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Role of the Wasteform in Deep Borehole Disposal

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Wasteform definition

Waste package: passively safe and physically robust, ensuring waste containment and handling (e.g. transport)

Compatible with safety case for operational and post closure period of disposal system

Safety function is provided by both wasteform and waste container

Waste container: assures waste containment during storage, transport and emplacement

Wasteform: a passively safe material which assures physical containment and chemical retention of the waste.

Note: this implies some element of engineering design and material processing.

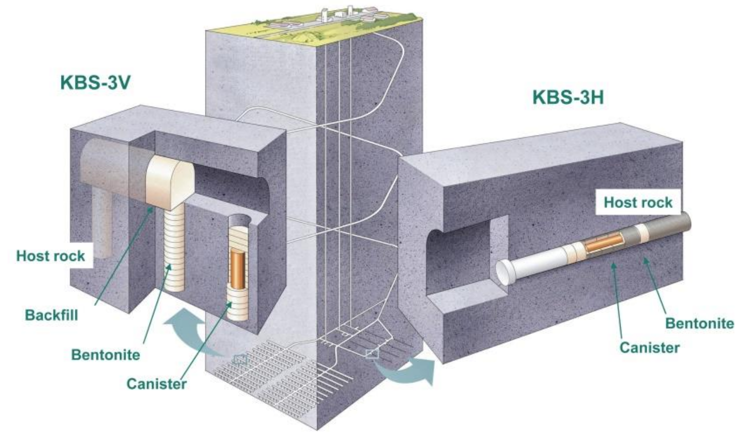
Example wasteforms: glass, ceramic, SNF.

http://www.irsn.fr/
http://www.rsc.org

Wasteform in a multibarrier concept

Conventional, relatively shallow, mined disposal facilities (<1km) utilise a multi barrier concept:

- Geological barrier – the excavated and overlying rock burden
- Engineered barrier – the wasteform, container, overpack, buffer and backfill

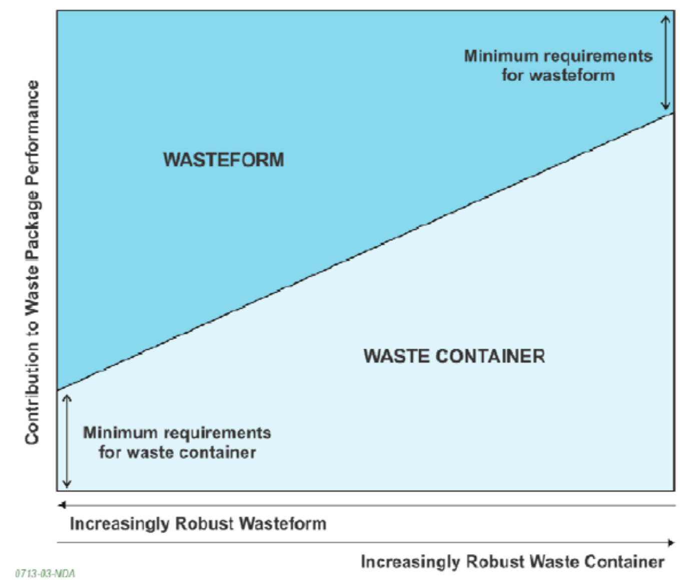


Safety concept takes credit for the EBS

- Groundwater flux is not negligible
- High uncertainty on matrix diffusion, sorption, redox conditions

The wasteform is the primary barrier to radionuclide release: low solubility

To some extent wasteform performance can be traded off against other parts of EBS



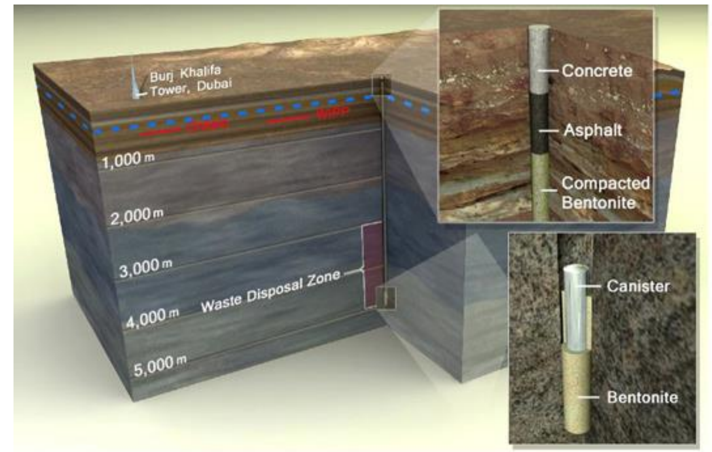
http://www.posiva.fi http://www.nda.gov.uk

Wasteform in a deep borehole concept

Characteristics of deep borehole concept:

- Static groundwater – density stratified
- Long return pathway (and return time)
- Reliably reducing geochemistry

In essence, deep borehole disposal relies primarily on the natural geological barrier.



In this context of deep borehole disposal, what is the role of the wasteform?

- Concept flexibility / reduce constraints
- Robustness of operational safety case
- Robustness of post closure safety case
- Efficiency of disposal system resource
- Public confidence

Deep borehole disposal offers potential advantages regarding confidence in performance of the natural barrier system… and potential for direct disposal of some waste forms without the need for further waste treatment.

SAND2014-17430R

Potential wastes for borehole disposal

Waste stream	Solubility	Dispersibility	Fissile
CsCl and SrF_2 capsules	High	High	No
Untreated Idaho calcines	Low-Med*	High	No
Unconsolidated sodium bearing TRU wastes	Low-Med*	High	No
Plutonium: not currently considered (also UK)	Low*	(High)	Yes

* More dependent on local geochemistry

What could be the drivers for having a robust and passively safe wasteform?

- Minimise radionuclide source term – enhance post closure safety
- Reduced impact of container damage (transport, handling, emplacement)
- Confidence in recovery of maloperations (stuck container)
- Confidence in waste package passive safety (e.g. gas evolution)
- Confidence in post closure criticality (e.g. addition of neuron poisons)
- Facilitates retrivability – if desirable

However: need to consider overall risk and benefit associated with waste processing

Alteration and solubilities in brines at realistic temperature (≤200^{o}C) require further R&D

Passive safety of wasteforms

WIPP release – 14 February 2014

Exothermic reaction involving the mixture of organic materials and nitrate salts in one drum processed at LANL in December 2013.

Activity release on and off site, worker exposure.

Recovery program: minimum 2y / $242M.

Goiânia accident – 13 September 1987

Theft of abandoned hospital radiation source: ca. 50 TBq / 93 g of $^{137}CsCl$ in IAEA standard capsule.

Estimated 44 TBq accounted for in contamination.

At least 250 people contaminated, 4 deaths; 3,500m^3 of radioactive waste.

Highlights the need for passively safe and non-dispsersible waste forms.

http://www.wipp.energy.gov/
http://www-pub.iaea.org/

Wasteforms for CsCl & SrF_2 - I

Iron phosphate glasses

Based on Na_2O-Al_2O_3-Fe_2O_3-P_2O_5

Up to 34 mol% CsCl and SrF_2 combined

Processing 950°C / 2h

Very little Cs and Sr volatilized

Almost all Cl and F inventory lost in processing

Highly durable: initial dissolution rate at least 10^2 lower than borosilicate HLW

Durability improved with higher Fe/P ratio, but relatively insensitive to wide composition range

Performance of iron phosphate glasses in concentrated brines unknown

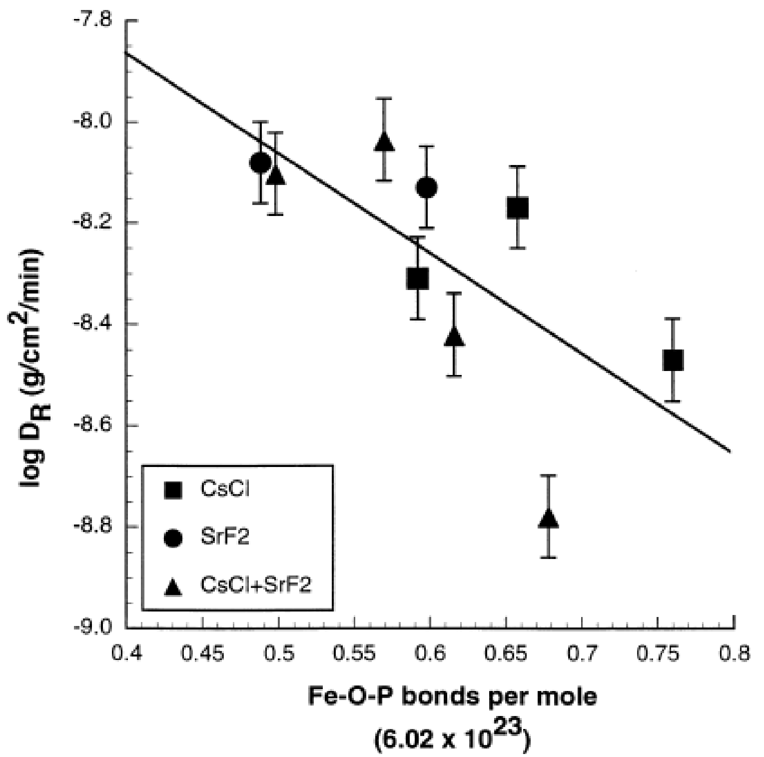


Fig. 3. Dependence of the log of the dissolution rate (D_R) on the number of Fe–O–P bonds per mole calculated from the batch composition of the glasses listed in Table 2. D_R measured in distilled water at 90°C for 16 days.

Mesko *et al.*, Waste Man., 20 (2000) 271.

Wasteforms for CsCl & SrF_2 - II

Ion exchange and consolidation

Use a highly specific commercially available ion exchanger to selectively extract Cs and / or Sr

- SrTreat®
- Ionsiv® (crystalline silicotitantate)
- Clinoptilolite

Wasteform would either be a stable glass (by vitrification) or ceramic (by HIP)

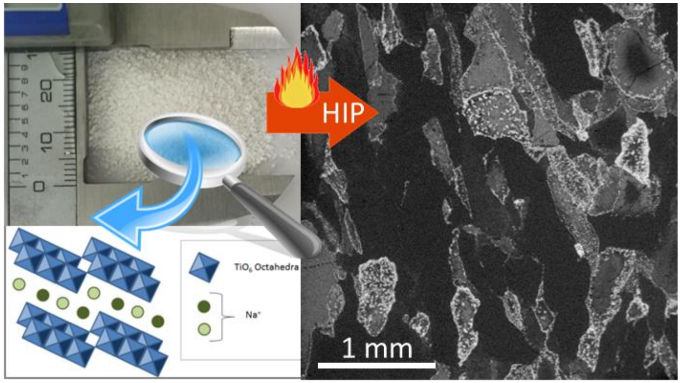
HIP has advantage of zero additives and complete volatile containment

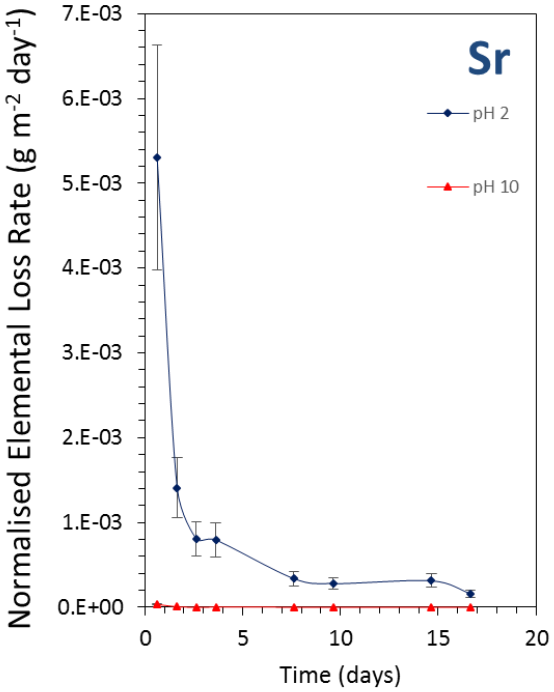
Probably increase waste volume by x 2-10

Dissolution rates: $10^{-1} – 10^{-4}$ g/m²/d at 90°C pH2

Performance in concentrated brines unknown

P.G. Heath, PhD Thesis, University of Sheffield (2015).





Wasteforms for CsCl & SrF_2 - III

Apatite related materials

Based on fluorapatite: $(Ca,Sr)_5(PO_4)_3F$

Proposed for $PuCl_3$ / $AmCl_3$ UK defence wastes as a glass bonded composite, processed at ≤800°C; Donald et al., J. Nucl. Mater., 361 (2007) 78.

Iodoapatite for I-129 $Pb_5(VO_4)_3I$; Stennett et al., J. Nucl. Mater., 31 (2011) 352.

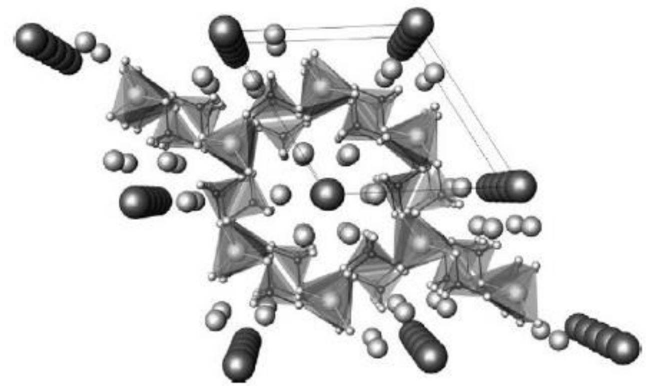


Fig. 1. Schematic representation of the crystal structure of $Pb_5(VO_4)_3I$ determined from X-ray powder diffraction data (Section 3.3), highlighting VO_4 tetrahedra and Pb1 polyhedra; large dark grey spheres represent I, large light grey spheres represent Pb, small black spheres represent V, and small grey spheres represent oxygen; viewed down [0 0 1]. The unit cell is highlighted.

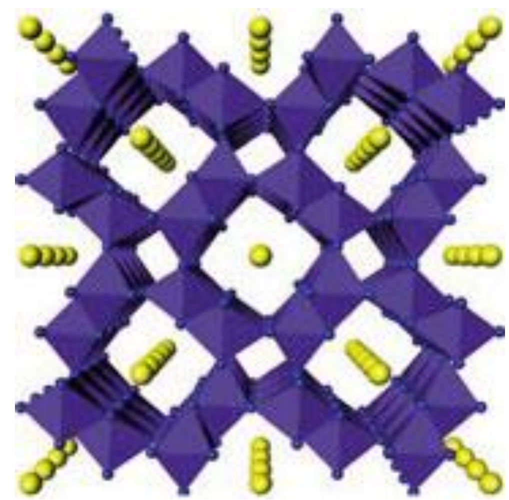
Hollandite and perovskite

Precipitation with titanium isopropoxide, followed by calcination and HIP

Hollandite = $(Ba,Sr,Cs)(Al,Ti)_2Ti_6O_{16}$

Perovskite = $SrTiO_3$

Both structures contain tunnels / cavities ideally suited to Cs / Sr incorporation



Wasteforms for Calcine, SBW, Pu

Glass / ceramic material tailored to chemistry and radiological risk

Illustrative example: glass ceramic developed for immobilisation of scrap contaminated plutonium

Ceramic phase: zirconolite - $CaZrTi_2O_7$, incorporates Pu, known to be highly resistant to radiation damage and dissolution from natural analogues

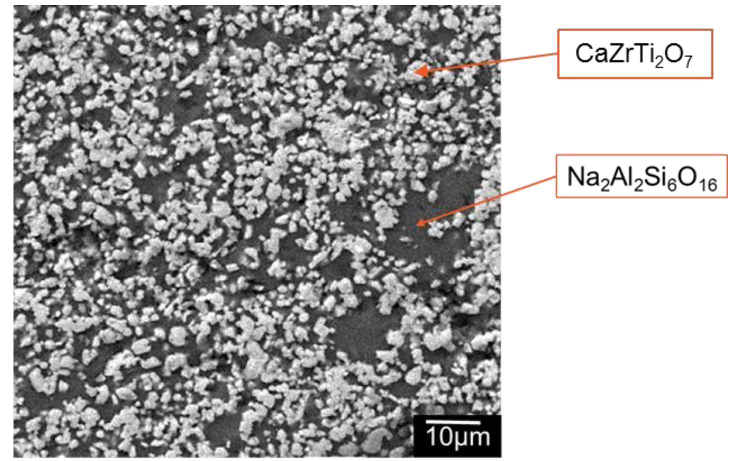
Glass phase: albite - $Na_2Al_2Si_6O_{16}$, scavenges all other radionuclides

Neutron poisons / U-238 can be to mitigate potential criticality

Fabricated by Hot Isostatic Pressing, demonstrated up to 20 kg scale

Recent work by Georoc Ltd. to demonstrate application to $Mg(OH)_2$ sludges up to 100 litre scale, final package ca. 30 litres

Maddrell *et al.*, J. Nucl. Mater., 456 (2015) 461.



Granite encapsulation concept

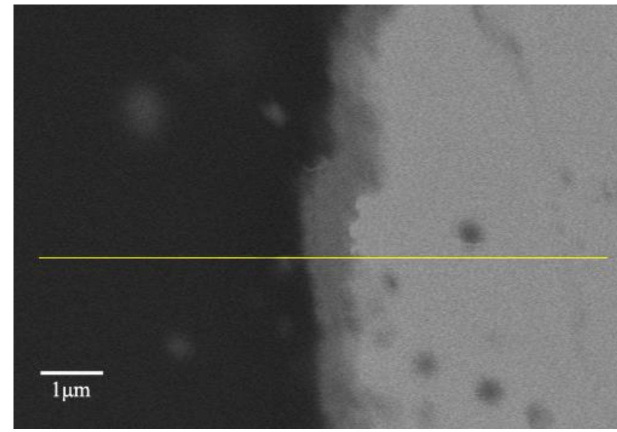
Approach is to encapsulate a host material within a partially melted and recrystallised granite from the borehole location.

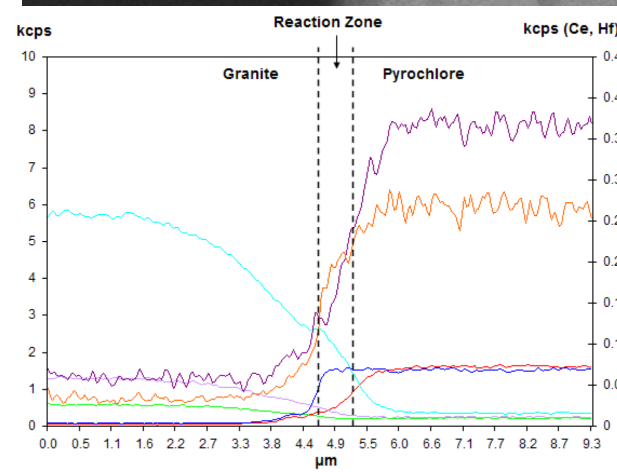
The recrystallised granite will be in equilibrium with the saline groundwater at emplacement depth, hence release rates will be very low.

Preliminary investigation has shown that envisaged wasteforms for Pu disposition are essentially stable under processing conditions.

Note – in this case, Ce is Pu surrogate; Hf and Gd present as neutron poisons.

$Gd_2Zr_{1.6}Ce_{0.2}Hf_{0.2}O_7$

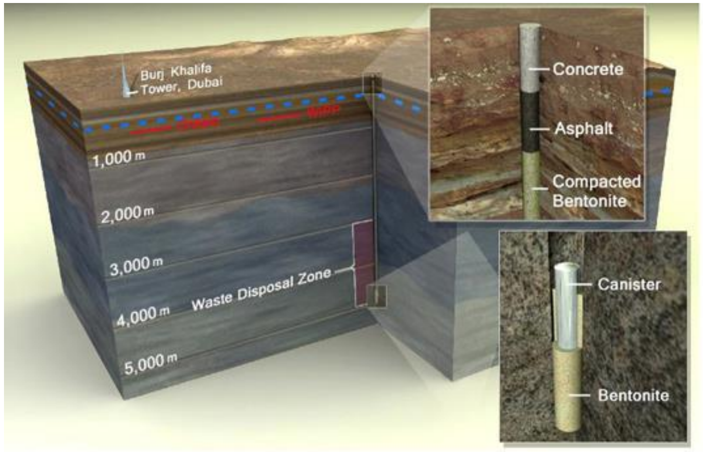




Gibb *et al.*, J. Nucl. Mater., 374 (2008) 364.

T.W. Ng, M. Eng. Thesis, University of Sheffield, (2010).

Summary



Deep borehole disposal concepts place greater reliance on the geological barrier.

Plausible materials and processes exist for treating and packing potential borehole wastes.

A robust wasteform, as the radionuclide source term, will mitigate against residual uncertainties in the disposal system.

A robust wasteform should help to make a more robust operational safety case – improved passive safety and waste package integrity.

A credible post closure safety case should feature a mechanistic model of wasteform evolution.

R&D challenge to understand wasteform evolution and soubilities under disposal conditions.