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European Joint Programme
on Radioactive Waste Management

INCREASED TEMPERATURE EFFECTS ON NA WYOMING BENTONITE : SIGNIFICANCE FOR DEEP GEOLOGICAL RADIOACTIVE WASTE DISPOSAL

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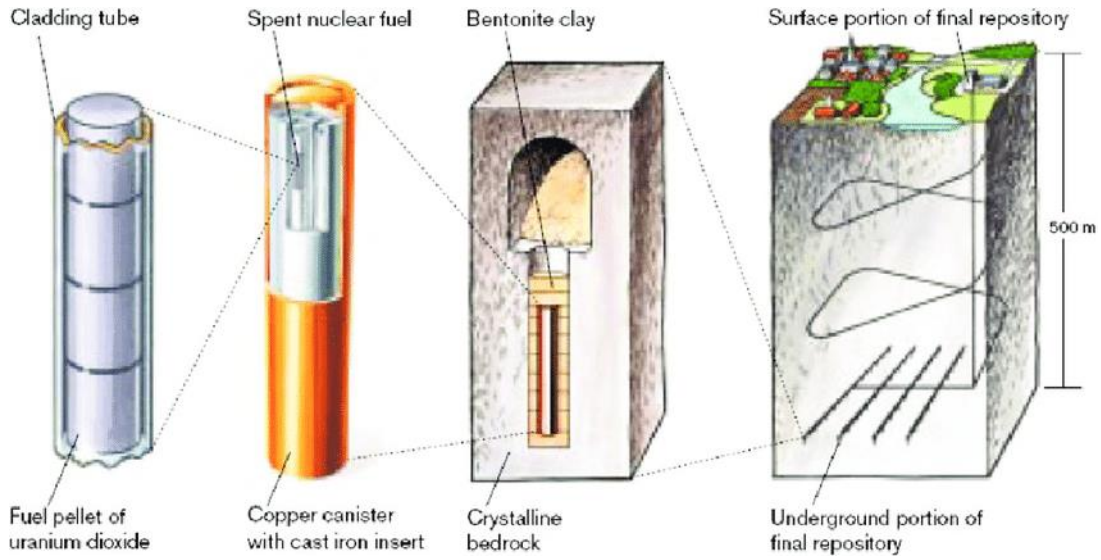
INTRODUCTION

- Spent nuclear fuel in Finland for deep geological disposal in KBS-3V style after a period of cooling.
- At the end of 2019 there was 2,261 tons of heavy metal (tHM) spent fuel in pool storages at NPP locations and an estimates 4,200 (tHM) in 2050.
- Are the safety limits feasible for a variety of geological disposal concepts for high heat generating wastes or does high temperature inhibit the buffers safety functions?



The KBS-3V final disposal concept for spent nuclear fuel.

[1] SKB - Swedish Nuclear Fuel and Waste Management Company (2006) *Long-term safety for KBS-3 repositories at Forsmark and Laxemar – a first evaluation. Main Report of the SR-Can project, October*. Available at: www.skb.se (Accessed: 11 May 2022).

AIMS AND OBJECTIVES

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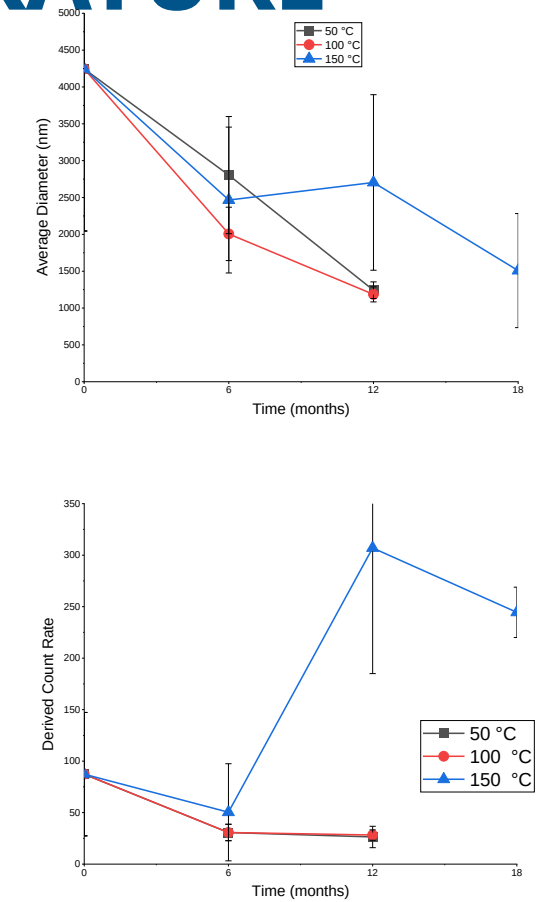
Understand the effects of elevated heat loadings on buffer material to be used in Finnish spent nuclear fuel disposal.

- **Task 1:** Characterization of Wyoming Na Bentonite (BARAKADE) exposed to high temperature (150 °C) over extended duration (24 months) (**HITEC**).
- **Task 2:** Understand the consequences of potential changes to the material following 2 years of heat treatment.
- **Task 3:** (Specific to SAFER) Highlight novel robust and repeatable experimental setup for column studies at The University of Helsinki – possibilities for future work.

CHARACTERIZATION OF MATERIAL TREATED BY HIGH TEMPERATURE

- Over 18 months we have monitored changes in the characteristics of NaWyoming Bentonite heated at 150 ^{0}C.
- Techniques used: XRD, FESEM, BET, MPAES, DLS & Zeta potential.
- Reaction to reach completion in August ‘22 (2 year).
- XRD & FESEM show no observable change in sample over 18 months.
- Colloidal size does not change but amount increases (See Right)



Top: Average colloid diameter (/nm).
Bottom: Derived count rate of colloidal solutions

Water Composition:
$CaCl_2.2H_2O$ 111 mM
NaCl 32 mM
pH = 5.3

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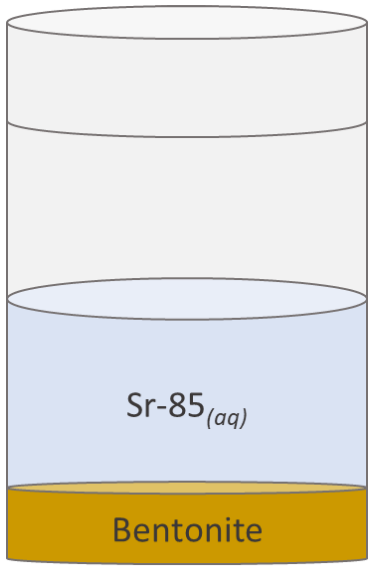
BUT we can do more than just characterise the changes to the bentonite. We can **QUANTIFY** these effects in terms of radionuclide sorption.

SR SORPTION ISOTHERMS



3 days

Centrifuge

1. Measure sorption as a function of
   a) Time
   b) pH (8 & 13)
   c) [Sr]

2. Determine supernatant composition (MP-AES / DLS / Zeta potential)

Ultra-centrifuge

Determine supernatant composition (MP-AES / DLS / Zeta potential

Repeat using heat exposed bentonite

SR SORPTION ISOTHERMS

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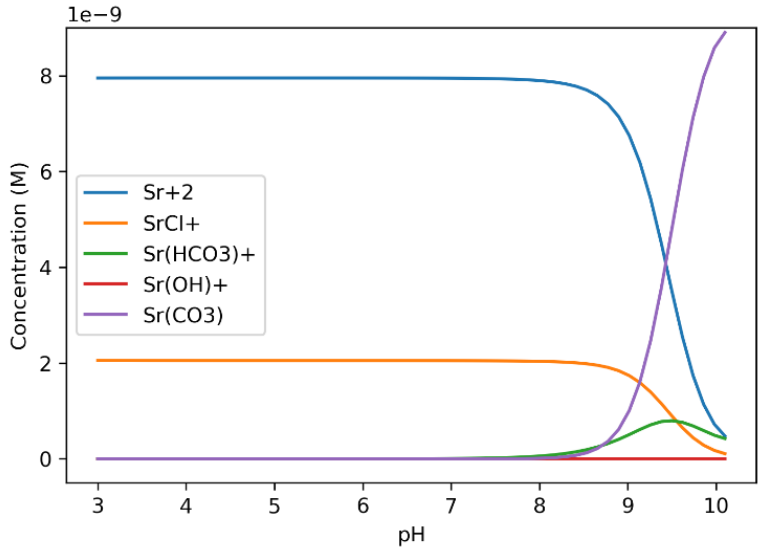
Current Work

Understand if the experiments must be carried out anaerobically

Modelled Sr-85 (10^{-10} to 10^{-3} M) sorption to Bentonite w/o CO_2

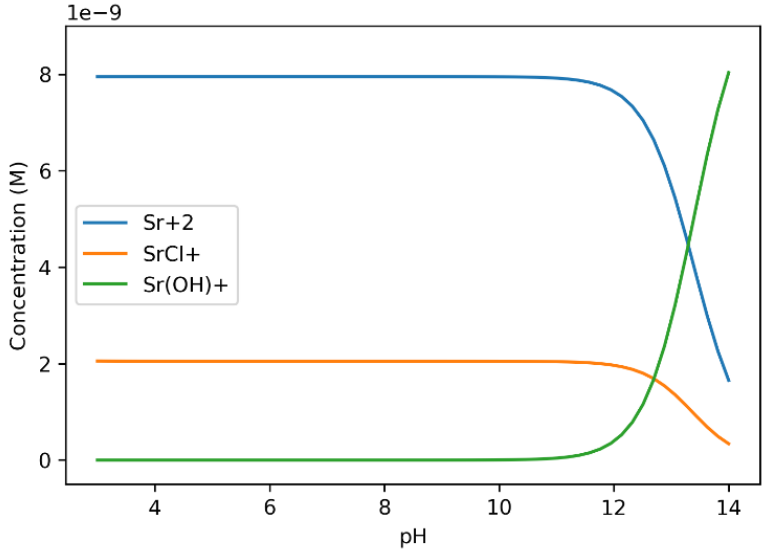
Sr-85 radiolabelled sorption isotherms are underway using unheated bentonite.

CEC determination on the unheated bentonite



$SrCO_{3(s)}$ phases dominate above pH 9

Sr speciation in reference solution with CO_2.



$Sr(OH)^+$ phases dominate above pH 12

Sr speciation without CO_2

CONCLUSIONS & FUTURE WORK

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Conclusions:

Characterization of changes in NaWyoming bentonite following heat exposure (150 ^{o}C) for up to 18 months.
Although XRD & SEM show no obvious changes, DLS + Zetapotential measurements suggest increased colloid production.

Full analysis possible upon reaction completion in August ’22.

Future Work:

Characterisation of 24 month sample.
Sr-85 radiolabelled sorption isotherms on bentonite materials.
CEC determination of exposed (150 ^{o}C) bentonite.
XAS analyses to confirm Sr speciation at molecular scale.

Presenting the sorption data at ICC ‘22 – Istanbul (July 25-29 2022).

WITH THANKS TO:

Dr. Xiaodong Li (The University of Helsinki) for PHREEQC modelling

Dr. Stellan Holgersson (Chalmers University) for BET analyses.

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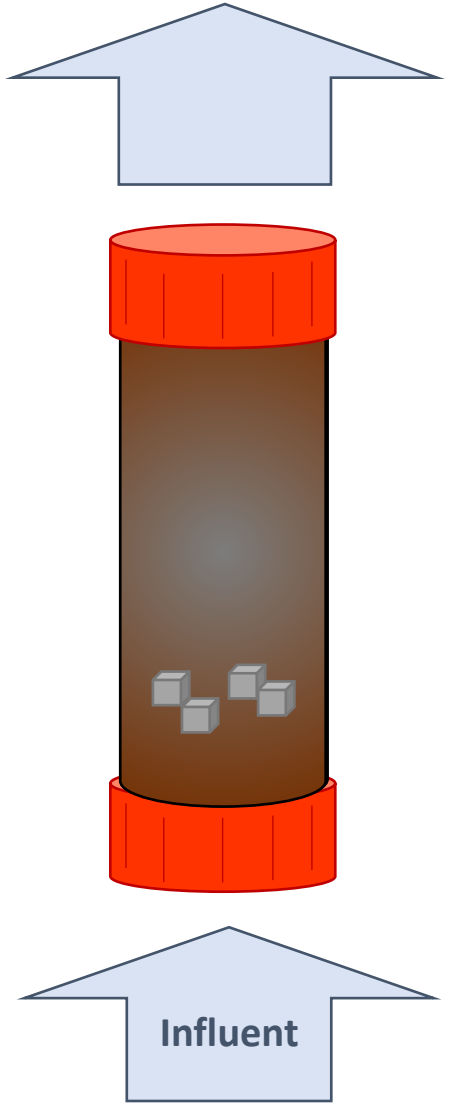
IDEAS FOR SAFER 28

- Microcosm experiments not representative of actual subsurface [1][2].
- Novel column setup at HY allows facile study of environmentally relevant conditions.
- Here we can :
  - Tailor fill material / influent
  - Monitor effluent
  - Test remobilisation
  - Post-mortem analyses



[1] Ho, M. S. *et al.* (2022) 'Retention of immobile Se(0) in flow-through aquifer column systems during bioreduction and oxic-remobilization', *Science of The Total Environment*, 834, p. 155332. doi: 10.1016/j.scitotenv.2022.155332. [2] Ho, M. S. et al. (2022) 'Redox behaviour of Technetium in sediment columns', Environmental Science & Technology, *manuscript in preparation*.