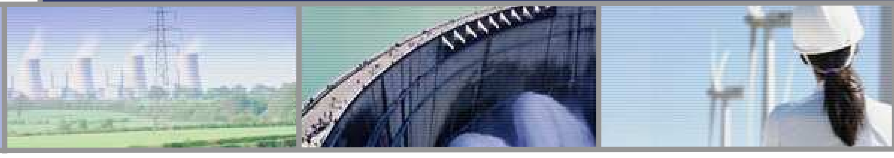
PAUL SCHERRER INSTITUT

NUCLEAR ENERGY AND SAFETY



Dynamics of ion diffusion in clays: a proposed probe - probe XPCS experiment

S. V. Churakov[1], R. Dähn[1], A. Froideval[1], B. Pedrini[2]

[1] Nuclear Energy and Safety (NES), Paul Scherrer Institute, 5232 Villigen PSI, Switzerland

[2] SwissFEL Project, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland

Context of the study

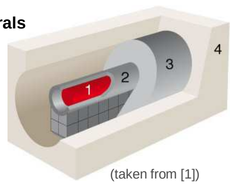
- Deep geological repositories: safe long-term containment of the radioactive waste

- Safety barriers in a repository for high-level waste:

engineered barriers:
1. glass matrix containing radioactive material
2. steel container
3. backfill with bentonite

geological barrier:
4. host rock

3., 4. = **clay minerals**



(taken from [1])

Main characteristics of the clay barrier:

- Dimensions:
- **bentonite:** 70 cm thick = montmorillonite (75 %) + quartz / calcite
- **host rock:** 100 m thick

- Low hydraulic conductivity: 2×10^{-14} m/s

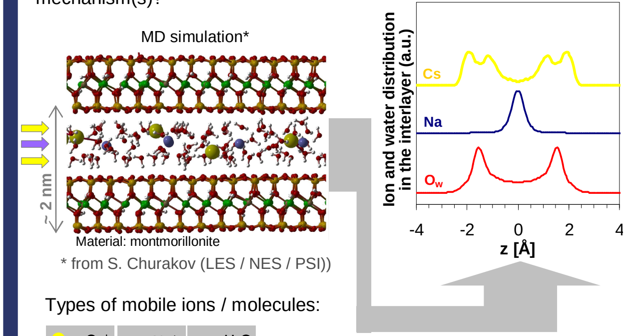
→ mass transport by diffusion

Motivation of the study

Understanding of:

- the ion diffusion (Na^+, Cs^+) mechanisms – timescale: ~ 10 - 100 ps – in the interlayer of clay minerals in order to increase our confidence in numerical models.

Is the ionic transport in the interlayer governed by non-Brownian diffusion mechanism(s)?



* from S. Churakov (LES / NES / PSI))

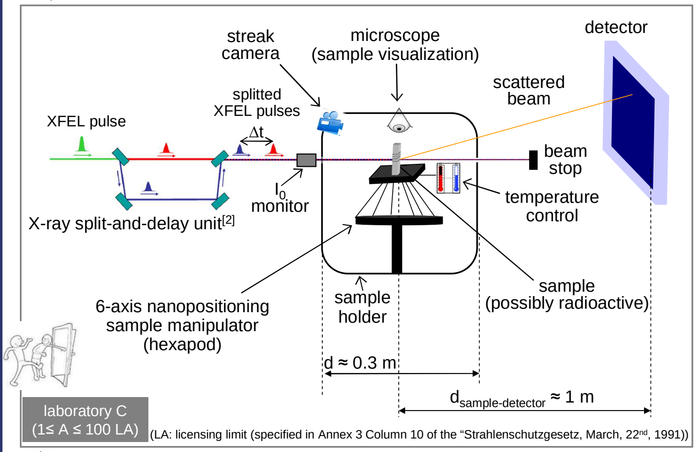
Types of mobile ions / molecules:

Cs^+ Na^+ H_2O

Proposed experiment

▪ Sketch of the experiment

- Proposed **PROBE-PROBE** X-ray photon correlation spectroscopy (XPCS) experiment:



(LA: licensing limit (specified in Annex 3 Column 10 of the "Strahlenschutzgesetz, March, 22nd, 1991))

▪ Expected results

- *in situ* measurements of the ionic mobility in the interlayer of clay minerals;
- ion transport mechanism(s) in the interlayer space, namely Brownian versus non-Brownian diffusion mechanisms.

→
- experimental validations of MD simulations
- scientific basis for performance assessments of the clay barrier in the deep geological disposal of radioactive waste.

▪ Key parameters of the experiment

Parameter		Unit	Requirement	Motivation / Remarks
▪ Key parameters of the X-ray FEL radiation				
Energy		keV	> 10.0	required by X-ray split-and-delay unit
	stability	%	< 0.05	required by X-ray split-and-delay unit
Bandwidth		%	0.1 / 0.01	1 nm / 1 Å resolution
	stability	%	< 10	stable incoming beam is wished
Position	stability		- - -	
Size		µm	~ 1	microfocus
Photons / pulse		#ph	~ 10^{11}	taking maximal fluence of 0.5 mJ/µm² and assuming 1% transmission of the split-and-delay unit
	stability	%	< 10	data to be normalized with I_0
Pulse length		fs	< ~ 200	mechanisms of ion diffusion on (few) tens ps timescale
	stability	%	10	
Pulse arrival time			- - -	
▪ Beam parameter changes during experiment				
Energy	range / step	eV		variable photon energy desirable (for resonance studies) e.g. at Na K-edge and at Cs L_{III}-edge; in the future: additional studies foreseen at L_{III}-edges of actinides (up to 20 keV)
	rate	%	- - -	
Size	range / step	µm	- - -	
	rate	µm/min	- - -	
Pulse length	range/step		100 - 500 fs	test at ≠ values for consistency check
	rate		- - -	no scanning
▪ Beam geometry				
Slope	max.	µrad	- - -	
Working distance	min.	mm	~ 300	space for sample holder
▪ Other				
Delay line			yes	split-pulse XPCS (delay: up to 3 ns with hard X-rays)
Transverse coherence			yes	required for coherent X-ray diffraction
Laboratory C			yes	for similar future studies on radionuclides (1 ≤ A ≤ 100 LA, with LA: licensing limit)

References

[1] from http://www.nagra.ch/, NAGRA: National Cooperative for the Disposal of Radioactive Waste.
[2] G. Grübel et al., Nucl. Instr. and Meth. in Phys. Res. B, 262 (2007) 357-367.

Acknowledgments

The authors wish to thank B. Patterson, E. Wieland from PSI, and G. Grübel from DESY for helpful discussions and interest in the work.