THE USE OF ANALOGUES IN ASSESSING THE CHEMICAL EVOLUTION OF HLW DISPOSAL CELLS

9 May 2023 • Erika Neeft, Guido Deissmann, Diederik Jacques

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EURAD Vision, SRA, Roadmap

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EURADSciences

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Waste Producers (NUGENIA)
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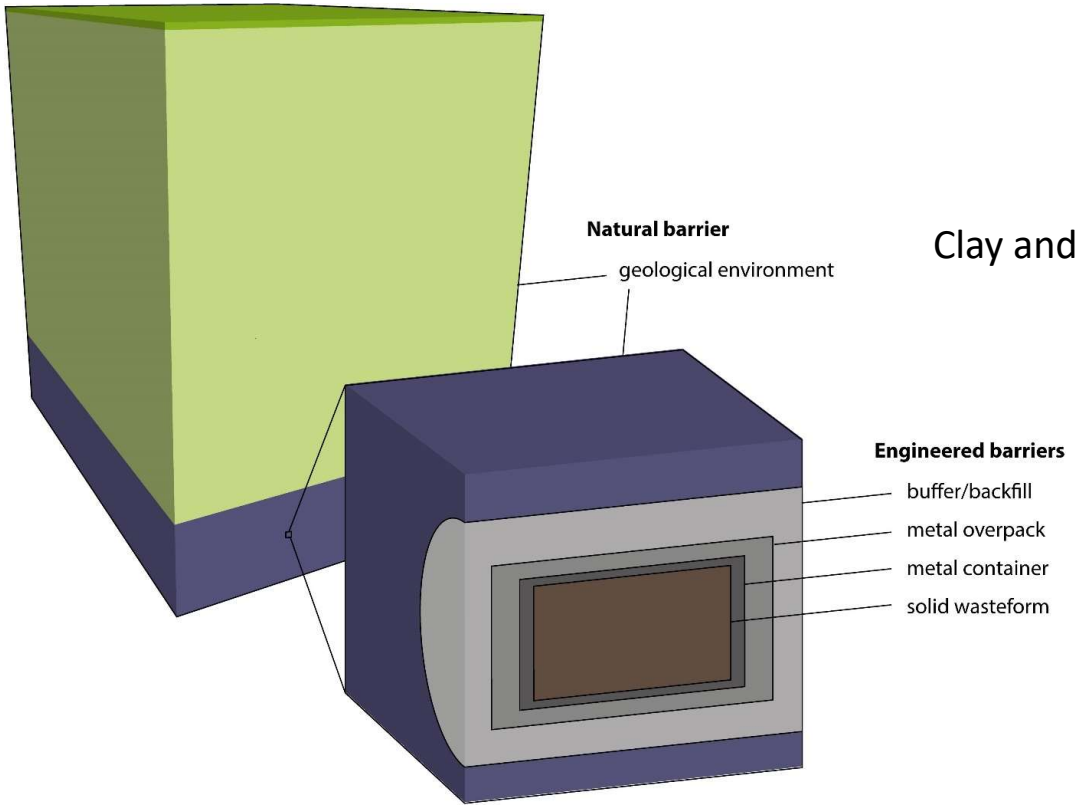
EURAD ACED

Assessment of Chemical Evolution of ILW and HLW Disposal Cells



Clay and granitic host rock

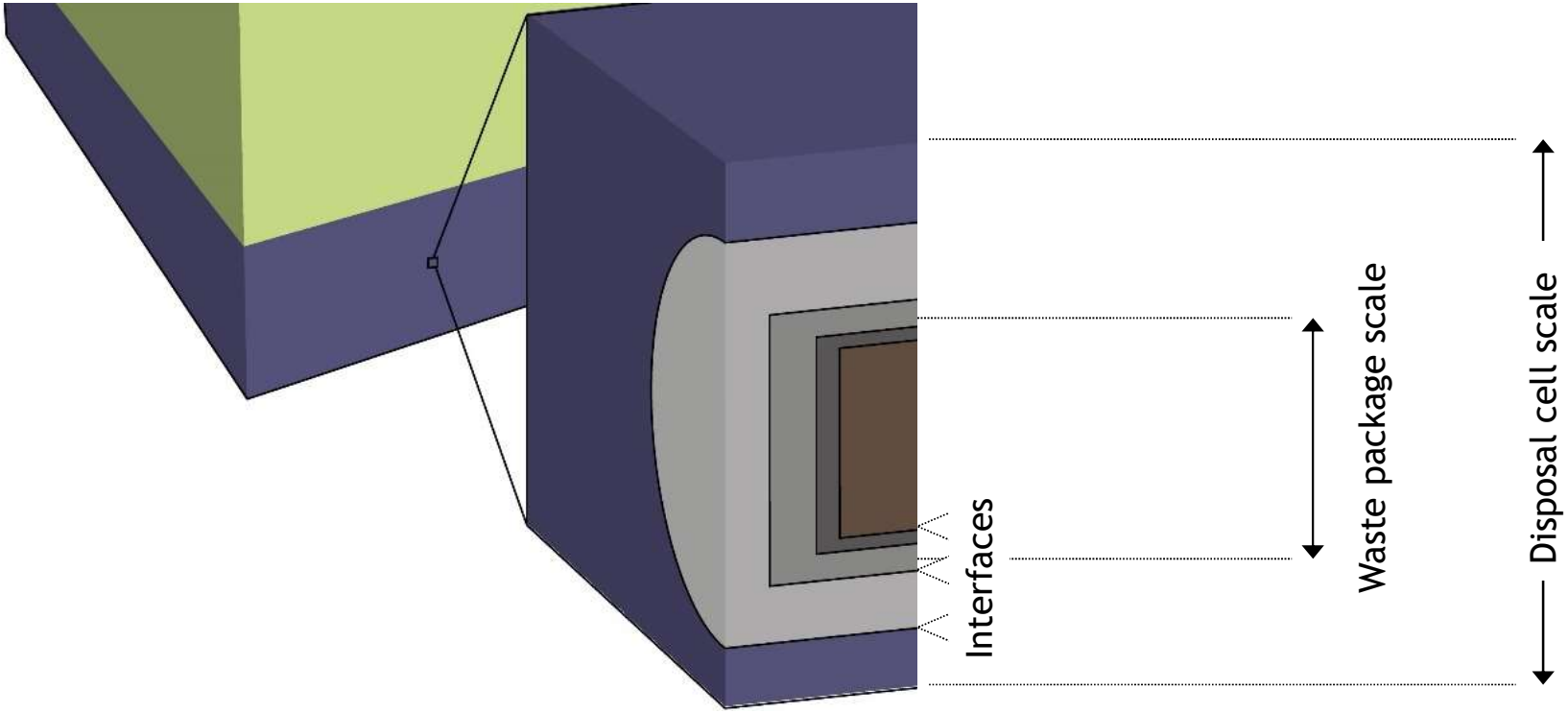
For HLW
Bentonite or Concrete

Steel

vitrified HLW

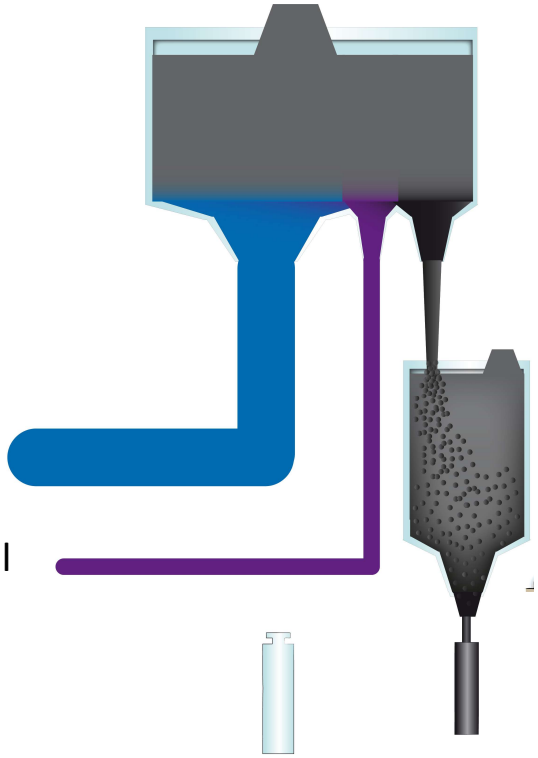
EURAD ACED

Assessment of Chemical Evolution of ILW and HLW Disposal Cells



VITRIFIED HLW

Separation of metallic parts from spent fuel assemblies



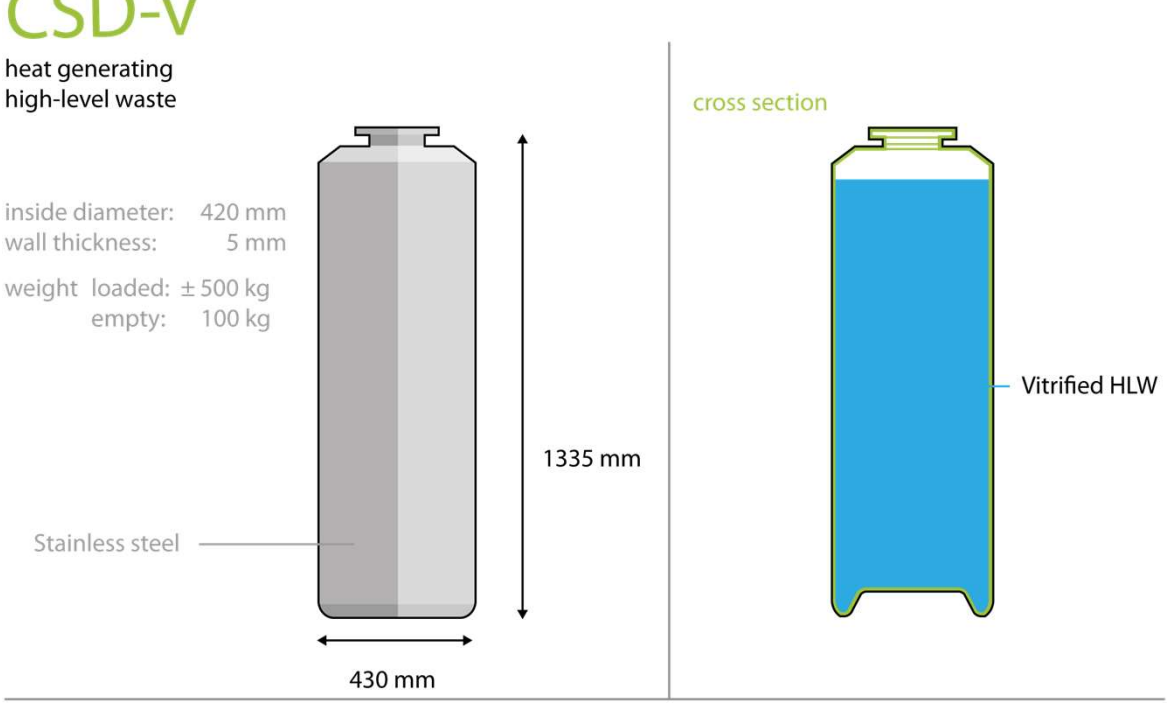
Extraction of uranium from dissolved spent fuel

Extraction of plutonium from dissolved spent fuel

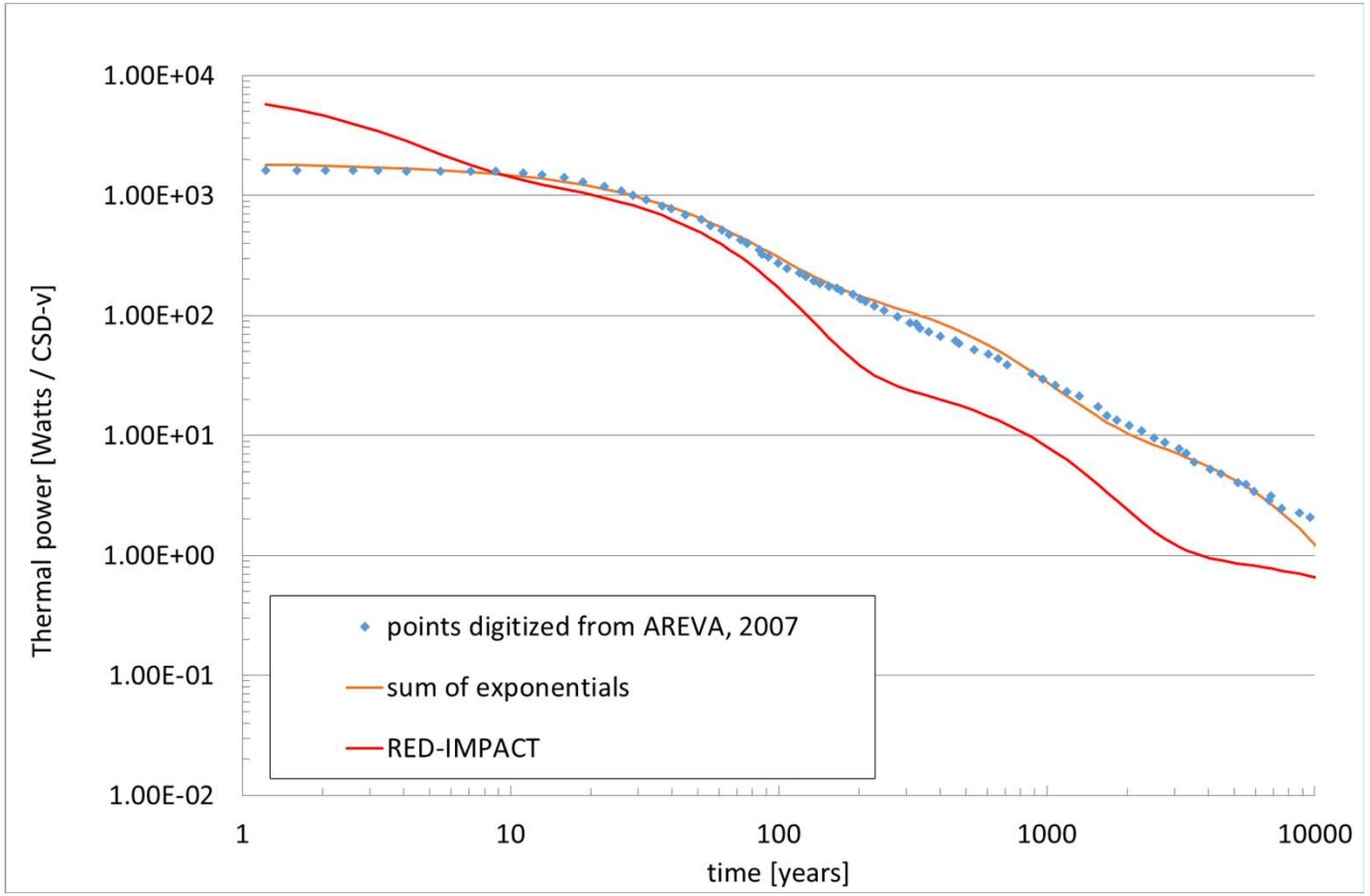
Other actinides than Pu & U
mixed with fission products
Immobilized in glass

VITRIFIED HLW

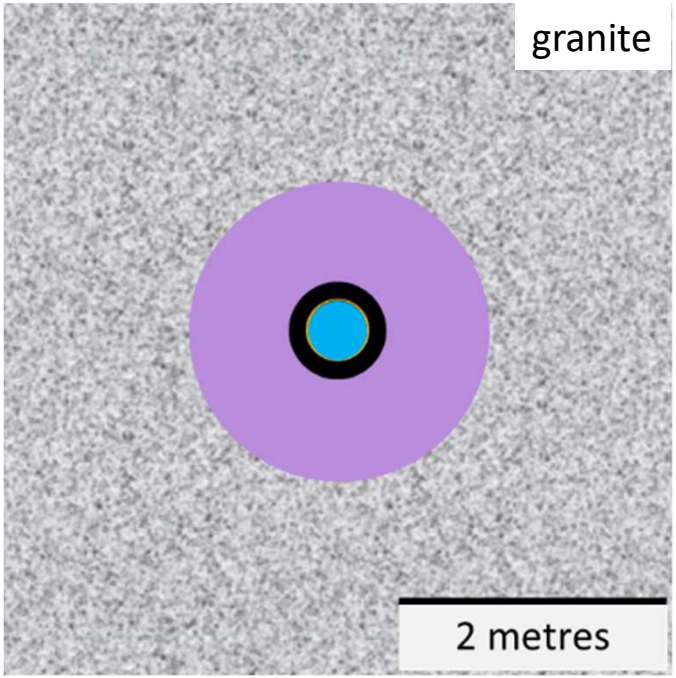
- **Only one waste processor in EU: France (past UK, Russian not yet known)**
  - **CSD-V: Colis Standard de Déchets - Vitrifiés**
    - **standardized waste characteristics with guaranteed properties**
    - **uniform waste package**

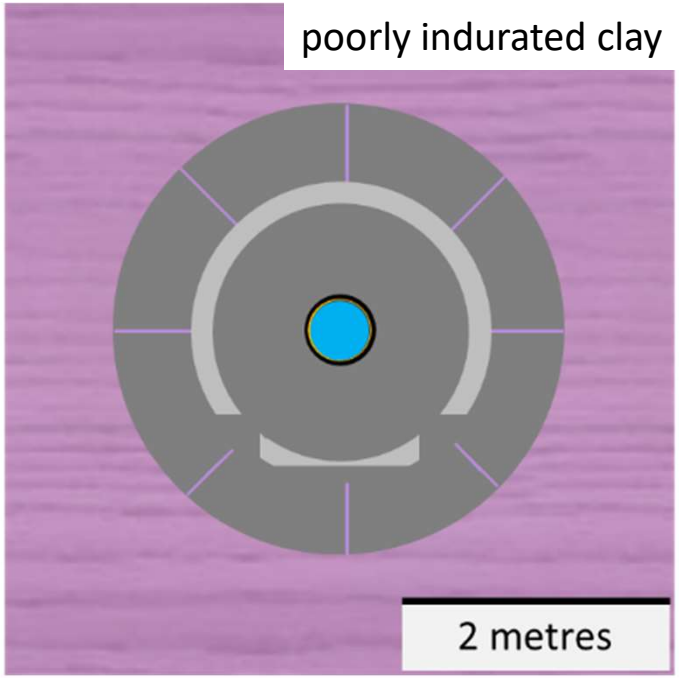


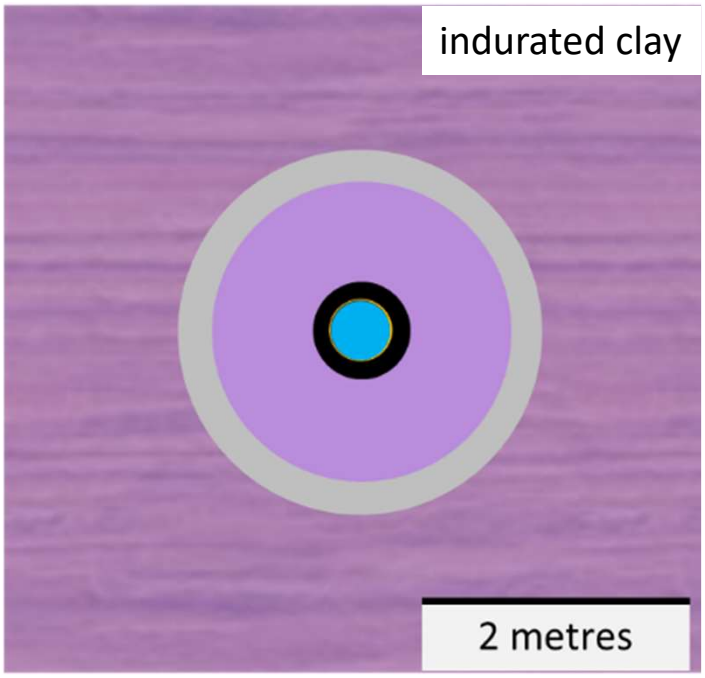
VITRIFIED HLW



VITRIFIED HLW IN DISPOSAL CELLS







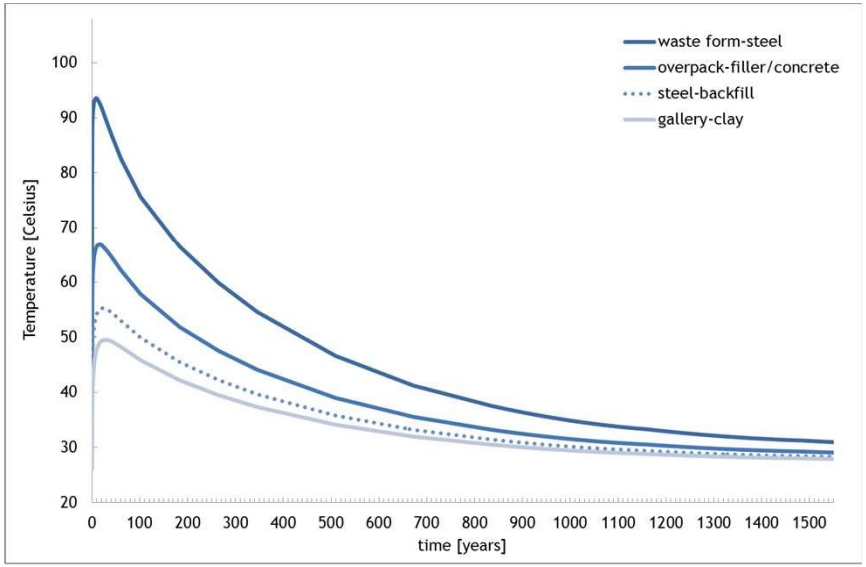
vitrified HLW steel bentonite concrete mortar

THERMAL EFFECT BENTONITE

- **NaNet (EURATOM FP5) available from natural-analogues.com**
  - Transformation of montmorillonite into illite, if sufficient supply of potassium
    - Isle of Skye in Scotland
    - Col du Perthus in France (Lodève, Herault)
    - Busachi in Sardinie
    - Kinnekulle in Sweden

THERMAL EFFECT BENTONITE

- Clay and granitic pore waters are deficient in potassium
- Concrete pore water may not be deficient in potassium



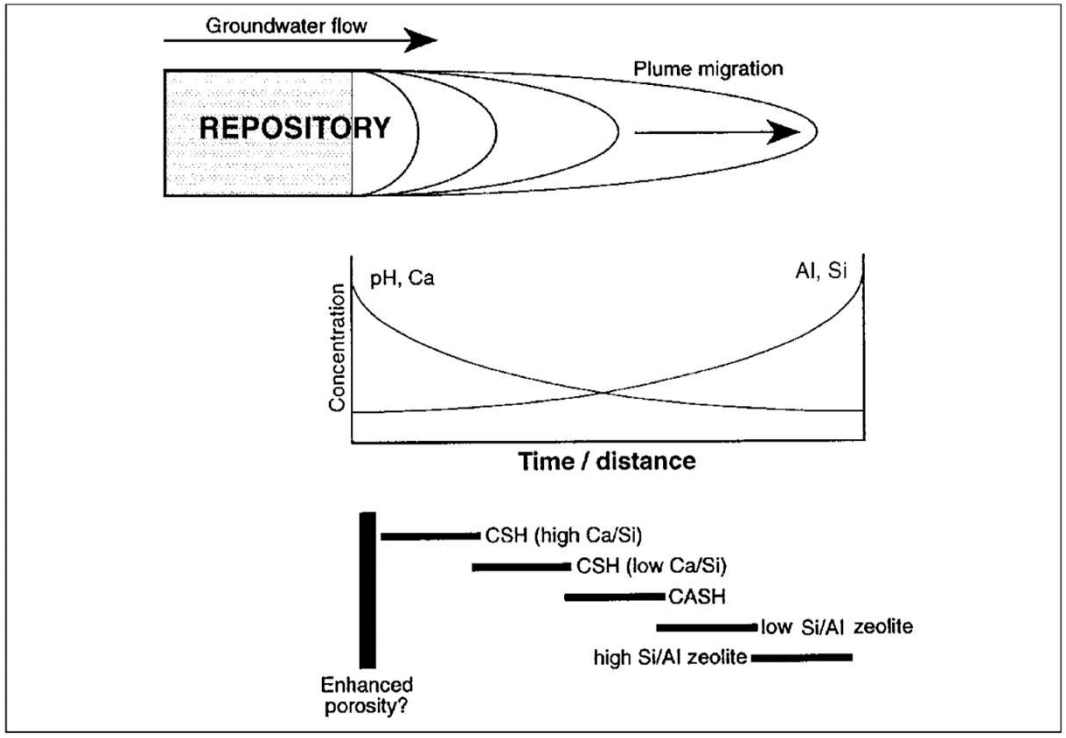
Modelled clay pore water EURAD ACED SOTA

Parameter	Unit	Belgian	Dutch	French	Swiss (reference)	Swiss (sea case)
Temperature	ºC	16	26	25	25	25
pH	-log(H^+)	8.5	6.9	7.28	7.203	7.009
pe	-log(e^-)	-4.7	-2.8	-2.64	-2.781	-2.563
pCO_2	log(bar)	-2.62	-1.5	-1.96	-2.2	-2.5
Na^+	mmol/kg	15.6	460.9	32.1	164.4	527.5
K^+	mmol/kg	0.2	9.8	7.10	2.604	1.77
Ca^{2+}	mmol/kg	0.05	13.2	15.0	12.51	49.71
Mg^{2+}	mr					
Sr^{2+}	mr					
Fe^{2+}	mr					
Al^{3+}	mr					
SiO_2 (aq)	mr					
Cl^-	mr					
SO_4^{2-}	mr					
HCO_3^-	mr					

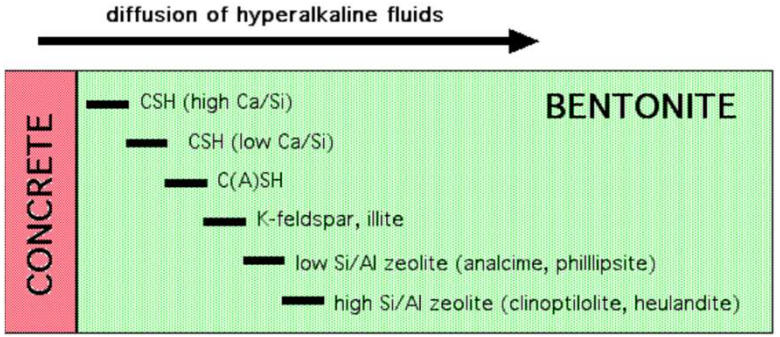
granitic pore water

Parameter	Unit	Czech (600 m)	Czech (1000 m)	Spanish	Swedish (512 m)	Finnish (446.0-558.5 m)
Temperature	ºC	25	25	30	11.35	11
pH	-log(H^+)	8.2	9.4	7.9	7.2	7.9
pe	-log(e^-)	+4.00	+4.00	-2.907	-2.54	-4.33
Na^+	mmol/kg	0.865	3.81	4.350	89	122
K^+	mmol/kg	0.0537	0.0179	0.05371	0.9	0.35
Ca^{2+}	mmol/kg	0.864	0.0324	0.1522	23	41.2
Mg^{2+}	mmol/kg	0.342	0.00412	0.1604	9.3	2.35
Sr^{2+}	mmol/kg					0.171
Fe^{2+}	mmol/kg	0.00179	0.00179	0.8953	0.033	0.00013
Al^{3+}	mmol/kg	0.00371	0.00371			0.002
SiO_2 (aq)	mmol/kg	0.520	0.481	0.3761		0.072
Cl^-	mmol/kg	0.0931	0.528	0.3949	153	214
SO_4^{2-}	mmol/kg	0.219	0.109	0.01561	5.2	0.114
HCO_3^-	mmol/kg	2.77	2.68	5.048	2.2	

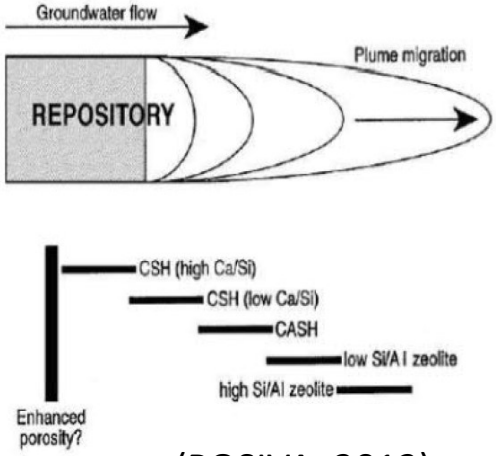
ALTERATION OF CONCRETE INTERFACING CLAY OR GRANITIC HOST ROCK



Savage, 1998

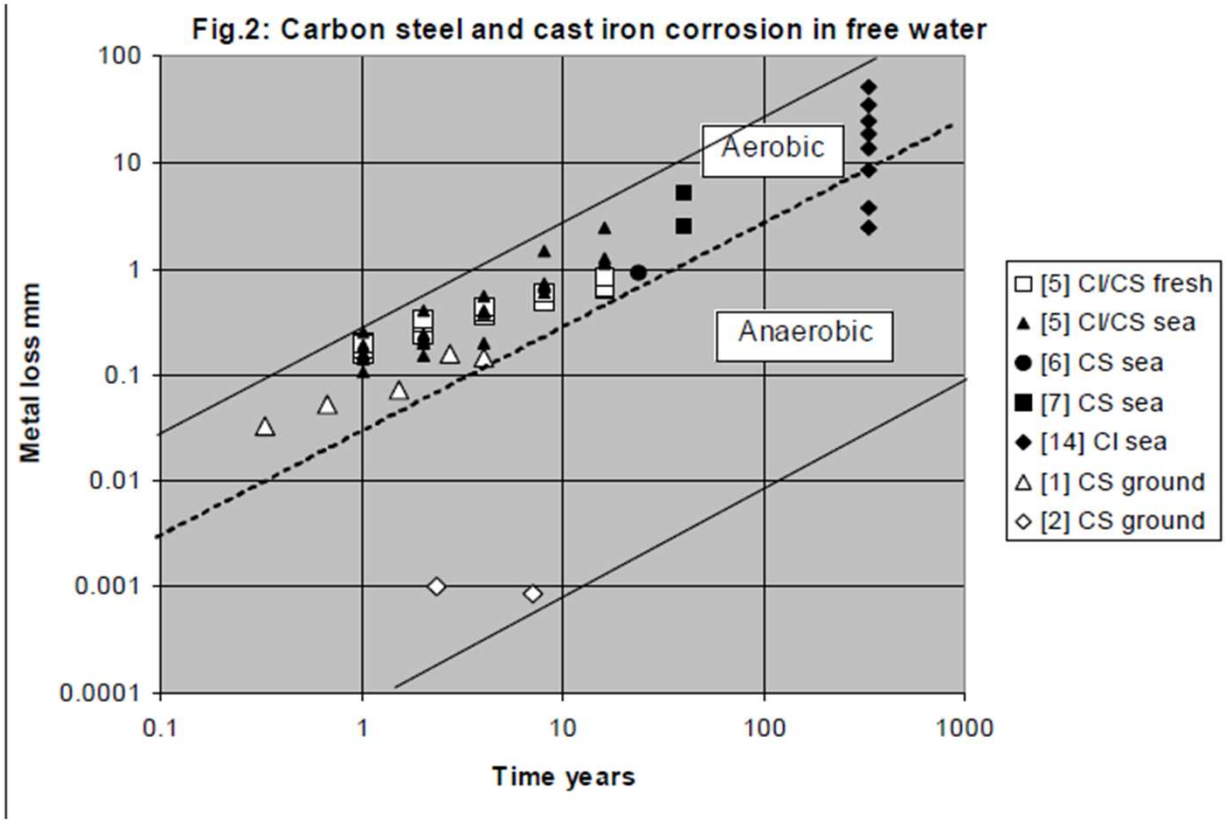


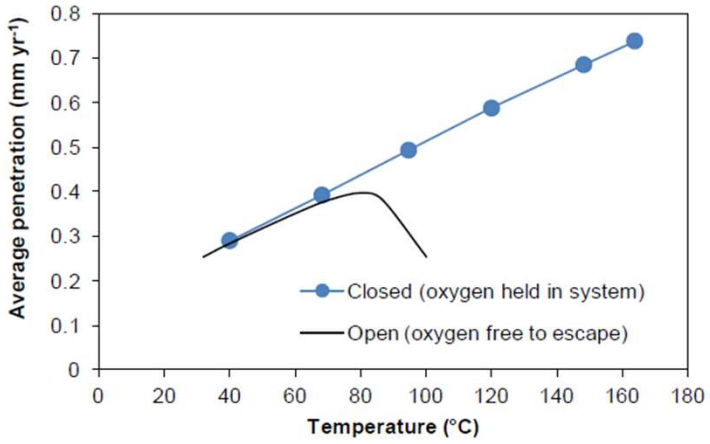
(Savage and Benbow, 2007; Bamforth et al., 2012).



Length in 'plume' may be less than 1 metre after 25.000 years (POSIVA, 2012)

INTERFACES WITH STEEL

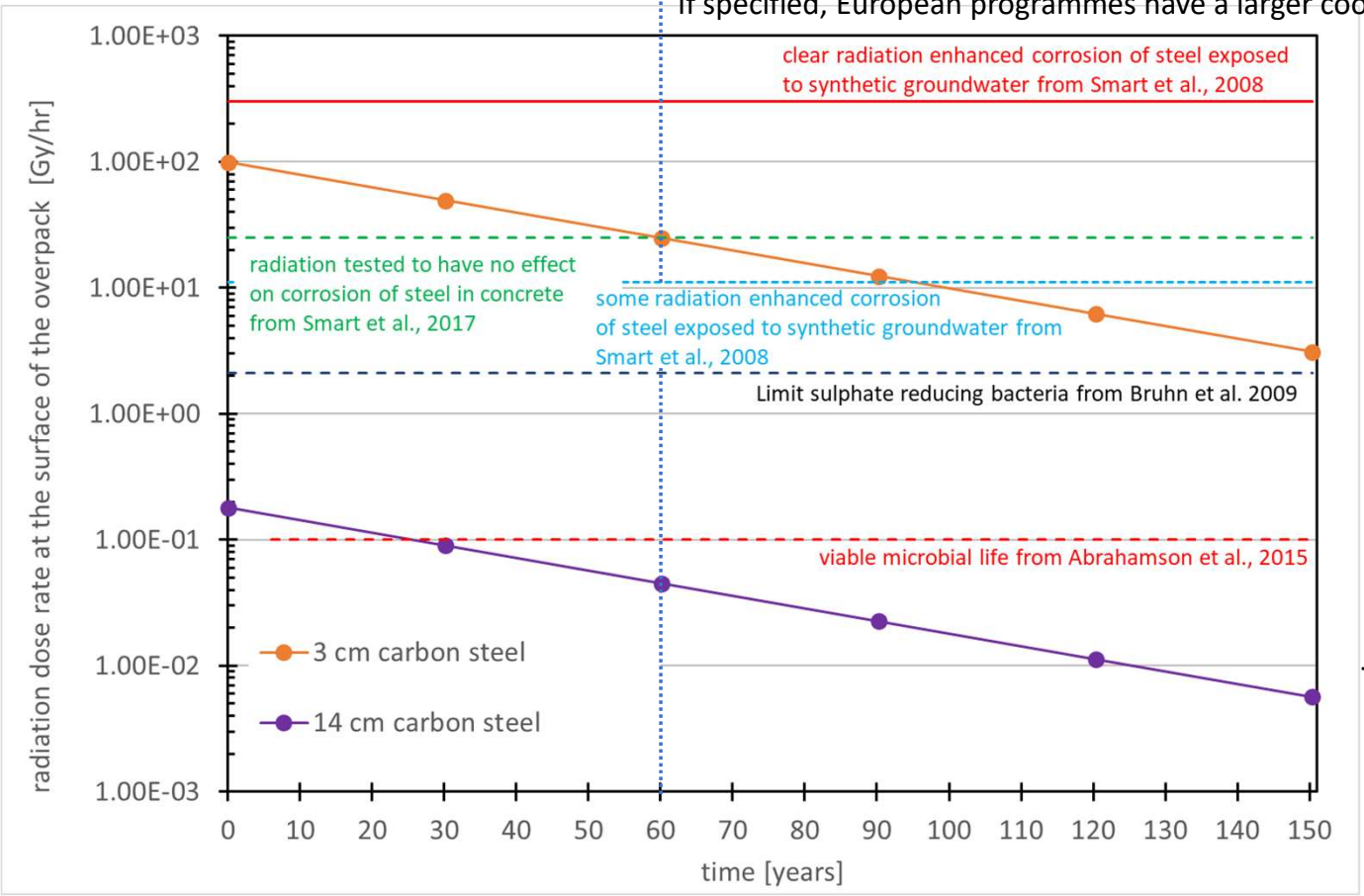




VITRIFIED HLW + STEEL (CANISTER & OVERPACK)

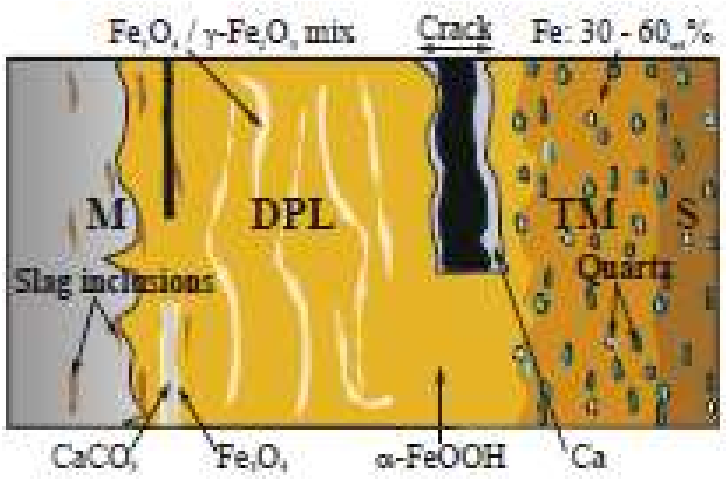
If specified, European programmes have a larger cooling period of 60 years

Max guaranteed
6600 ^{137}Cs (^{137m}Ba)



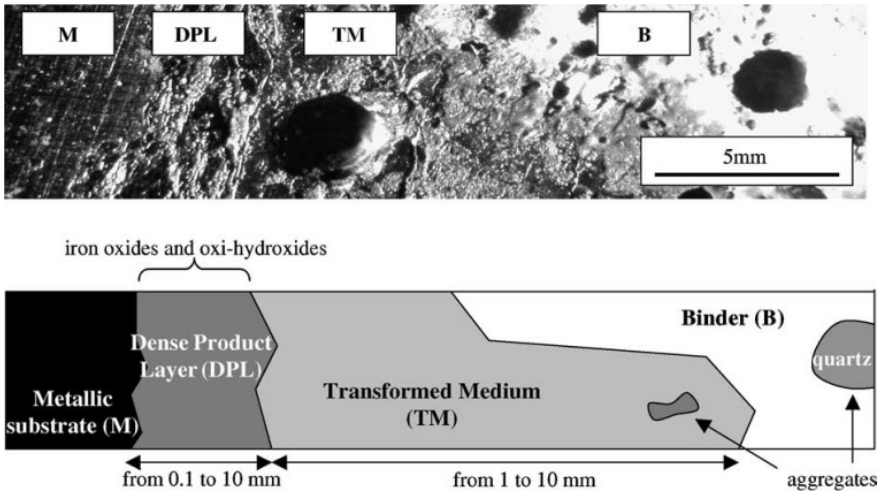
Buffer
Dunarobbe forest
Trees protected against microbial degradation

INTERFACES WITH STEEL



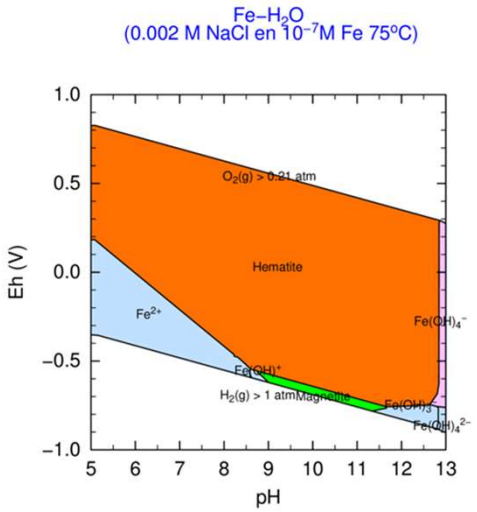
steel-soil: several hundred years old

but less focus on clay minerals in soils

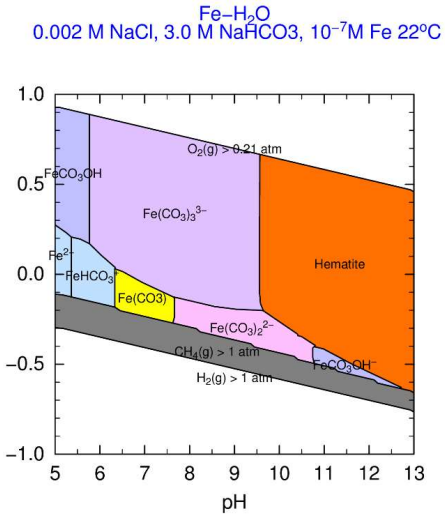


steel-binder: 350 years old

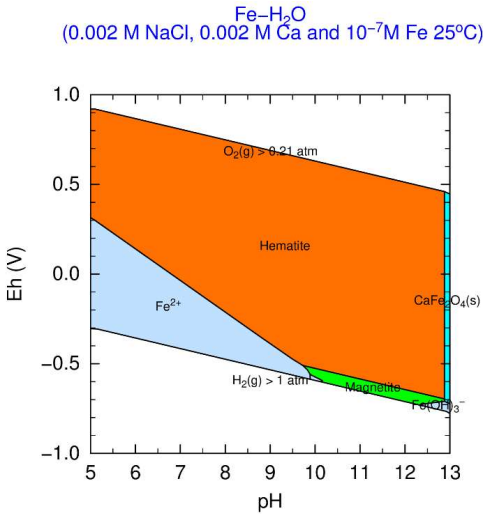
INTERFACES WITH STEEL



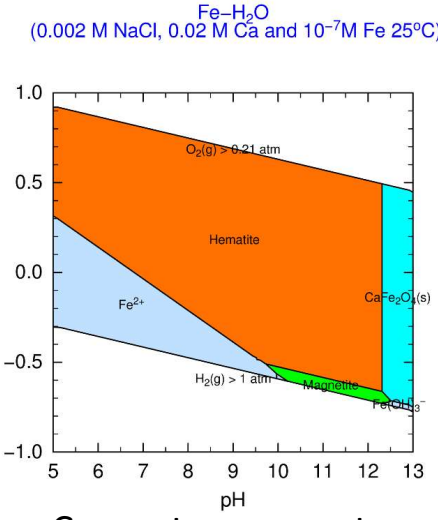
Tap water



Clay pore water



Tap water



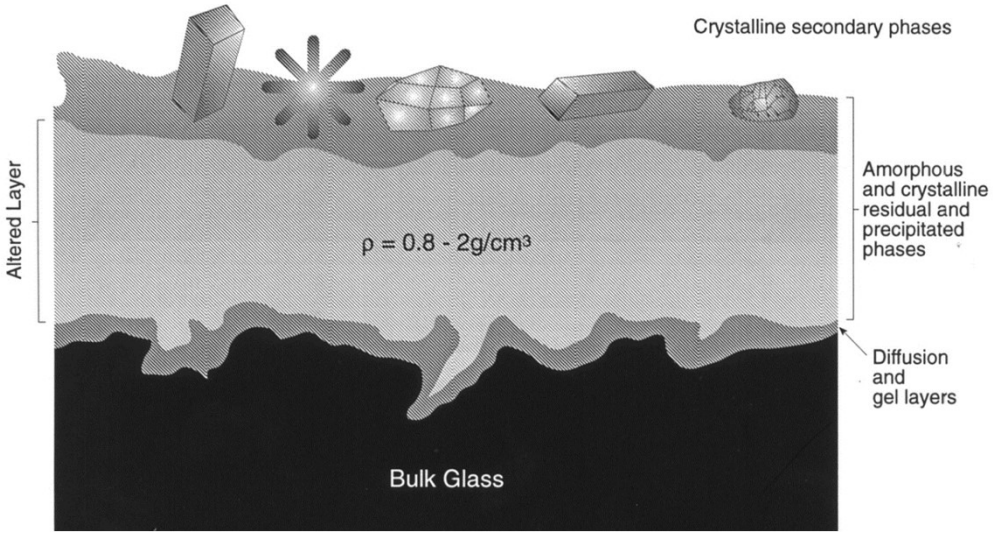
Concrete pore water

$$3Fe + 4H_2O \rightarrow Fe_3O_4 + 4\ H_2$$

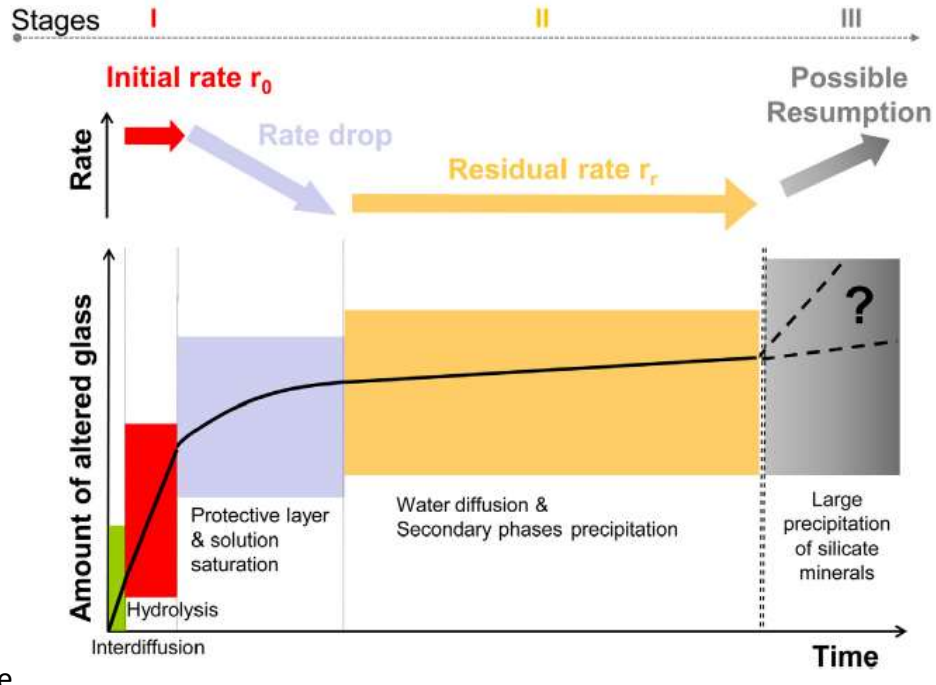
$$Fe\ (s) + H_2O \rightarrow Fe^{2+}\ (aq) + 2OH^-\ (aq) + H_2\ (g)$$

due to insufficient dissipation OH^-, magnetite formation

VITRIFIED HLW



from Lutz and Ewing (Havlova et al., 2007) (Milodowski et al., 2015



Gin et al., 2013

Alteration rates of basaltic glass, the natural analogue for a borosilicate waste form, have estimated to be 0.1 µm per 1000 years in silicon saturated environments (Lutze et al., 1987).

Of course some glass is interfacing steel but interfacing area negligible compared to the bulk

CONCLUSION

- **In ACED, representative disposal cells are studied**
- **Representativeness of the characteristics of disposal cells (pore water, radiation, thickness overpack) allows assessing the exclusion of processes**
  - Radiation enhanced steel corrosion is not a relevant process in the chemical evolution of European representative disposal cells with vitrified HLW
- **Natural and archeological analogues are used to identify the relevant processes for the chemical evolution of disposal cells**
  - The ‘old’ is good!

ANY QUESTIONS?

Thank you for attention

References used in this presentation are in the next slides

REFERENCES USED IN THIS PRESENTATION

Abrahamsen, L., Arnold, T., Brinkmann, H., Leys, N., Merroun, M., Mijnendonckx, K., Moll, H., Polvika, P., Ševců, A., Small, J., Vikman, M. and Wouters, K. 2015 A review of anthropogenic organic wastes and their degradation behaviour - Deliverable 1.1 MIND

AREVA (2007). Specification for standard vitrified waste residue (CSD-v) with high actinide content produced at la Hague AREVA (ed), p. 23, (Confidential document not published openly).

Bamforth, P.B., Baston, G.M.N., Berry, J.A., Glasser, F.P., Heath, T.G., Jackson, C.P, Savage, D., Swanton, S.W. (2012). Cement materials for use as backfill, sealing and structural materials in geological disposal concepts. A review of current status. Serco Report SERCO/005125/001 Issue 3, Serco, Harwell, Didcot, United Kingdom.

Bruhn, D.F., Frank, S.M., Roberto, F.F., Pinhero, P.J. and Johnson, S.G. (2009). Microbial film growth on irradiated, spent nuclear fuel cladding. Journal of Nuclear Materials 384(2), 140-145

Chapman, N.A., Hooper A. (2012). The disposal of radioactive wastes underground, Proceedings of the Geologists' Association 123 (1), 46-63.

Chitty, W.-J., Dillmann, P., L'Hostis, V. and Lombard, C. (2005). Long-term corrosion resistance of metallic reinforcements in concrete—a study of corrosion mechanisms based on archaeological artefacts. Corrosion Science 47, 1555–1581

Crossland, I. (2005). Long-term corrosion of iron and copper, The 10th International Conference on Environmental Remediation and Radioactive Waste Management, Glasgow (Scotland), ICEM05-1272, ASME.

Dunarobbe forest: Lombardi, S. and Valentini, G. (1996). The Dunarobbe forest as natural analogue: analysis of the geoenvironmental factors controlling the wood preservation, pp. 127-133, Sixth EC Natural analogue working group meeting Proceedings of an international workshop held in Santa Fe, New Mexico, USA. EUR 16761
De Putter, T., Grainger, P., Lombardi, S., Manfroy, P. and Valentini, G. (1997). Fourth European Conference on Management and Disposal of Radioactive waste, pp. 609-626, EUR 17543. Luxembourg
Hooker, P. (2003). Dunarobba, http://www.natural-analogues.com/nawg-library/na-overviews/analogue review

REFERENCES USED IN THIS PRESENTATION

Gin, S., Abdelouas, A., Criscenti, L.J., Ebert, W.L., Ferrand, K., Geisler, T., Harrison, M.T., Inagaki, Y., Mitsui, S., Mueller, K.T., Marra, J.C., Pantano, C.G., Pierce, E.M., Ryan, J.V., Schofield, J.M., Steefel, C.I., Vienna, J.D. (2013). An international initiative on long-term behaviour of high-level nuclear waste glass. Materials Today 16, 243-248

Greneche, D., Quiniou, B., Boucher, L., Delpech, M., Gonzalez, E., Alvarez, F., Cunado, M.A., Serrano, G., Cormnezana, J.L., Kuckshinrichs, W., Odoj, R., von Lensa, W., Wallenius, J., Westlén, D., Zimmerman, C. and Marivoet, J. (2007) **RED-IMPACT** Impact of Partitioning, Transmutation and Waste Reduction Technologies on the Final Nuclear Waste Disposal - Synthesis report, Schriften des Forschungszentrums Jülich - Reihe Energie & Umwelt / Energy & Environment Band - Band / Volume 15

Havlova, V., Knight, L., Laciok, A., Cervinka, R. and Vokal, A. (2007). Analogue evidence relevant to UK HLW glass waste forms p. 37, NIREX 509009 made by Nuclear Research Institute Řež; natural-analogues.com/nawg-library

Lutze, V., Grambow, B., Ewing, R.C. and Jercinovic, M.J. (1987). Natural analogues in radioactive waste disposal. Côme, B. and Chapman, N.A. (eds), Commission of the European Communities - Radioactive Waste Management Series EUR 11037

Milodowski, A.E., Alexander, W.R., West, J.M., Shaw, R.P., McEvoy, F.M., Scheidegger, J.M., Field, L.P. (2015). A catalogue of analogues for radioactive waste management. British Geological Survey Commissioned Report CR/15/106, British Geologcial Survey, Keyworth, Nottingham, United Kingdom

Neeft E., Jacques D., Deissmann G., (2022): Initial State of the Art on the assessment of the chemical evolution of ILW and HLW disposal cells. D 2.1 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593.

Neeft, E., Weetjens E., Vokal A., Leivo M., Cochepin B., Martin C., Munier I., Deissmann G., Montoya V., Poskas P., Grigaliuniene D., Narkuniene A., García E., Samper J., Montenegro L., Mon A. (2019): Treatment of chemical evolution in National Programmes, D 2.4 of the HORIZON 2020 project EURAD. EC Grant agreement no: 847593

Neff, D., Reguer, S., Bellot-Gurlet, L., Dillmann, P. and R., B. (2004). Structural characterisation of corrosion products on archaeological iron. An integrated analytical approach to establish corrosion forms. Journal of Raman Spectroscopy 35(8-9), 739-745.

POSIVA (2012). Safety case for the disposal of spent nuclear fuel at Olkiluoto - Features, events and processes 2012. Posiva Report 2012-07, Posiva, Olkiluoto, Finland

REFERENCES USED IN THIS PRESENTATION

Savage, D. (1998). Maqarin natural analogue study: Phase III. Smellie, J.A.T. (ed), pp. 281-316, SKB Technical report TR-98-04.

Savage, D., Benbow, S. (2007). Low pH cement. SKI Report 2007:32, Swedish Nuclear Power Inspectorate, Stockholm, Sweden

Smart, N.R., Rancea, A.P. and Wermeb, L.O. (2008). The effect of radiation on the anaerobic corrosion of steel. Journal of Nuclear Materials 379(1-3), 97-104

Smart, N.R., Rance, A.P., Nixon, D.J., Fennell, P.A.H., Reddy, B. and Kursten, B. (2017). Summary of studies on the anaerobic corrosion of carbon steel in alkaline media in support of the Belgian supercontainer. Corrosion Engineering, Science and Technology 52, 217-226.

Swanton, S.W., Baston, G.M.N. and Smart, N.R. (2015). Rates of steel corrosion and carbon-14 release from irradiated steels – state of the art review D2.1 from CArbon-14 Source Term project from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779.

Trotignon, L., Techer, I. and Kohler, E. (2004). Col du Perthus, http://www.natural-analogues.com/nawg-library/na-overviews/analogue review.

Wouters, L. and Verheyen, A. (2004). Isle of Skye, http://www.natural-analogues.com/nawg-library/na-overviews/analogue review.

Wouters, L., Verheyen, A. and Dalton, J. (2005). Busachi (Sardinia), http://www.natural-analogues.com/nawg-library/na-overviews/analogue review.

Wouters, L., Verheyen, A. and Dalton, J. (2005). Kinnekulle, http://www.natural-analogues.com/nawg-library/na-overviews/analogue review.