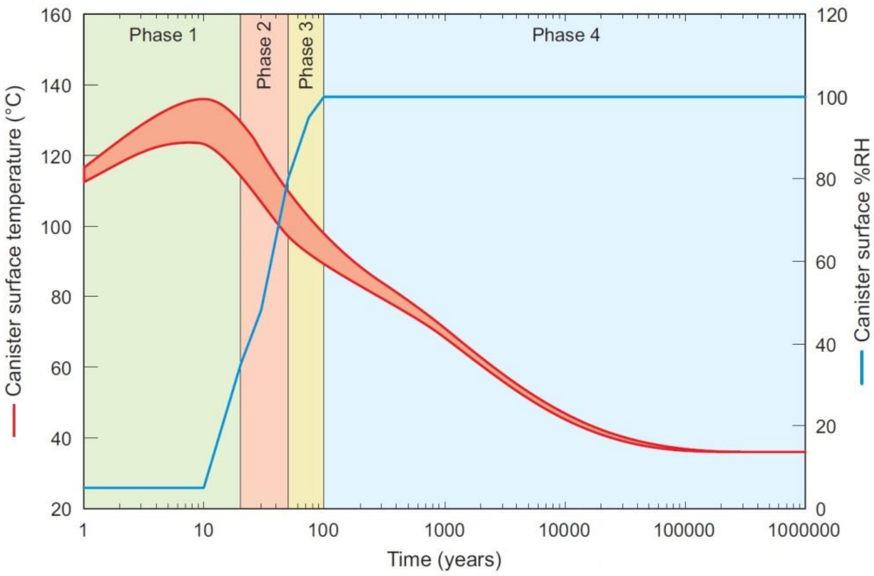


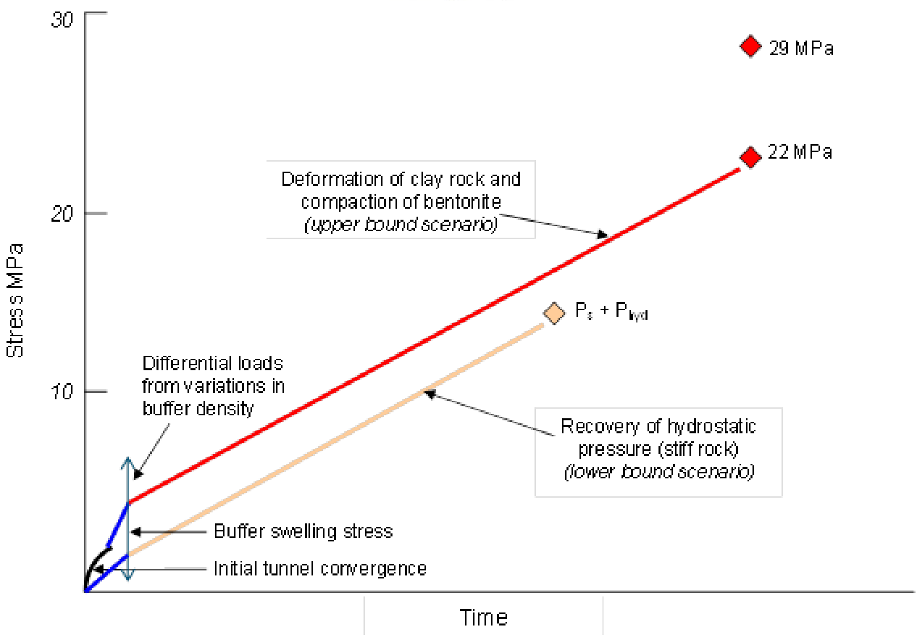


Ceramic solutions for nuclear waste disposal canisters

NAGRA canister review: NAB-14-90 evaluation categories

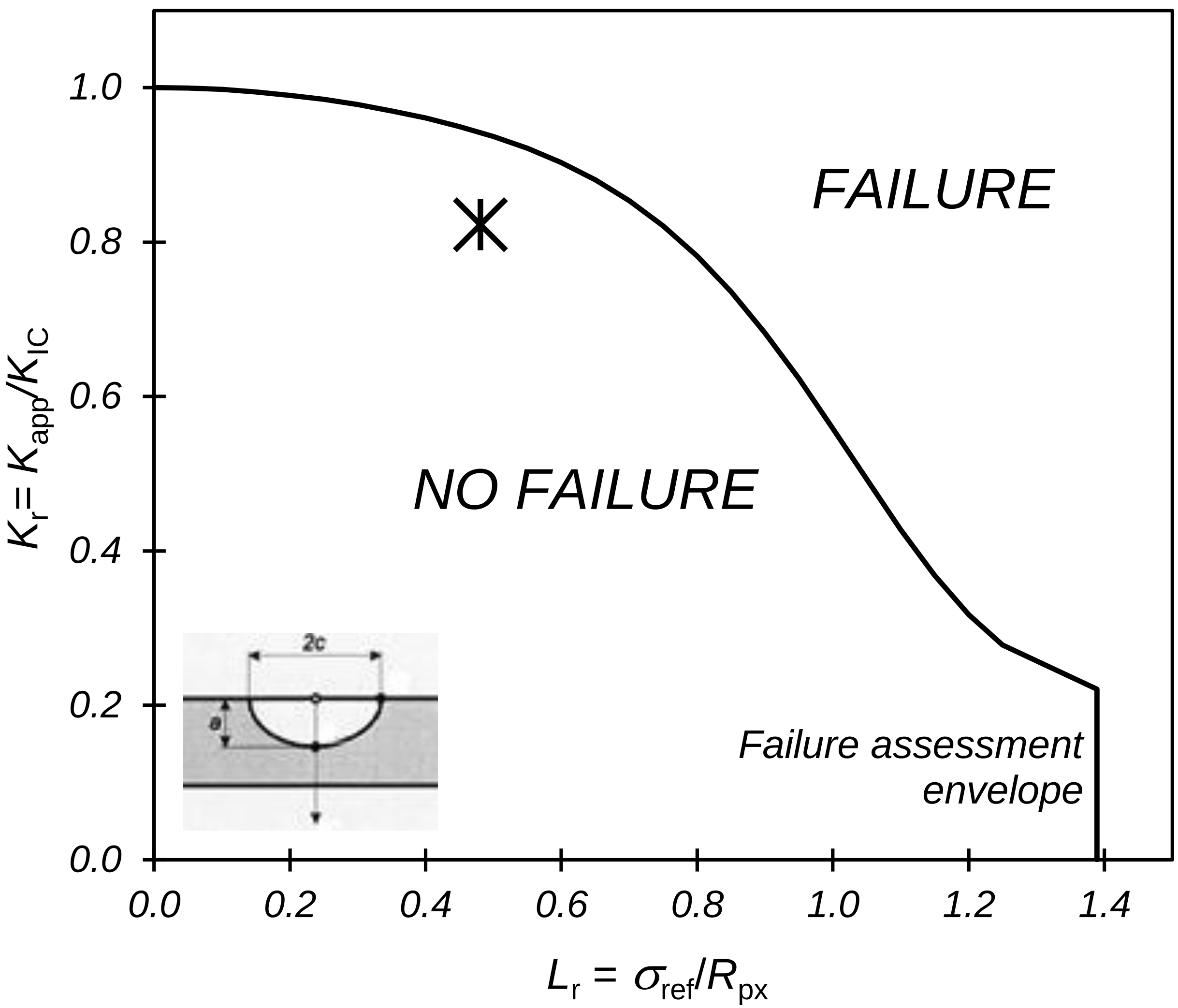
- **Mechanical integrity**
  - *Load cases: handling, disposal*
- **Environmental damage**
  - *'Short-time' aerobic (dry) phase; 'Long-time' anaerobic (moist) phase*
  - *General corrosion, localised corrosion, microbial induced corrosion, stress corrosion and hydrogen induced cracking*
- **Impact on geological barrier**
- **Robustness of lifetime prediction**
  - *Very long time (>10,000y) corrosion damage predictions*
- **Fabrication**
  - *Canister manufacture, final sealing, inspection*
- **Costs**
  - *Development costs, unit costs*

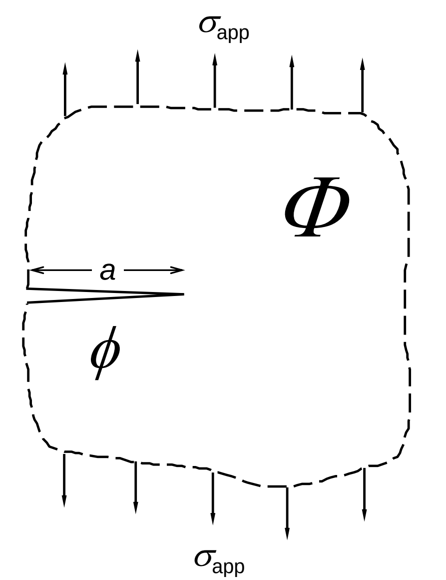




Ceramic solutions for nuclear waste disposal canisters

Defect integrity: Failure assessment diagram



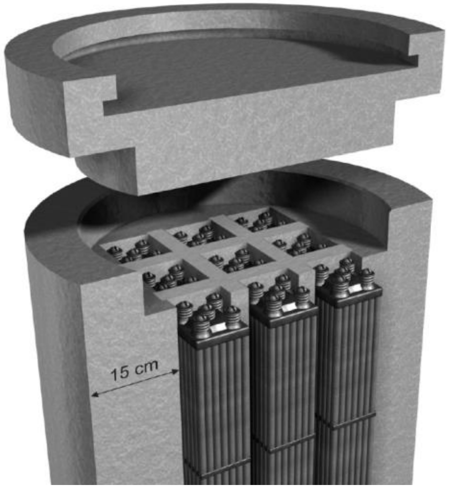


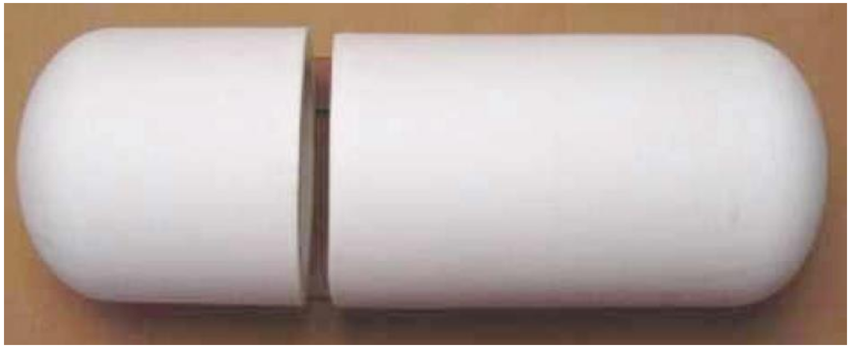
$$K_{app} = \sigma_{app}.\sqrt{a}.Y(a,\phi,\Phi)$$

Ceramic solutions for nuclear waste disposal canisters

NAGRA canister review: Candidate material solutions

- Carbon steel (with corrosion allowance)
- Copper shell with internal cast iron support
- Copper (or nickel alloy) coating of carbon steel
- Titanium or nickel alloy shell with carbon steel support
- Ceramics





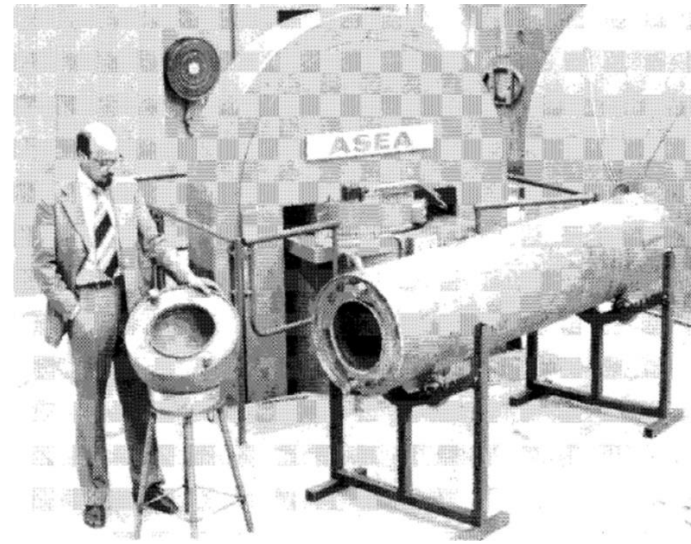
Ceramic solutions for nuclear waste disposal canisters

Candidate ceramics

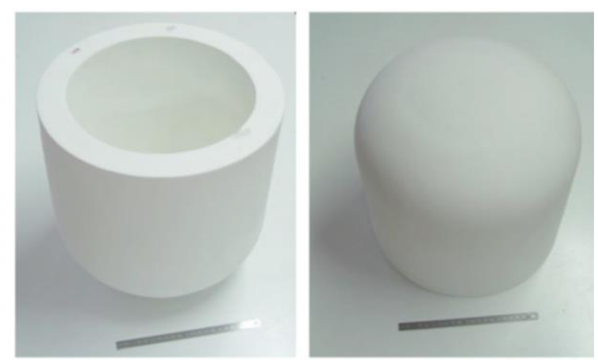
	SiO_2-MgO	Al_2O_3 (96-99.1%)	Al_2O_3 (99.8%)	ZrO_2-MgO	ZrO_2-Y_2O_3	TiO_2	SiC	Si_3N_4-Y_2O_3
Gross density (g/cm^3)	2.2-2.8	3.80-3.82	3.96	5.74	6.08	4.26	3.10	3.21
Flexural strength (MPa)	110-180	280-350	500	500	1000	69-103	350	750
Compressive strength (MPa)		2000	4000	1600	2200		2000	3000
Fracture toughness (MPa√m)		4.0	4.3	8.1	10.0	2.5	3.8	7.0
Elastic modulus, dynamic (GPa)	70-120	270-340	380	210	210	283	350	305
Vickers hardness (GPa)		14-17	18	13	13		25	16
Thermal conductivity (W/mK)	2-5	24-28	30	3	2.5	8.8	100	21
Thermal expansion, CTE (10^{-6}/°C)	4-7	7.1-7.3	7.5	10.2	10.4	9.4	3.5	3.2
Maximum operating temperature (°C)	1000	1400	1500	850	1000		1800	1600
Melting point (°C)		2015		2700		1840	2700	2700

Ceramic solutions for nuclear waste disposal canisters

Previous experience

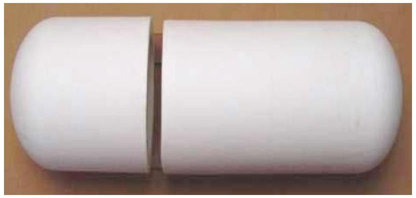


- Swedish evaluation programme
  - *Al_20_3 fabricated by sintering under isostatic pressure*
  - *Sealing by diffusion bonding with TiO_2 powder*
  - *Some environmental damage evaluation*
  - *Programme terminated in ~1980 due to progress with copper canister development*
- German evaluation programme
  - *Focus on Al_2O_3*
  - *Pre-stressed mechanical solution adopted for sealing*
  - *Significant environmental damage testing*
  - *No indication of programme continuing after 1990s*
- BNL/Nucon (US) evaluation programme
  - *$MgAl_2O_4$ spinel (one of Yucca Mountain solutions)*
  - *Sealing by diffusion bonding with local microwave heating*
  - *No environmental damage test results identified*
  - *Evaluation programme no longer continuing*
- NAGRA feasibility studies
  - *ANDRA evaluation of Al_2O_3-SiO_2 solutions (P72)*
  - *EMPA first review [NAB 12-45]*
  - *EMPA second review (as part of [NAB 14-90])*
    - *Greater focus on SiC*

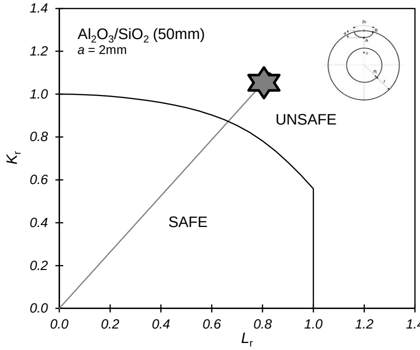


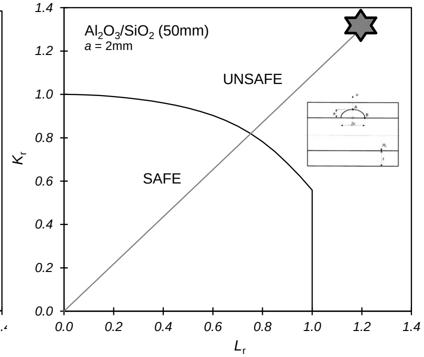
Ceramic solutions for nuclear waste disposal canisters

Al_2O_3-SiO_2 and SiC solutions

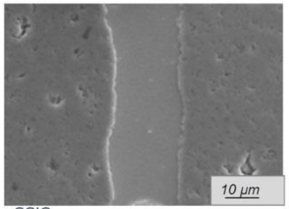


- **ANDRA evaluation**
  - *Main focus on Al_2O_3-SiO_2 HLW canister feasibility*
  - *Half scale body manufacture of this size feasible*
  - *Sealing is still an unresolved problem, requiring significant R&D activity – diffusion bonding is promising except for very high temperatures required*

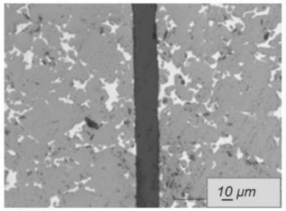




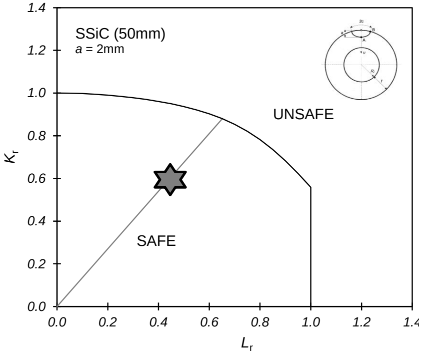
- **Silicon carbide (SiCeram)**
  - *There are a number of variants: SSiC, SiSiC, RSiC, LPSSiC, Si_f/Si_3N_4*
  - *SSiC exhibits better properties, but prone to high porosity levels (up to 20%), in particular in large sections (could be overcome by Hipping)*
  - *SiSiC – low porosity, lower mechanical properties, prone to leaching*
  - *Sealing by laser beam heating with glass ceramic solders (Y_2O_3-Al_2O_3-SiO_2) – currently only feasible for small sections*

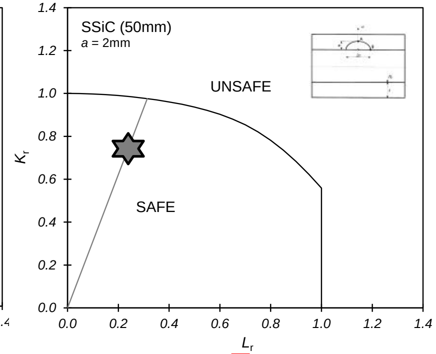


SSiC



SiSiC





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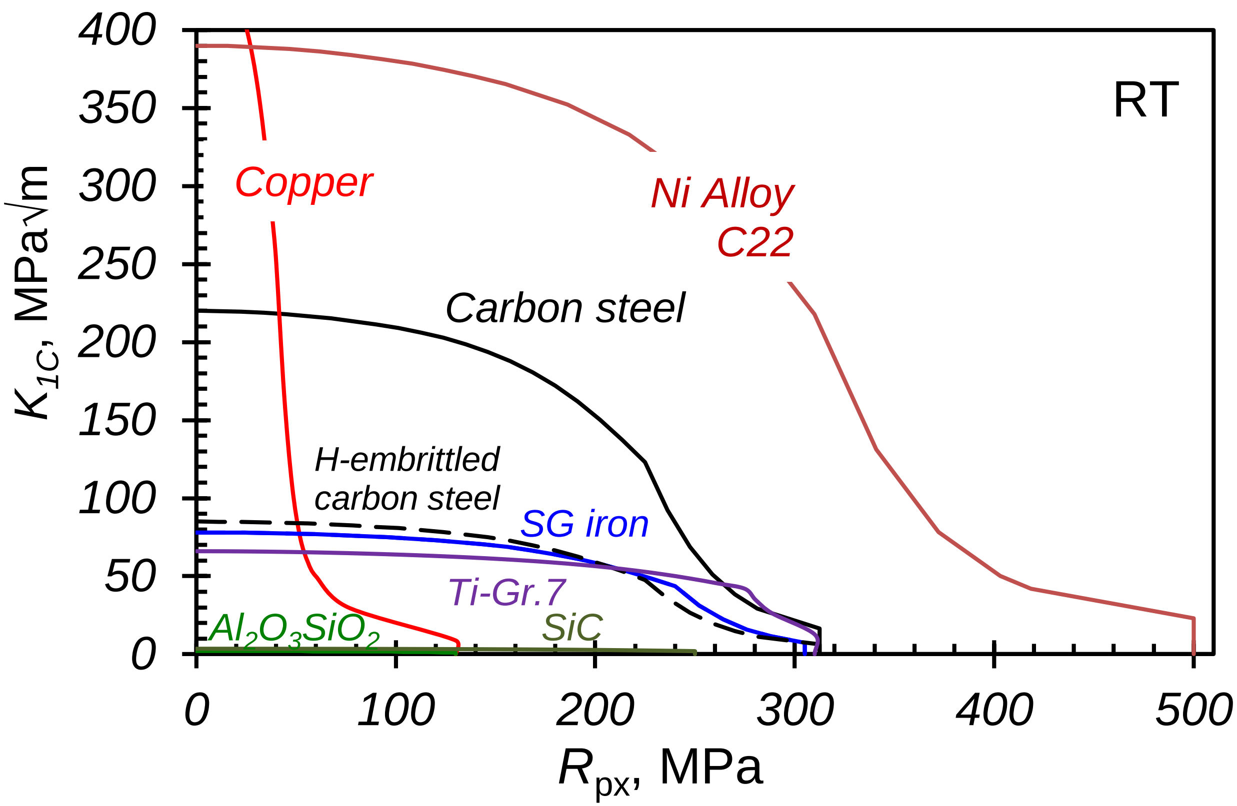
Ceramic solutions for nuclear waste disposal canisters

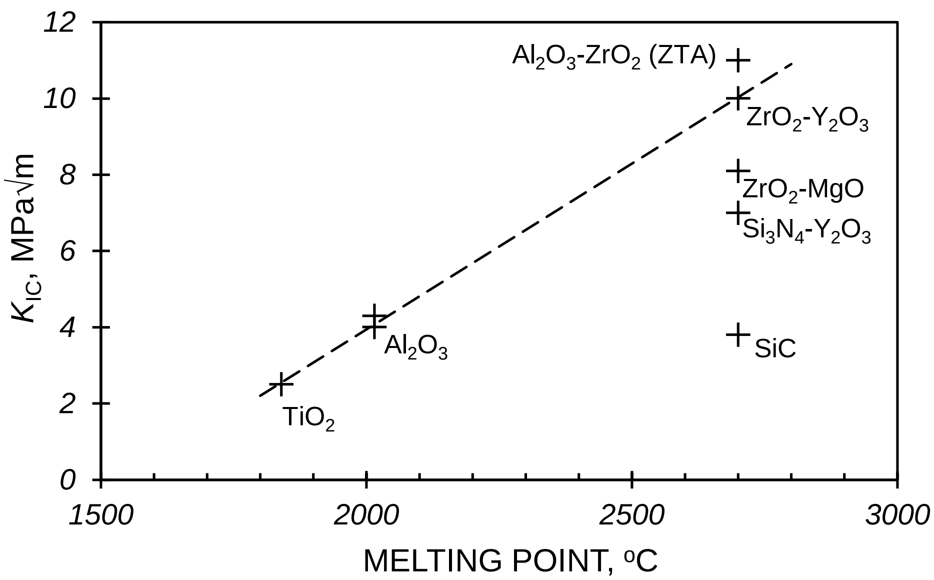
Mechanical property overview

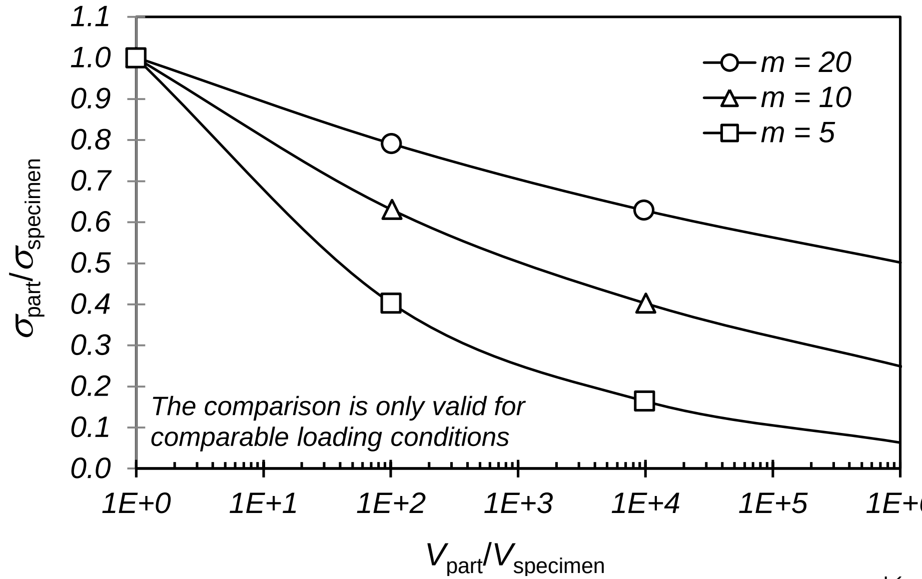


Ceramic solutions for nuclear waste disposal canisters

Mechanical Integrity

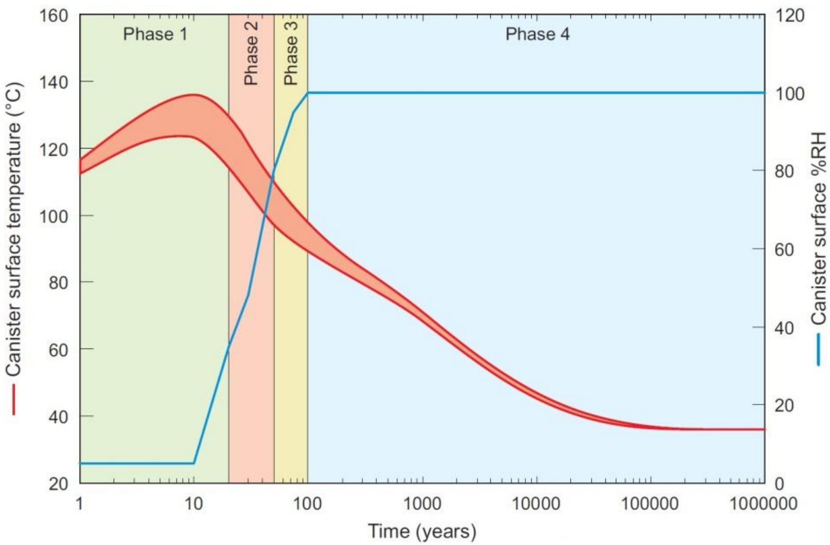
- The highest toughness ceramics are typically the most refractory (and thereby, the most difficult to sinter)
- The mechanical properties of ceramics are notoriously variable and size dependent
  - Strength and fracture toughness properties determined on small specimens are not representative of larger parts, when Weibull modulus is low
    - *For ceramics, m ≤ 3 is not unusual*
    - *10 < m < 20 is feasible for modern ceramics*
    - *For metals, m ≥ 100)*

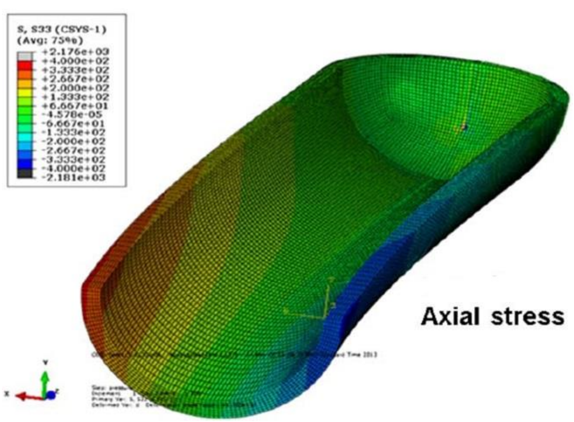




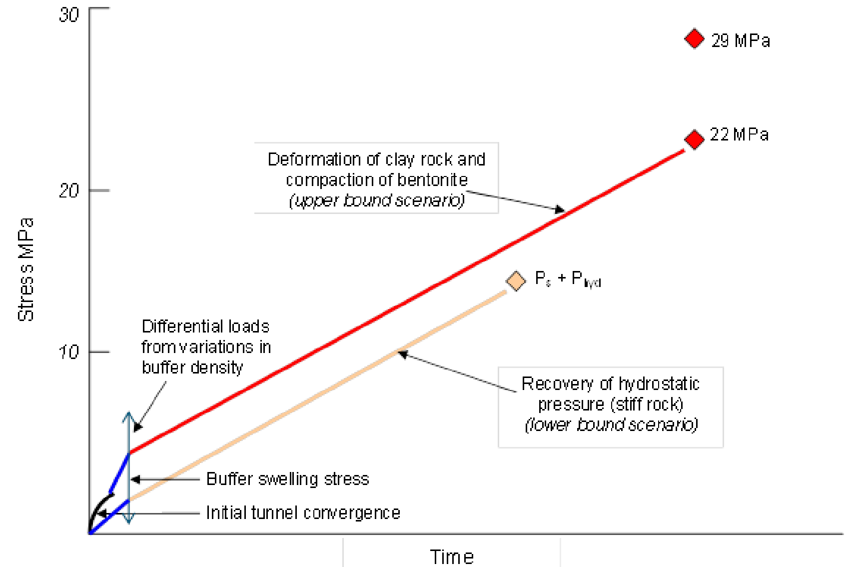
Ceramic solutions for nuclear waste disposal canisters

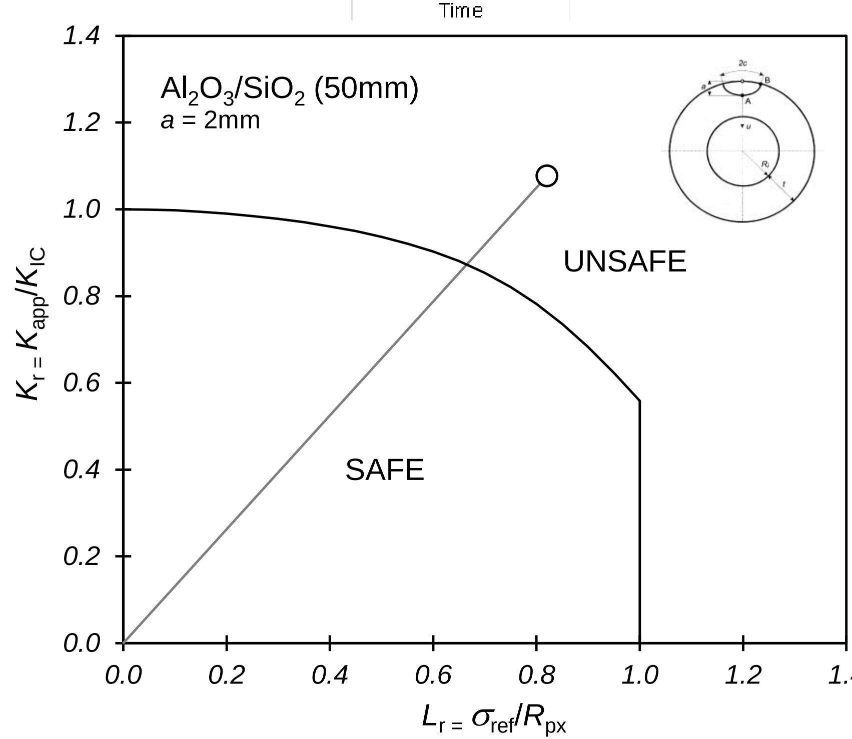
Mechanical integrity





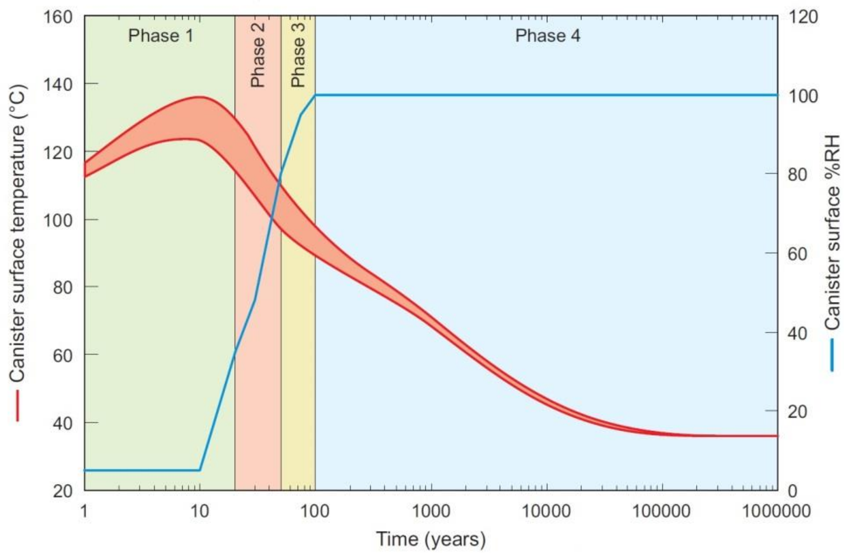
'Short-time' stress distribution due to variable buffer density along, and around, emplaced canister

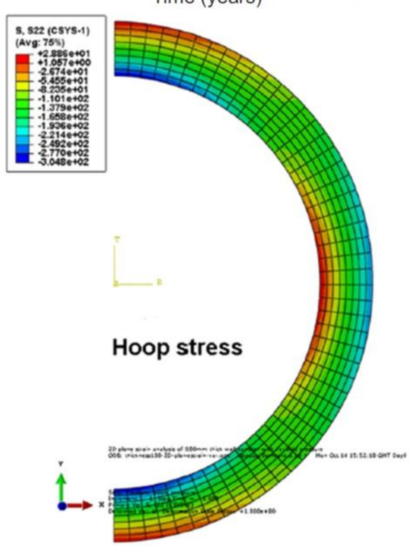




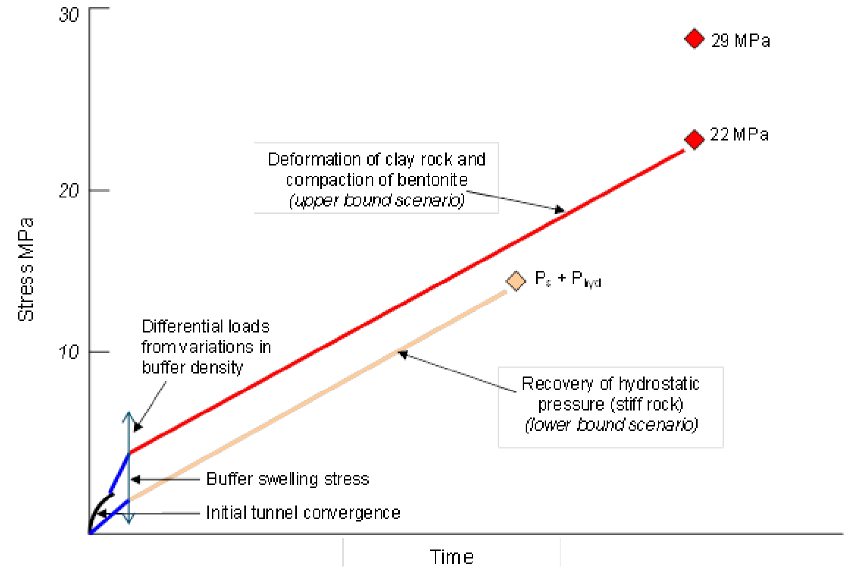
Ceramic solutions for nuclear waste disposal canisters

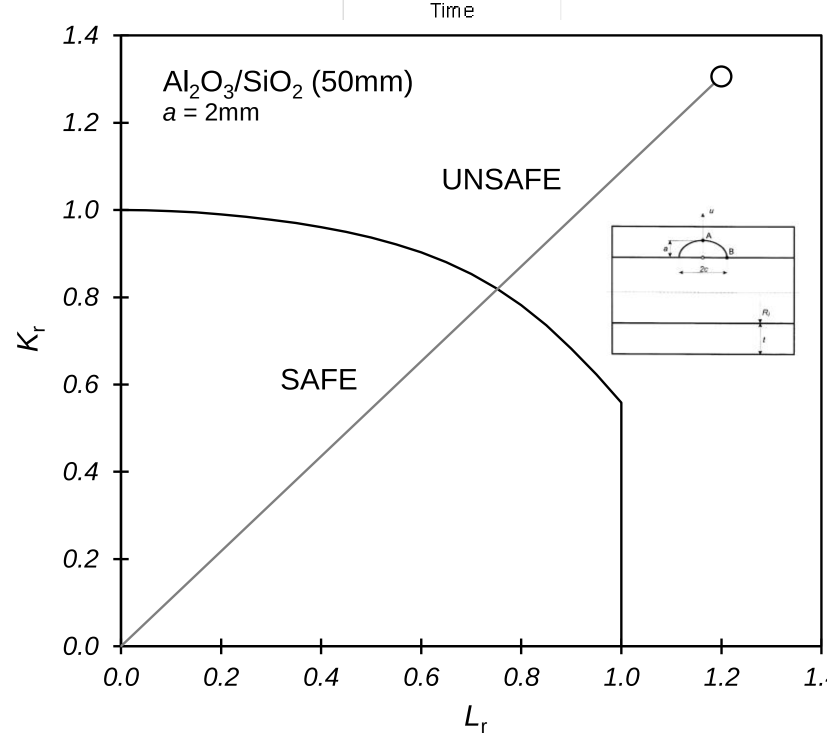
Mechanical integrity





'Long-time' stress distribution due to variable bentonite compaction around, emplaced canister

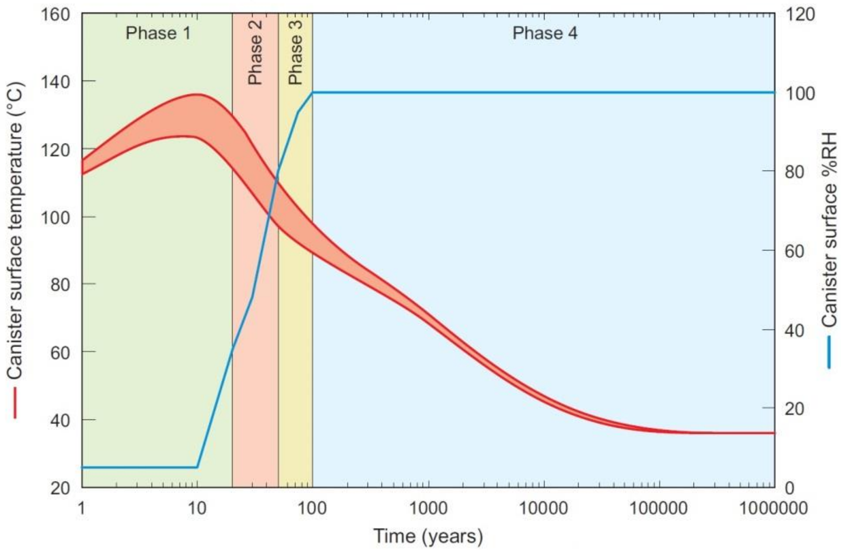


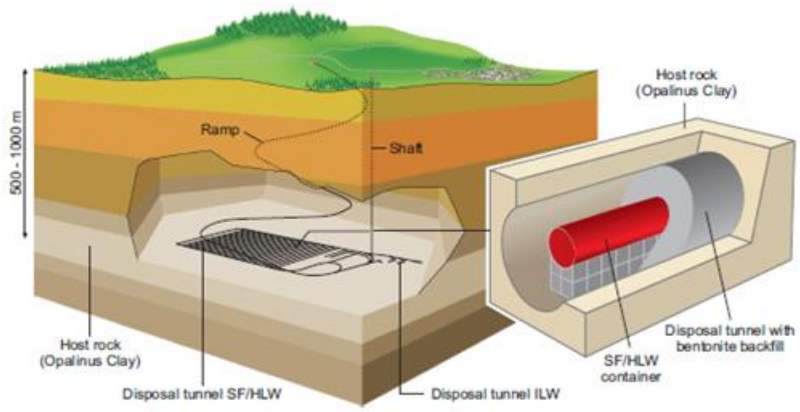


Ceramic solutions for nuclear waste disposal canisters

Susceptibility to environmental damage and impact on geological barrier

- The main advantage of a ceramic solution for nuclear waste disposal canisters is the very high resistance to environmental damage with low impact on the geological barrier
- Environmental damage
  - *'Short-time' aerobic (dry) phase; 'Long-time' anaerobic (moist) phase*
  - *General corrosion (dissolution/leaching in the case of ceramics), localised corrosion (intergranular leaching), microbial induced corrosion, stress corrosion and hydrogen induced cracking*
- Impact on geological barrier
  - *Impact on structure of compacted bentonite backfill and low permeability host rock*
  - *No hydrogen evolution to induce host rock cracking*
  - *No metal ion formation to influence the swelling capacity of the bentonite*





Ceramic solutions for nuclear waste disposal canisters

Susceptibility to environmental damage and impact on geological barrier

- Ceramics apparently not susceptible to H_2 generation in the presence of saturated bentonite
- Ceramics can be prone to local radiation damage
  - α-Al_2O_3 may be prone to minor swelling (~1%) to a depth of <1mm at relatively low temperatures
  - $MgAl_2O_4$ spinel is the most radiation resistant ceramic known, exhibiting zero swelling after neutron irradiation at 400-550ºC
    - *A number of pottery materials have also been shown to be immune to γ-radiation*
- Ceramics are prone to environmental damage in repository simulation media after significant time periods
    - *But not to the same extent as metals*
  - After 2 year autoclave tests in German repository simulated environment, even Al_2O_3 and ZrO_2 are shown to be susceptible to weight gain (solution ingress) or weight loss (dissolution/leaching) and, most importantly,
    - *Intergranular cracking to depths of ≥2mm (which was not observed after 35 day tests)*
  - Even Andra predictions based on relatively short duration (≤90 day) tests on Al_2O_3:SiO_2 in Callovian-Oxfordian deep groundwater solutions at 50 and 80ºC indicate the requirement for a 1,000 year 9mm 'corrosion' allowance

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Ceramic solutions for nuclear waste disposal canisters

Manufacturing challenges

- Manufacturing processes
  - Pressing capacity
    - *To achieve adequate density (porosity levels) in section thicknesses of ≥50mm*
    - A single-cylinder HLW canister is about the limit of current capacity
  - Effective handling of large pieces in the green state
    - *The technology would need to be developed*
  - Sintering furnace capacity
    - *A single-cylinder HLW canister is about the limit of current capacity*
- Manufacturing development activity
  - In the absence of a demand for large thick section ceramic pressure vessels from any other industrial sector, the funding of such R&D could only come from the nuclear waste disposal community
    - *Current forecast is for ~1900 SF canisters, and for ~300 HLW canisters*

Hipping Plant
FCT Hartbearbeitungs GmbH

Ceramic solutions for nuclear waste disposal canisters

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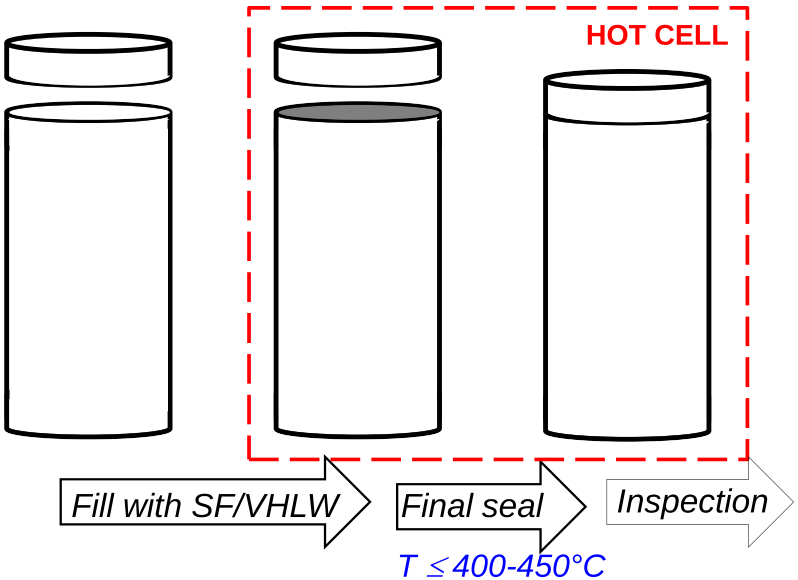
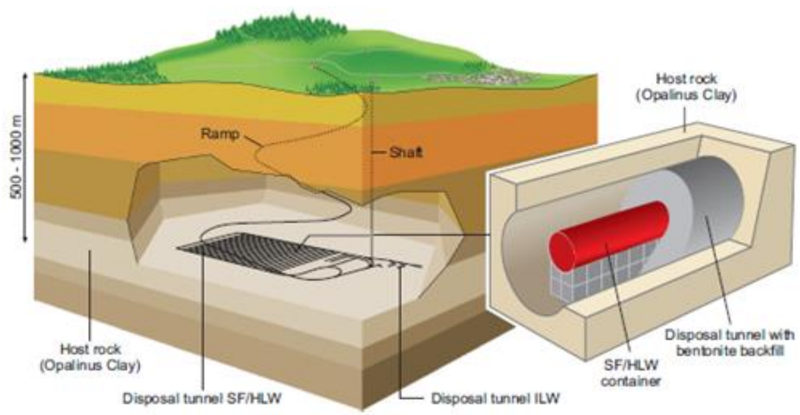
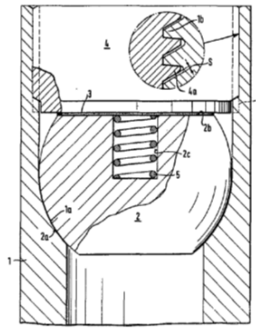




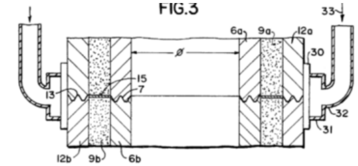
Ceramic solutions for nuclear waste disposal canisters

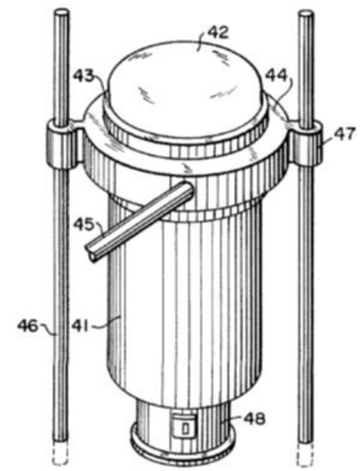
Container sealing solutions



- General requirements
  - *Secure containment of all matter and the prevention of water penetration*
  - *Final closure joint must be made and inspected remotely in a hot cell*
  - *During process, temperature of contents is ≤~400ºC*
- Candidate solutions
  - Mechanical joints
  - Glass ceramic solders
    - *For Al_2O_3: Al_20_3, La_2O_3, ZrO_2, SiO_2-BaO-B_2O_3-A where A is CaO, MgO, SrO, ZnO, …..*
    - *For SiC: Y_2O_3-Al_2O_3-SiO_2, CaO-Al_2O_3-SiO_2, MgO-Al_2O_3-SiO_2*
  - Metallic solders
    - *Ni-Ta, Ni-Ti, Cu-Ag, …..*
  - Brazes
  - Diffusion bonding
    - *Involves the growth of crystals across a carefully matched interface*
    - *Requires the application of mechanical pressure, and a sufficiently high temperature for sintering*
    - *Swedish evaluation programme adopted this technique with TiO_2 powder*
  - Cements
  - Sol Gels

FIG.3

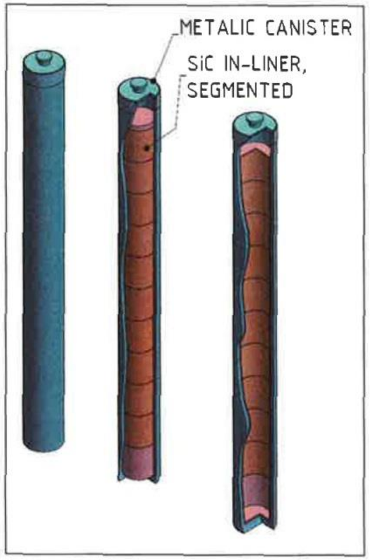


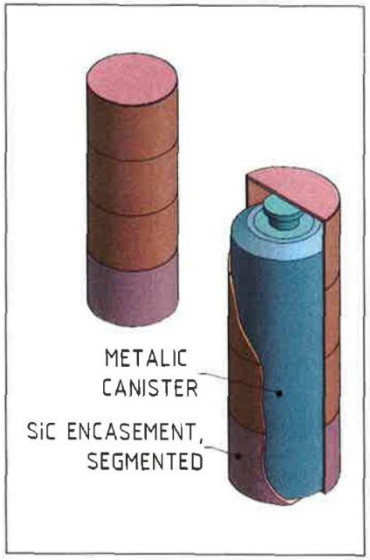


Ceramic solutions for nuclear waste disposal canisters

Example of NAB-14-90 manufacturer survey: SiCeram

- SiCeram
  - Published proposals are conceptual
    - *SiCeram visited in June 2014 (refer to NAB 14-90)*
    - *SiCeram does not have the current capability to manufacture full size SSiC SF or HLW canisters without (significant external) investment programme – requirement said to be >100 M€*
    - *Only FCT Hartbearbeitungs GmbH currently have the potential capability to manufacture a SSiC single-cylinder HLW canister in Europe (and their SSiC contains 0.5 wt% B)*
    - *Concerns about acceptable density above 50mm*
    - *Importantly, SiC in-liner solution for SF disposal is not helpful, from the point of view of avoiding hydrogen evolution*
  - No sealing solution
    - *Current TUD laser welding technology (on which SiCeram rely) would need to be significantly upscaled*
  - No inspection experience of thick walled ceramics
    - *Technology would have to be developed*





Ceramic solutions for nuclear waste disposal canisters

Coatings

- Ceramic coating of steel provides a potential solution for eliminating/reducing the rate of hydrogen generated by anoxic corrosion in saturated bentonite
  - *Only an effective solution if coatings are free of defects and pores to avoid localised corrosion*
  - *Porosity must be <6% to avoid a continuous path between the metal surface and the external environment*
  - *Susceptibility to porosity increases with increasing melting point.*
  - *A close to matching coating CTE with the steel is required to avoid thermal cracking*
- Candidate coating solutions
  - Al_2O_3, $MgAl_2O_4$ spinel and Al_2O_3/TiO_2 have been extensively evaluated for HLW steel container applications
    - *Plasma spray, Detonation-gun (DGUN), High velocity oxy-fuel (HVOF)*
    - *Lower porosity obtained with DGUN and HVOF processes*
    - *Coating thicknesses of over 1mm evaluated*
    - *Evidence indicates that greater thicknesses will be required in certain environments*
    - *6 years environmental testing in concentrated salts solution at 90°C*
    - *But remember Andra recommended 1,000 year 9mm dissolution allowance*
  - Chemical vapour deposition (CVD) can be used to make dense impervious ceramic coatings of TiC, Al_2O_3 and Y_2O_3
    - *Not usually applied to large parts because of high temperatures required to bond refractory ceramics*

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Ceramic solutions for nuclear waste disposal canisters
Concluding remarks

- The main advantages of a ceramic nuclear waste disposal canister solution are:
  - A high resistance to environmental damage, and
  - No gas generation (H_2) during long time emplacement
- Disadvantages include:
  - Low mechanical strength (in tension) and very low fracture toughness
  - Major manufacturing challenges requiring focussed R&D activity to achieve:
    - *effective handling of very large pieces in the green state, and*
    - *adequate density (porosity levels) in section thicknesses of ≥50mm*
  - Limiting press and sintering furnace capacities, worldwide
  - No effective large ceramic container sealing solution
    - *in particular with temperature limitation of nuclear waste*
- It is possible that the disadvantages could be overcome with appropriate investments in research and infrastructure, but
    - *in the absence of a significant demand for large thick section ceramic pressure vessels from any other industrial sector, the funding of such R&D could only come from the nuclear waste disposal community*