DEMONI - DEcommisioning Material characterizatiON and final dIsposal studies

KYT and SAFIR final seminar
24.1.2023

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Introduction

- Focus on LLIW waste management and final disposal.
- Coordinated project between VTT and HU
  - Partially funded by NKS and EU
- Four tasks:

1) Developing activity measurement methods for difficult to measure radionuclides (DTM)
2) Effect of radiation on mechanical properties of concrete
3) Concrete leaching in simulated final disposal conditions
4) Steel leaching in simulated final disposal conditions
   - Tasks are connected by same samples and partially same methods
   - Activated samples of the FiR1 research reactor are utilized, but developed methods are universal
   - Connections to various projects KYT/TERKOR, EU/RADWASTE, SAFIR/CONAGE

Task 1: Developing activity measurement methods

- Alpha and beta active radionuclides are referred as difficult to measure (DTM)
- DTM analysis require destructive analyses and radiochemistry
- Method development and/or validation of several materials
  - Betas (C-14, Fe-55, Ni-63) and gammas in activated steel in 2019
  - Betas (H-3, C-14, Fe-55, Ni-63, Ca-41, Cl-36) and gammas in activated concrete in 2020
  - Betas (H-3, C-14, Fe-55, Ni-63, Sr-90, Tc-99) and gammas in spent resin in 2021
  - Alphas (U/Pu/Cm-isotopes, Am-241, gross alpha) in spent ion exchange resin in 2022
  - Ni-59 in activated steel (2019-2023) in EU-PREDIS project
- Intercomparison exercises with international partners

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VTT
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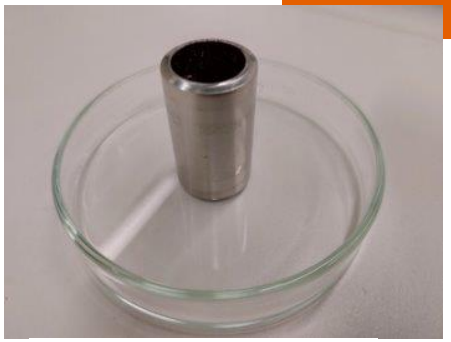
Task 1: Developing activity measurement methods

- Implementation of lessons learned in sampling and characterisation of highly active stainless steel
  - Researcher visit from UH to VTT in 2022
  - Chemical composition of the stainless steel (inactive)
    - ICP-MS, ICP-OES, GD-OES, nitrogen analyser
  - Sub-sampling of 2 mSv/h sample in hot cells
  - Trilateral intercomparison exercise being finalised early 2023
    - VTT, UH, and international partner
    - Co-60, Fe-55, Ni-63
  - Drafting of a publication on-going (foreseen submission spring 2023)
  - Ni-59 intercomparison in PREDIS in 2023?

Task 1 publications
- Bachelor thesis
  - 1 in Metropolia
- NKS-reports
  - 3 published, 1 (to be published in January 2023)
- Peer-reviewed publications in Journal of Radioanalytical and Nuclear Chemistry
  - 4 published, 3 writing on-going (foreseen submissions in spring 2023)

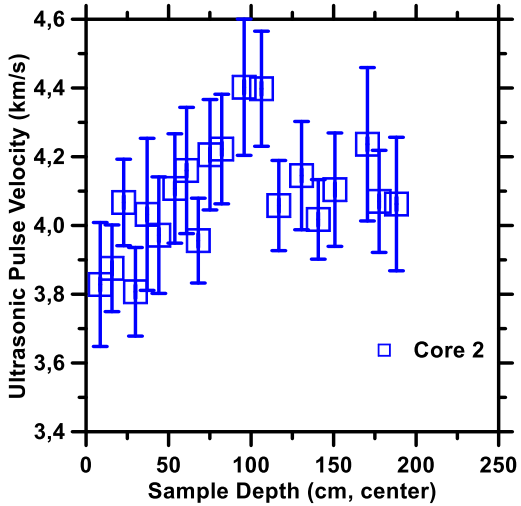


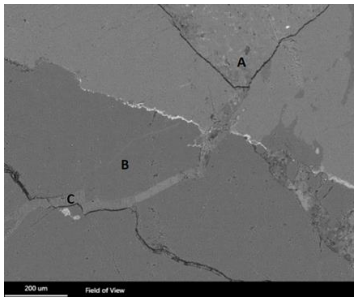
Picture T1.1. Low activity steel sample

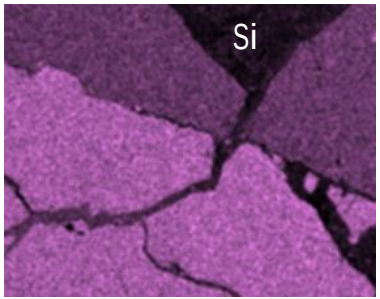
Picture T1.2. Sub-samples of highly activated steel samples ready for DTM analyses

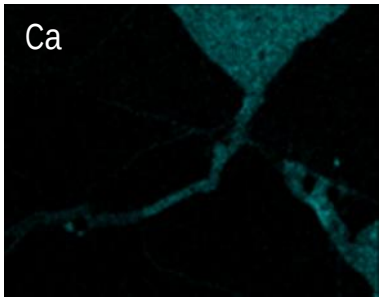
Task 2: Radiation effect on concrete mechanical properties

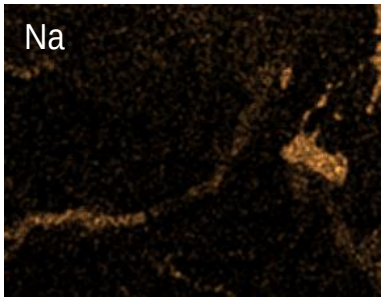
- Concrete mechanical integrity is important because this material is used as a physical barrier structure during final disposal of radioactive waste
- However, radiation degrades concrete's mechanical properties in many ways
- Non-destructive testing indicates a lower mechanical strength of concrete (slower ultrasound pulse velocity) in radiation-exposed concrete core
- Scanning electron microscopy shows formation of cracks within the concrete aggregates (rocks) caused by increased reactivity of Si containing material
- These results suggest that chemical changes to the concrete by radiation lead to a subsequent deterioration in its mechanical properties
- Such a finding is consistent with the focus of other projects, e.g., ACES, SAFFIR, etc., on radiation-induced changes to long-term concrete durability





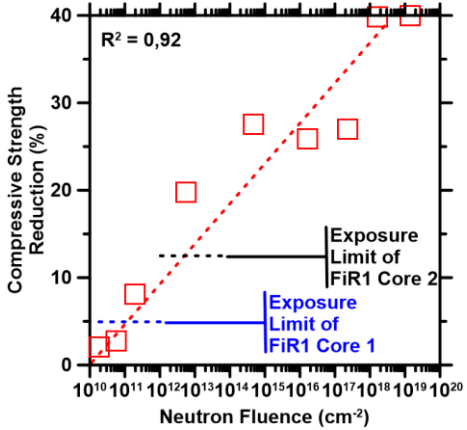


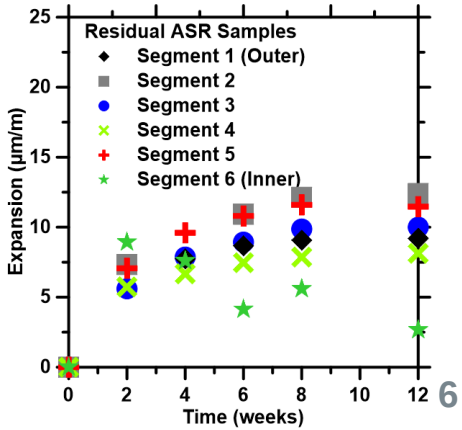




Task 2: Radiation effect on concrete mechanical properties

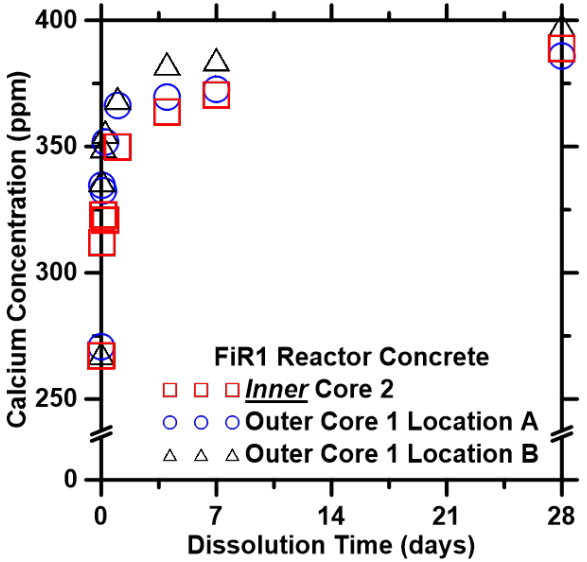
- The limited available data from the literature that match the lower radiation dose of the FiR 1 samples suggest a possible reduction in concrete strength of up to 30%
- This is consistent with the results of non-destructive testing, and the equivalent observed reduction in strength for concretes from the outer parts of each core
- However, the primary damage mechanism for these low radiation doses remains poorly understood, compared to radiation-induced volumetric expansion that dominates at higher radiation dose (>10^{19} cm^{-2} neutron fluence)
- The possibility of residual expansion from increased aggregate reactivity (ASR) has been largely ruled out, based on very low residual expansion from such tests on the FiR 1 samples, indicating more direct changes to the concrete/cement system
- The possibility of changes focused on the cement paste matrix, by gamma radiation as well as neutron radiation, remains to be investigated in later studies
- This also highlights a potentially important consideration for long-term waste disposal, during which barrier materials could be subjected over long periods of time to sufficient (gamma) radiation to induce changes in mechanical stability





Task 3: Concrete leaching in simulated final disposal conditions

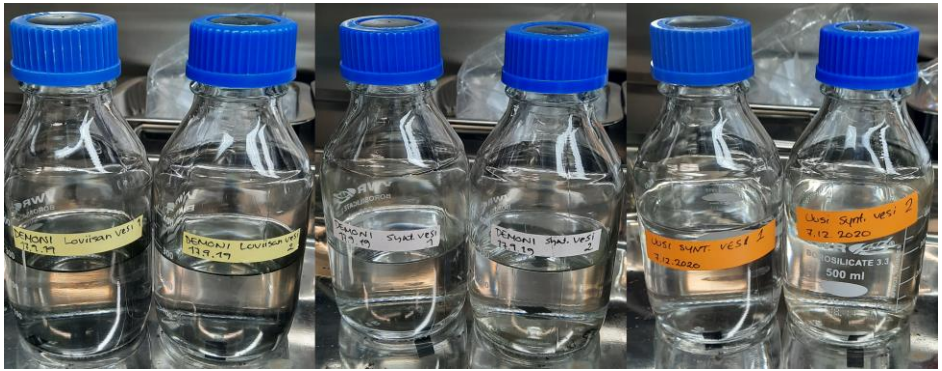
- Leaching tests on the samples from portions of the outer and inner FiR 1 concrete cores demonstrated similar release of the expected elements typically present in cement paste, such as calcium, silicon, aluminum, etc.
- This indicates that radiation-induced changes to chemical stability of the cement paste matrix, for the lower radiation dose of FiR one samples, do not significantly impact known deterioration processes such as de-calcification
- Furthermore, all leaching samples were devoid of radionuclides over the course of the experiment (below detection)
- These results support the idea that for such LILW concrete waste, the direct impact of irradiation on leaching is not a concern for disposal
- However, the potential coupling between mechanical deterioration and accelerated leaching, i.e., leaching along preferred pathways such as through cracks that form due to mechanical instability, remains to be investigated in future studies (as mentioned in Task 2)



Task 4: Steel leaching in simulated final disposal conditions

VTT: C-14, with long half-life and high mobility in geosphere and biosphere, is important radionuclide in the safety assessment

-C-14 release from irradiated pressure vessel steel
-long-term dissolution experiment (2019 – 2022)
-gas phase experiments (2020 – 2022)
-simulated anoxic repository conditions (Ar atmosphere glove box)
-Loviisa ground water and two simulant waters: regular sampling during experiments
-Analyses: C-14 (LSC), stable elements (ICP-OES), TOC-14 (acidification + Ar purging and LSC), TOC (UV-Vis), TIC and anions (IC), gas analysis (GC)
-steel sample analysis with SEM-EDS



Dissolution experiments in glove box

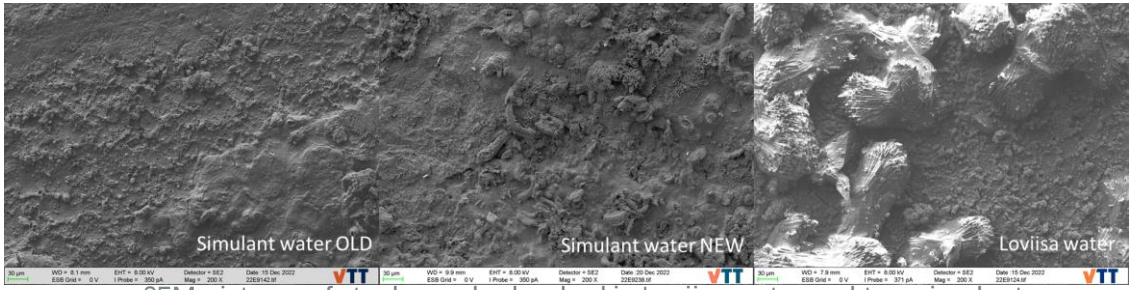
Irradiated and leached steel sample (1 cm x 1 cm)

Gas phase experiments and sampling in glove box

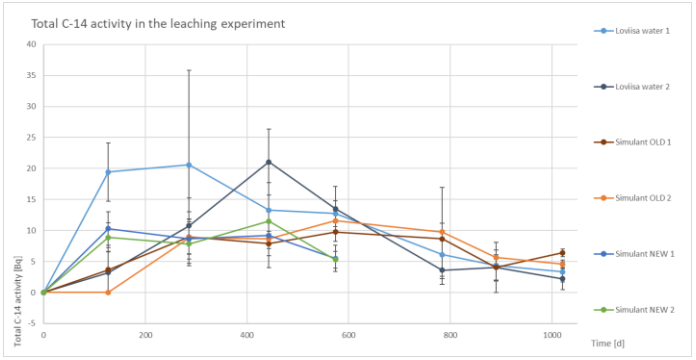
Task 4: Steel leaching in simulated final disposal conditions

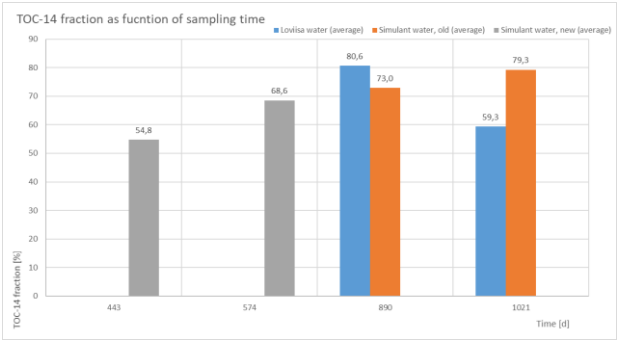
VTT:

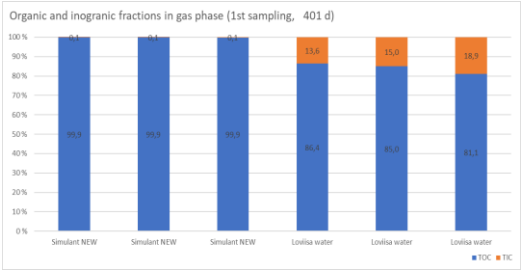
-Low C-14 release detected: in Loviisa water and in simulant about 0,011 % per year of the total C-14 inventory
-C-14 mainly detected in organic form in the solution phase
-SEM pictures show differences in the surfaces of the steel samples leached in Loviisa water and simulant waters (results in analysis)
-CH_4 was the main component in the gas phase
-Co-operation with KYT-TERKOR project
-Final results will be published in 2023



SEM pictures of steel samples leached in Loviisa water and two simulants



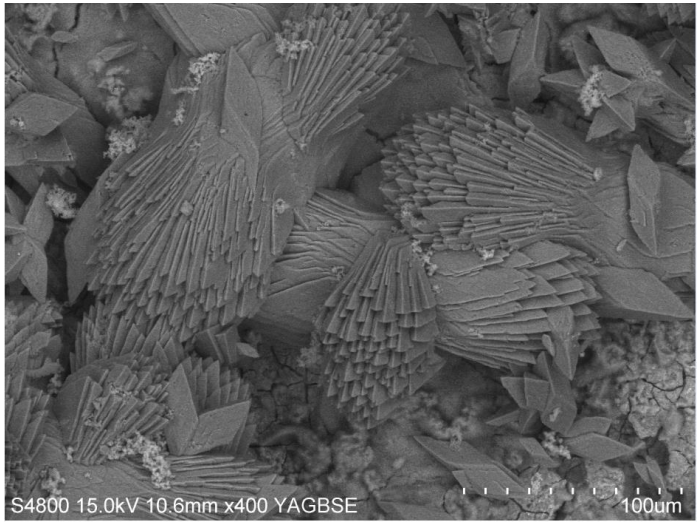




Task 4: Steel leaching in simulated final disposal conditions

University of Helsinki:

-long-term dissolution experiment in 2019-2022
-anoxic environment (glove box)
-Loviisa ground water and a saltier simulant water
-> differences in steel leaching and corrosion products after leaching?
-60Co, 55Fe and 63Ni determined from the water regularly
-steel samples have been analysed in late 2022: Raman spectroscopy, SEM-EDS
- differences in radionuclide dissolution and corrosion products on steel surfaces between two corrosion environments
-XRD analyses are still pending, will be done in 01/2023



SEM-picture of a steel sample that has been corroded in Loviisa ground water

-strong co-operation with KYT-TERKOR project
-Master thesis work is at the final stage
-one previous Bachelor thesis supervised in this task (2020-2021)
-data from different analyses are now being summarised
-> to be published in the Master thesis in early 2023 and as an article in 2023