Effect of high saline alkaline conditions onto radionuclide transport in a CEM V/A hardened cement paste

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Context and Objectives

This study is a cooperative work between CEA and Subatech, in the framework of the CTEC laboratory group project managed by Andra. The main objective is the **determination of transport parameters** of radionuclides and toxic species (**HTO, Cs^+**, Ni^{2+}, Cl^-, SeO_3^{2-}) **in Hardened Cement Paste** (HCP) as a function of ionic strength from 0.2 to 4.3 mol/$kg_{solution}$ reached by using **$NaNO_3$, Na_2SO_4 or a mixture $NaNO_3$/Na_2SO_4** added to an Artificial Cement poreWater (ACW).

Results and Modelling for HTO and Cs-137 diffusion experiments

HCP is prepared with CEM V/A (S-V)42.5N, Rombas, Calcia ; water:cement = 0.40; curing time in ACW = 6m minimum
ACW is prepared with **79 mM Na^+, 291 mM K^+, 0.6 mM Cl^-, 1 mM SO_4^{2-} + excess CaO (pH = 13.5)**
HTO and Cs-137 are measured by liquid scintillation counting in dual mode after regular samplings of the solution in both reservoirs (In-Diff + T-diff)
Cs-137 activities are measured by gamma-counter after HCP-post-diffusion-micro-grinding (In-Diff)

Diffusion model

Fick second law equation

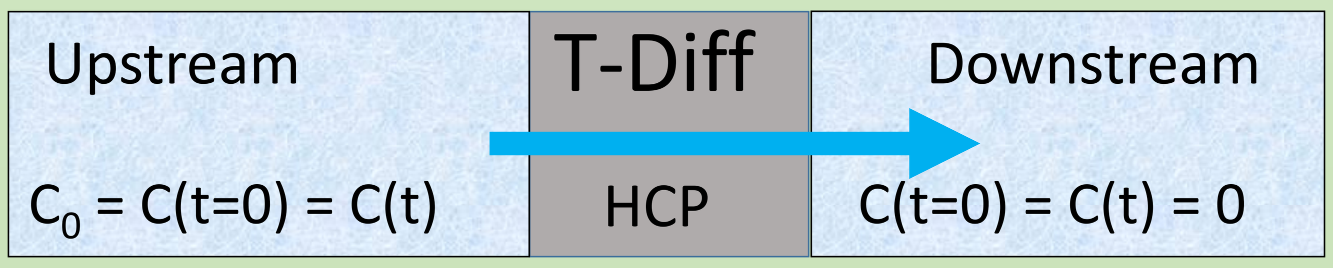
$$\frac{\partial C}{\partial t} = D_a \frac{\partial^2 C}{\partial x^2} = \frac{D_e}{\alpha} \frac{\partial^2 C}{\partial x^2}$$

D_a: apparent diffusion coefficient [m²/s]
D_e: effective diffusion coefficient [m²/s]
α: material capacity factor [-]

T-diff: through diffusion experiment / **In-diff**: in-diffusion experiment

Diffusion experiments carried out at Subatech

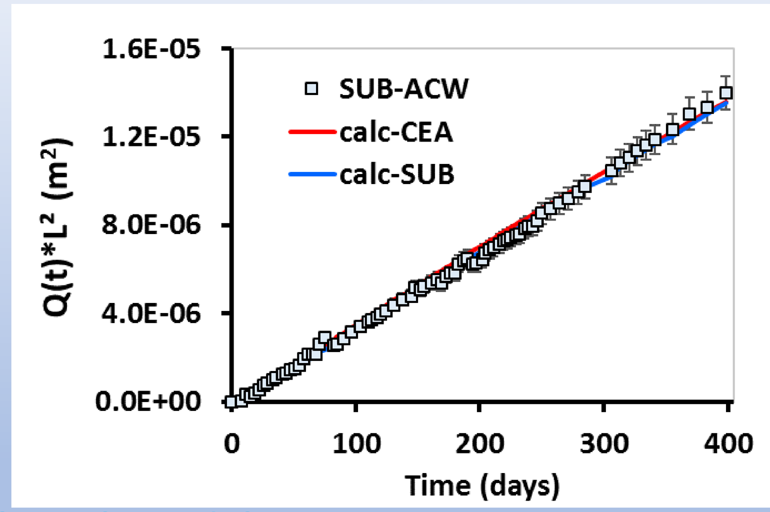
	$NaNO_3$ added to ACW [mol/$kg_{solution}$]	Na_2SO_4 added to ACW [mol/$kg_{solution}$]	HTO	Cs-137 + 1mM CsCl
SUB-ACW	-	-	T-diff	T-diff
SUB-MIX1	-	0.3	T-diff	T-diff



Subatech model: least-square fitting on Crank's analytical solution

a) HTO in SUB-ACW

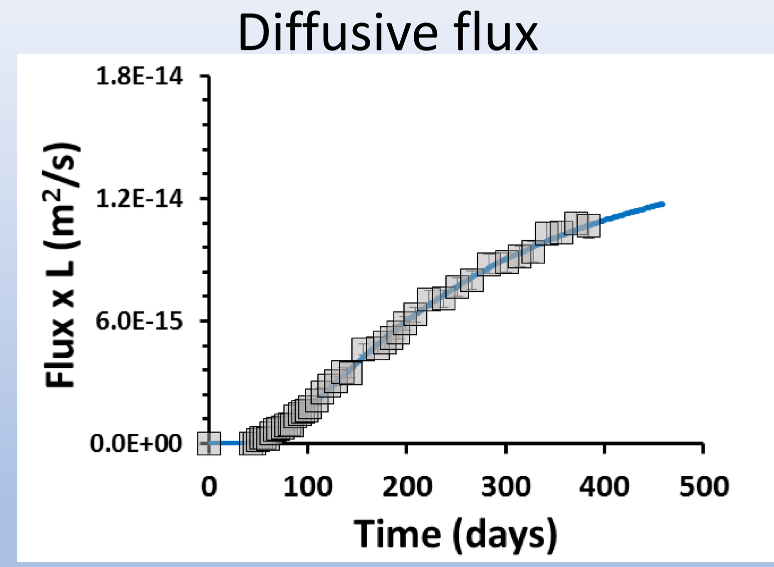
Cumulative activity in downstream



Subatech model: D_e = 4.1 10^{-13} m²/s; α = 0.33
CEA model: D_e = 4.4 10^{-13} m²/s; α = 0.35

c) Cs-137 in SUB-MIX1

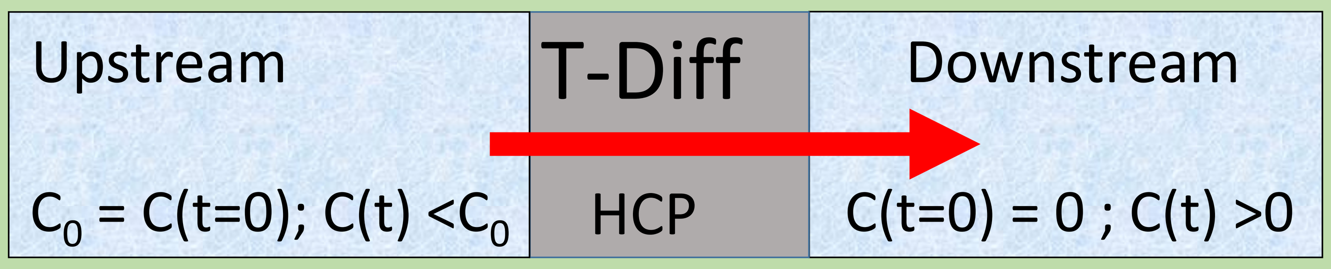
Diffusive flux

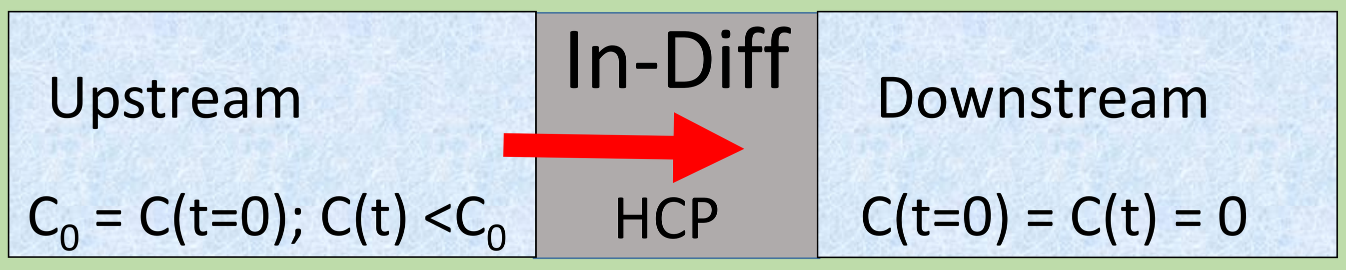


Subatech model: D_e = 1.8 10^{-14} m²/s; α = 0.35

Diffusion experiments carried out at CEA

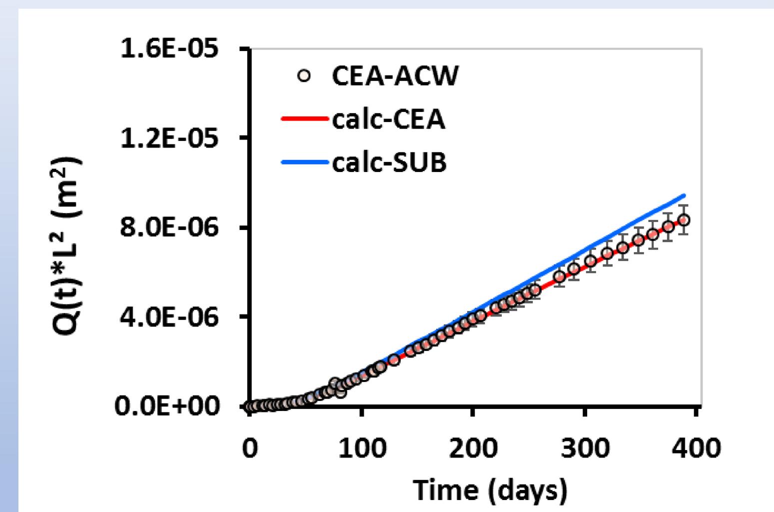
	$NaNO_3$ added to ACW [mol/$kg_{solution}$]	Na_2SO_4 added to ACW [mol/$kg_{solution}$]	HTO	Cs-137 + 1mM CsCl
CEA-ACW	-	-	T-diff	T-diff
CEA-MIX1	1.4	-	T-diff	In-diff





b) HTO in CEA-ACW

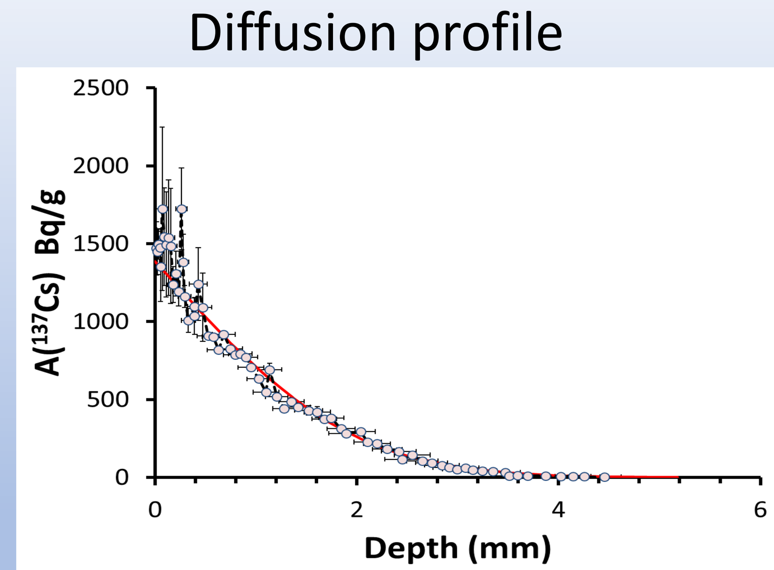
Cumulative activity in downstream



Subatech model: D_e = 3.2 10^{-13} m²/s; α = 0.34
CEA model: D_e = 3.0 10^{-13} m²/s; α = 0.33

d) Cs-137 in CEA-MIX1

Diffusion profile



CEA model: D_e = 1.2 10^{-14} m²/s; α = 0.55

CEA model: numerical tool *I-Mode* (Interpretation Model of Diffusion Experiment) based on a Laplace-transform analytic-element (Moridis (1999), Furman and Neuman (2003), Didierjean *et al.* (2004)).

Conclusions

HTO diffusion: For most of the experiments, HTO steady-state diffusive flux is reached after around 200 days. Benchmarking exercises for the reference sample in ACW give similar results.
Whatever the salt mixture and considering the measurements uncertainties, the mean value of D_a(HTO) is (8.6 ± 2.7).10^{-13} m²/s.
Cesium diffusion: Cs experiments are still on-going due to a very low diffusivity compared to that obtained for HTO. All experiments confirm that Cs migrates slower than HTO, then considered as conservative for cesium transfer.

Finally, no significant effect of salt could have been evidenced yet on HTO and Cs diffusion parameters.

References

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