

# The EURAD EU Project Work Package FUTURE: Fundamental Understanding of radionuclide RETention



Sergey V. Churakov<sup>1</sup>, Vaclava Havlova<sup>2</sup>, Dirk Bosbach<sup>3</sup>, Norbert Maes<sup>4</sup>, Martin A. Glaus<sup>1</sup>, Cornelius Fischer<sup>5</sup>, Rainer Dähn<sup>1</sup>, Sylvain Grangeon<sup>6</sup>, Maria Marques Fernandes<sup>1</sup>

<sup>1</sup>PSI, Switzerland; <sup>2</sup>Řež a.s., Czech Republic; <sup>3</sup>FZ-JUELICH, Germany; <sup>4</sup>SCK.CEN, Belgium; <sup>5</sup>HZDR, Germany; <sup>6</sup>BRGM, France.

The work package (WP) FUTURE is dealing with radionuclides retention and transport in repository systems in argillaceous and crystalline rocks. FUTURE is organized in two main tasks: (1) MOBILITY, comprising work on mobility of radionuclides in compacted clay, the mobility of radionuclides in crystalline rock and the reversibility of sorption as well as (2) REDOX, comprising work on redox reactivity of radionuclides on mineral surfaces. Twenty European Research Entities (RE), Waste Management Organizations (WMO) and Technical Support Organizations (TSO) are contributing to the WP.

## Main Objectives

The main research objectives of the WP FUTURE comprise the:

- Impact of chemical boundary conditions (pH, ionic strength) and of the rock microstructures on radionuclide (RN) speciation, mobility and sorption reversibility in argillaceous and crystalline rocks (C).
- Quantitative and mechanistic understanding of the impact of specific surface properties of materials (diffusive double layer, surface potential), the role of grain boundaries, and structural heterogeneity on the mobility of chemical species at pore scale (A).
- Relationships between fracture/pore structures and transport as well as the feedback of mineral reactions on RN mobility/retention (B).
- Fundamental understanding of surface induced (heterogeneous) redox processes relevant for uptake of redox-sensitive radionuclides at Fe<sup>II</sup>/Fe<sup>III</sup> bearing minerals surfaces (D).

## Systems:

- Pure clay minerals (e.g. illite) or clay rocks (e.g. Opalinus or Callovian-Oxfordian Clay), both in dispersed and compacted state
- Fractured crystalline rocks
- Porous fracture fillings, including sheet silicates, iron oxides, etc.
- Ni/Zn/Se/Ra on purified clay minerals (illite/smectite) and hematite
- U/Ra on natural rocks (Opalinus Clay, Callovo-Oxfordian Clay)
- Fe<sup>II</sup>/Fe<sup>III</sup> bearing minerals: Pure clays and iron oxides

## Nuclides

- Weakly, moderately and strongly sorbing radionuclides (Se, Ni, Zn, Ra, Eu, U)
- Redox sensitive nuclides U, Pu, Tc, Np, Se

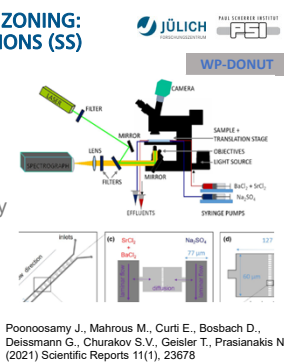
## A ENIGMA OF OSCILLATORY CRYSTAL ZONING: RN RETENTION BY SOLID SOLUTIONS (SS)

### A lab-on-a-chip approach:

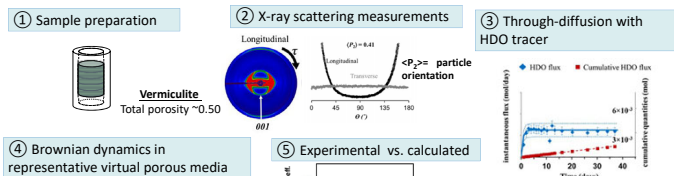
- in-situ characterization
- reactive transport modelling

### Results

- Composition of nucleating SS is predicted by Classical nucleation theory
- Crystal zoning is controlled by the limited diffusion of solutes and crystallization kinetics



## B EFFECT OF PARTICLE ORIENTATION ON WATER DIFFUSION



### Take Home message

Particle orientation and porosity are the primary parameters controlling water diffusion.

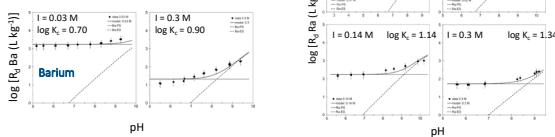
Asaad A., Hubert F., Ferrage E., Dabat T., Paineau E., Porion P., Savoye S., Gregoire B., Dazas B., Delville A., Terture E. (2021) Applied Clay Science, 207, 106089.

## C ADSORPTION OF BARIUM AND RADIUM ON MONTMORILLONITE

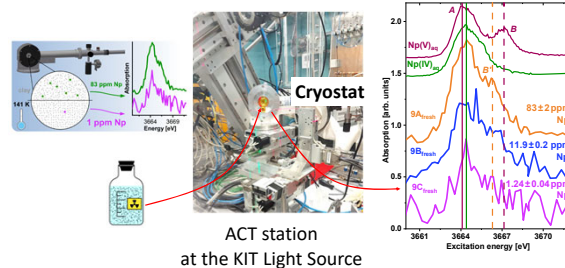
### Is Ba a good inactive analogue for <sup>226</sup>Ra?

- At low ionic strength: selectivity coefficients for Ba and Ra in good agreement ( $\log K_d$  values vary between 0.70 and 0.84). At high ionic strength, the  $K_d$  (Ba-Na) slightly higher compared to the low ionic strength data.
- $K_d$  (Ra-Na) exhibits a clear dependency on ionic strength

Ba is a good analogue for <sup>226</sup>Ra at I < 0.1 M and pH < 8



## D PROBING THE NP OXIDATION STATE BY XAS AT 1 – 80 ppm



Schacherl B., Joseph C., Lavrova P., Beck A., Reitz C., Pruessmann T., Feilhauer D., Lee J., Dardenne K., Rothe J., Geckeis H., Vitova T. (2002) Anal. Chim. Acta 1202, 339636.

## Contributing organization

Organisation	Function	Country	Organisation	Function	Country
BRGM	RE	France	Andra	WMO	France
CEA	RE	France	RAWRA/SÚRAO	WMO	Czech Republic
CNRS	RE	France	UIJV	LTP/RE	Czech Republic
UGrenoble	RE	France	TNO	RE	Netherlands
UPoitiers	RE	France	MTA EK	RE	Hungary
FZJ	RE	Germany	RATEN/ICN	RE	Romania
HZDR	RE	Germany	SCK-CEN	RE	Belgium
KIT	RE	Germany	UHELSINKI	RE	Finland
JGU INC MAINZ	RE	Germany	CIEMAT	TSO	Spain
PSI	RE	Switzerland	GRS	TSO	Germany

## Conclusions:

- Significant progress has been made in understanding the RN transport and retention at repository relevant conditions. These include consideration of chemical analogues and relations between microstructure and ions/solvent mobility
- To support quantitative assessment of sorption mechanism detection limits of high resolution X-ray spectroscopy have been pushed down to ppm level
- New way for approaching crystallization phenomena at pore scale provide deep inside in the mechanism of RN retention by Solid Solution

## WP-Coordination Team



The project leading to this application has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement n° 847593.