Politecnico di Torino

Master of Science in Energy and Nuclear Engineering
Course of Advanced Materials for Nuclear Applications

Advanced Materials for the Immobilization of High Level Nuclear Waste: The Role of Borosilicate Glasses in Long Term Containment

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NUCLEAR WASTE

> *Radioactive waste is any radioactive material in gaseous, liquid or solid form that is not going to be used any longer. The material has to be controlled as radioactive waste by a regulatory body under the legislative and regulatory framework.*
>
> *All EU countries generate radioactive waste, and 17 of them also manage spent fuel on their territory. Owing to its radiological properties and the potential hazard it poses, it is important to ensure the safe management of radioactive waste and spent fuel at all stages. It requires containment and isolation from humans and the living environment over a long period of time.*
>
> *European Commission*

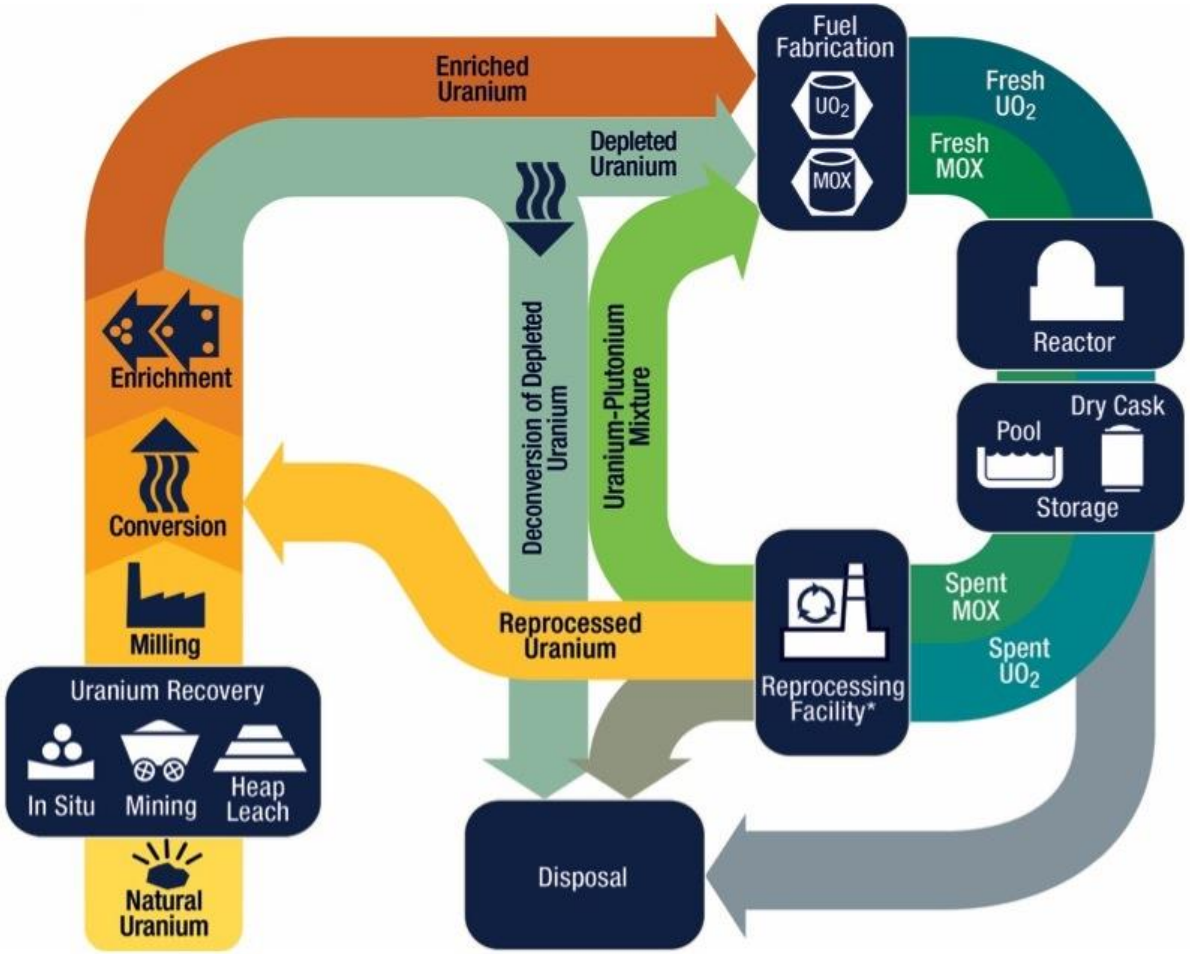

Fig.01 – Closed Fuel Cycle – source: https://en.wikipedia.org/wiki/Nuclear_fuel_cycle - author: Nuclear regulatory commission

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DISPOSAL

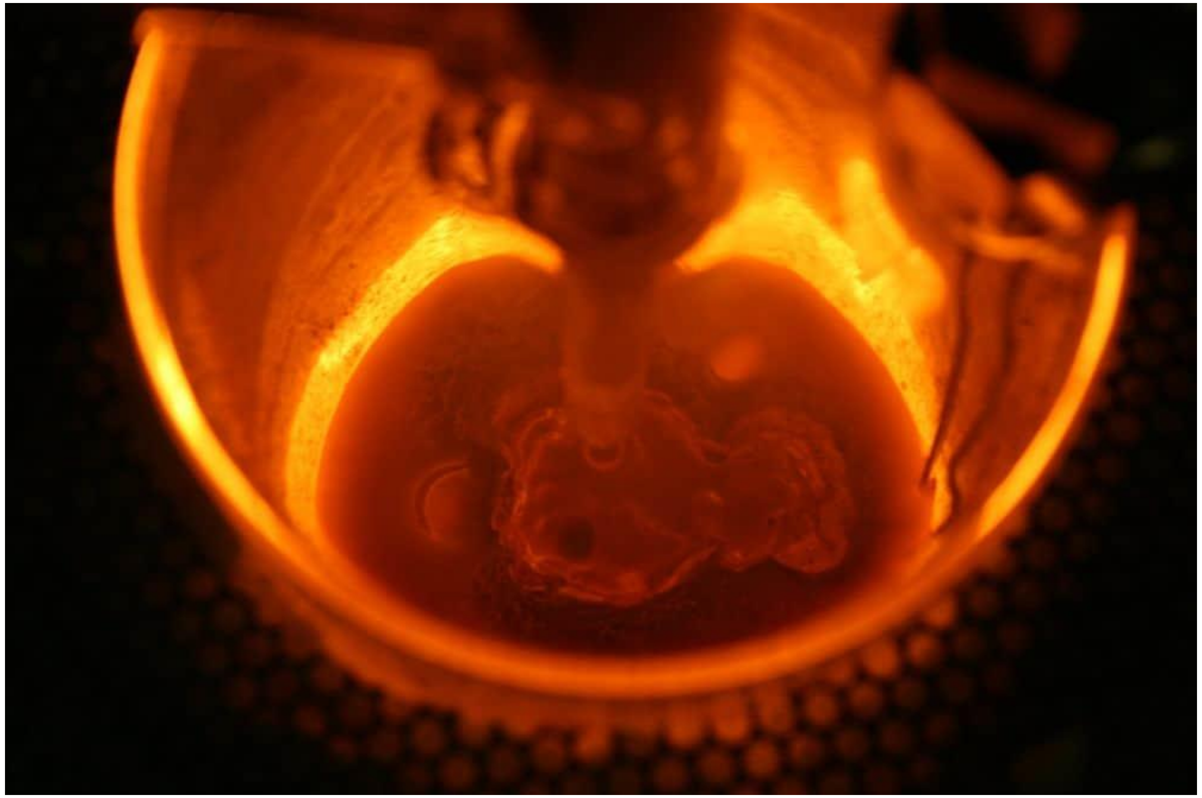

Fig.02 – Molten nuclear waste glass – sorce: https://ceramics.org/acers-spotlight/introduction-to-glass-for-nuclear-waste-disposal-for-glass-then-and-now/ - author: Courtesy of Pacific Northwest National Laboratory

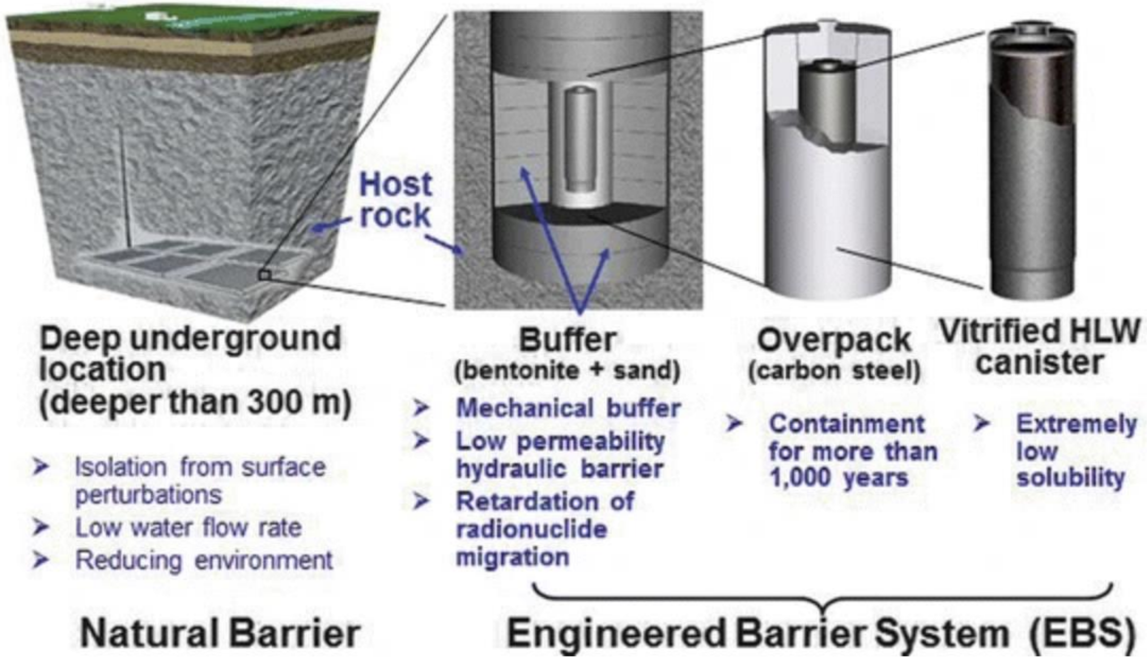

Fig.03 – Multibarrier concept – source: https://link.springer.com/chapter/10.1007/978-4-431-55111-9_24/figures/4 - author: NUMO

THE EFFECTS OF RADIATIONS

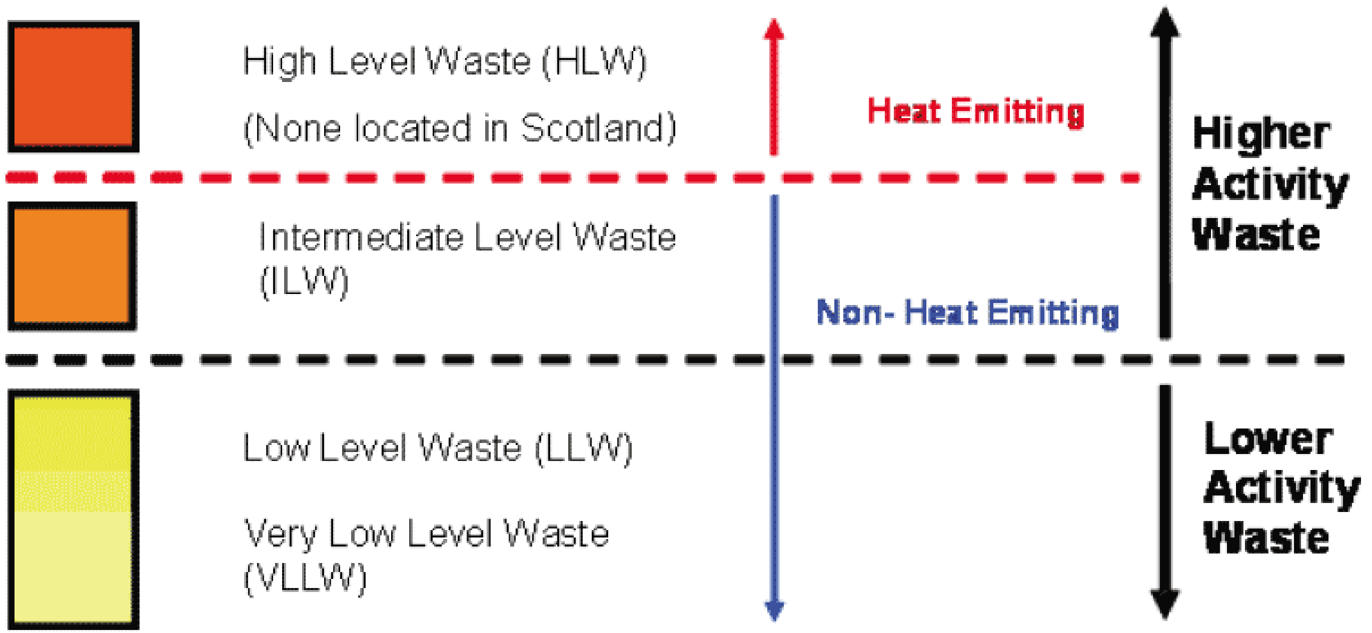

Fig.08 - Diagram of Waste Categories – source: Scottish Government, https://www.gov.scot/publications/scotlands-higher-activity-radioactive-waste-policy-supplementary-information-2010/pages/3/

- α
- β
- γ
- Neutrons

- Structural damage
- Amorphization
- Mechanical degradation
- Chemical corrosion
- Ionization

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GLASS LATTICE

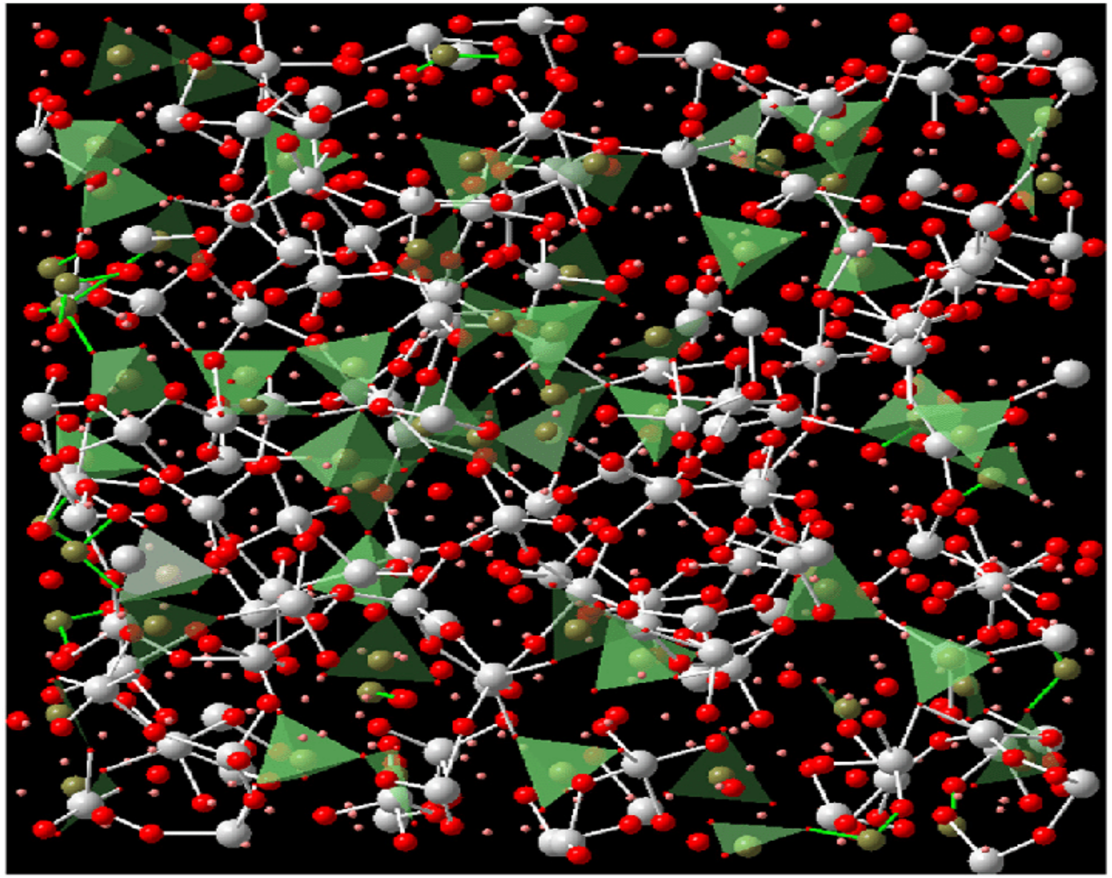

Fig.04 – Structure of borosilicate glass obtained with azTotMD software – source: https://www.researchgate.net/publication/332554502_azTotMD_Software_for_non-constant_force_field_molecular_dynamics - author: Raskovalov, Anton

Network Formers		
Si	40 – 50 [%]	Structure
B	10 – 20 [%]	HLW solvent
Network Modifiers		
Na	10 – 20 [%]	Viscosity
Network Intermediates		
Al	5 – 15 [%]	Durability

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THE ROLE OF BORON IN GLASSES

1. **Structural network former**: increases the network polymerization
2. **Reduces viscosity and melting temperatures***: improves industrial processibility and reduce energy consumption
3. **Mechanical hardness and strength***: improves fracture resistance and mechanical durability
4. **Leaching resistance**: enhances hydrolytic stability and chemical durability
5. **Ionic radionuclide solvent**: improves solubility and dispersion of radionuclides within the glass matrix
6. **Optical**: increases optical transparency
7. **Phase stability**: prevents formation of crystalline or phase-separated regions that could reduce waste retention

* Crucial aspect for the so called "Boron conversion"

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THE ROLE OF MODIFIERS AND INTERMEDIATES

$$B_2O_3 \rightarrow 2[BO_3]$$

$$[BO_3] + Na_2O \rightarrow [BO_4]^- + 2Na^+$$

$$[BO_4]^- + [AlO_4]^- + 2Na^+ \rightarrow stable\ B - O - Si - Al$$

$$N_4 = \frac{[BO_4]}{[BO_3] + [BO_4]} = 0.4 - 0.8$$

$\boldsymbol{BO_3}$

Lower melt viscosity
Enhances the solubility of many radionuclides
VS
Low network connectivity
Reduces the long-term durability

$\boldsymbol{BO_4}$

Increase network polymerization
Improve chemical durability and mechanical rigidity
VS
Risk of generating NBOs if alkalis are excessive
Must be balanced to avoid network over-stabilization or phase separation

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RADIATION INTERACTIONS

Fig.09 - sample nuclear glass – source: https://www.ans.org/news/article-4737/locked-in-glass-the-vitrification-of-llw-streams/ author: Veolia

α

β

γ

Neutrons

- ✓ Amorphous structure
- ✓ High bond flexibility
- ✓ Displacement absorption
- ✓ No bubble-induced cracking
- ✓ Chemical durability

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LONG TERM DURABILITY PREDICTION

Objective: ensure that the glass continues to immobilize radionuclides for tens of thousands of years, without the need to directly observe processes that occur over geological timescales.

Accelerated Laboratory Test

Predictive Modelling

Performance Assessment

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ACCELERATED LABORATORY TEST

- Leach tests (static and dynamic)
- Stress tests
- Radiation damage simulations
- High temperature annealing
- Vapor hydration tests

The results does NOT reproduce the thousands years effects, but are useful for predictive modelling!

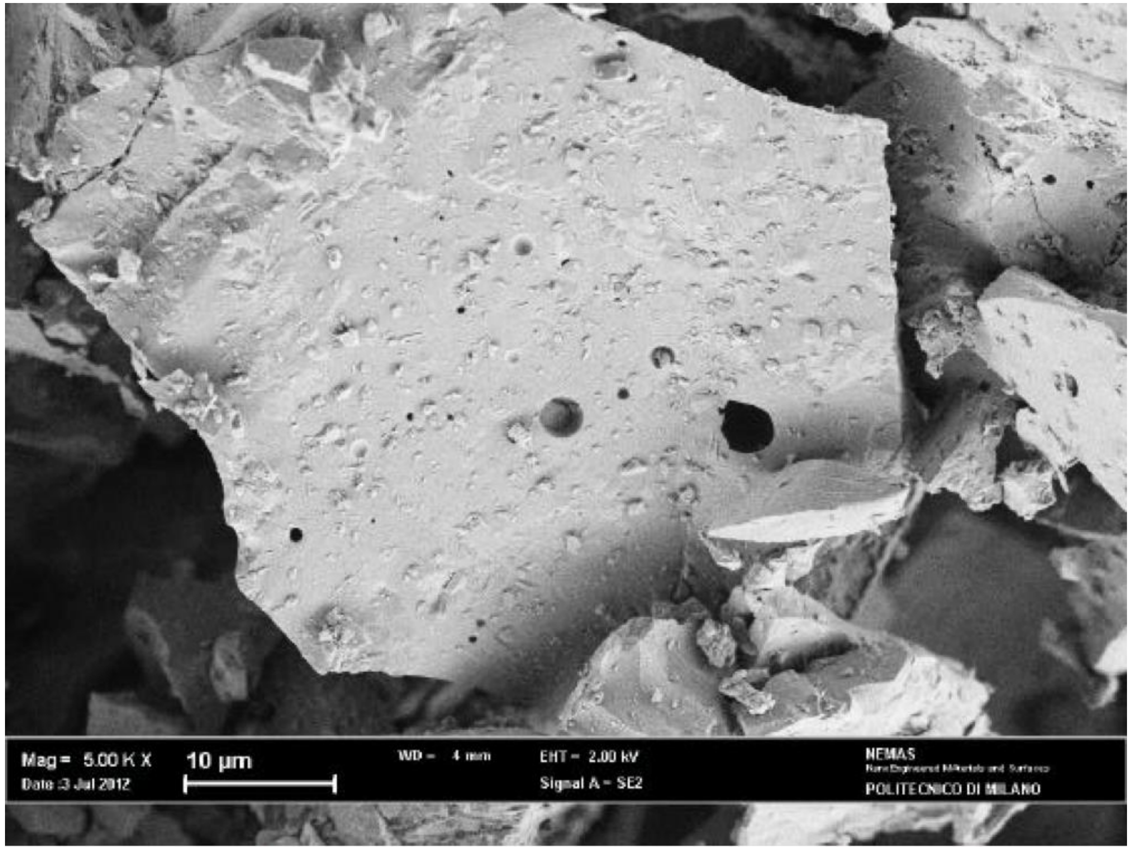

Fig.05 – static leach test result (pH 10) – source: https://www.ricercasistemaelettrico.enea.it/archiviodocumenti.html?task=download.send&id=1880:condizionamento-di-rifiuti-radioattivi-in-matrici-vetro-ceramiche-e-studio-delle-interazioni-rifiuto-terreno&catid=368 author: Paride Meloni, ENEA

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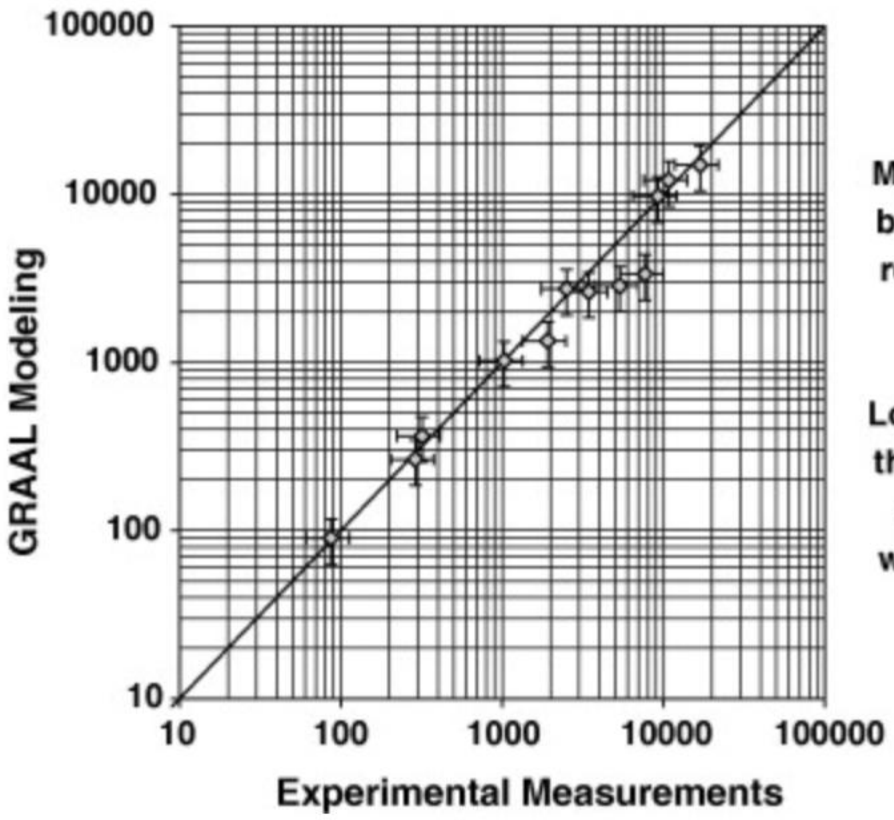
PREDICTIVE MODELLING

- Glass dissolution/corrosion models
- Data driven models
- Nano-/micro-structural model
- Safety assessment models



Mass of altered glass based on boron total release at the end of each experiment (mg of glass)
Log scale is chosen so that each experiment can have the same weight on the figure.

Fig.06 – Typical GRAAL output – source: https://www.sciencedirect.com/science/article/pii/S0022311509005996
author: P.Frugier

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PERFORMANCE ASSESSMENT

- Deterministic approach
- Statistic approach (Monte Carlo method)

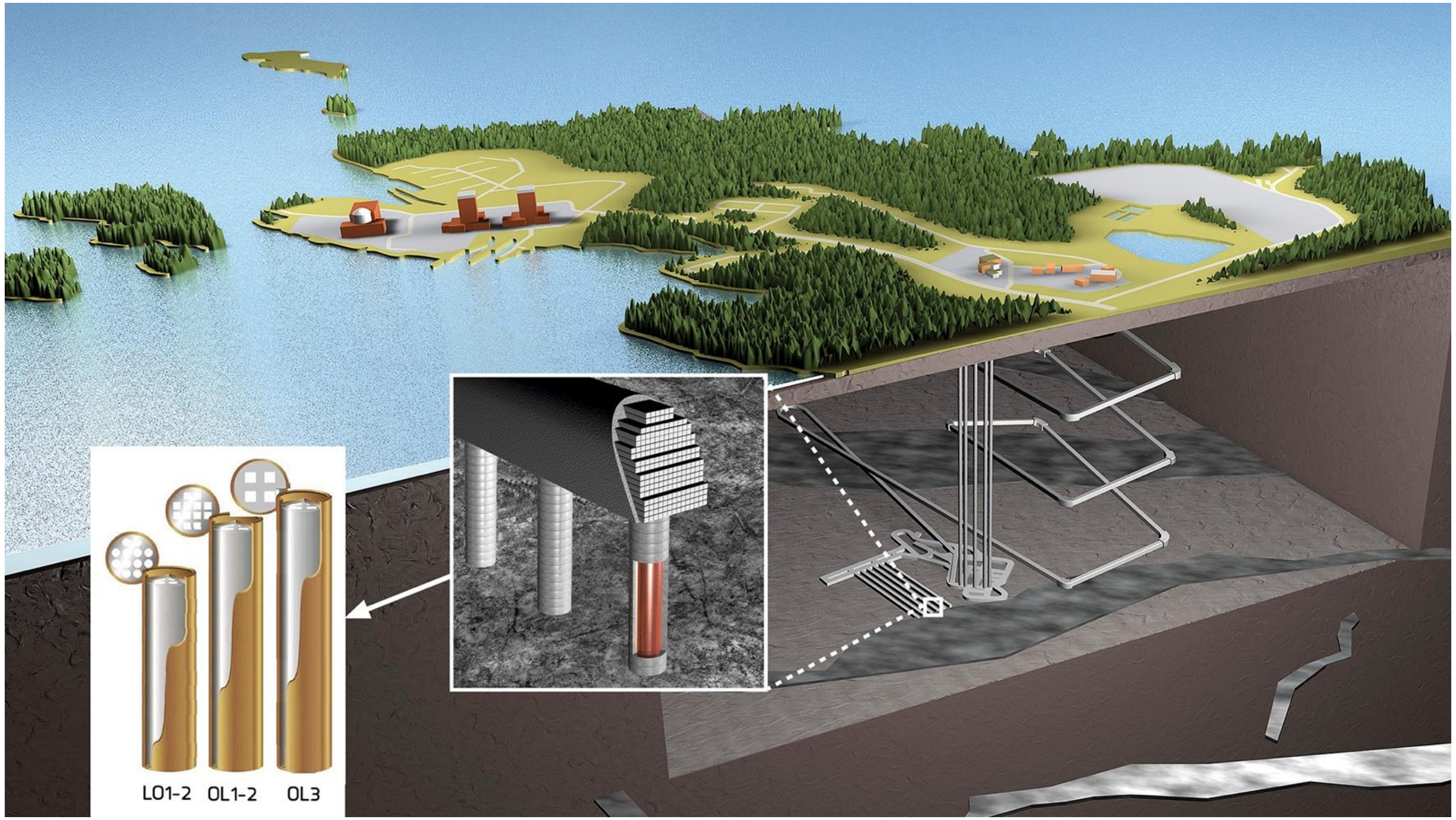

Fig.07-The Onkalo spent nuclear fuel repository (Posiva, Finland) – source: https://www.oecd-nea.org/jcms/pl_31093/expert-group-on-operational-safety-egos author: Soufiane Mekki

OTHER POSSIBLE SOLUTIONS

- Ceramic Crystals
- Alternative Glasses (phosphate, sulphate, alkali-silicate)
- Cements and Geopolymers

CONCLUSIONS

Borosilicate glasses provide an optimal balance between:

- chemical stability
- compositional tolerance
- high HLW loading capacity
- industrial manufacturability

Alternatives may be suitable for selective radionuclides or medium-level waste, but they do not achieve the versatility and long-term durability of borosilicate glass

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THANKS FOR YOUR ATTENTION

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