



21ST EXCHANGE MEETING

Cement materials for radioactive waste management: new insights and current RD&D programme

Monday 29 May 2017

13:30 Welcome

13:40 Introduction: Cement chemistry and cementitious materials

Seif Ben Hadj Hassine and Erik Coppens (ONDRAF/NIRAS)

14:10 Conditioning of radioactive waste by cementation

Erik Coppens and Seif Ben Hadj Hassine (ONDRAF/NIRAS)

14:40 Long-term durability of cementitious materials: degradation mechanisms

Phung Quoc Tri (SCK•CEN)

15:10 Coffee break

15:30 Modelling of concrete structures: the role of cracks

Janez Perko (SCK•CEN)

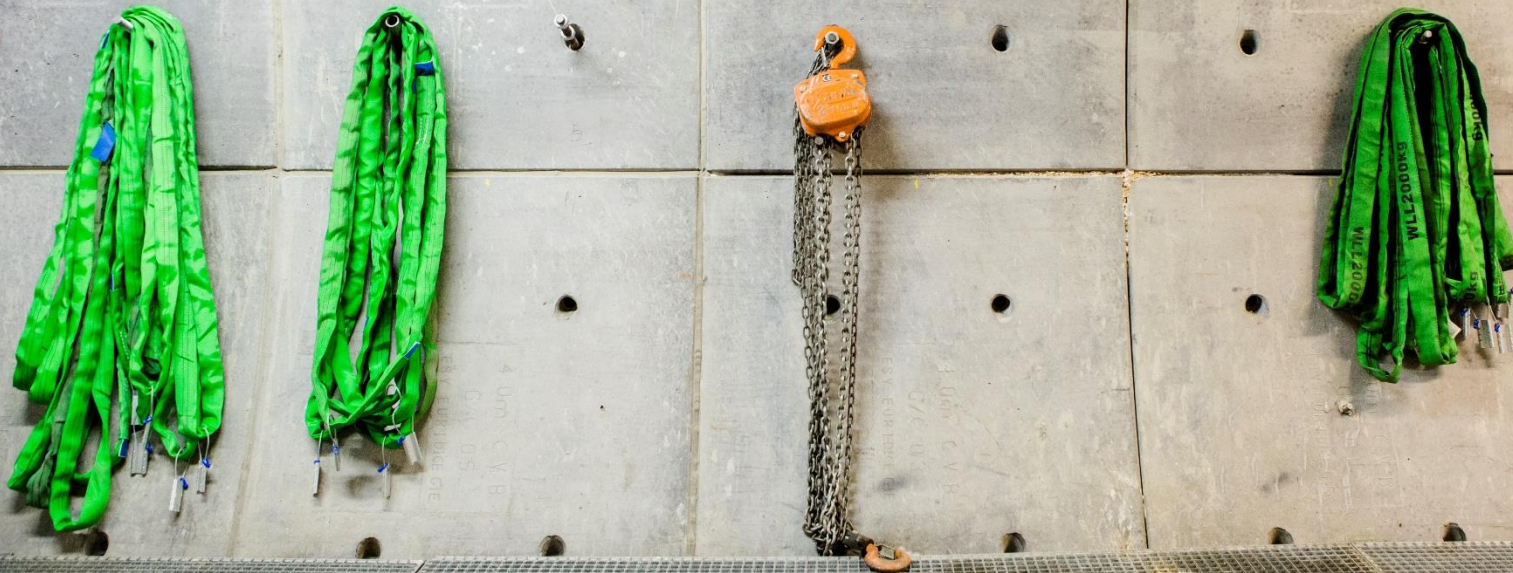
16:00 RD&D priorities on cement within the Belgian National Programme

Xavier Sillen (ONDRAF/NIRAS)

16.30 Conclusions

Closure drink

Cement materials for radioactive waste management: *New insights and current RD&D programme*



21st Exchange Meeting

29 May 2017 - Mol





ONDRAF/NIRAS

Cement chemistry and cementitious materials

**Seif Ben Hadj Hassine
Erik Coppens**

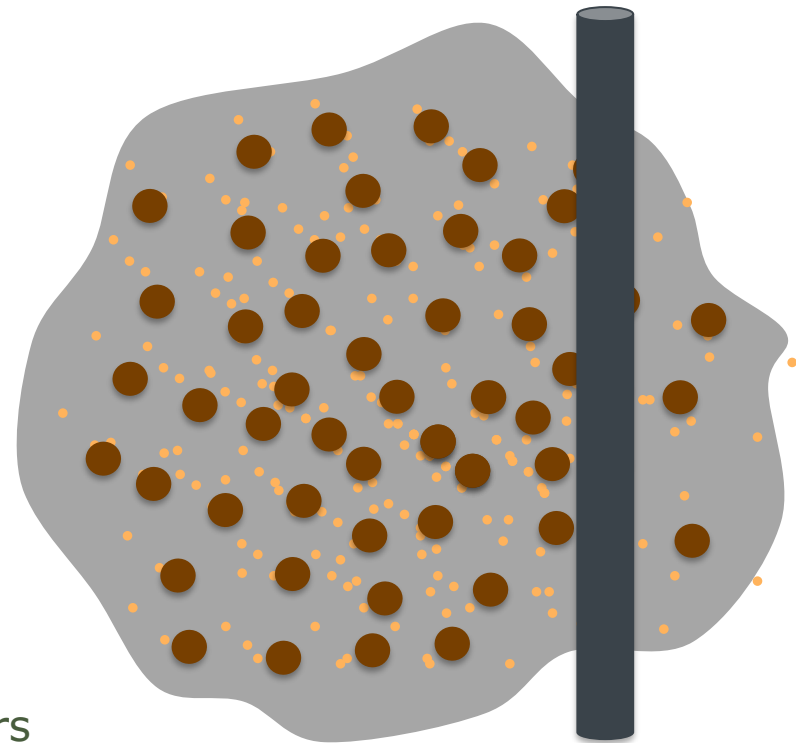


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- **Cement-based materials: what, where?**
 - A reminder of what the concrete ingredients are
 - cAt surface repository design
 - B&C underground repository design
- **General introduction to cement chemistry**
 - Introduction to cements
 - Cement hydration mechanisms and reactions
 - Characterization of cement-based materials
- **Examples of highlighted phenomenon**
 - Delayed Ettringite Formation (DEF)

Concrete materials

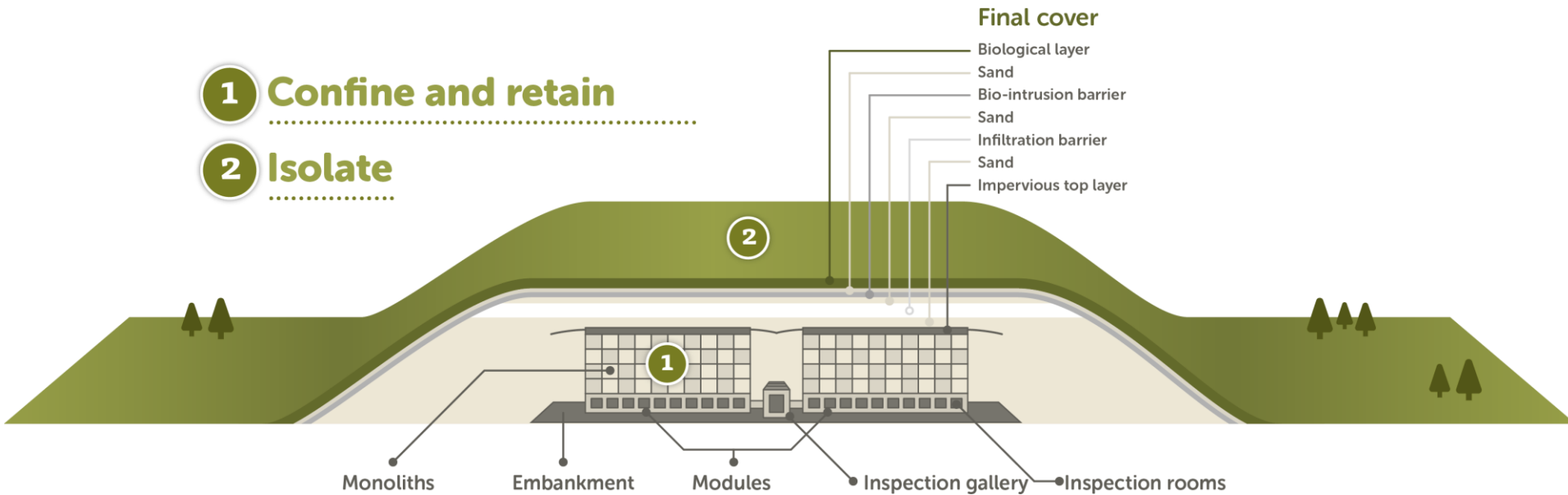
- **Cement-based materials are appropriate for the EBS**
 - Good mechanical performances
 - Acts as chemical and physical barriers
 - (Relatively) durable material
- **Cement paste**
 - Water + cement
- **Mortar**
 - Water + cement + sand
- **Concrete**
 - Water + cement + sand + gravel
- **Reinforced concrete**
 - Water + cement + sand + gravel + bars



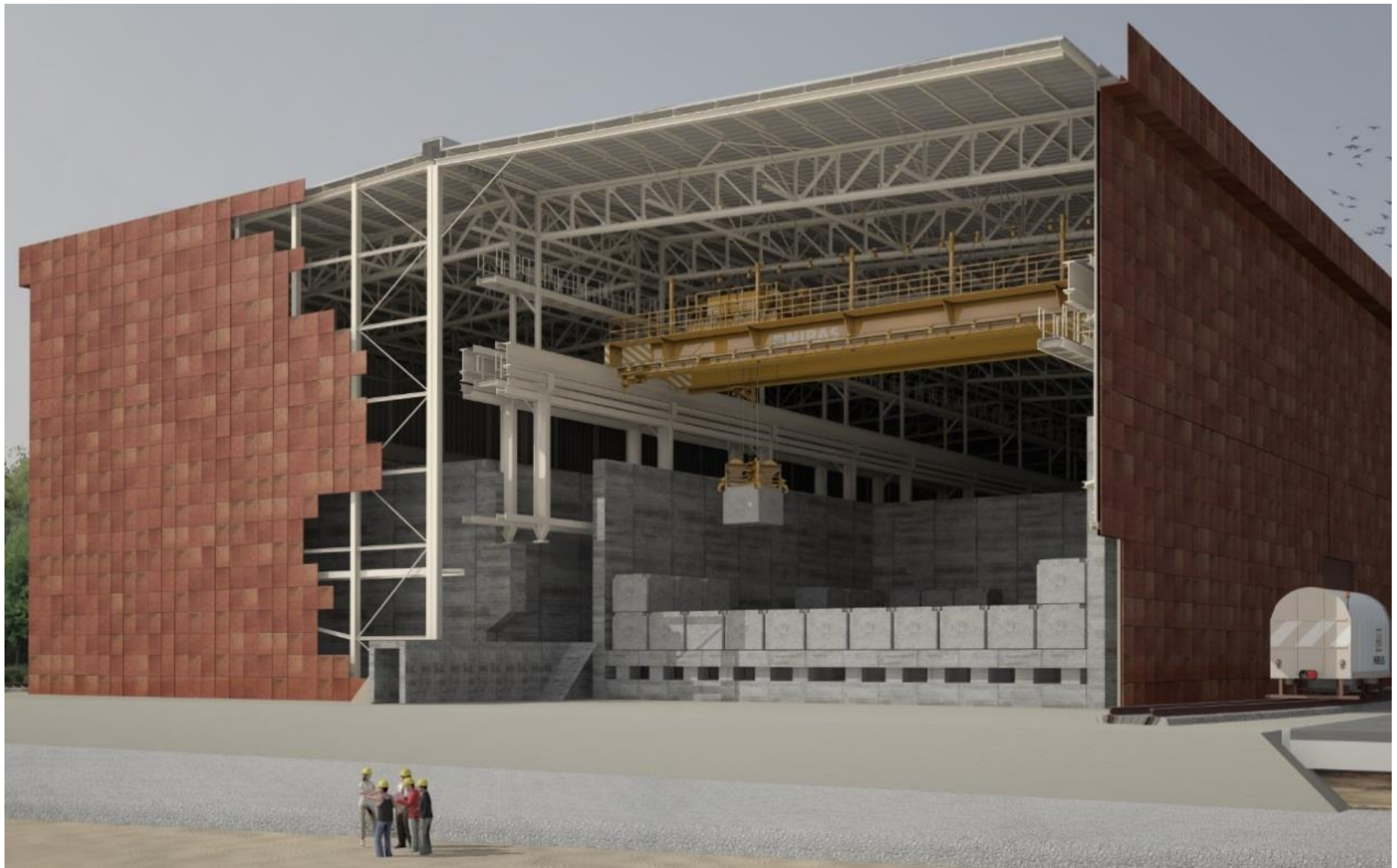
The cAt surface repository design

1 Confine and retain

2 Isolate



The cAt surface repository design



The B&C underground repository design

Monolith B

Supercontainer

Concrete segments

Concrete lining

Backfill

Buffer

Concrete container

Host rock

Mortar filler

Cement-based immobilization matrix

Concrete

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Introduction to cement



- **Cement is an hydraulic binder**

- Hydraulic: a material able to react with water to form a hardened end-product.
- Binder: Other materials (e.g. sand and gravel) can be incorporated to this hardened end-product in a rigid way.

- **Various cement types exist**

- Calcium-Silicate-Cement
- Calcium-Aluminate-Cement
- *Magnesium-Phosphate-Cement*
- Calcium-Sulfate-Aluminate-Cement

NBN EN 197

NBN EN 14647

} Different chemistry

Introduction to cement



- **Cement is an hydraulic binder**

- Hydraulic: a material able to react with water to form a hardened end-product.
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- | | |
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| ▪ Calcium-Silicate-Cement | NBN EN 197 |
| ▪ Calcium-Aluminate-Cement | NBN EN 14647 |
| ▪ <i>Magnesium-Phosphate-Cement</i> | |
| ▪ Calcium-Sulfate-Aluminate-Cement | |

} Different chemistry

Introduction to cement

■ NBN EN 197

EN: European standard carrying the obligation to be implemented at national level.
NBN: Published by the Belgian Bureau for standardization.

ICS: 91.100.10

**Geregistreeerde
Belgische norm**

NBN EN 197-1

2e uitg., oktober 2011

Normklasse: B 12

**Cement - Deel 1 : Samenstelling, specificatie en
overeenkomstigheidscriteria voor gewone cementsoorten**

Ciment - Partie 1 : Composition, spécifications et critères de conformité des ciments courants
Cement - Part 1 : Composition, specifications and conformity criteria for common cements

Toelating tot publicatie: 21 oktober 2011

Vervangt NBN EN 197-1 (2000), NBN EN 197-1/A1 (2004), NBN EN 197-1/A3 (2007) en NBN EN 197-4 (2004).

Deze Europese norm EN 197-1:2011 heeft de status van een Belgische norm.

Deze Europese norm bestaat in drie officiële versies (Duits, Engels, Frans).



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Prijsgroep: 19

CEM I
« Portland
cement »

CEM II

CEM III

CEM IV

CEM V

Hoofd- typen	Aanduiding van de 27 producten (gewone cementsoorten)	Samenstelling (gehalten in massaprocent) ^{a)}											Nebenbestanddelen
		Hoofdbestanddelen											
		Klinker	Hoog- ova- slak	Micro- silica	Puzzolanen		Vliegas		Gebran- de leesteen	Kalksteen			
					Natuur- lijke	Gebrande natuurlijke	Silicium- houdend	Calcium- houdend		T	L	LL	
		K	S	D ^{b)}	P	Q	V	W	T	L	LL		
CEM I	Portland- cement	CEM I	95-100	-	-	-	-	-	-	-	-	-	0-6
	Portland- slakcement	CEM III/A-S	80-94	6-20	-	-	-	-	-	-	-	-	0-5
		CEM III/B-S	65-79	21-35	-	-	-	-	-	-	-	-	0-5
	Portlandmic- rosilica-cement	CEM III/A-D	90-94	-	6-10	-	-	-	-	-	-	-	0-5
	Portland- puzzolaan- cement	CEM III/A-P	80-94	-	-	6-20	-	-	-	-	-	-	0-5
		CEM III/B-P	65-79	-	-	21-35	-	-	-	-	-	-	0-5
		CEM III/A-Q	80-94	-	-	-	6-20	-	-	-	-	-	0-5
		CEM III/B-Q	65-79	-	-	-	21-35	-	-	-	-	-	0-5
		CEM III/A-V	80-94	-	-	-	-	8-20	-	-	-	-	0-5
	Portland- vlieg- as- cement	CEM III/B-V	65-79	-	-	-	-	21-35	-	-	-	-	0-5
		CEM III/A-W	80-94	-	-	-	-	-	6-20	-	-	-	0-5
		CEM III/B-W	65-79	-	-	-	-	-	21-35	-	-	-	0-5
		CEM III/A-T	80-94	-	-	-	-	-	-	6-20	-	-	0-5
	Portlandlei- steen- cemen- t	CEM III/B-T	65-79	-	-	-	-	-	-	21-35	-	-	0-5
	Portlandkalk- steen- cemen- t	CEM III/A-L	80-94	-	-	-	-	-	-	-	6-20	-	0-5
		CEM III/B-L	65-79	-	-	-	-	-	-	-	21-35	-	0-5
		CEM III/A-LL	80-94	-	-	-	-	-	-	-	-	6-20	0-5
CEM III/B-LL		65-79	-	-	-	-	-	-	-	-	21-35	0-5	
CEM III/A-M		80-94	←					6-20	→				0-5
Portlandcomposit- cement ^{c)}	CEM III/B-M	65-79	←					21-35	→				0-5
CEM III	Hoogoven- cement	CEM III/A	35-64	36-65	-	-	-	-	-	-	-	-	0-5
		CEM III/B	20-34	66-80	-	-	-	-	-	-	-	-	0-5
		CEM III/C	5-19	81-85	-	-	-	-	-	-	-	-	0-5
	Puzzolaan- cement ^{c)}	CEM IV/A	65-80	-	←					11-35	→	-	-
CEM IV/B		45-64	-	←					36-55	→	-	-	0-5
CEM V	Composit- cement ^{c)}	CEM V/A	40-64	18-30	-	←			18-30	→	-	-	0-5
		CEM V/B	20-36	31-50	-	←			31-50	→	-	-	0-5

^{a)} De waarden in de tabel zijn uitsluitend ten opzichte van de som van hoofd- en nevenbestanddelen.

^{b)} Het gehalte microsilica mag niet meer dan 10% bedragen.

^{c)} In het geval van Portlandcompositcement CEM III/A-M en CEM III/B-M, Puzzolaancementen CEM IV/A en CEM IV/B en Compositcement CEM V/A en CEM V/B, moeten de hoofdbestanddelen, anders dan klinker, in de aanduiding van het cement vermeld zijn.

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- **NBN EN 197**

NBN: Published by the Belgian Bureau for standardization.

Main types	Notation of the 27 products (types of common cement)		Composition [proportion by mass ¹⁾]										
			Main constituents										Minor additional constituents
			Clinker	Blastfurnace slag	Silica fume D ²⁾	Pozzolana		Fly ash		Burnt shale	Limestone*		
						natural	natural calcined	siliceous	calcareous		L	LL	
			K	S		P	Q	V	W	T			
CEM I	Portland cement	CEM I	95-100	-	-	-	-	-	-	-	-	-	0-5
CEM II	Portland-slag cement	CEM II/A-S	80-94	6-20	-	-	-	-	-	-	-	-	0-5
		CEM II/B-S	65-79	21-35	-	-	-	-	-	-	-	-	0-5
	Portland-silica fume cement	CEM II/A-D	90-94	-	6-10	-	-	-	-	-	-	-	0-5
	Portland-pozzolana cement	CEM II/A-P	80-94	-	-	6-20	-	-	-	-	-	-	0-5
		CEM II/B-P	65-79	-	-	21-35	-	-	-	-	-	-	0-5
		CEM II/A-Q	80-94	-	-	-	6-20	-	-	-	-	-	0-5
		CEM II/B-Q	65-79	-	-	-	21-35	-	-	-	-	-	0-5
	Portland-fly ash cement	CEM II/A-V	80-94	-	-	-	-	6-20	-	-	-	-	0-5
		CEM II/B-V	65-79	-	-	-	-	21-35	-	-	-	-	0-5
		CEM II/A-W	80-94	-	-	-	-	-	6-20	-	-	-	0-5
		CEM II/B-W	65-79	-	-	-	-	-	21-35	-	-	-	0-5
	Portland-burnt shale cement	CEM II/A-T	80-94	-	-	-	-	-	-	6-20	-	-	0-5
		CEM II/B-T	65-79	-	-	-	-	-	-	21-35	-	-	0-5
	Portland-limestone cement	CEM II/A-L	80-94	-	-	-	-	-	-	-	6-20	-	0-5
		CEM II/B-L	65-79	-	-	-	-	-	-	-	21-35	-	0-5
		CEM II/A-LL	80-94	-	-	-	-	-	-	-	-	6-20	0-5
CEM II/B-LL		65-79	-	-	-	-	-	-	-	-	21-35	0-5	
Portland-composite cement ³⁾	CEM II/A-M	80-94	<----- 6-20 ----->						----->				0-5
	CEM II/B-M	65-79	<----- 21-35 ----->						----->				0-5
CEM III	Blastfurnace cement	CEM III/A	35-64	36-65	-	-	-	-	-	-	-	-	0-5
		CEM III/B	20-34	66-80	-	-	-	-	-	-	-	-	0-5
		CEM III/C	5-19	81-95	-	-	-	-	-	-	-	-	0-5
CEM IV	Pozzolanic cement ³⁾	CEM IV/A	65-89	-	<----- 11-35 ----->					-	-	-	0-5
		CEM IV/B	45-64	-	<----- 36-55 ----->					-	-	-	0-5
CEM V	Composite cement ³⁾	CEM V/A	40-64	18-30	-	<----- 18-30 ----->			-	-	-	-	0-5
		CEM V/B	20-38	31-50	-	<----- 31-50 ----->			-	-	-	-	0-5

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CEM III

CEM IV

CEM V



ONDRAF/NIRAS

Introduction to cement

■ NBN EN 197

EN: European standard carrying the obligation to be implemented at national level

NBN: Published by the Belgian Bureau for standardization.

Most used in Belgium

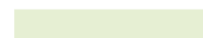
Samenvatting (gebruik in massaproductie)											
Hoofdstuk	Aanduiding van de 21 producten (geen overeenkomst)	Hoofdbestanddelen								Bemerkingen	Norm
		CEM I	CEM II	CEM III	CEM IV	CEM V	CEM VI	CEM VII	CEM VIII		
CEM I	Portlandcement	CEM I	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I
	Portlandkalksteencement	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J
	Portlandcomposietcement	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J	CEM II/K
	Hoogovencement	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J
	CEM III	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J
CEM II	Portlandcement	CEM I	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I
	Portlandkalksteencement	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J
	Portlandcomposietcement	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J	CEM II/K
	Hoogovencement	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J
	CEM III	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J
CEM III	Portlandcement	CEM I	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I
	Portlandkalksteencement	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J
	Portlandcomposietcement	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J	CEM II/K
	Hoogovencement	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J
	CEM III	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J
CEM IV	Portlandcement	CEM I	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I
	Portlandkalksteencement	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J
	Portlandcomposietcement	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J	CEM II/K
	Hoogovencement	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J
	CEM III	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J
CEM V	Portlandcement	CEM I	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I
	Portlandkalksteencement	CEM II/A	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J
	Portlandcomposietcement	CEM II/B	CEM II/C	CEM II/D	CEM II/E	CEM II/F	CEM II/G	CEM II/H	CEM II/I	CEM II/J	CEM II/K
	Hoogovencement	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J
	CEM III	CEM III/A	CEM III/B	CEM III/C	CEM III/D	CEM III/E	CEM III/F	CEM III/G	CEM III/H	CEM III/I	CEM III/J

Cementtype

		Hoofdbestanddelen	
Portlandcement	CEM I	100	
Portlandkalksteencement	CEM II/A-LL	6	94
		20	80
Portlandcomposietcement	CEM II/B-M (S-V)	21	79
		35	65
Hoogovencement	CEM III/A	36	64
		65	35
	CEM III/B	66	34
		80	20
	CEM III/C	81	19
		95	5



Clinker



Fly Ash



Calcareous
filler



GGBS
blast-furnace slag

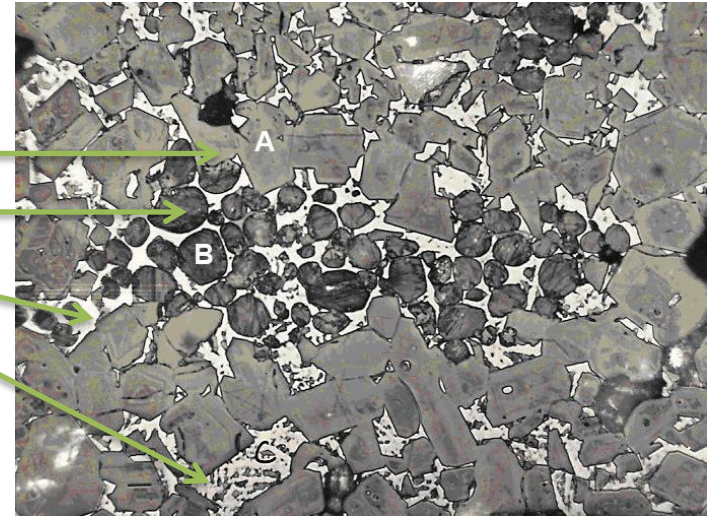
Introduction to Cement

■ Clinker composition

Clinker = calcareous and argillaceous rocks, ground and calcined

Polished section of clinker (OM) [NONAT, 2008]

matrix {	Anhydrous phase	Notation	% weight
	Tricalcium silicates	C_3S (Alite)	60-65
	Bicalcium silicates	C_2S (Belite)	10-20
	Tricalcium aluminates	C_3A	8-12
	Tetracalcium aluminoferrite	C_4AF	8-10



■ Supplementary Cementitious Materials (SCM)

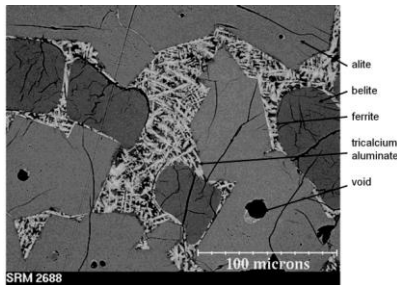
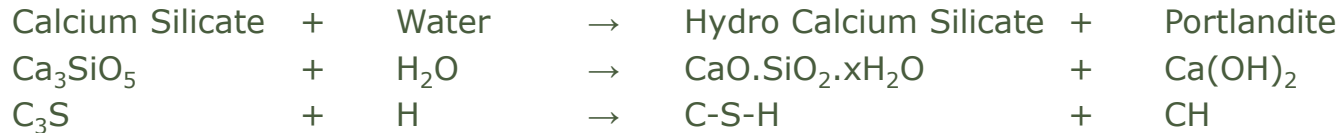
- Calcareous fillers, mainly calcium carbonate,
- Pouzzolanic admixtures, mainly siliceous or siliaceous and aluminous materials derived from silicon or coal industries (e.g. Silica Fumes, Fly Ash) or ground volcanic rocks.
- Latent hydraulic binders, their composition is similar to the clinker (CaO , SiO_2 , Al_2O_3), but in different proportions. Their hydraulic properties are activated in specific conditions. (e.g. Blast Furnace Slag are activated in high pH solution).

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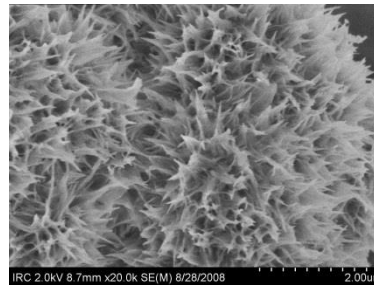
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Cement Hydration

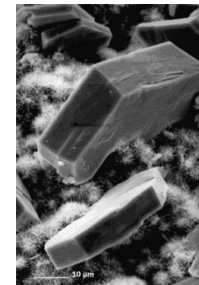
Calcium silicates hydration [Not chemical reactions]



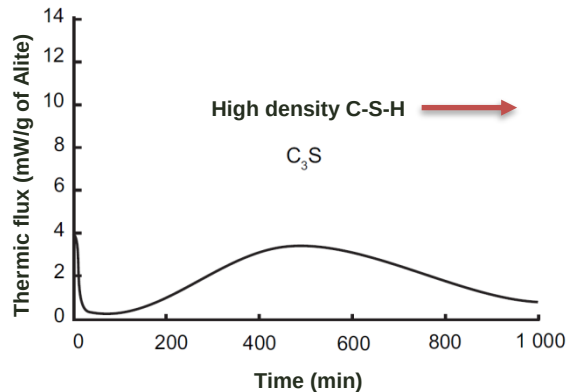
NIST SRM 2688, SEM
Backscattered Electron Image



CSU, CementLab, [ALIZADEH, 2007]



Portlandite, [DAMIDOT, 2008]



C-S-H

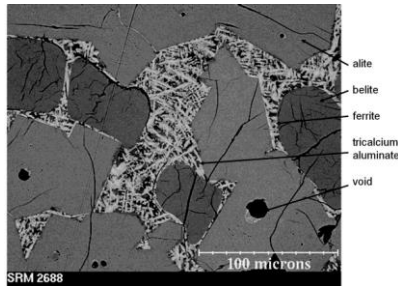
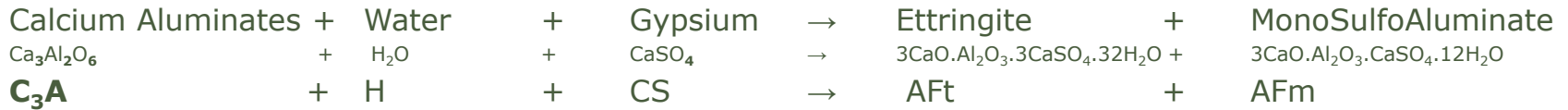
- Gel-like amorphous phase
- Main component of the hardened paste
- Characterized by the C/S ratio and its specific surface
- High electric surface density
- Stable at high pH, dissolved at pH 10

Portlandite

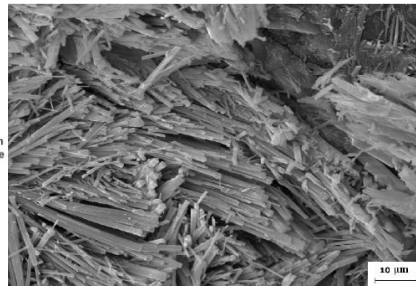
- Hexagonal crystal structure
- Most soluble phase
- Guarantees a high pH ~12,6

Cement Hydration

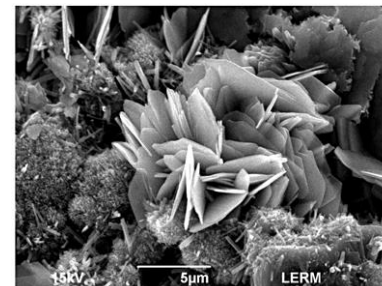
Calcium aluminates hydration [Not chemical reactions]



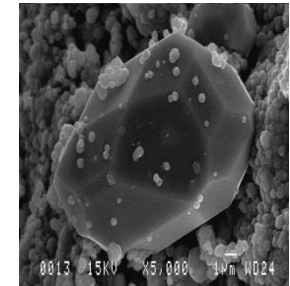
NIST SRM 2688, SEM
Backscattered Electron Image



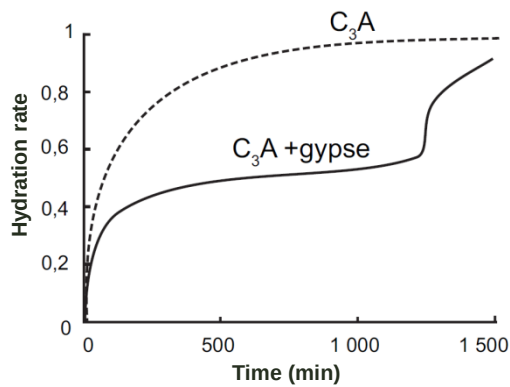
Ettringite, [BENICI, 2012]



AFm and Aft phases, [LERM, 2010]



Hydrogrenat, [DAMIDOT, 2008]



Step 1: Hydration of aluminates with formation of ettringite (Aft)

- Addition of gypsum regulates the hydration of aluminates
- Ettringites plays only a chemical role in the hydration process

Step 2: Once all gypsum is consumed, ettringite redissolves (partly) and forms monosulfoaluminates (AFm)

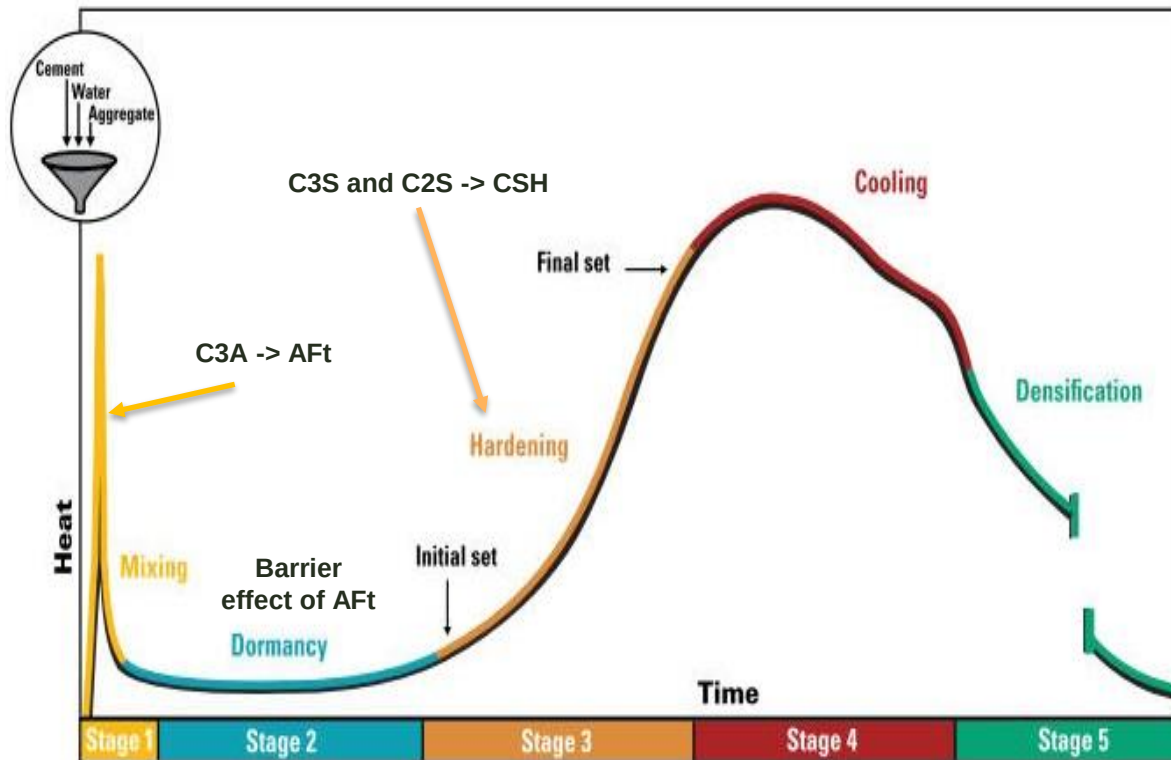
- AFm phases structure depends on the C/A ratio

Step 3: If all ettringite is consumed, anhydrous aluminates are hydrated and form hydrogrenats and hydrated aluminates phases (with no sulfates)

Cement Hydration

What does this mean in « real life » ?

- Heat generation from the hydration reaction (no drying!)

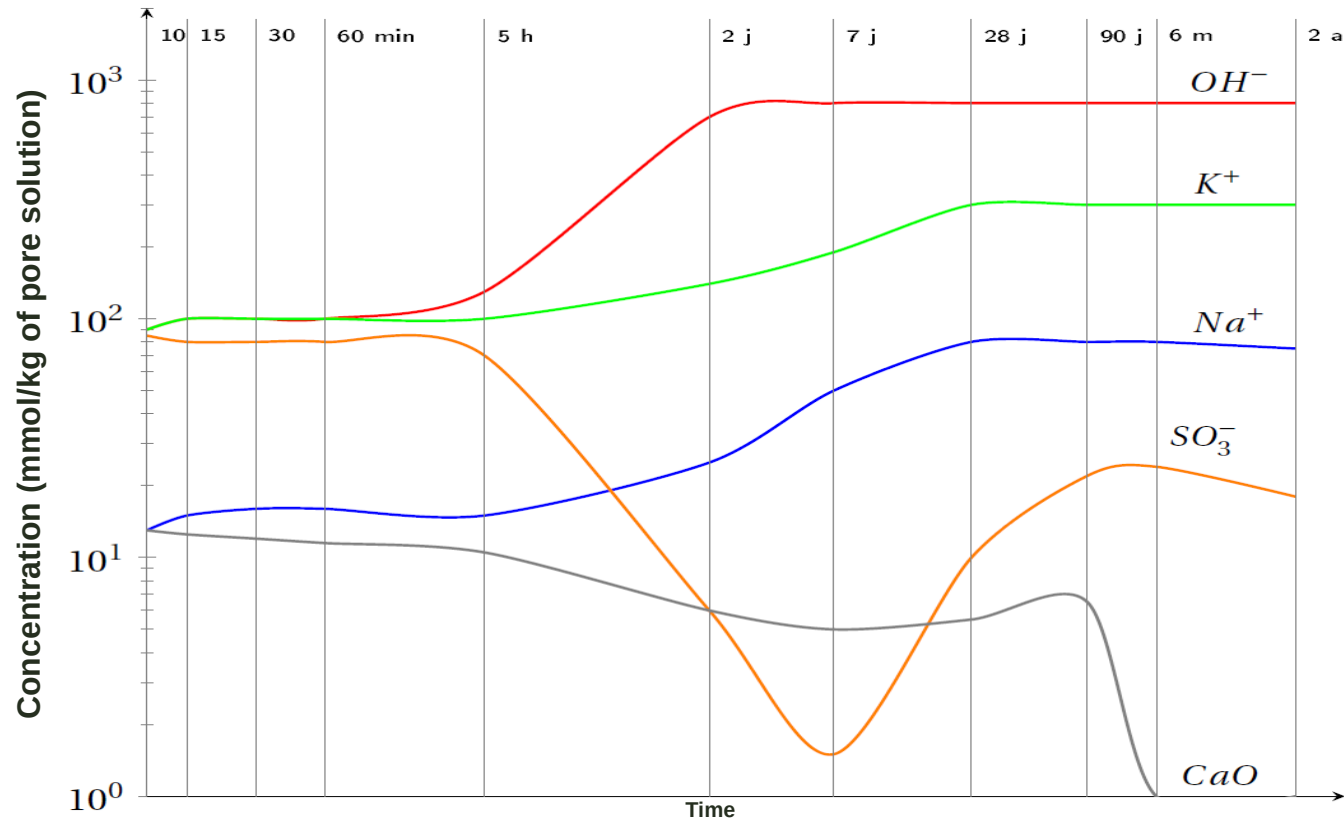


Heat generation curve from cement hydration, [J.Shilstone, 2012]

Cement Hydration

What does this mean in « real life » ?

- Heat generation from the hydration reaction (no drying!)
- Water is transferring into pore-water with specific chemistry



Cement Hydration

What does this mean in « real life » ?

- Heat generation from the hydration reactions (no drying!)
- Water is transferred into pore-water with a specific chemistry
- Chemical shrinkage

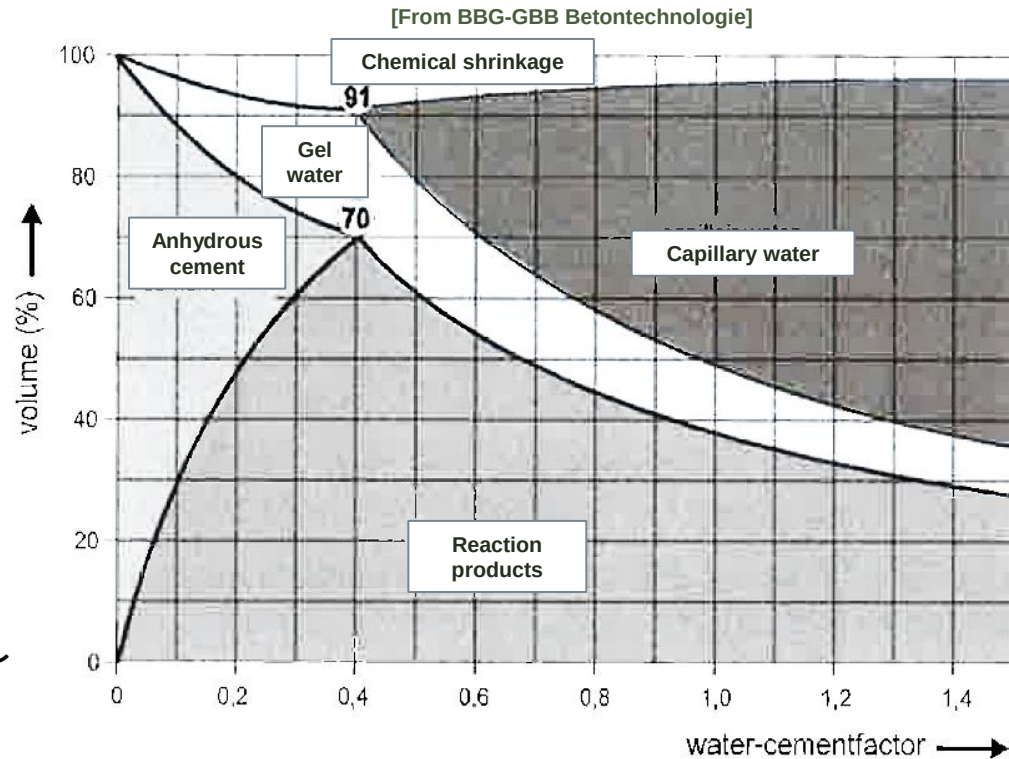
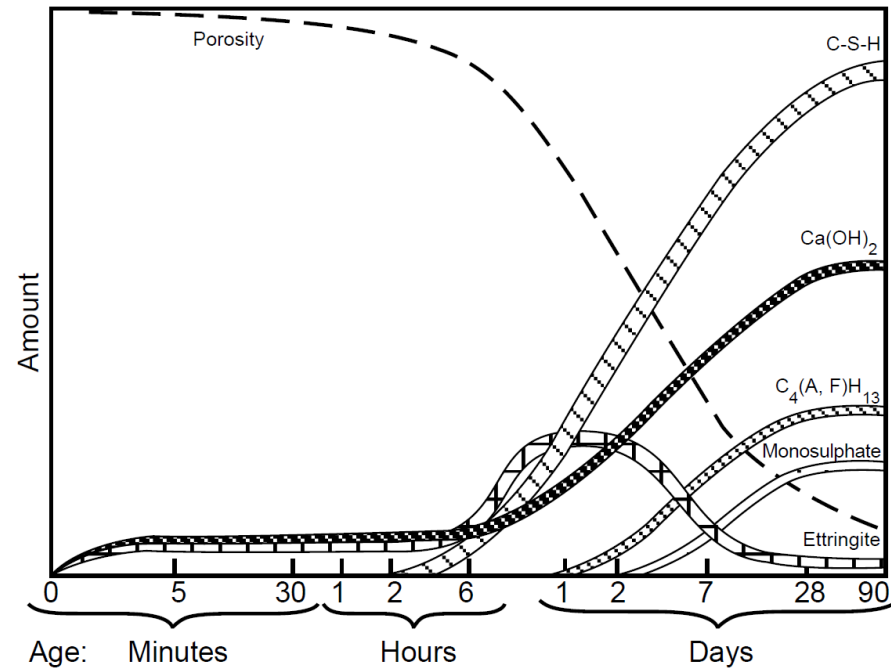


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- **Cement-based materials: what, where?**
 - A reminder of what the concrete ingredients are
 - cAt repository design
 - B&C underground repository design
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 - Introduction to cements
 - Cement hydration mechanisms and reactions
 - Characterization of cement-based materials
- **Examples of highlighted phenomenon**
 - Delayed Ettringite Formation (DEF)

Control and analysis techniques

Techniques and material

Porosity

Mercury Intrusion
BET

Chemical analysis

ICP, IC, AAS

μ -scale

characterization

Optic Microscopy
SEM

Time control

Calorimeter

Weight control

**Temperature
and relative
humidity
control**

Rheometer

Thermometer

Calorimeter

**Entrapped air
cell**

Weight control

**Climatic
chamber**

**Bleeding
measures**

Extensometers

Mech. Strength (Compression, traction, flexion)

Porosity (MIP, BET, Water porosity)

Pore water analysis

Sorptivity of chem. species

Diffusivity (Resistivity, diffusion, migration, Imp. Spectroscopy)

Permeability

Microstructure (MO, SEM, EDS, NMR)

Mineralogy (TGA, XRD, OM)

**Dry
ingredients**

t ~ s

Casting

t ~ min

**Fresh
material**

t ~ h

Curing

t ~ w, m

**Hardened
material**

**Degraded
material**

t ~ y

Aggregates

Porosity control
Chemical analysis
Mineralogy
Size and grading

Cement

Chemical
composition

Water

Tap water quality

Recipe design

Proportions (1m^3)
W/C ratio

Mix design

Mixer type
Mixing times

Setting

Workability
**Density and air
content**
Hydration heat

Bleeding

Shrinkage
**Curing
conditions**

Alkali-Silica Reactions

Corrosion
Leaching
Carbonation
Chlorides/sulfates ingress
Freeze/Thaw damage
DEF

Physical phenomena and quantities



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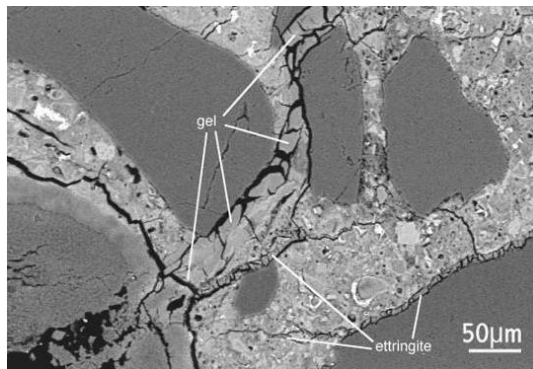
Highlight: Delayed Ettringite Formation

- DEF is the formation of ettringite while the cement (concrete) is rigid.

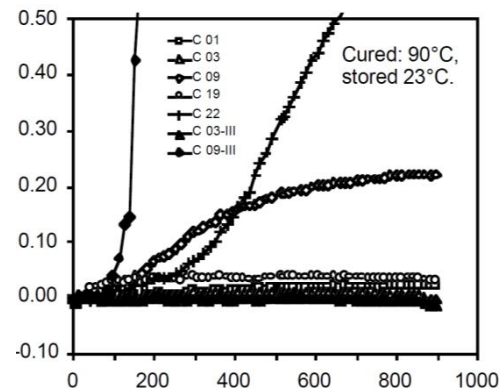


- **Consequences: Damage due to expansion**
 - Structure instability
 - Loss of mechanical strength
 - Increase of open paths for aggressive agents ingress

DEF in the inter-aggregates spaces result in trough cracks



Some mortars cured at 90°C show important expansions after 100 days

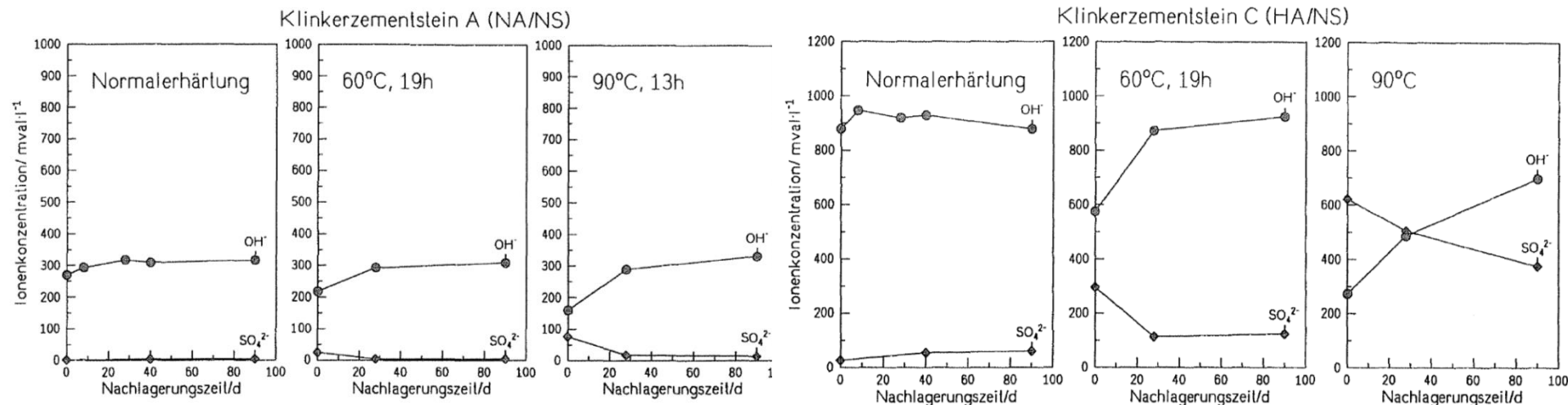


Cracks pattern can be the result of DEF and other affections (e.g. ASR). Lab experiments are needed.



Highlight: Delayed Ettringite Formation

- Ettringite stability is strongly affected by:
 - Temperature
 - Alkali-content



Wieker, W.; Hübert, C.; Schubert, H.: *Untersuchungen zum Einfluß der Alkalien auf die Stabilität der Sulfoaluminathydrate in Zementstein und -mörteln bei Warmbehandlung*. In: Schriftenreihe des Institutes für Massivbau und Baustofftechnologie, Uni Karlsruhe (1996), p.175-186

■ Cement-based materials: what, where?

■ General introduction to cement chemistry

■ Example of highlighted phenomenon

Introduction into Cement



■ Cement is an hydraulic binder

- Hydraulic: A material that is able to reaction both underwater as in air with water to form a stone-like end product.
- Binder: This stone-like end product is able to incorporate other materials (like sand and gravel) in a rigid way.

■ Various cement types exists

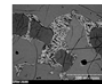
- Calcium-Silicate-Cement NBN EN 197
- Calcium-Aluminate-Cement NBN EN 14647
- Magnesium-Phosphate-Cement
- Calcium-Sulphate-Aluminate-Cement

Various chemistry

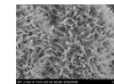


Cement Hydration

Calcium silicates hydration (not chemical reactions)



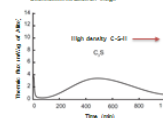
NIST SRM 2600, SEM Backscattered Electron Image



CSU, Cambridge, (ALBA/ACD), 2007



Portlandite, (DAMDOT), 2000



- C-S-H**
 - Gel-like amorphous phase
 - Main component of the hardened paste
 - Characterized by the C/S ratio and its specific surface
 - High electric surface density
 - Stable at high pH, dissolved at pH 10

- Portlandite**
 - Hexagonal crystal structure
 - Most soluble phase
 - Guarantees a high pH ~12.6



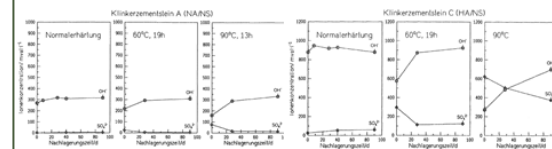
19/05/2017

15

Highlight: Delayed Ettringite Formation

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- Temperature
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Wielers, W.; Hübner, C.; Schubert, H.: Untersuchungen zum Einfluß der Alkalien auf die Stabilität der Sulfatoaluminate in Zementstein und Mörteln bei Warmbehandlung. In: Schriftenreihe des Institutes für Massivbau und Baustofftechnologie, Uni Karlsruhe. (1996), S.175-186



24

A short introductive video

JRAM Video

Video on cement chemistry by Arnaud MULLER and John ROSSEN, Laboratory for Construction Materials, from Ecole Polytechnique Fédérale de Lausanne.

Complete link if the hyper link doesn't work:

<https://www.youtube.com/watch?v=L4OLBNXMdHk&app=desktop>

Concrete materials in the B&C repositroy

- **Concrete segments and lining**

- Reinforced High Performance concrete (the current gallery lining was a CEM I cement)
- Mechanical performances

- **Backfill**

- *No fixed recipe yet* (the recipe can be different for the Monoliths B and the Supercontainer)
- High permeability, high porosity
- Retrievability

Monolith B

- **Concrete container**

- Concrete (*no fixed recipe yet*)

- **Mortar filler**

- Mortar (*no fixed recipe yet*)

- **Cement-based immobilization matrix**

- Depends on the waste form and the conditioning line

Supercontainer

- **Buffer**

- CEM I concrete with limestone aggregates
- Chemical role for the passivation of the metallic Overpack

- **Mortar filler**

- Mortar recipe based on the buffer formulation



The CEBAMA project

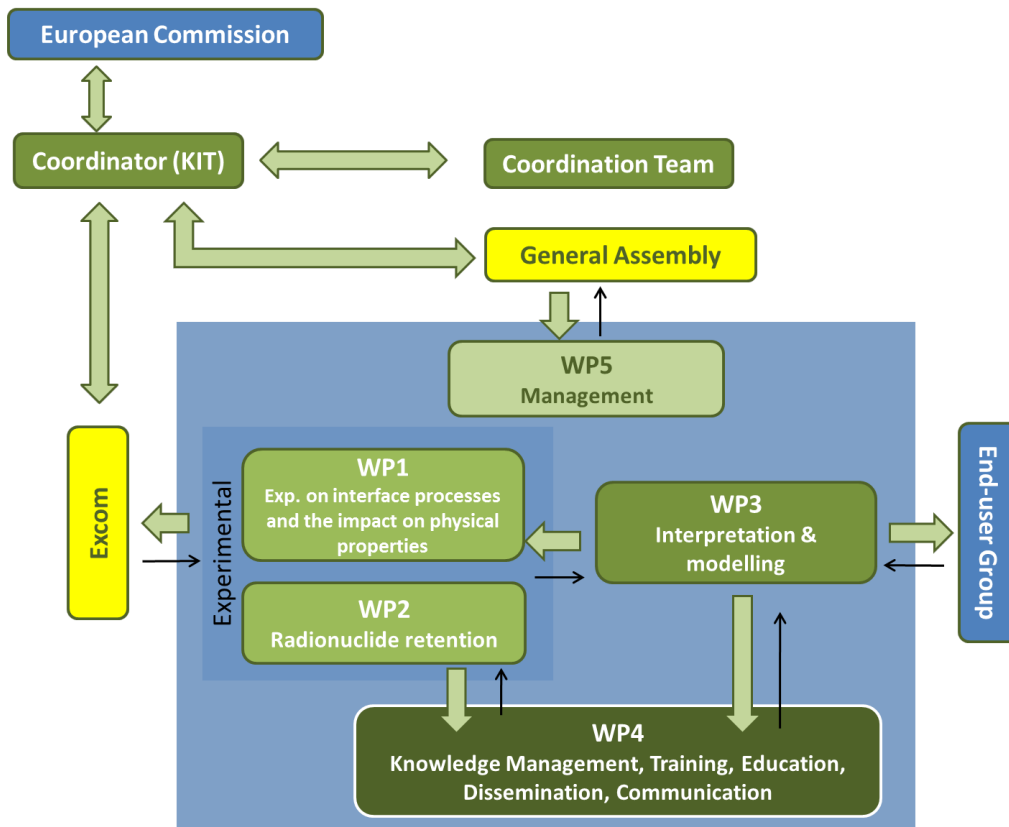


■ What is CEBAMA?

- A collaborative project related to the use of CEment-BASed MAterials in nuclear waste disposal applications
- Funded by the EC (Horizon 2020)
- 4 years started in June 1st 2015
- 27 Beneficiaries (SCK•CEN included), O/N part of the End-Users



The CEBAMA project



- **WP1:** Experiments on interface processes between the host rock and the concrete materials and the impact on physical properties
- **WP2:** Radionuclide retention in high pH concrete environments (Be, C, Cl, Ca, Se, Mo, I and Ra)
- **WP3:** Interpretation and modelling of transport processes in cement-based materials and interfaces with host rocks



ONDRAF/NIRAS

Conditioning of Radioactive Waste by Homogenous Cementation

Erik Coppens

21st Exchange Meeting ESV EURIDICE GIE

29/09/2025

Belgian Agency for Radioactive Waste and Enriched Fissile Materials



Conditioning of Radioactive Waste by Homogenous Cementation

- **Introduction**
- **The challenges and their origine**
- **How to develop a cemention process**
- **Conclusion**

Conditioning of Radioactive Waste by Homogenous Cementation

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Conditioning of Radioactive Waste by Homogenous Cementation - Introduction

Conditioning are those operations that produce a waste package suitable for handling, transport, storage or disposal.

This may include solidifaction, the conversion of a gaseous, liquid or liquid-like materials into a solid waste form, often done by homogenous intermixing with a cement

Homogenous Cementation : A conditioning process consisting of a solidification (=immobilisation) by homogenous intermixing with cement.

Conditioning of Radioactive Waste by Homogenous Cementation - Introduction

Homogenous cementation

1. (Aqueous) waste is fed
2. (gravel and sand is fed)
- 3. Cement** is fed
4. (In-drum) mixing
5. (Pouring)
6. (Transport to buffer)
7. Allow for hardening
8. Transport to storage



Photo: Westinghouse

Conditioning of Radioactive Waste by Homogenous Cementation - Introduction



Low-cost
Low-temperature
Mechanical strength
Compatible w aqueous flux
Elevated pH



Conditioning of Radioactive Waste by Homogenous Cementation - Introduction



Low-cost
Low-temperature
Mechanical strength
Compatible w aqueous flux
Elevated pH



It looks simple, but it isn't

Conditioning of Radioactive Waste by Homogenous Cementation

- Introduction
- The challenges and their origin
- How to develop a cementation process
- Conclusion

Homogenous Cementation – The Challenge

1. (Aqueous) waste is fed
2. (gravel and sand is fed)
3. Cement is fed
4. (In-drum) mixing
5. (Pouring)
6. (Transport to buffer)
7. Allow for hardening
8. Transport to storage

At all these (intermediate) process-steps the waste form should be compatible with the technical installation and in compliance with the waste acceptance criteria.

Homogenous Cementation – The Challenge

- 1. (Aqueous) waste is fed**
2. (gravel and sand is fed)
3. Cement is fed
4. (In-drum) mixing
5. (Pouring)
6. (Transport to buffer)
7. Allow for hardening
8. Transport to storage

Example: The waste is (after pretreatment) difficult to feed.

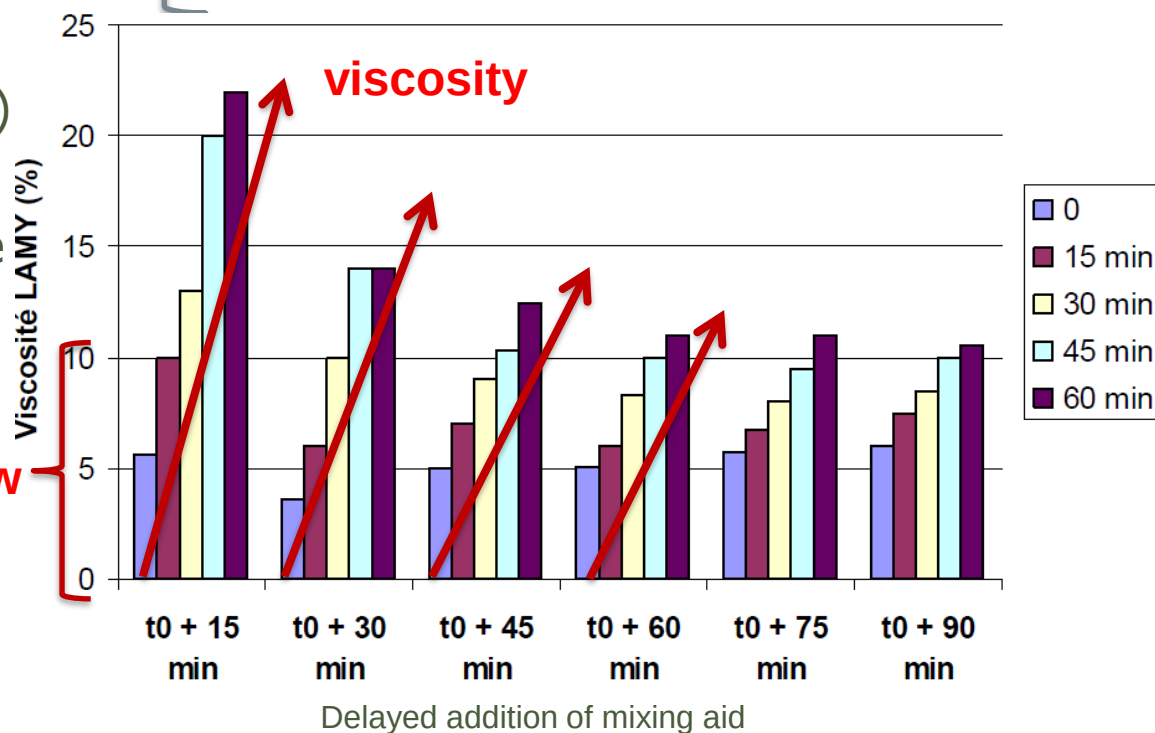


Homogenous Cementation – The Challenge

1. (Aqueous) waste is fed
2. (gravel and sand is fed)
3. Cement is fed
4. (In-drum) mixing
- 5. (Pouring)**
6. (Transport to buffer)
7. Allow for hardening
8. Transport to storage

Example: viscosity of the mixture is rising fast, not allowing for pouring.

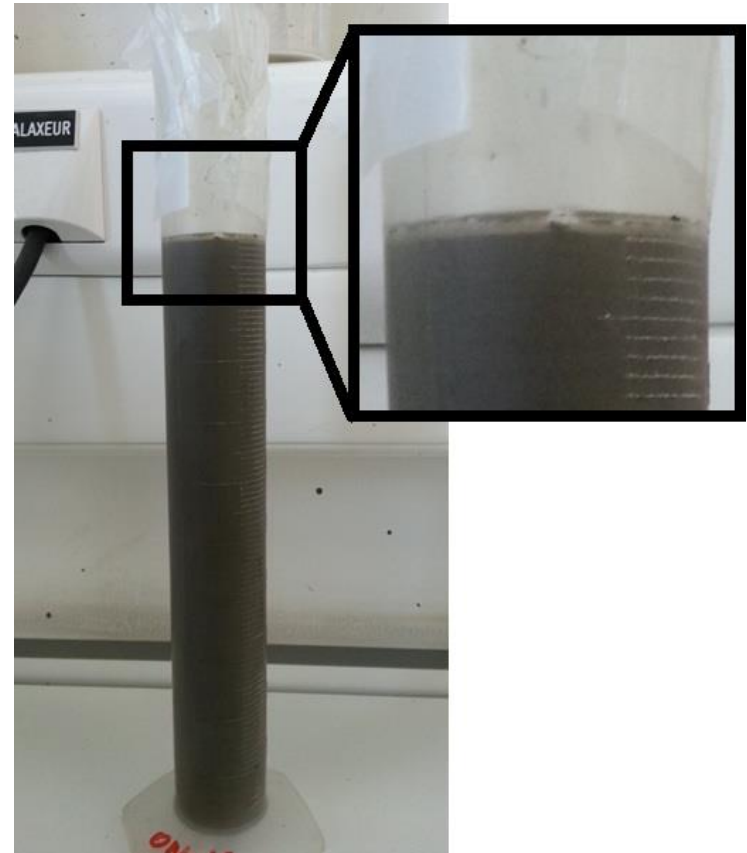
Sufficient low
for pouring



Homogenous Cementation – The Challenge

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8. Transport to storage

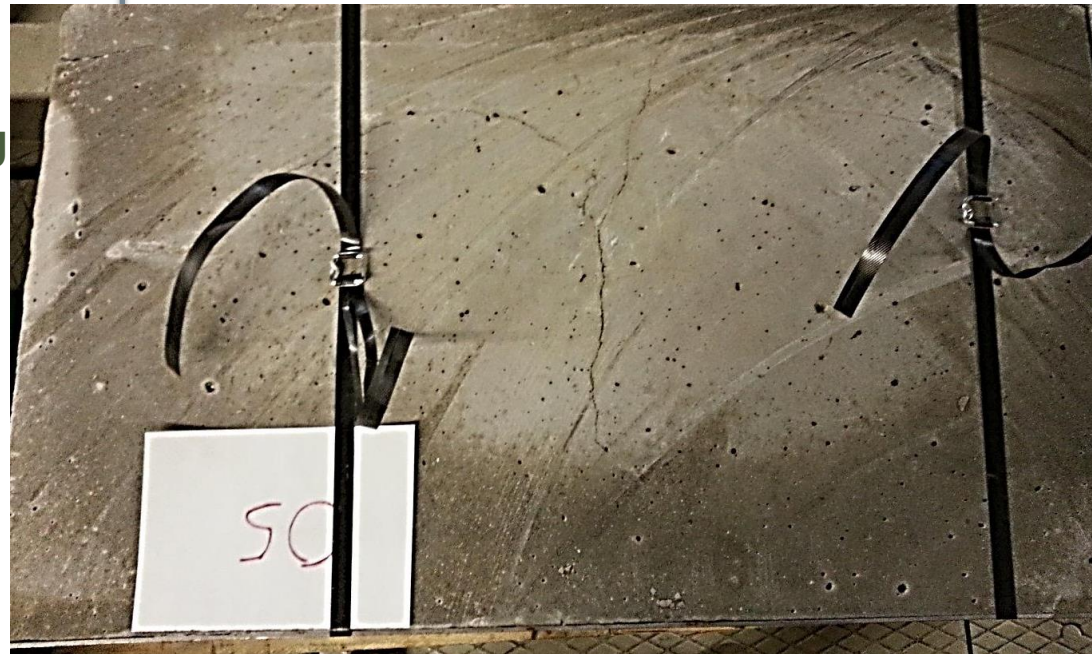
Example: The waste form is unstable during setting from a rheological point of view.



Homogenous Cementation – The Challenge

1. (Aqueous) waste is fed
2. (gravel and sand is fed)
3. Cement is fed
4. (In-drum) mixing
5. (Pouring)
6. (Transport to buffer)
- 7. Allow for hardening**
8. Transport to storage

Example: The waste form is cracked and fissured due to hydration heat.



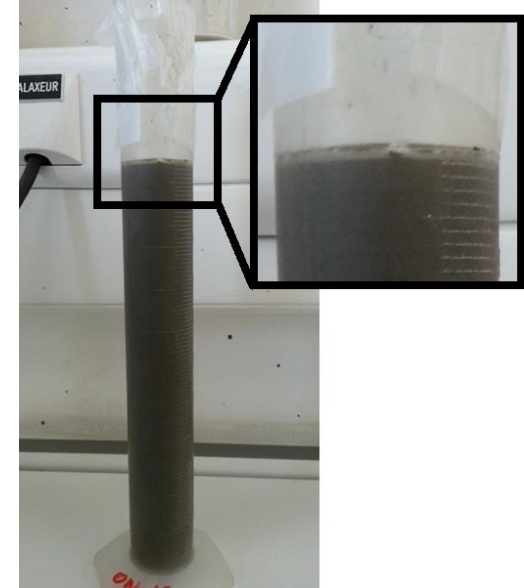
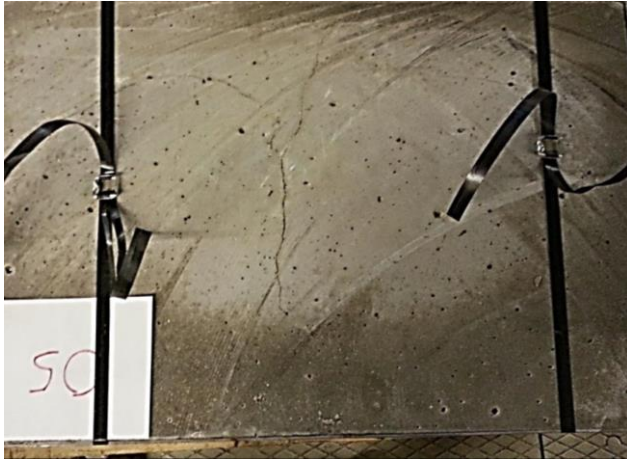
Homogenous Cementation – The Challenge

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4. (In-drum) mixing
5. (Pouring)
6. (Transport to buffer)
7. Allow for hardening
8. Transport to **storage**

Example: The waste form suffers from a pathology.

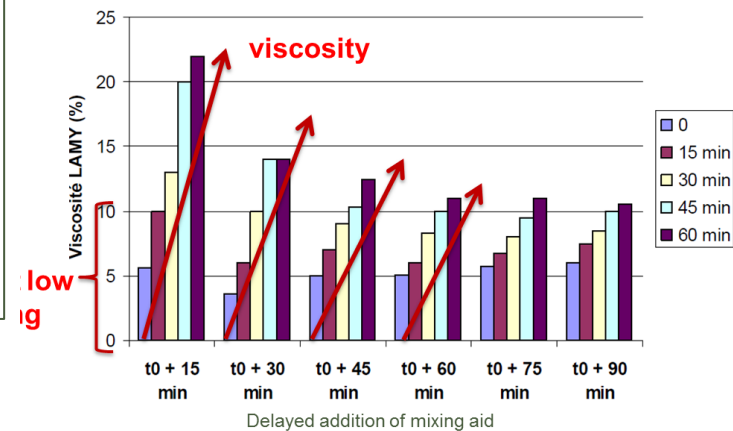


The Origine of Challenges



None of these problems is related to radioactivity.

Classic chemistry is dominating the behavior.



The Origine of Challenges

How come these problems are (often) unforeseen?

Knowledge of the effect of mixing cement(s) with concentrates of complex chemistry is still limited.

1. Most available data on the interaction of salts with cement are relative to each species taken separately from the others. Although interactions, synergy and antagonism exists.

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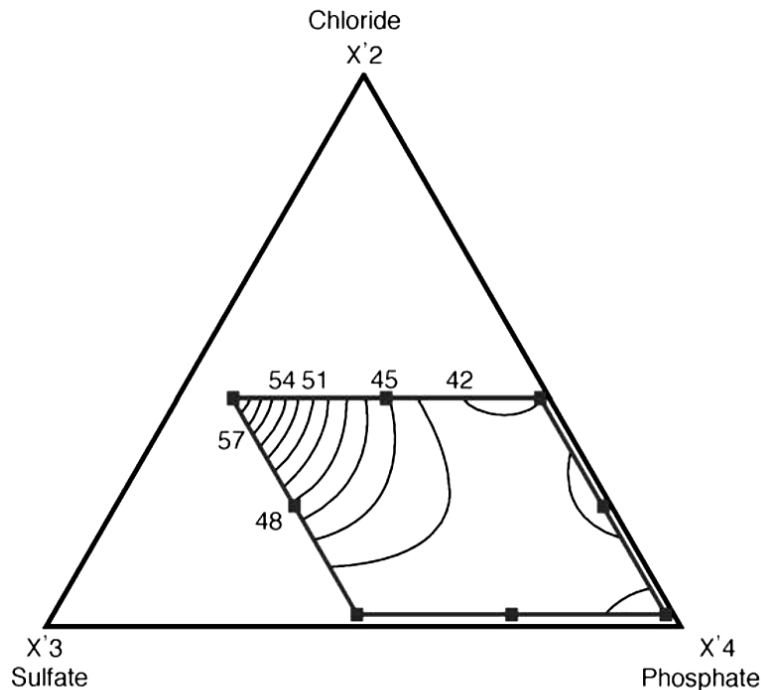
1. Most available data on the interaction of salts with cement are relative to each species taken separately from the others. Although interactions, synergy and antagonism exists.

Example: Compressive strength of waste form – contour plots

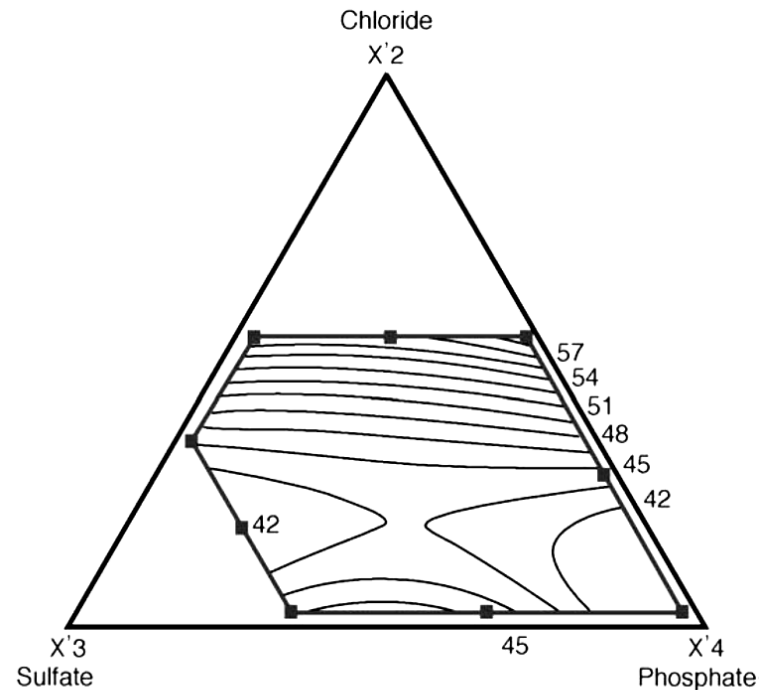
The Origine of Challenges

Example: interaction

(from C. Cau-dit-Coumes, S. Courtois / Cement and Concrete Research 33 (2003) 305-316)



$[B] = 0 \text{ g.L}^{-1}$



$[B] = 3 \text{ g.L}^{-1}$

The Origine of Challenges

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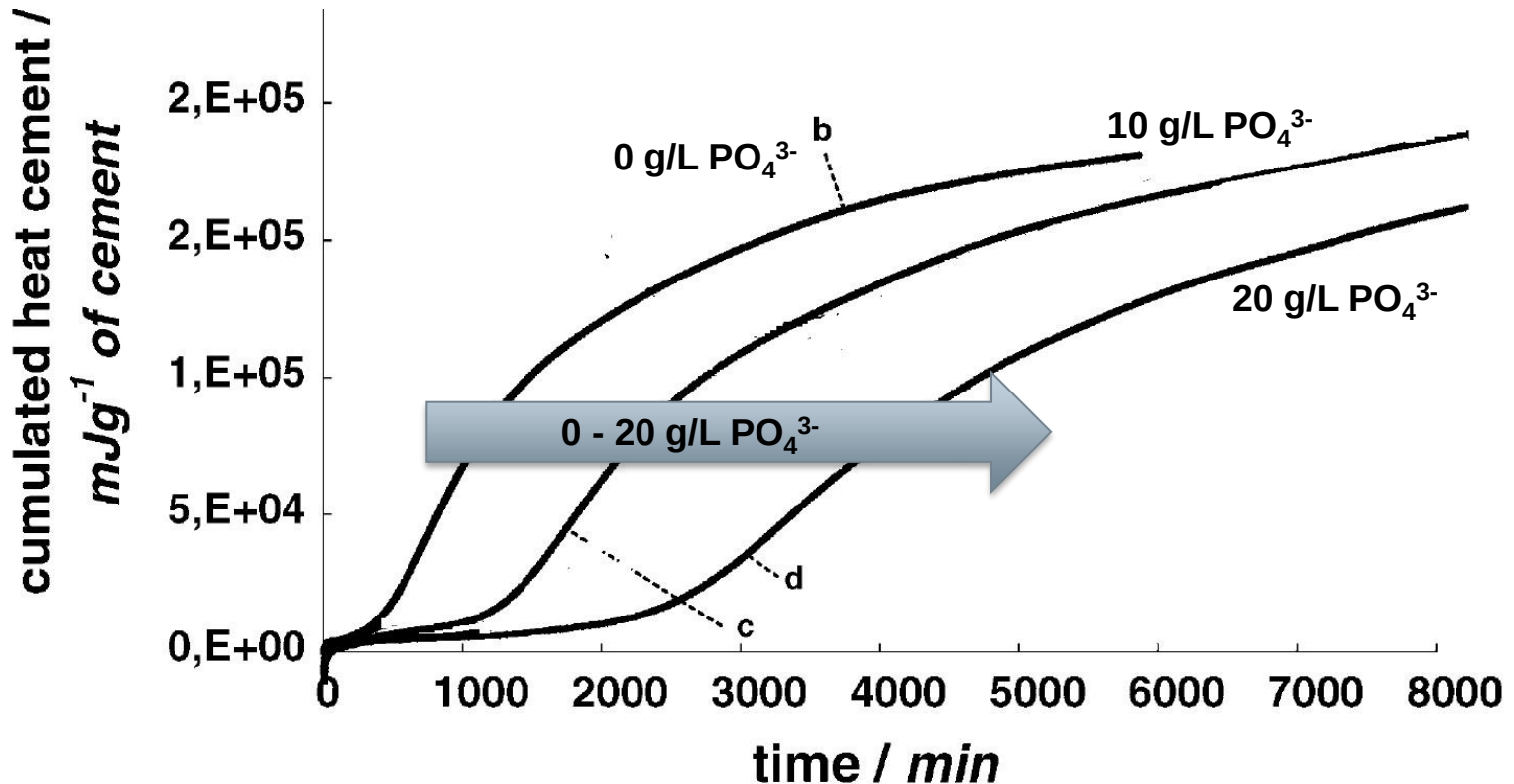
Example: The effect of phosphate on the hydration kinetics of Portland cement.

The Origin of Challenges

Example: phosphates – concentration effect

(from P. Bénard, S. Garrault, A. Nonat and C. Cau-dit-Coumes

Journal of the European Ceramic Society 25 (2005) 1877–1883)

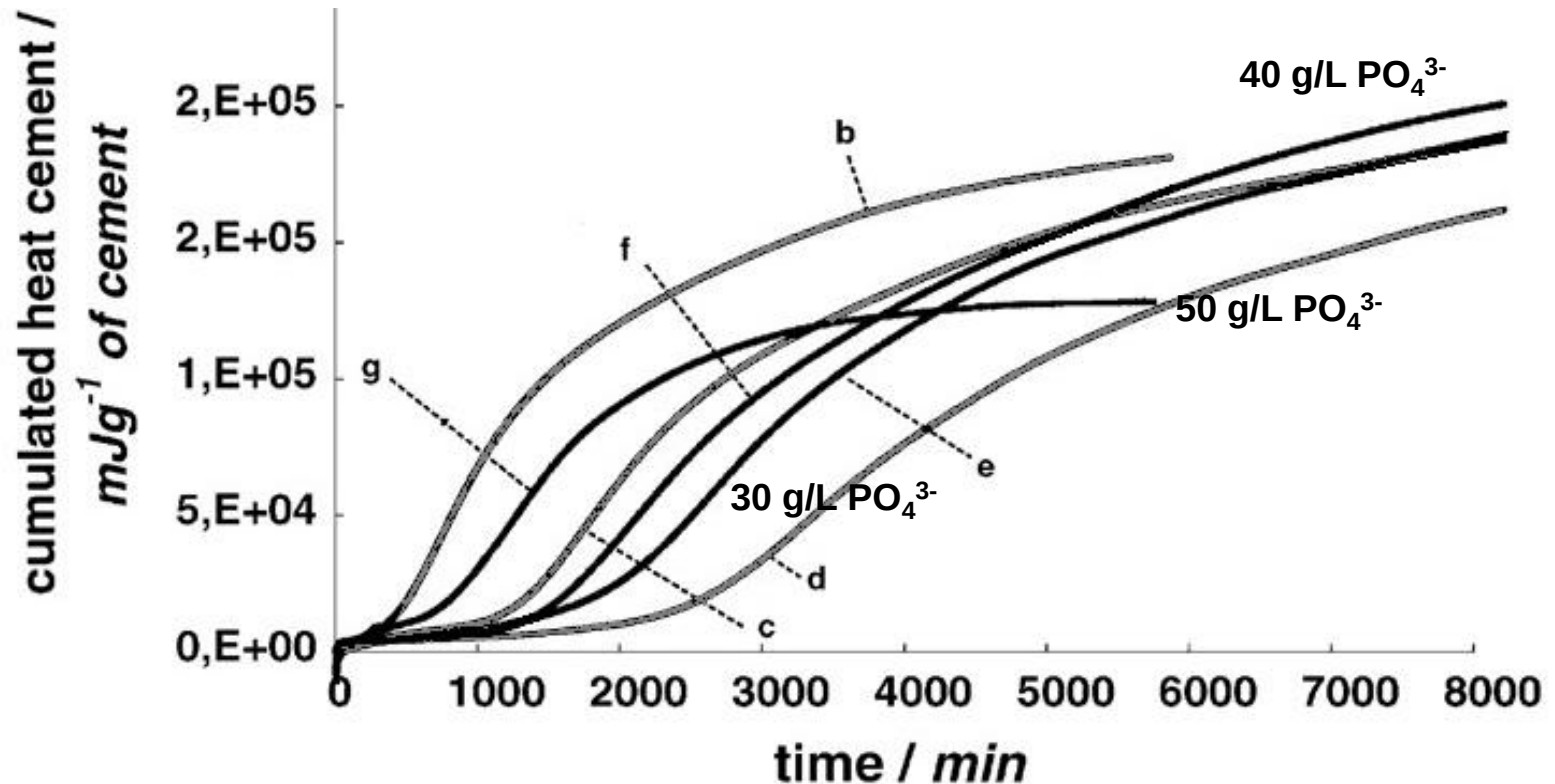


The Origin of Challenges

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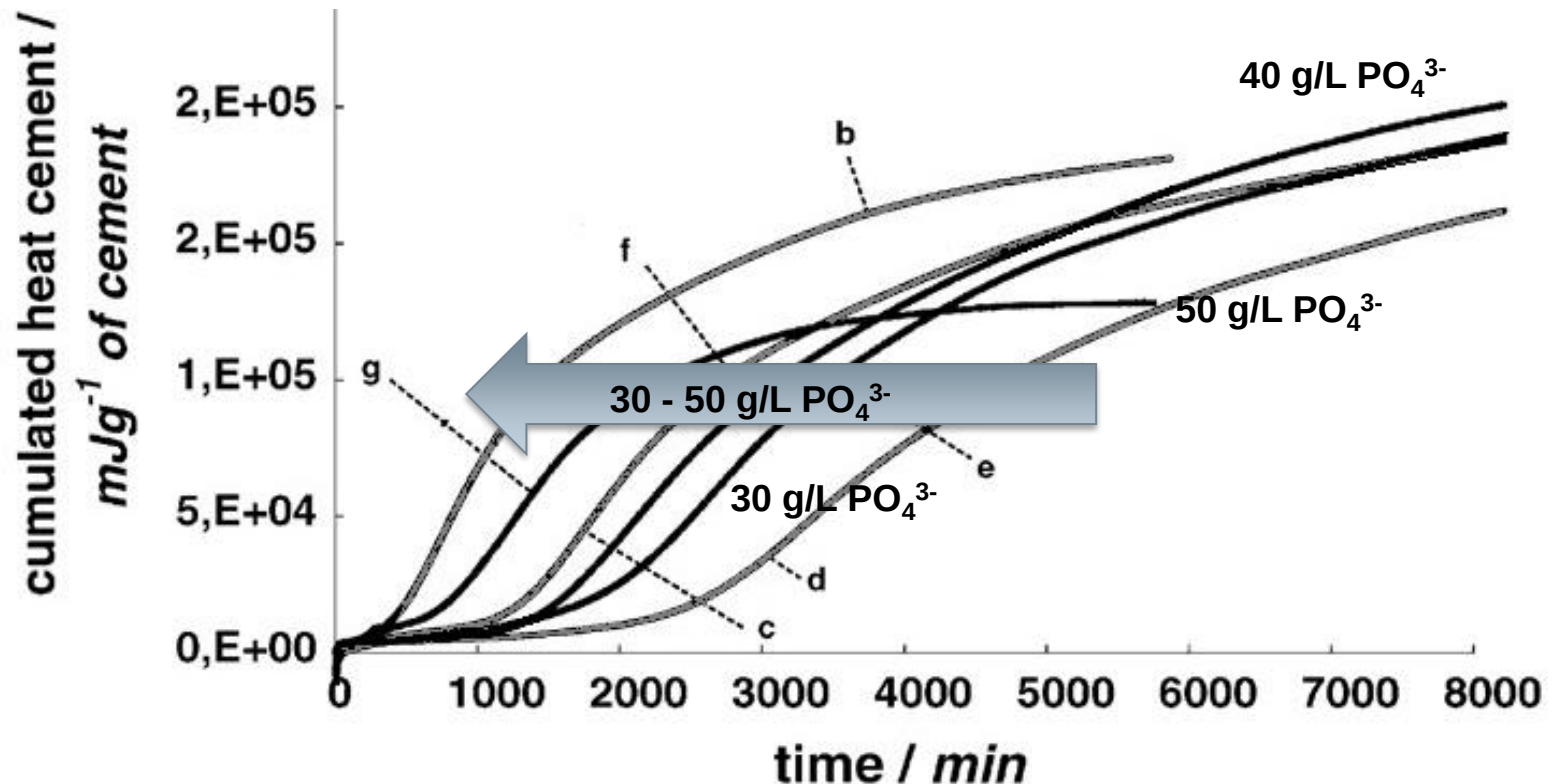


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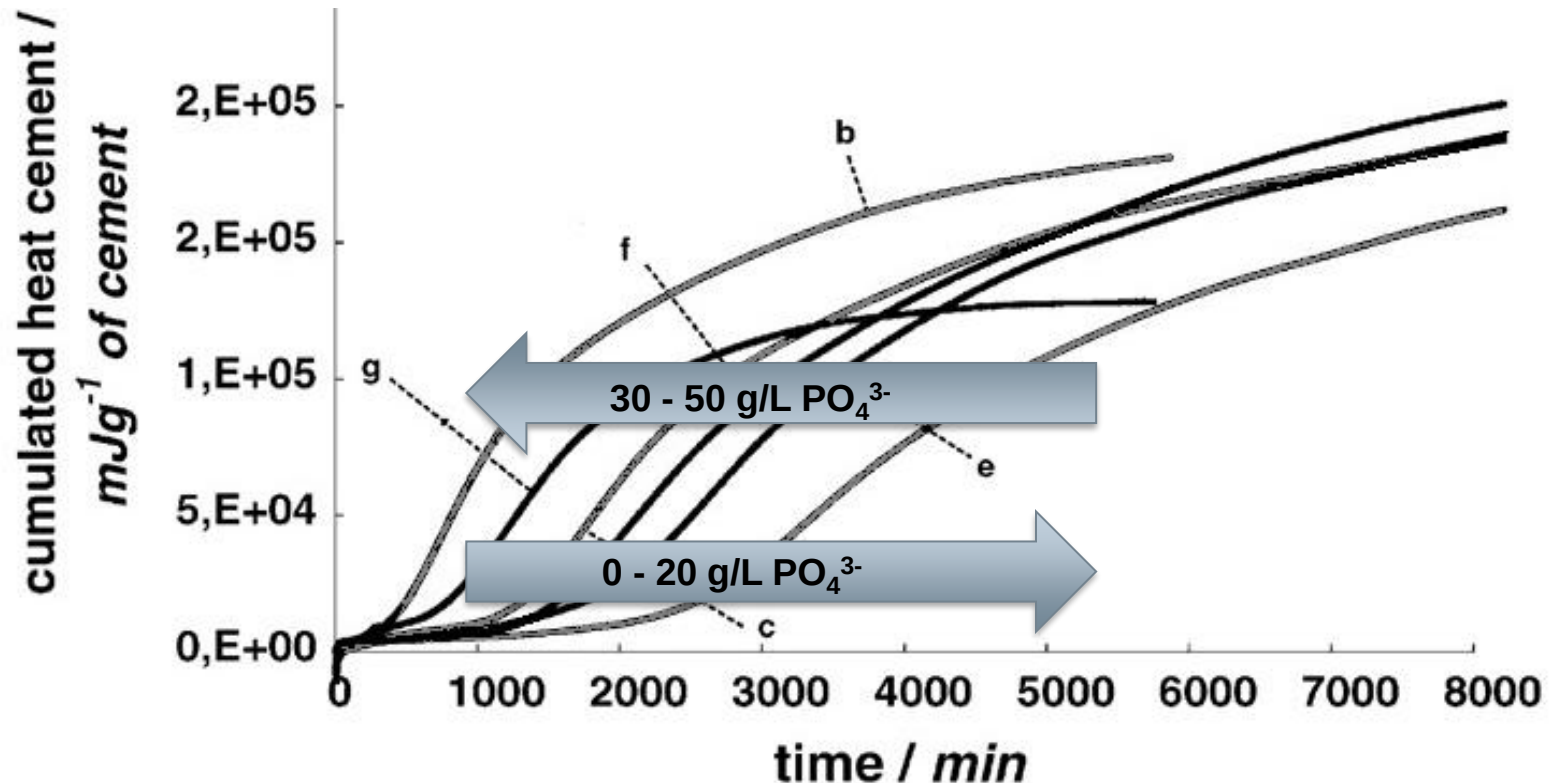


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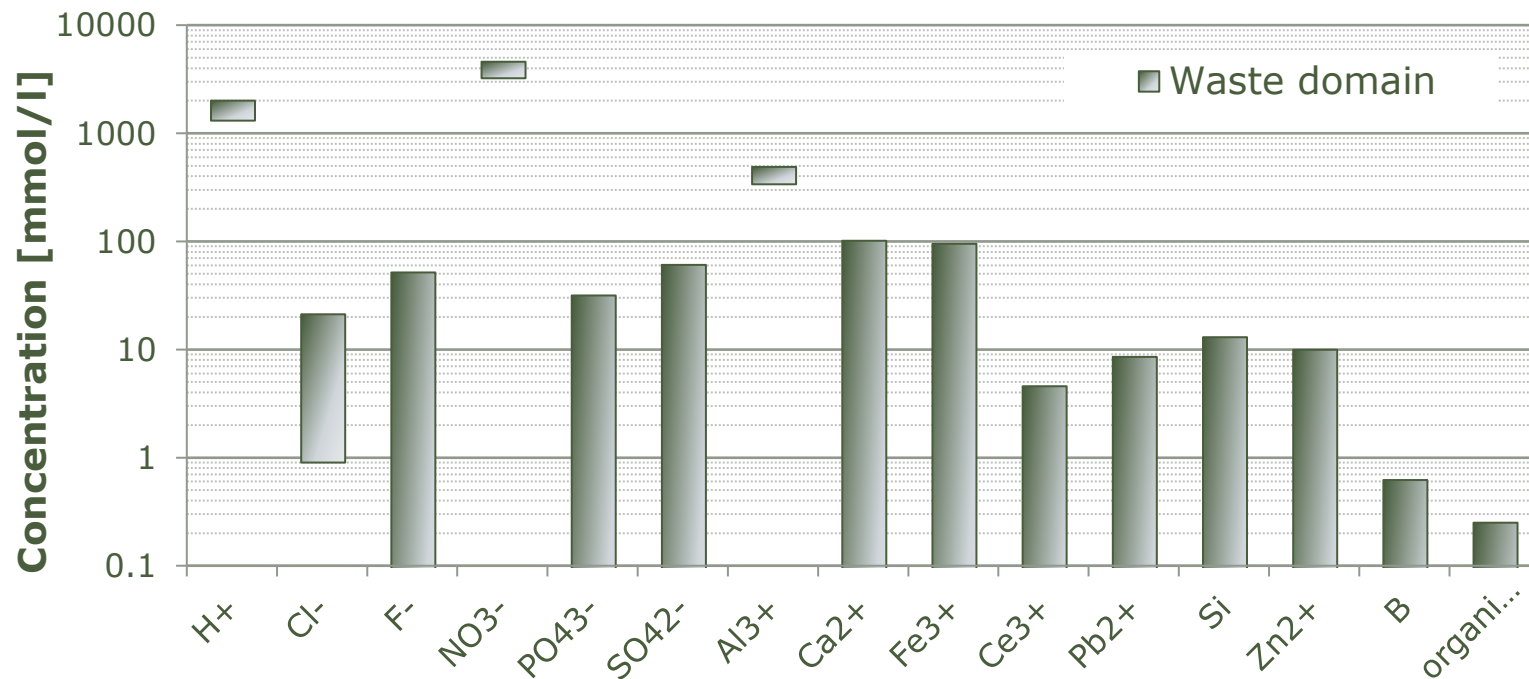
1. Most available data on the interaction of salts with cement are relative to each species taken separately from the others. Although interactions, synergy and antagonism exists.
2. Unexpected concentration effects are sometimes observed.
3. The system itself is not constant. Waste composition can vary in function of time, batch by batch, affecting the behavior of the (intermediate) waste forms. **Example: the concept of a waste domain.**

The Origin of Challenges

Example: Waste domain – variation in brute waste

(from E. Coppens, P. Antonucci and C. Cau-dit-Coumes

WM2016 Conference, March 6-10, 2016, Phoenix, Arizona, USA.)



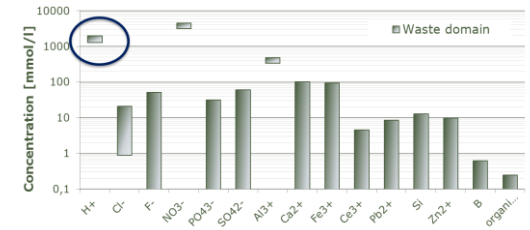
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4. Unexpected kinetic effects are sometimes observed. **Example: pretreatment of acidic waste before cementation.**

The Origin of Challenges



Example: Pretreatment of the waste - Kinetic effects

Raw waste + base $\xrightarrow{\text{Day after}}$ towards cementation

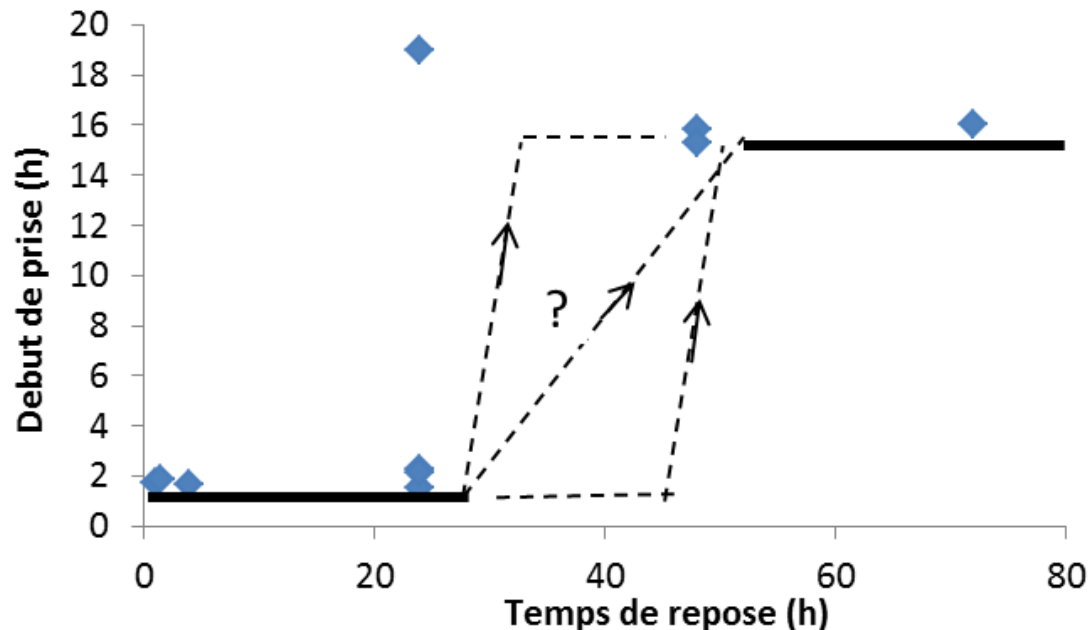
$\xrightarrow{\text{After the weekend}}$

$\xrightarrow{\text{After the prolonged weekend}}$

The Origin of Challenges

Example: Pretreatment of the waste before cementation

Raw waste + base $\longrightarrow$ towards cementation



(from CEA O/N-BP R&D on homogenous cementation)

The Origin of Challenges

How come these problems are (often) unforeseen?

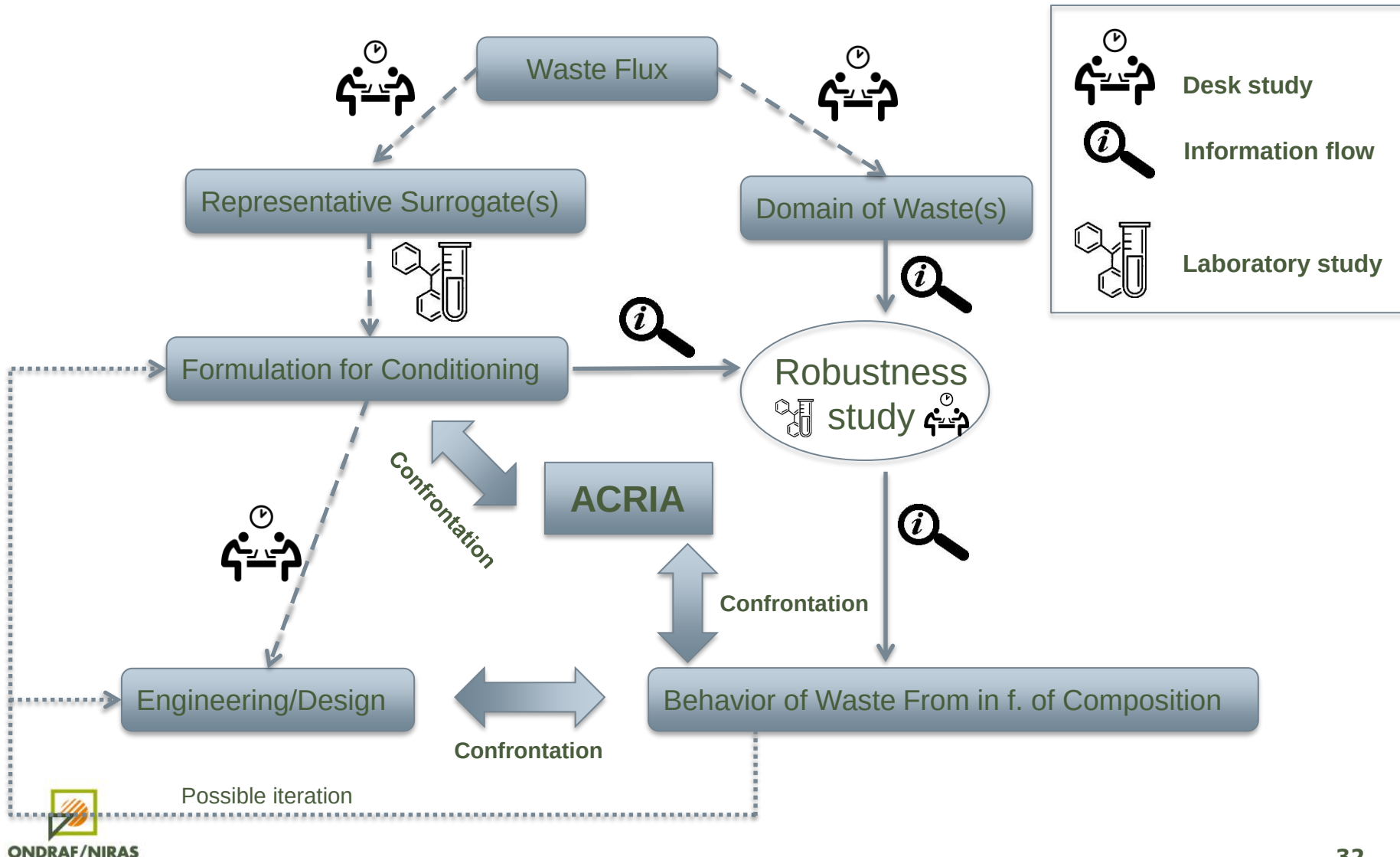
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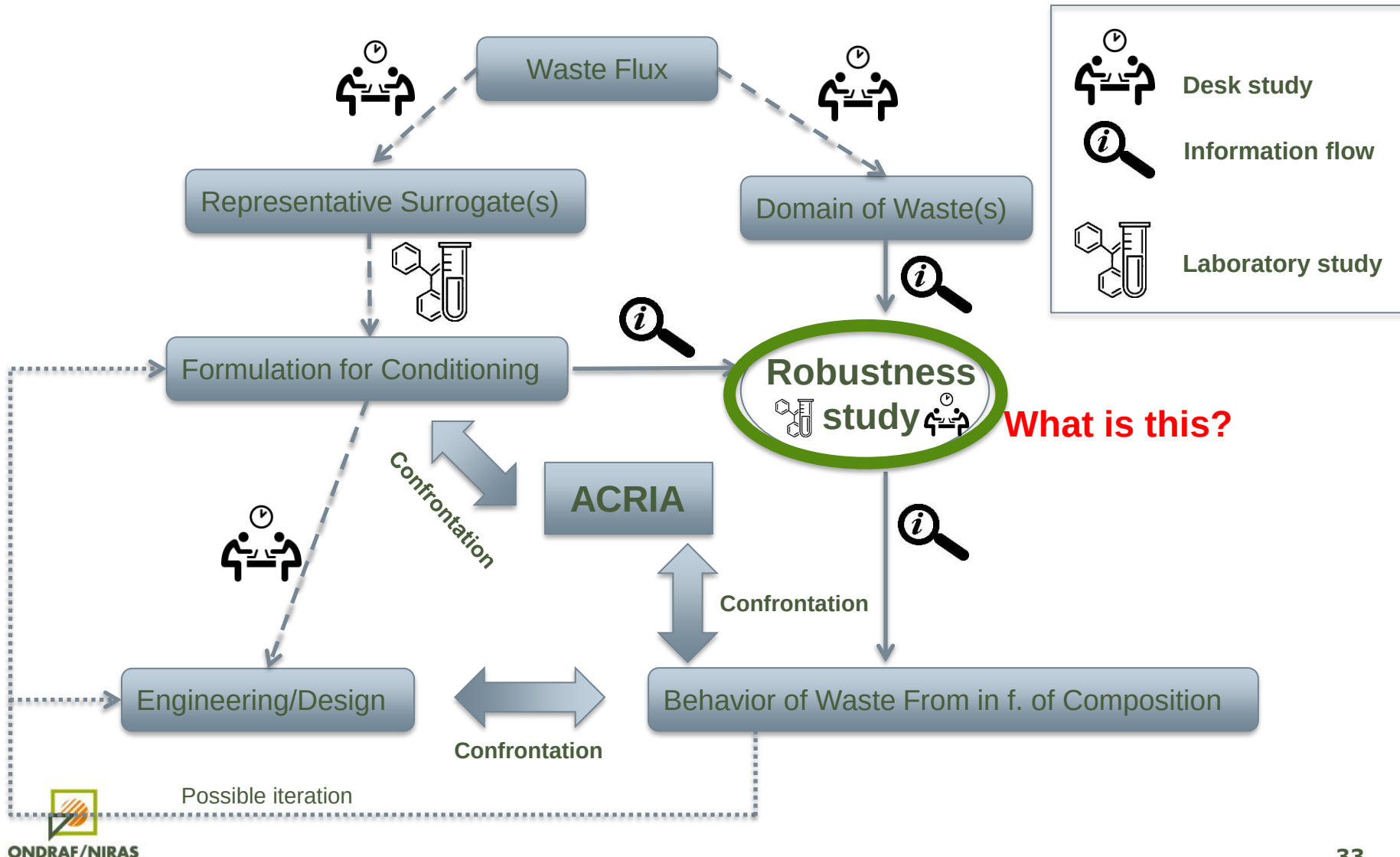
Conditioning of Radioactive Waste by Homogenous Cementation

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How to approach the developpement of a Homogenous Cementation Process



How to approach the developpement of a Homogenous Cementation Process



Robustness Study

Checking the whole waste domain for incompatibilities by making use of non-radioactive **surrogates**.

Often proposed approaches

- **Intuitive approach**
- **Study by changing one factor at a time**

Preferred approach

- **By DOE 'Design Of Experiments'**

Robustness Study

Checking the whole waste domain for incompatibilities by making use of non-radioactive **surrogates**.

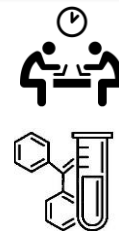
Often proposed approaches

- **Intuitive approach**
- **Study by changing one factor at a time**

Preferred approach

- **By DOE 'Design Of Experiments' in two stages**
 1. Identification of the **main influencing species** within the domain of study
 2. A surface response investigation, focused on the main influencing species to **build predictive models for predicting properties** of the cement-waste form as function of the waste composition.
 - to confront with imposed criteria
 - To confront with engineering / design

Robustness Study



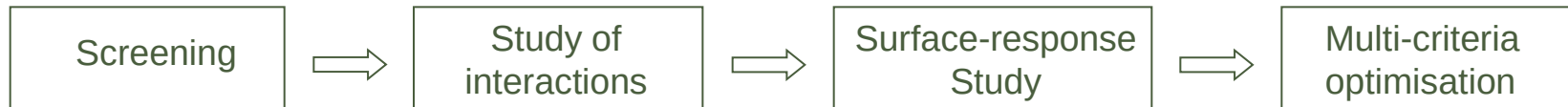
Desk study

Laboratory study

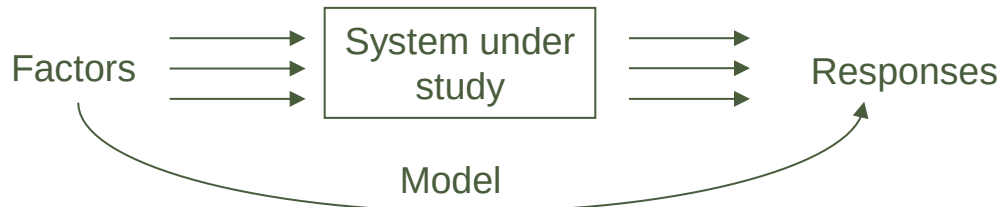
Main Principles of Design Of Experiments (DOE)



1. Defining your objective



2. Define factors, experimental domain, and responses

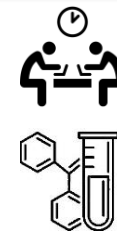


3. Choose a model

- Empirical
- In function of your objective

Objective	Postulated model
Screening	$\eta = \beta_0 + \sum_{i=1}^k \beta_i x_i$
Study of Interactions	$\eta = \beta_0 + \sum_{i=1}^k \beta_i x_i + \sum_{i=1}^{k-1} \sum_{j=i+1}^k \beta_{ij} x_i x_j$
Surface Response Study	$\eta = \beta_0 + \sum_{i=1}^k \beta_i x_i + \sum_{i=1}^{k-1} \sum_{j=i+1}^k \beta_{ij} x_i x_j + \sum_{i=1}^k \beta_{ii} x_i^2$

Robustness Study



Desk study

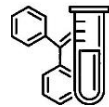
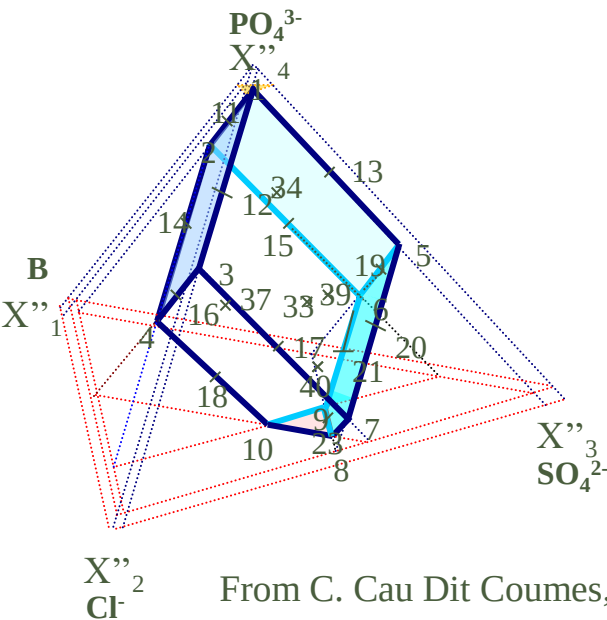
Laboratory study

Main Principles of Design Of Experiments (DOE)



4. Choosing the experimental matrix.

The experiments must be chosen / localized in the domain of interest (*the waste domain*) in such a way the coefficients of the imposed models can be estimated with significant confidence.



5. Realization of the experiments.



6. Calculation of the estimates for the models coefficients.



7. Validation of the model.



8. Interpretation of the results.

From C. Cau Dit Coumes, S. Courtois, *Cement and Concrete Research*, **33** [3] (2003) 305-316



Conditioning of Radioactive Waste by Homogenous Cementation

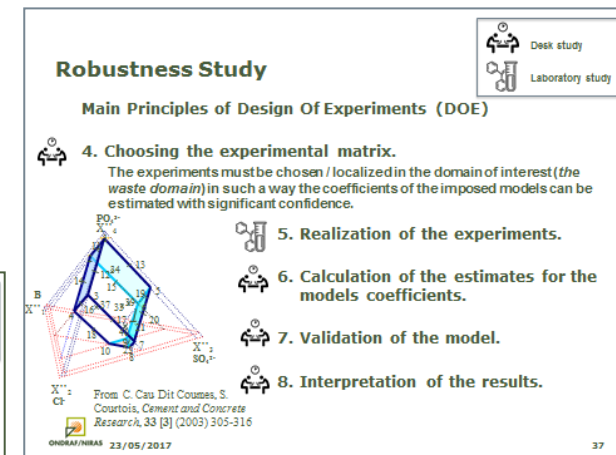
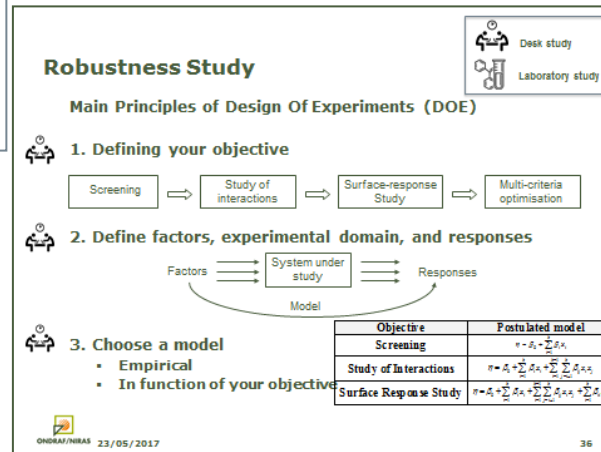
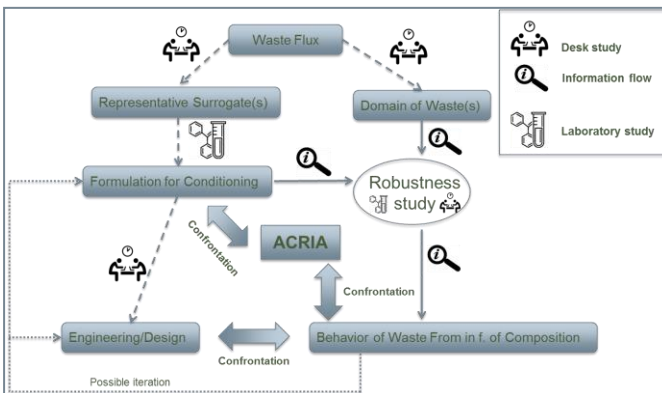
- Introduction
- The challenges and their origin
- How to develop a cementation process
- Conclusion

Conclusion

Main problem: Knowledge of the effect of mixing cement with concentrates of complex chemistry is very limited

Thus: Cementation might look easy, it is not.

However: A well thought-through approach allows for developing cementation processes in a rigid way.



Robustness Study

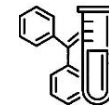
Example applied to homogenous cementation

From: S. Courtois, C. Cau Dit Coumes, *Proc. Chimiométrie 2007, Lyon, 29-30 Nov (2007)*

Forget about the chemistry and approach the problem as an empirical mathematical black box.

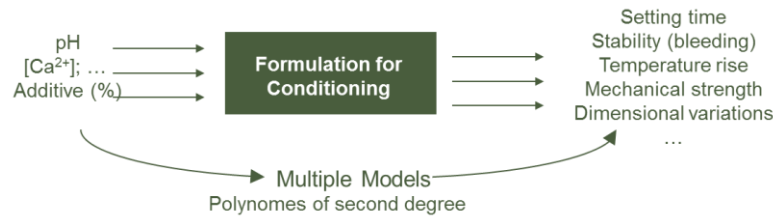


Desk study



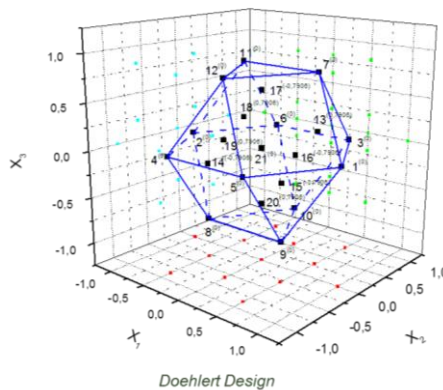
Laboratory study

Step 1



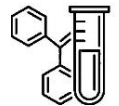
- 1.1 Define input and output of the 'black box'
- 1.2 Postulate an empirical mathematical model: polynomes of second order

Step 2



2. Situate the experiments in the waste domain to extract a maximum of information with a minimum amount of experiments.

Step 3



3. Execute the experiments and measure « output »

Step 4

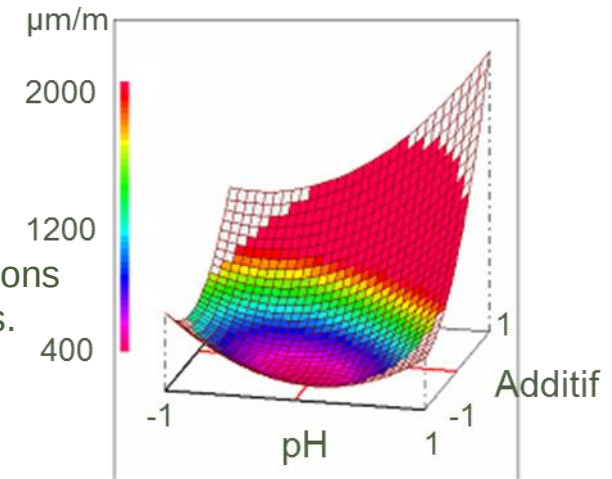


4. Calculate model-coefficients and validate the models

Step 5



5. Study the predictions made by the models.



Expansion after 120 days in water

The Origin of Challenges

Example: Waste domain – variation in brute waste



Information gathering



NIRAS Nationale instelling voor radioactief afval
an. verspreide activiteiten

BELG PROCESS

NIRAS Kunstlaan 14 1210 Brussel Belgoproces Gravenstraat 73 2480 Dessel

Document NOTA

Referentie	NIRAS	Belgoproces	NIRAS-Belgoproces
0	N/A		

Herziening	NIRAS	Belgoproces	NIRAS-Belgoproces
2		UB7/2015-00588	2016-3252

Raden herziening

- 1 H⁺ concentraties were exaggerated due to a misinterpretation of raw data
- 2 New information from additional measurements is incorporated. Changes to provisions are made due to a change in production of a third party and the closure of reservoir 540-13 from any other waste stream

Datum	Pagina's	Bijlage(n)
11/2016	24	71 p

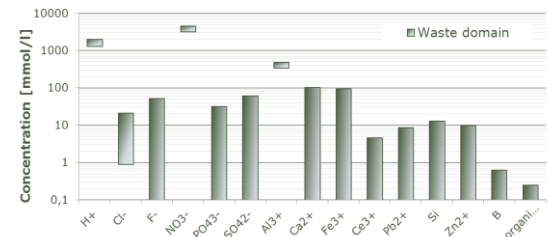
Titel / Onderwerp
Source term PD47 – Homogenous cementation

Samenvatting
This note describes the provisions of the content of Tank 540-13 in function of time. Tank 540-13 is expected to store all liquid intermediate level waste that is intended to be conditioned by homogenous cementation. A significant volume of waste is yet stored in this tank, but more waste streams shall be added in the (near) future. This note aims to deliver sufficient input towards R&D departments for the development and validation of a homogenous cementation process of these wastes.

Auteur(s)	Paraaf	Verificateur(s)	Paraaf	Goedgekeuring	Paraaf
Wouter Aerts		N. Van Hoof		W. Notelzeers	
Erik Coppens		B. de Blochouse		G. Vanzaelen	

Full description of unconditioned waste + uncertainties

Distillation of minimal and maximal concentrations



Long-term durability of cementitious materials: degradation mechanisms

21st Exchange Meeting ESV EURIDICE GIE

Quoc Tri Phung

R&D Disposal, Expert Group Waste & Disposal

qphung@sckcen.be



STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

Introduction

Concrete?

Degradation

Cement (11-13%)



Coarse Agg. (48-50%)



Fine Agg. (32-34%)



Water (6-7%)



SCMs, SP



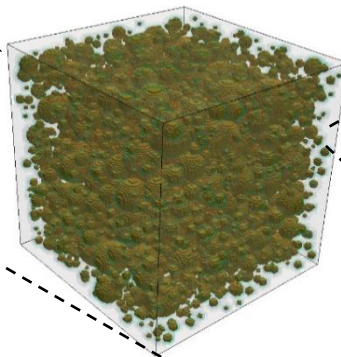
Casting/curing



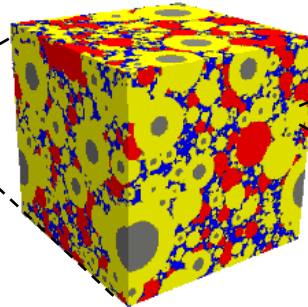
Fresh concrete



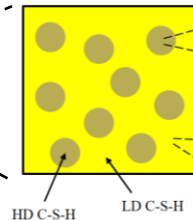
Macro



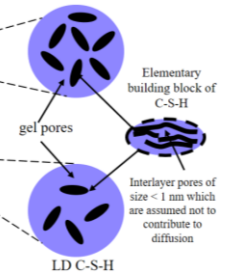
Meso



Micro



Nano



C-S-H



Pore

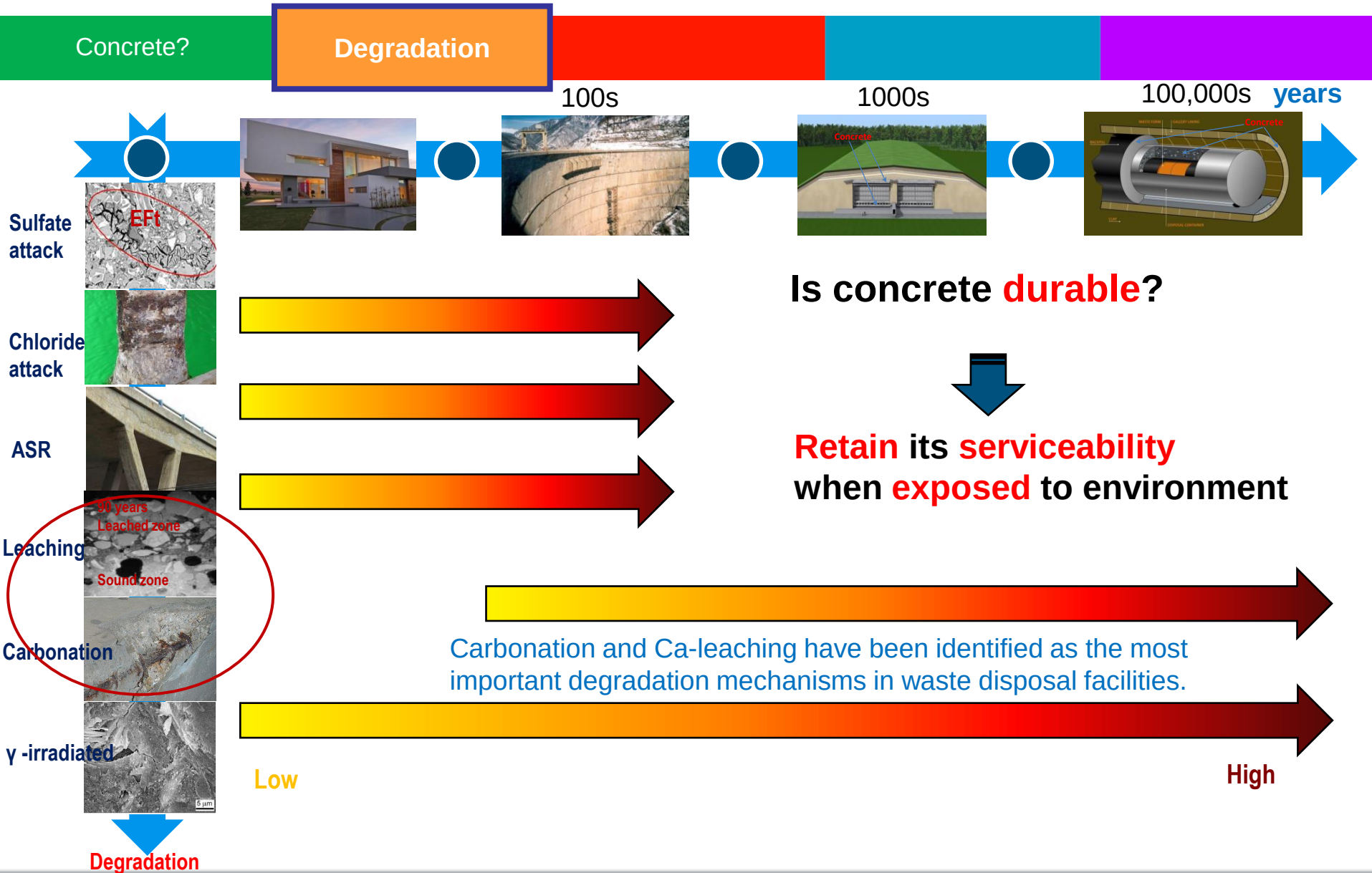


Clinker



Portlandite

Introduction

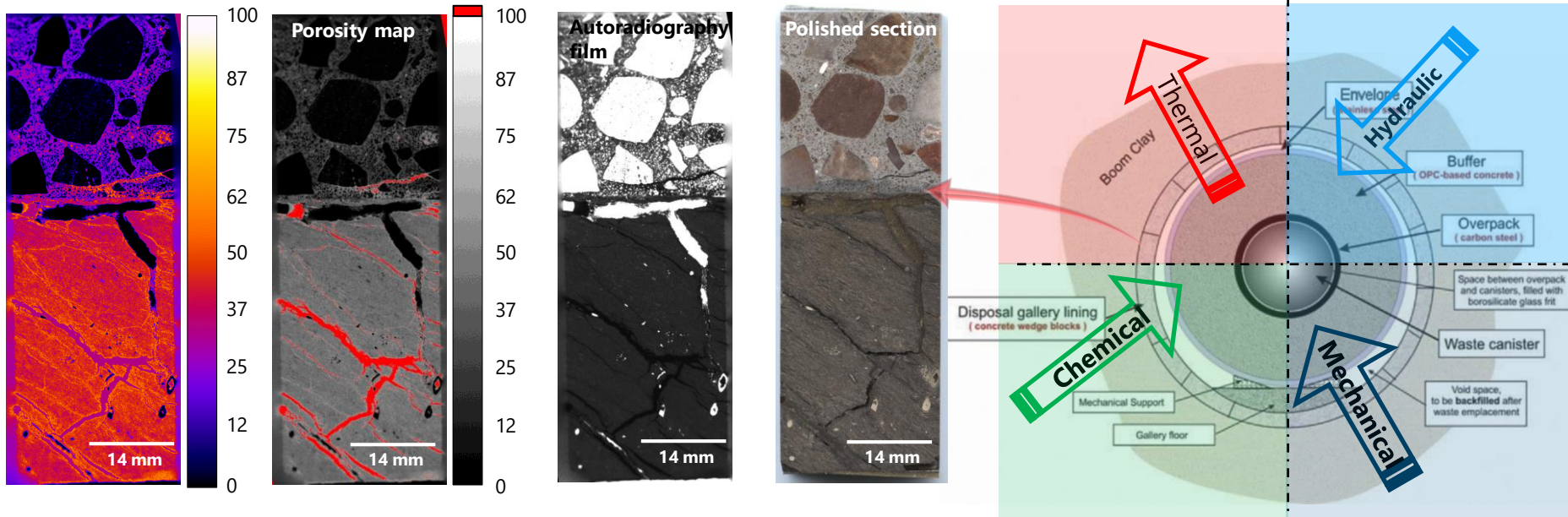


Concrete?

Degradation

● Challenges for concrete durability assessment in nuclear related structures

- Long-term performance (1000s yrs for waste disposal, up to 100 yrs for NPP)
- Coupled effects
- Exposed conditions changed
- Limited experimental timeframe vs. long-term processes



Leaching

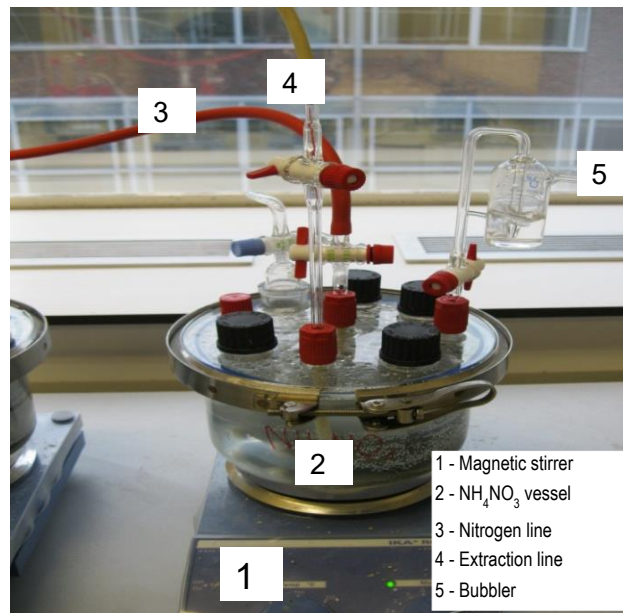
Carbonation

ASR & DEF

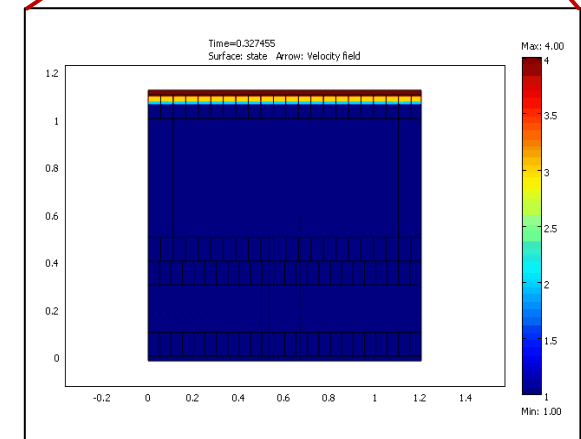
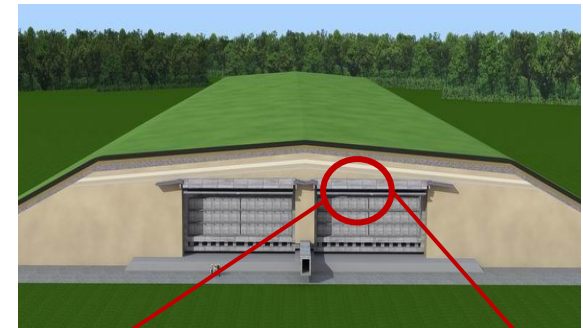
Radiation damage

Ca-leaching [Jacques 2008; Phung 2014a,b, 2015a,b]

- A process of Ca extraction from solid phases
- Relevant for extreme long lifetime structures: nuclear waste disposal
- Extremely slow process → accelerated Ca-leaching using NH_4NO_3



Near surface disposal @ Dessel, Belgium



state I state III
state II state IV

Degradation mechanisms

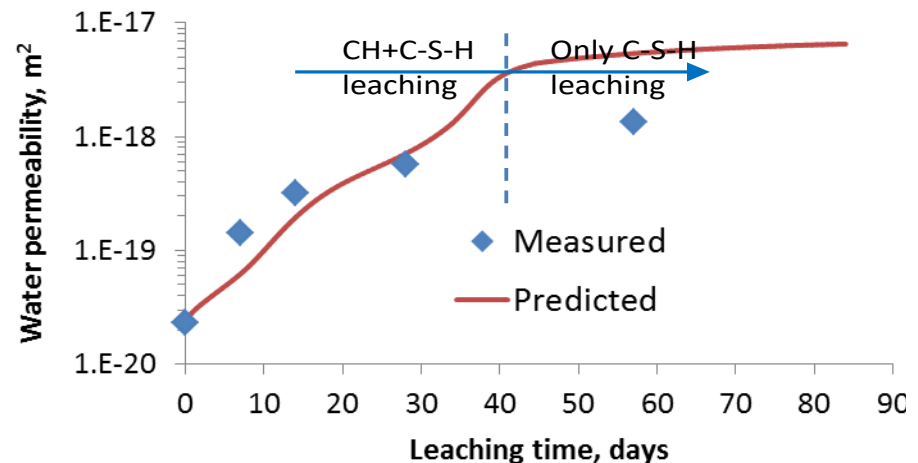
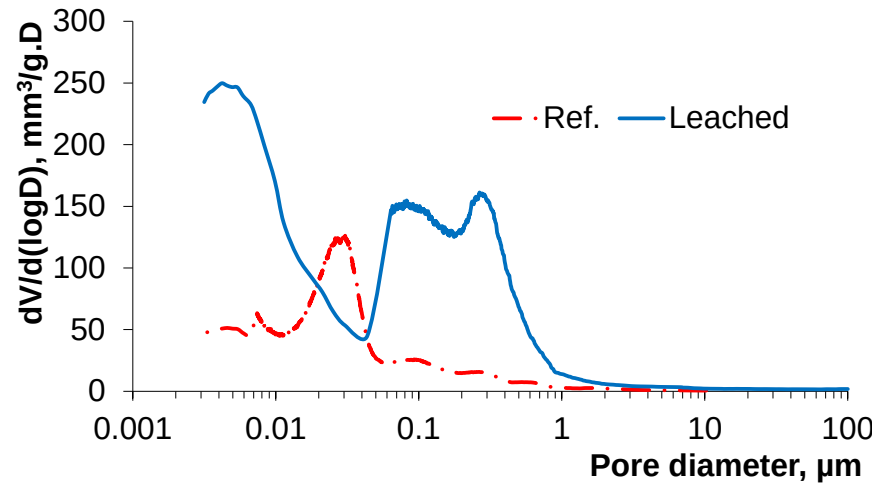
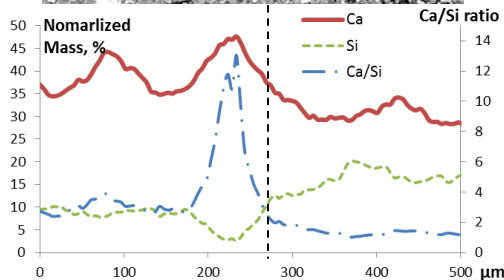
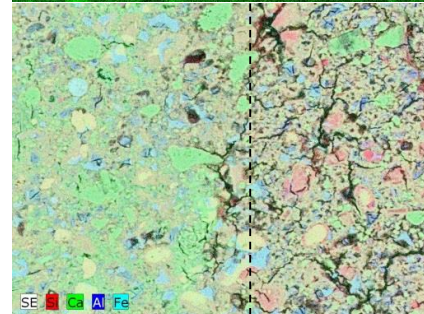
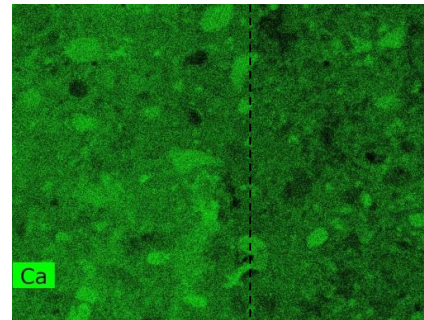
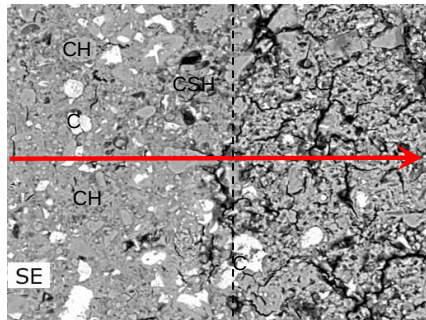
Leaching

Carbonation

ASR & DEF

Radiation damage

- Detrimental effects:
 - pH decrease;
 - Mechanical properties loss;
 - Coarser μ -structure; transport properties increase



Degradation mechanisms

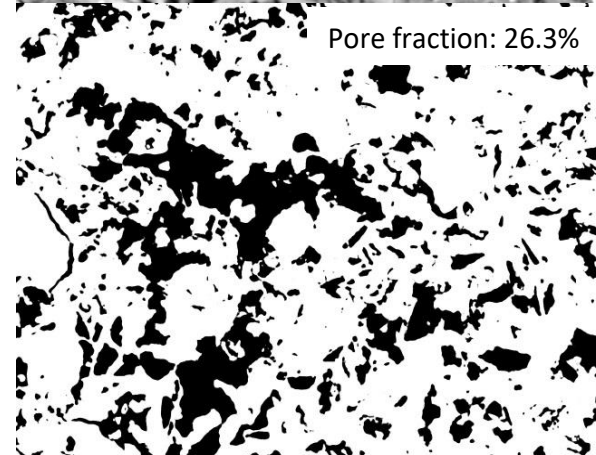
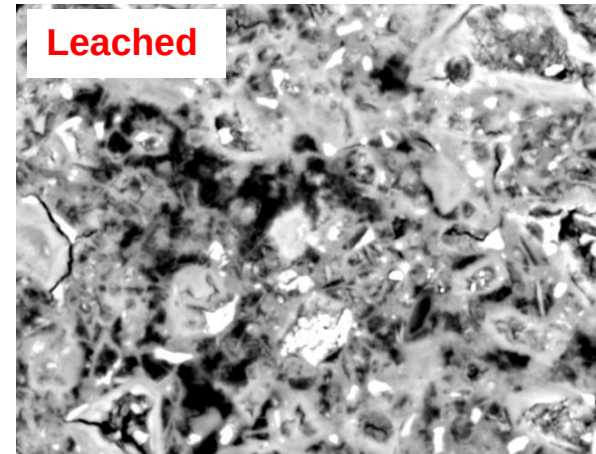
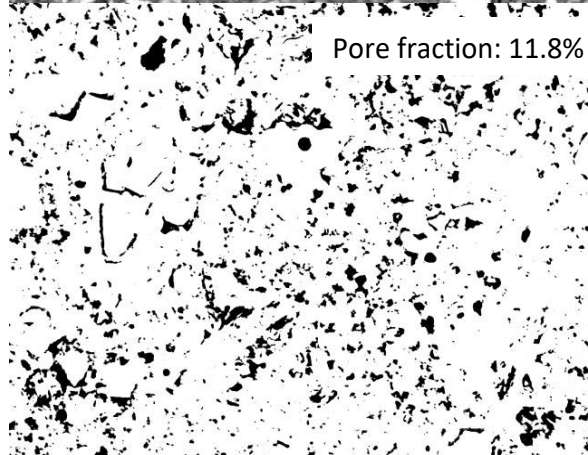
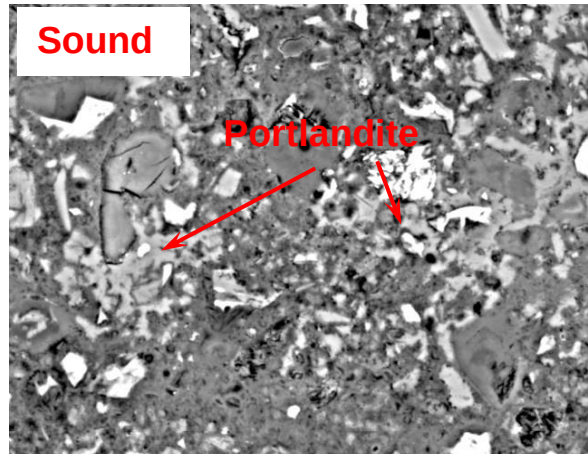
Leaching

Carbonation

ASR & DEF

Radiation damage

- Detrimental effects:



Leaching increases the pore sizes, porosity and percolation.

Leaching

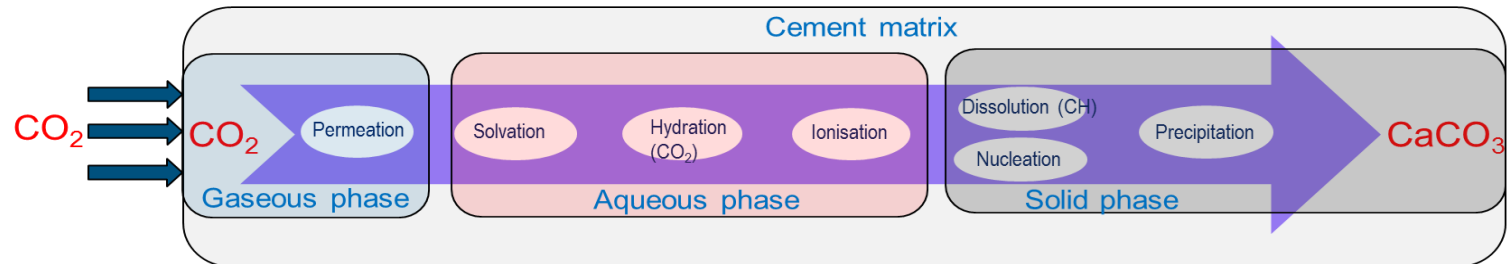
Carbonation

ASR & DEF

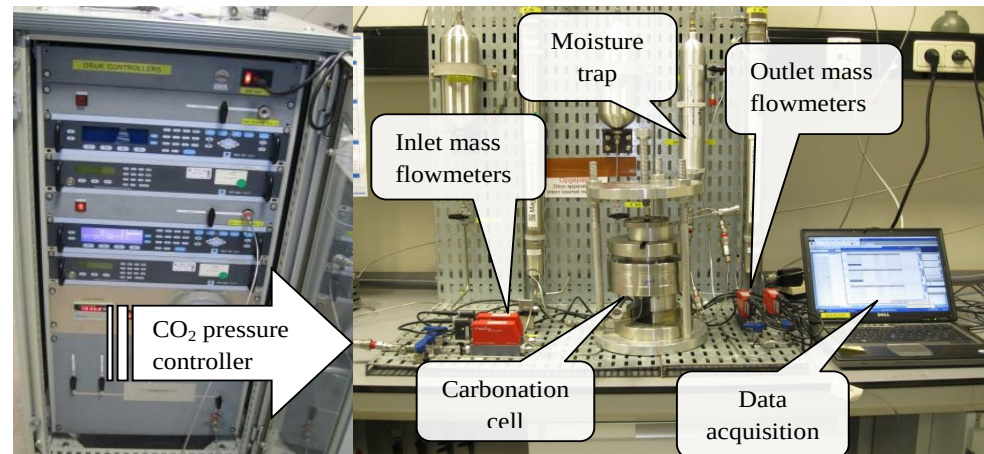
Radiation damage

Carbonation [Phung 2013a, 2015a,c]

- $\text{Ca}^{2+} + \text{CO}_3^{2-} \rightarrow \text{CaCO}_3$
- Complicated physicochemical phenomenon



- Slow process → accelerated carbonation under controlled CO_2 pressure



Leaching

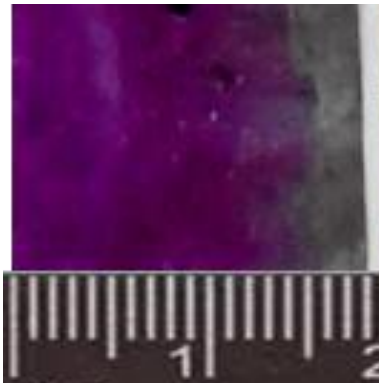
Carbonation

ASR & DEF

Radiation damage

Carbonation [Phung 2013a, 2015a,c]

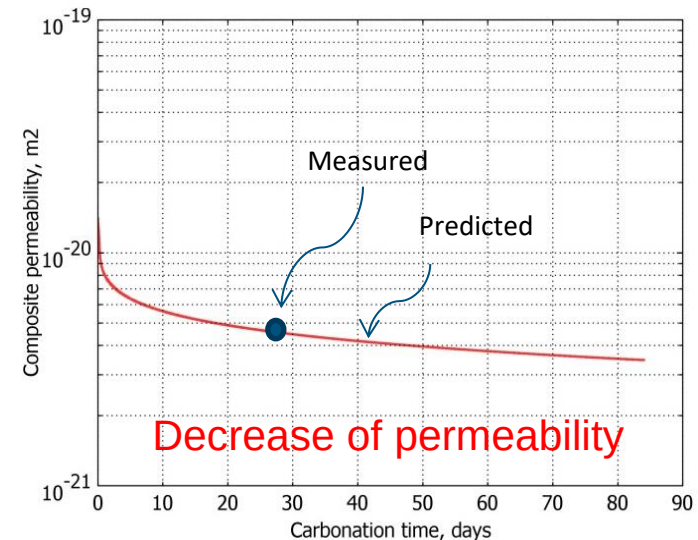
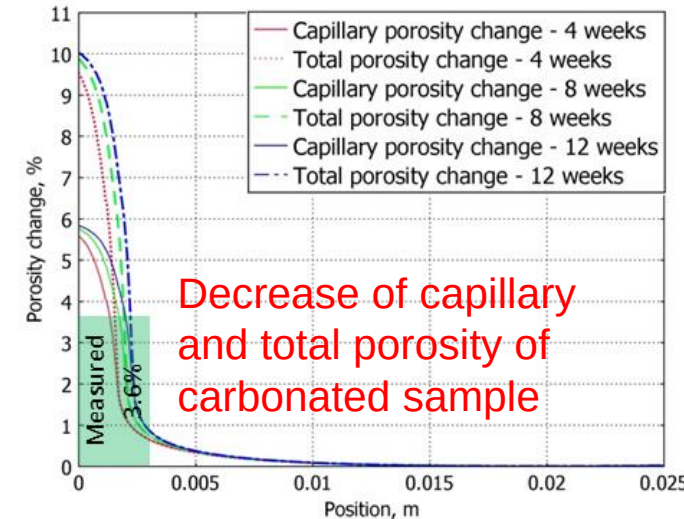
- Consequences:
 - pH decrease → corrosion,
 - Beneficial effects on the leaching/transport processes and strength



pH drop



Hippodrome Wellington, Belgium



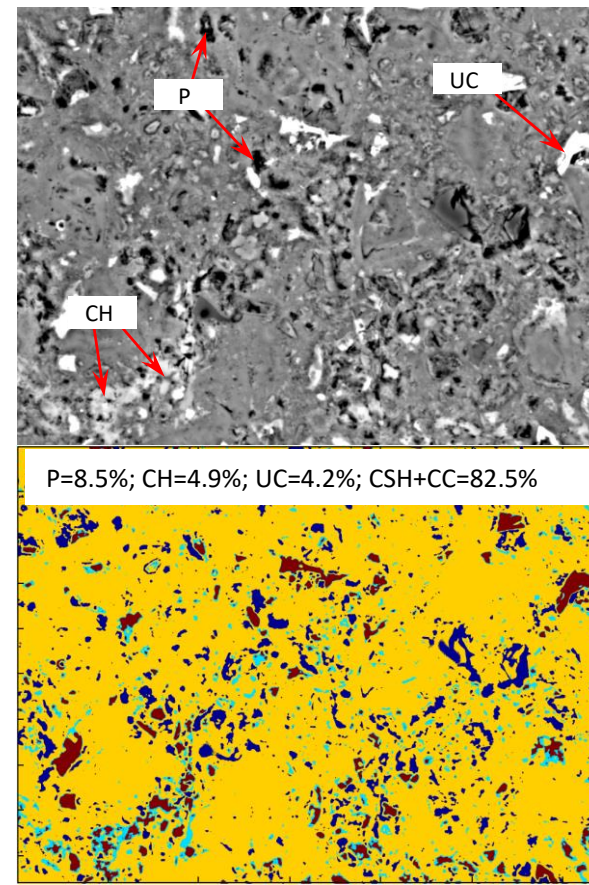
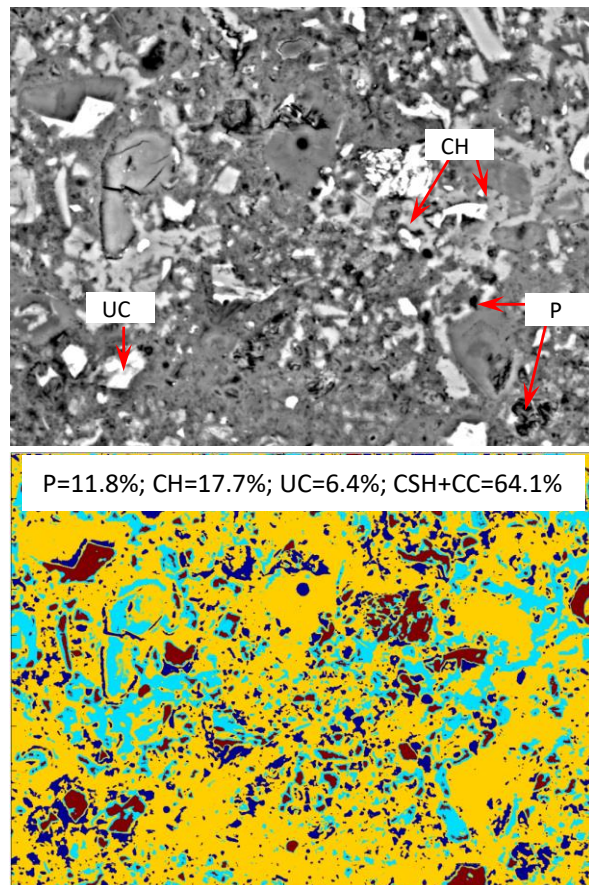
Leaching

Carbonation

ASR & DEF

Radiation damage

Carbonation [Phung 2013a, 2015a,c]



Carbonation slightly decreases the porosity, portlandite is carbonated but still observable after carbonation.

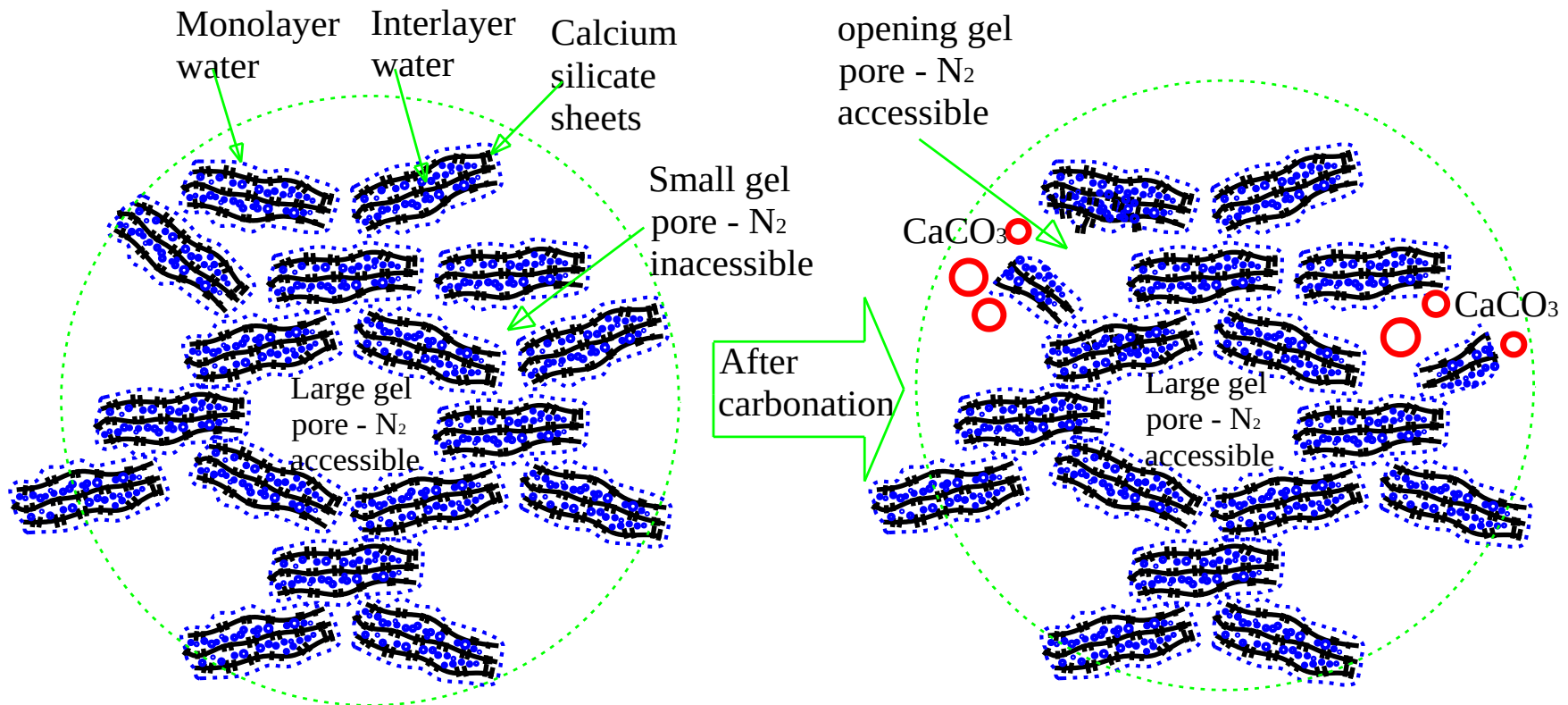
Leaching

Carbonation

ASR & DEF

Radiation damage

- **Contribution of C-S-H to degradation processes:**
 - C-S-H carbonation results in opening gel pores.



Leaching

Carbonation

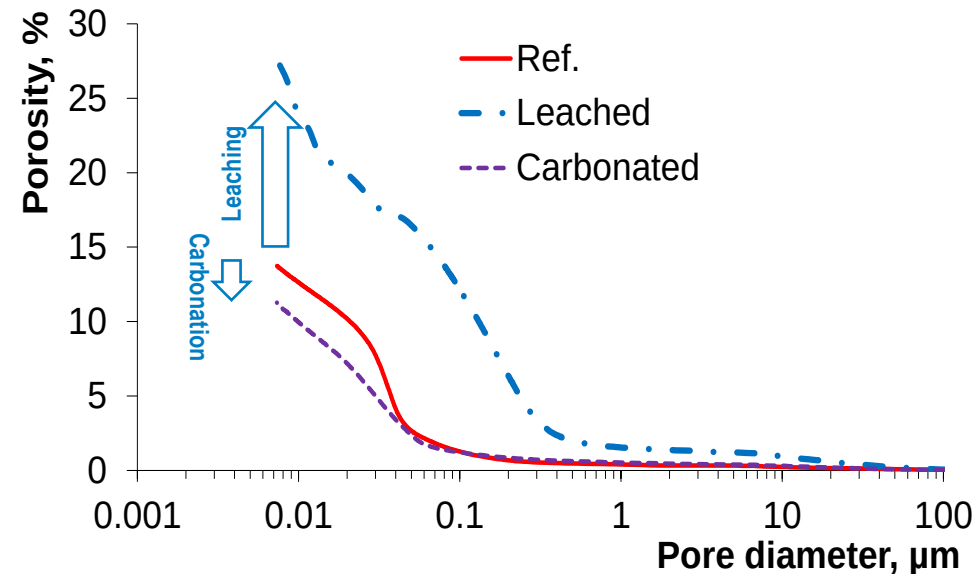
ASR & DEF

Radiation damage

● Carbonation vs. Leaching

- Carbonation and leaching significantly change the microstructure of cementitious materials but in opposite ways.
 - Finer microstructure for carbonated materials
 - Coarser microstructure for leached materials
 - C-S-H degradation significantly contributes to both processes

Microstructural parameters	Leaching	Carbonation
Porosity		
Pore size		
S. Surface area		 



Leaching

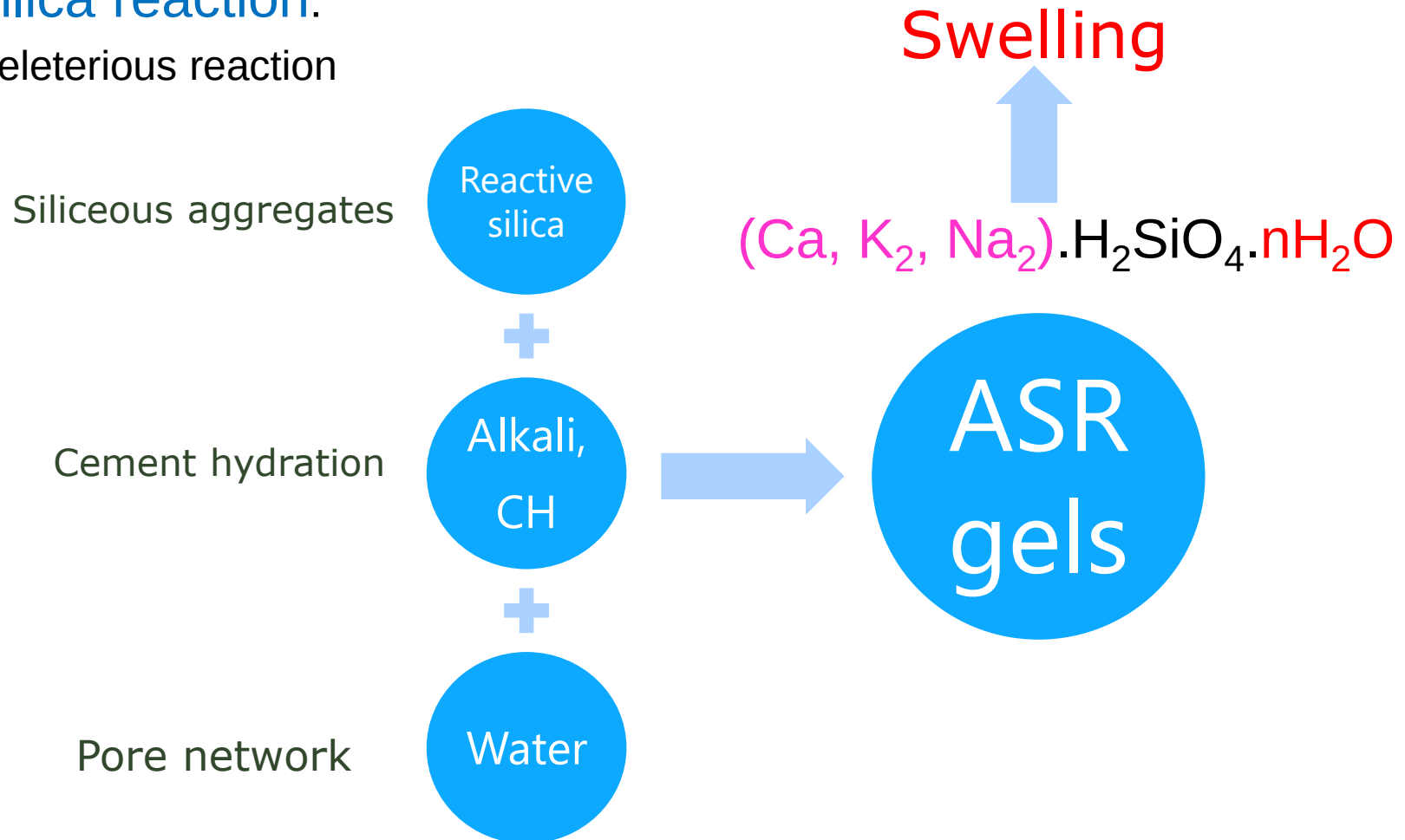
Carbonation

ASR & DEF

Radiation damage

Alkali-silica reaction:

- Deleterious reaction



Leaching

Carbonation

ASR & DEF

Radiation damage

Alkali-silica reaction

- Consequences:
 - Cracking
 - Loss of mechanical properties
 - Aggressive substances ingress
- No feasible method available to stop the reaction but possible to prevent (e.g. limit alkali content, no reactive agg.) → **Concrete cancer**
- Neutron radiation can increase the reactivity of silica-rich aggregates



ASR at containment building of Tihange NPP (a) [FANC 2013], Seabrook NPP (b) [Saouma et al. 2014] and cemented waste drum from Doel NPP (c) [FANC 2014]

Leaching

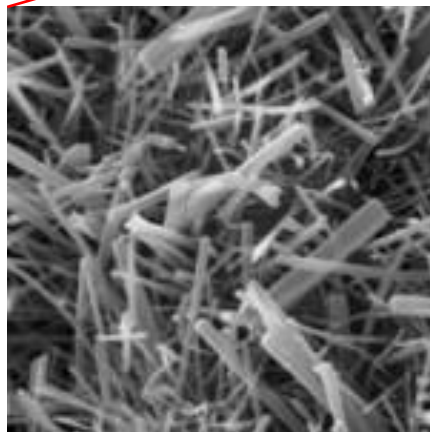
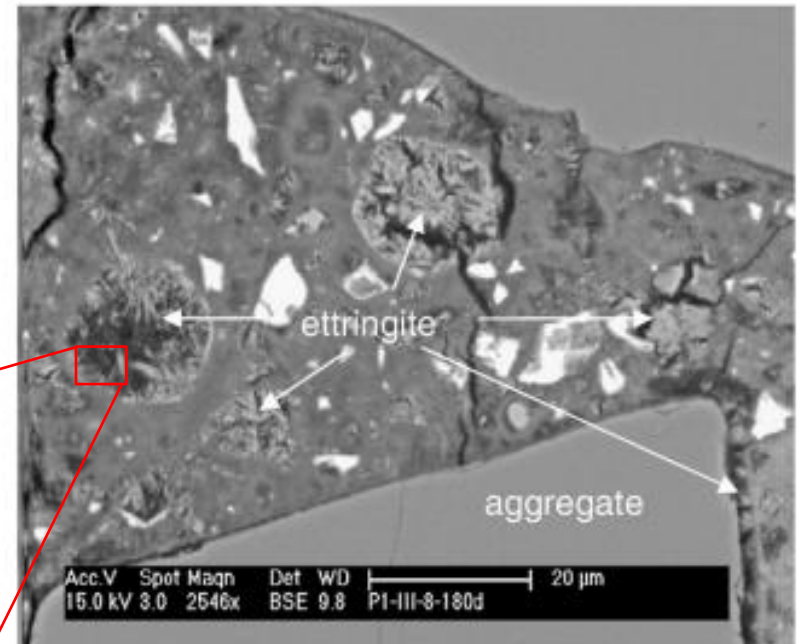
Carbonation

ASR & DEF

Radiation damage

Delayed ettringite formation

- Internal or external sulfates + anhydrous or hydrated calcium aluminates after hardening
- Elevated temperature + humid conditions + high alkali content → accelerated



Leaching

Carbonation

ASR & DEF

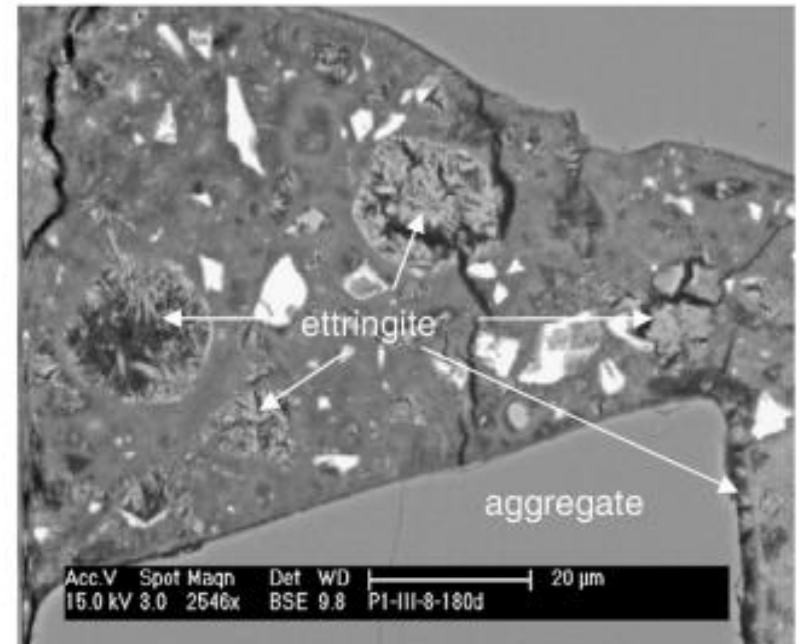
Radiation damage

Delayed ettringite formation

- Consequences:
 - Cracking
 - Loss of mechanical properties
 - Increase transport properties
- High risk for cemented wastes



[Civildigital.com]



Leaching

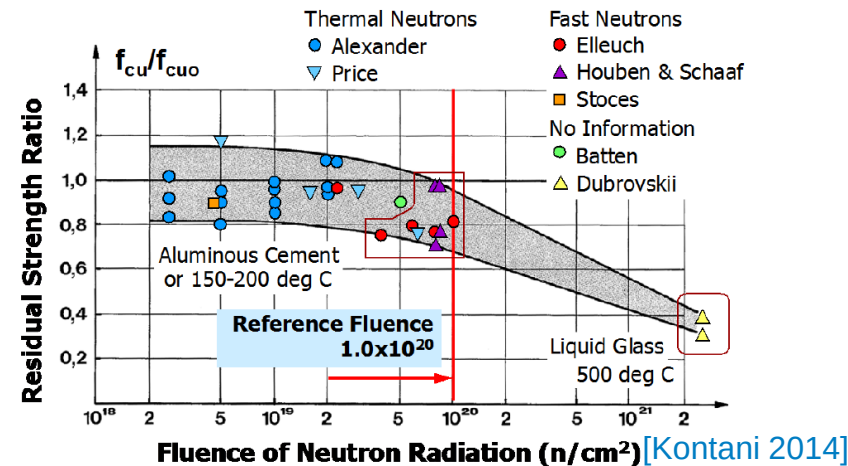
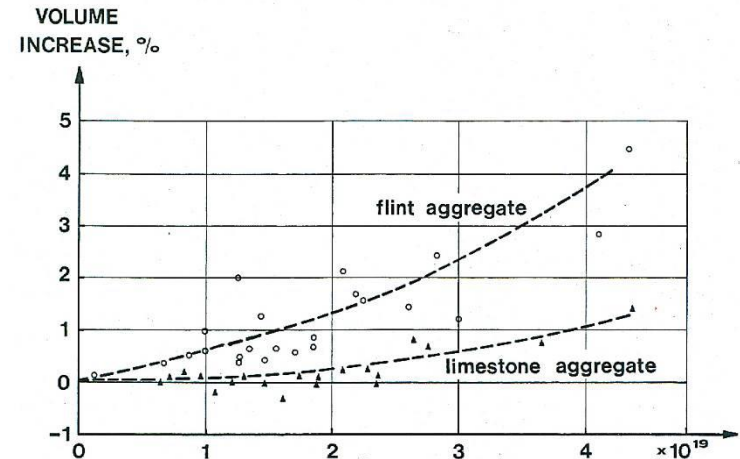
Carbonation

ASR & DEF

**Radiation
damage**

Neutron radiation

- Influence on the solid phases
- Structure of some aggregates ordered $\rightarrow$ disordered crystalline $\rightarrow$ increase in volume.
- Elevated temperature $\rightarrow$ accelerated
- Reactivity of silica-rich quartz can be significantly increased $\rightarrow$ produce ASR in concretes containing nonreactive aggregates



Leaching

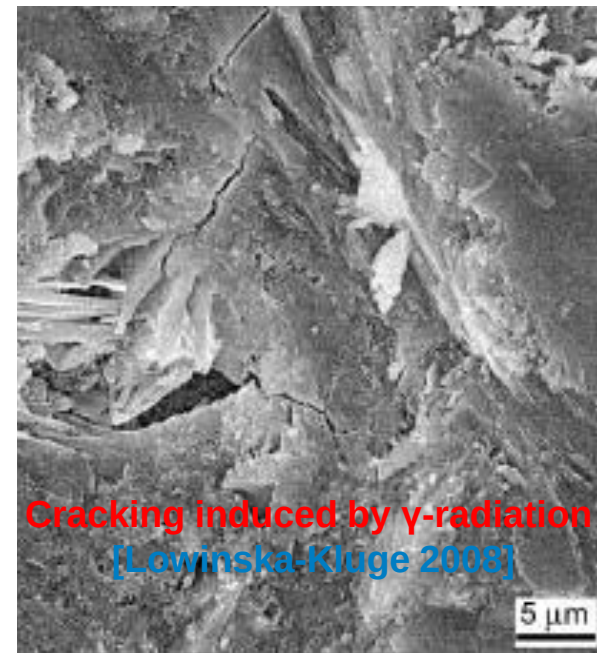
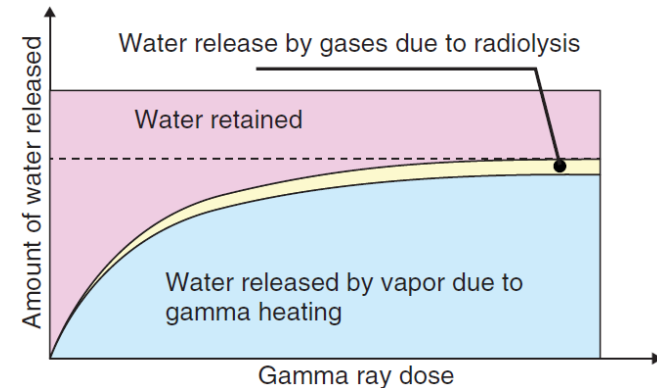
Carbonation

ASR & DEF

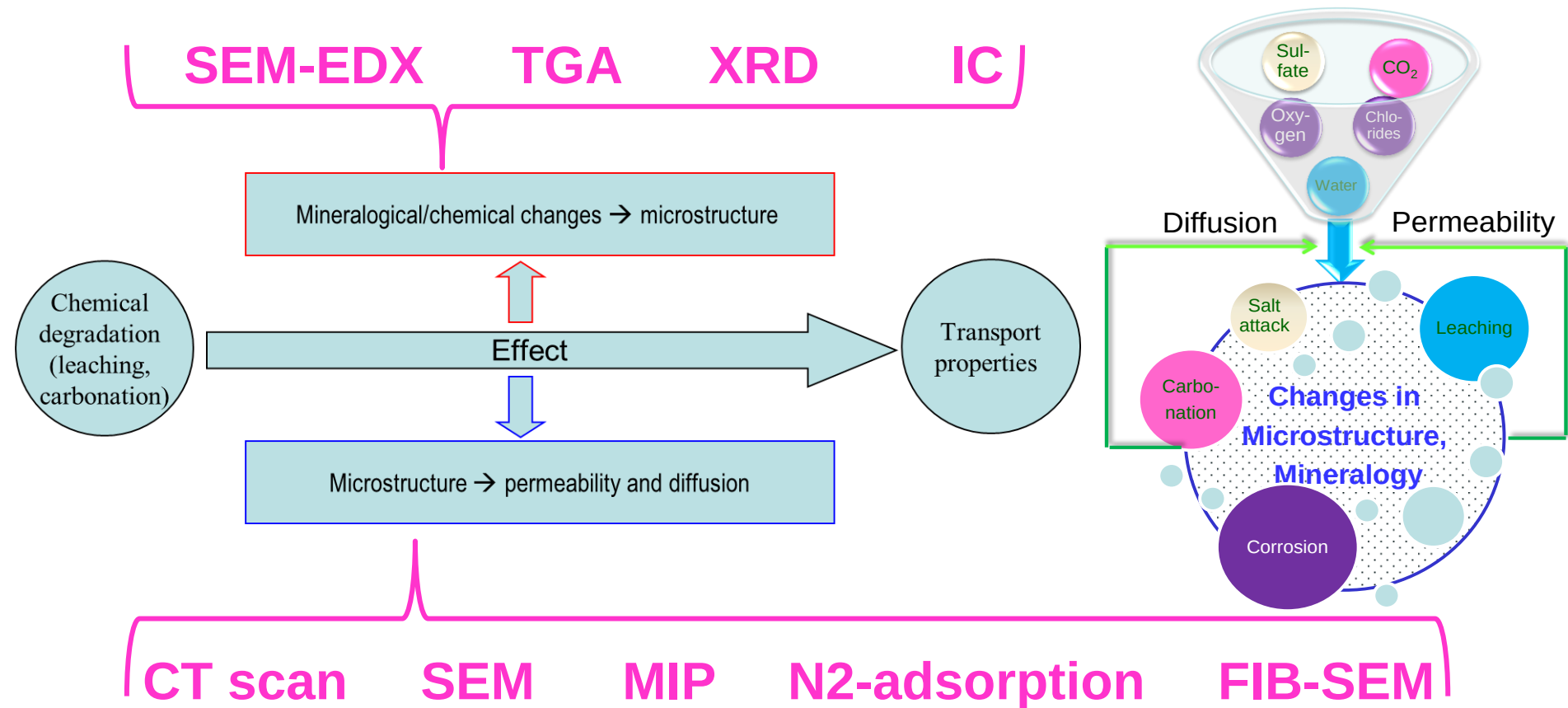
Radiation
damage

Gamma radiation

- Significantly influence the pore solution
- Radiolysis: Water → converted to hydrogen, oxygen, and hydrogen peroxide → react with the cement paste.
- Water evaporation due to heat generated by gamma radiation → shrinkage cracking
- Radiation carbonation (in middle)



Multiple analysis techniques → provide comprehensive information on microstructural and mineralogical changes



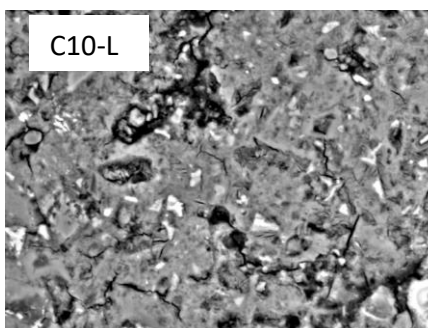
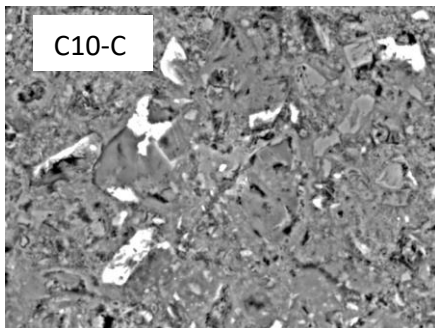
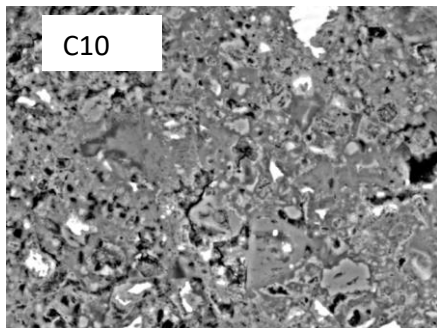
Material characterization

μ-structure & mineralogy

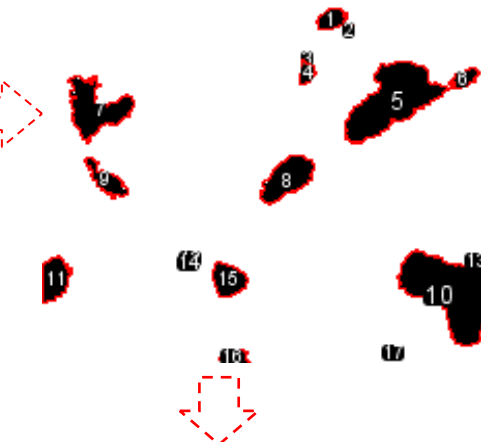
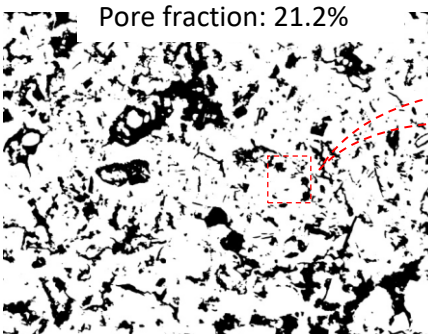
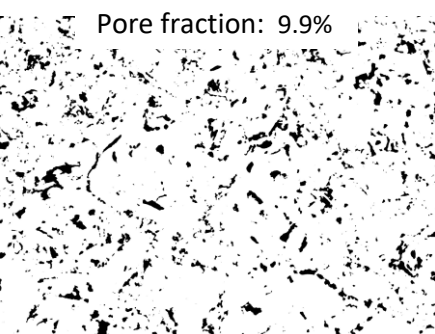
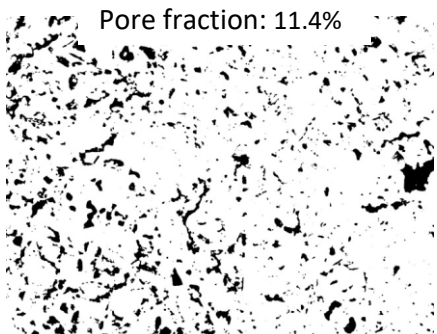
Transport properties

Size range	1 nm	10 nm	100 nm	1 μ m	10 μ m	100 μ m	1 mm
Name of pores according to IUPAC							
Name of pores according to concrete science terminology							
Method							
Mercury intrusion porosimetry							
Gas adsorption							
Water absorption							
Helium pycnometry							
Thermoporometry							
Nuclear magnetic resonance							
Small-angle scattering							
Optical microscopy							
Electron microscopy							

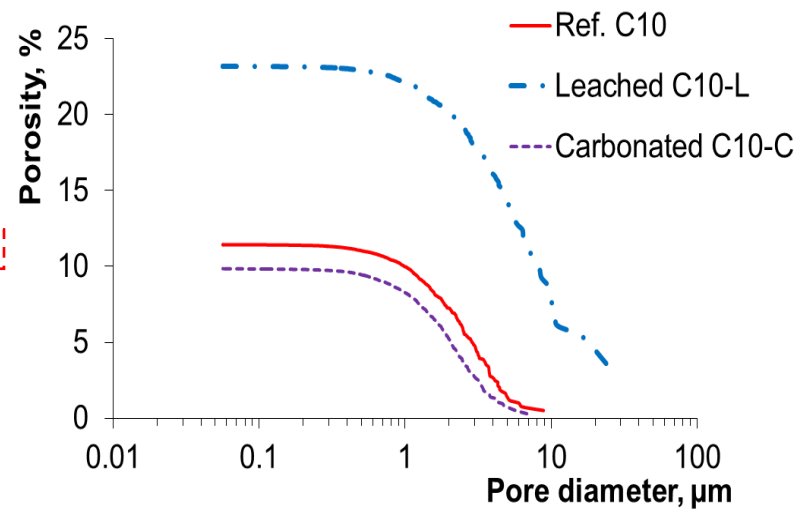
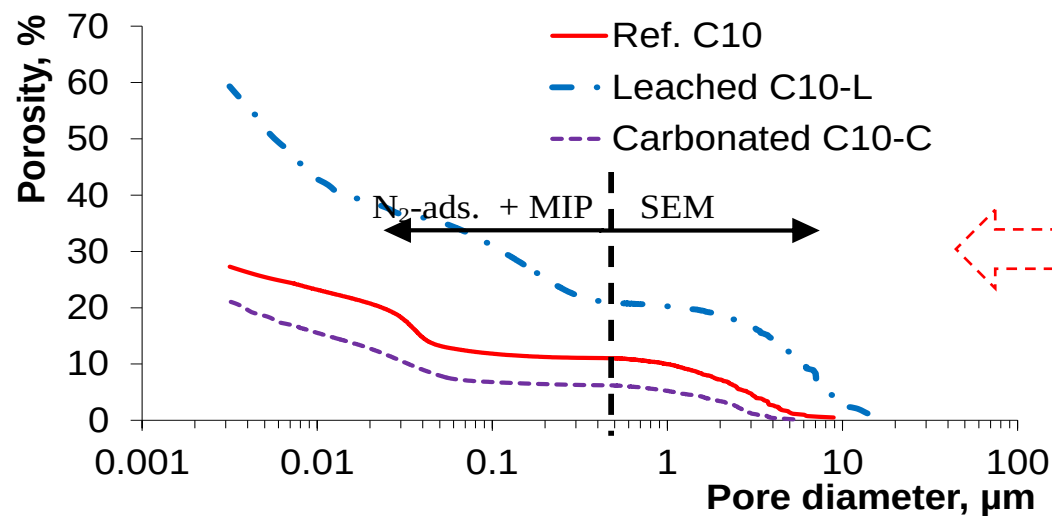
[Kalliopi 2006]



“L” = Leached
“C” = Carbonated

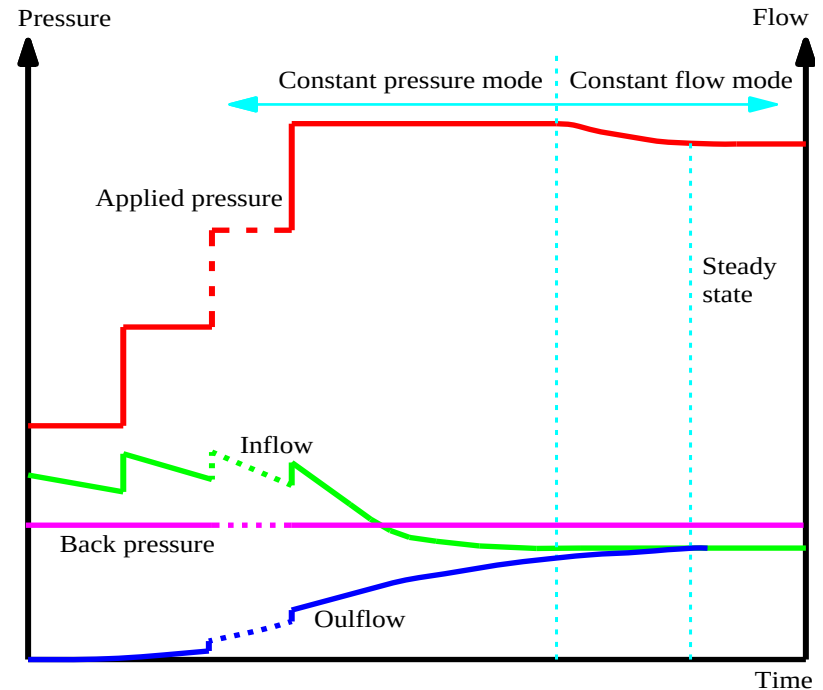
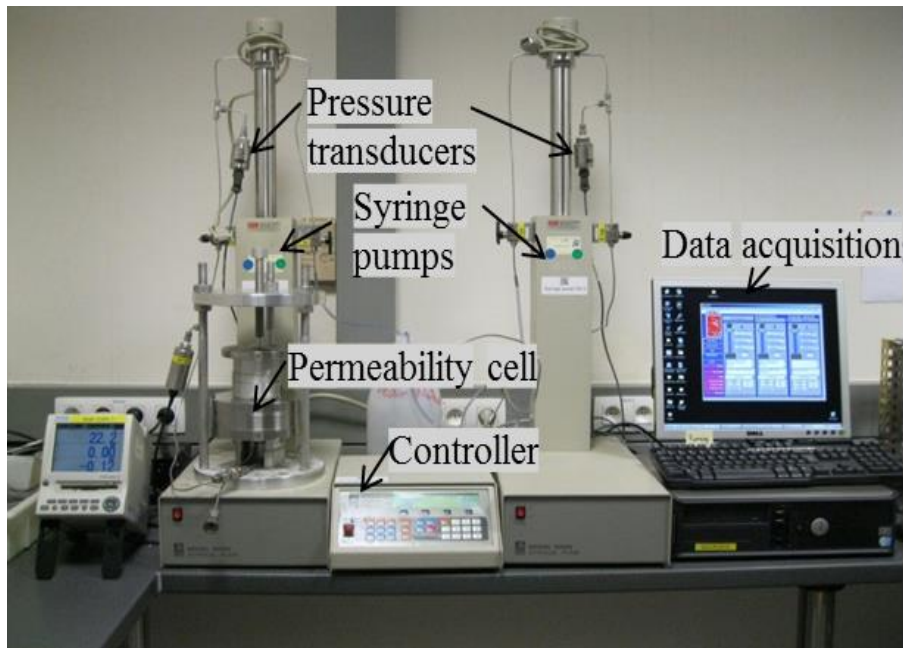


N_2 – adsorption + MIP + SEM



Improved permeability measurement: controlled **constant flow** [Phung 2013b]

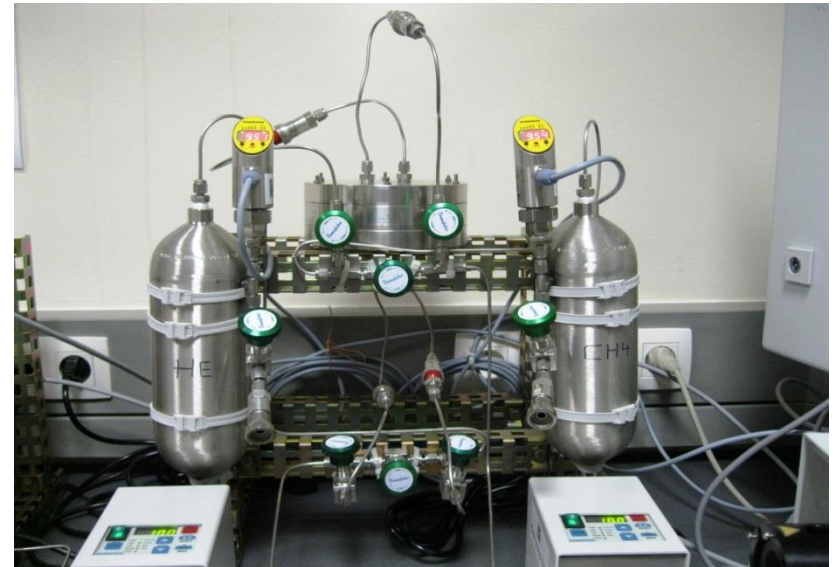
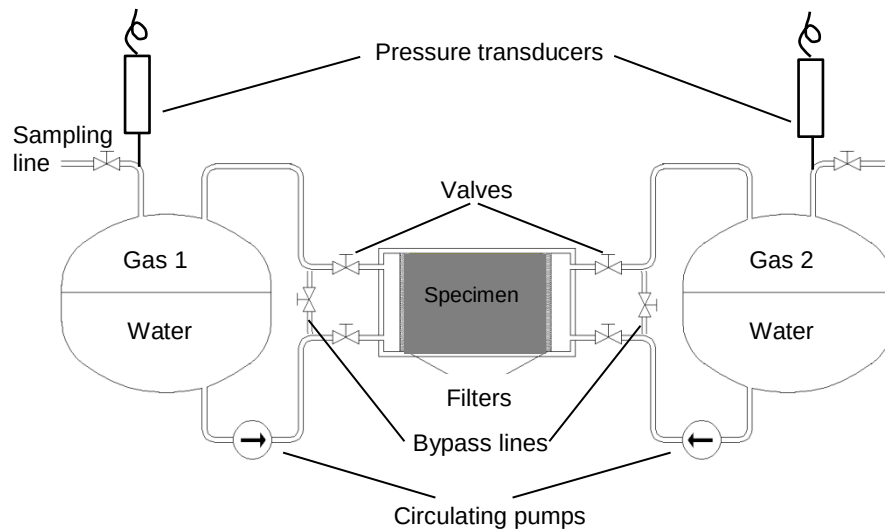
- Novelty: steady-state conditions are controlled by applying a constant water flow → increase the accuracy

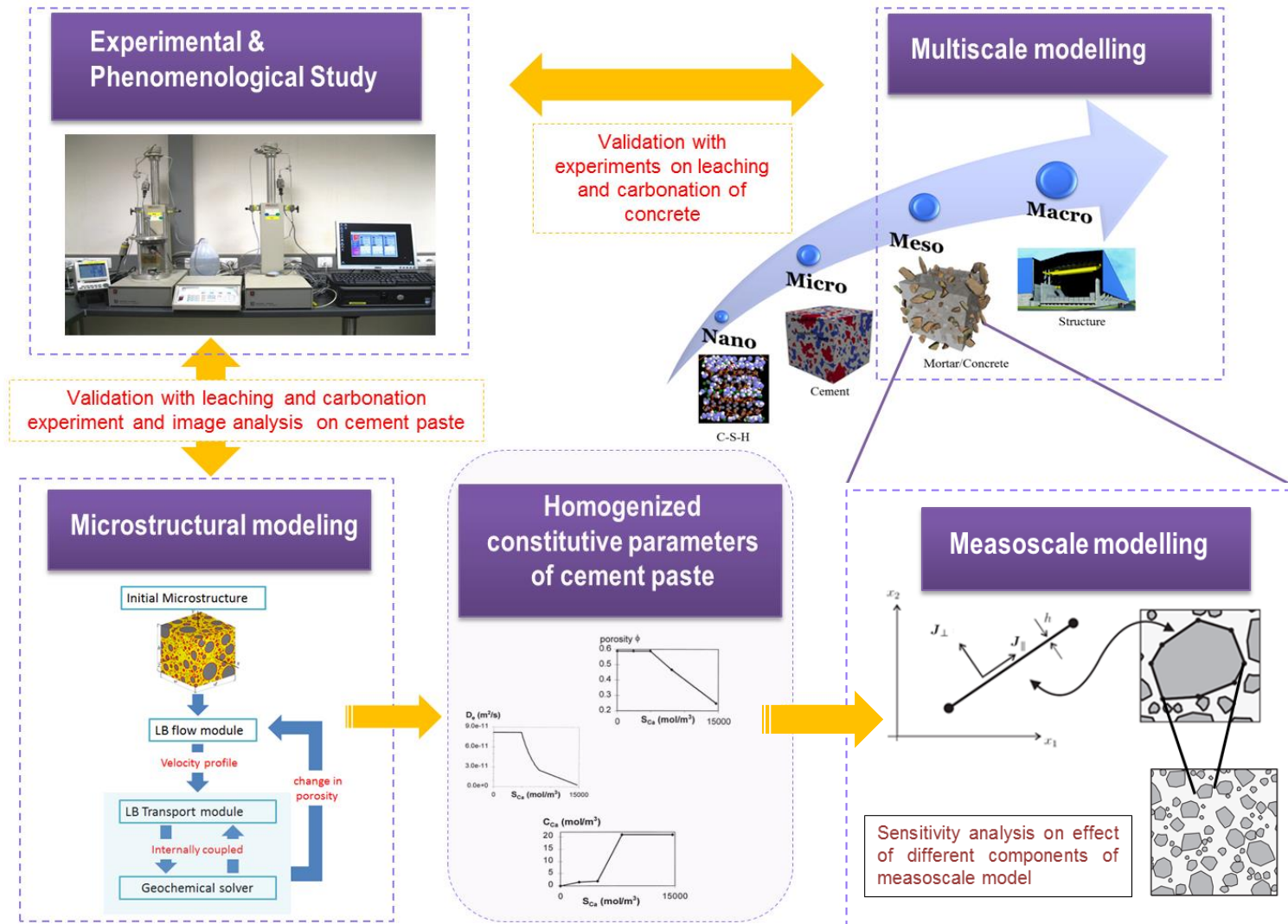


Diffusion of 2 dissolved gases: adapted from clay, **first time for concrete**

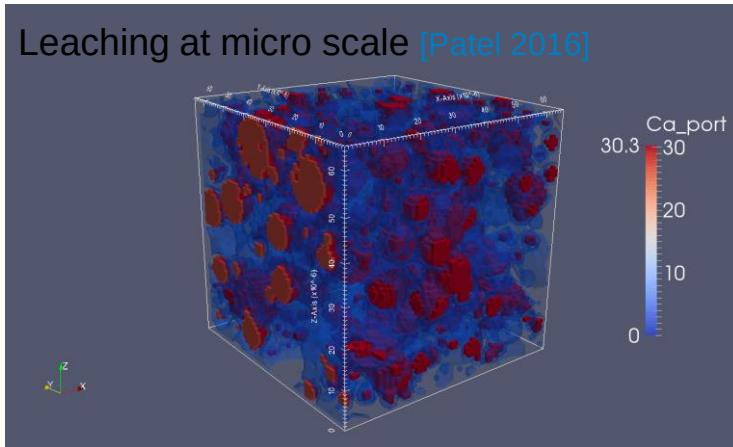
[Jacops 2013, Phung 2015d]

- Single experiment → diffusion coefficients of 2 gasses

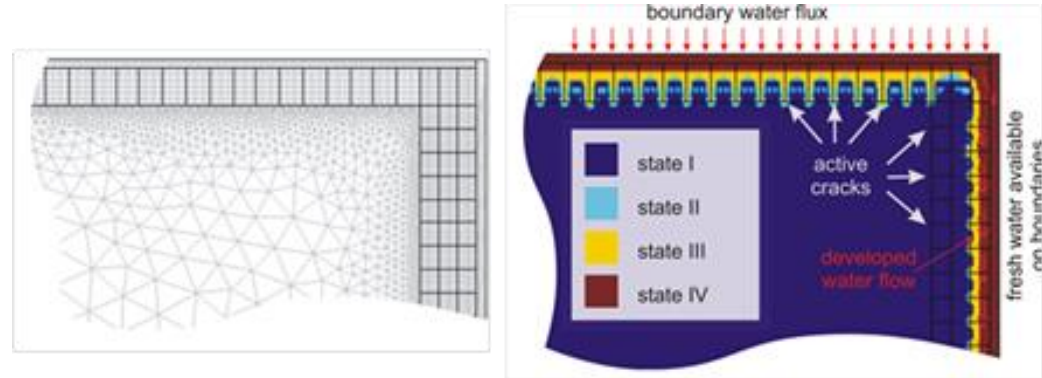




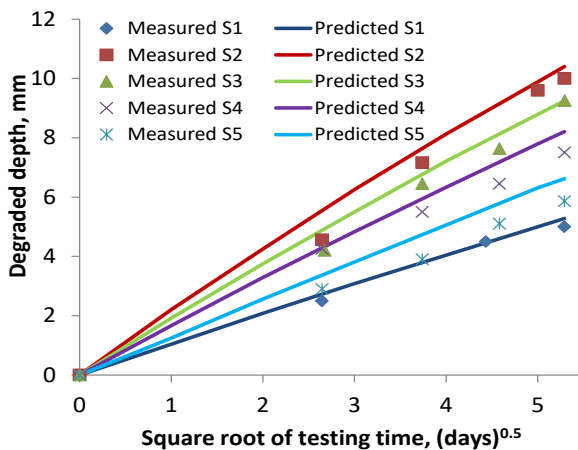
Leaching at micro scale [Patel 2016]



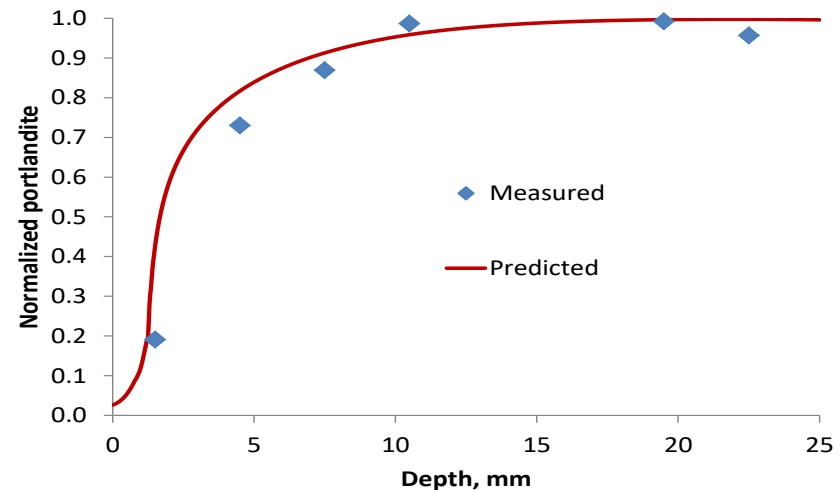
Leaching of monolith [Perko 2011]



Leaching at continuum scale [Phung 2016a]

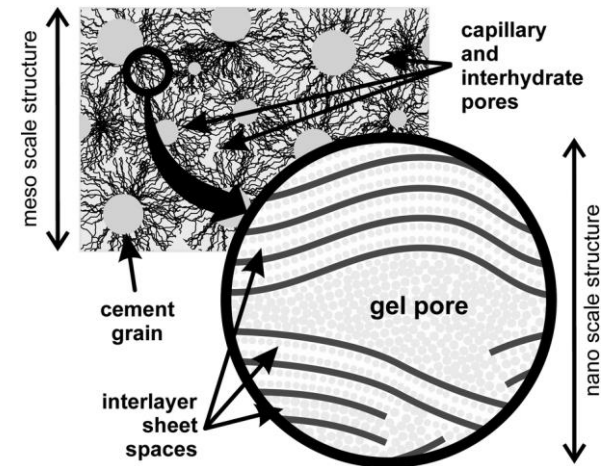


Carbonation at continuum scale [Phung 2016b]



Conclusions and Perspectives

- The assessment of the long-term durability is very important in waste management.
- Degradation processes: very slow but may significantly change the physical integrity and the chemistry of the structures
 - Changes in microstructures and mineralogy
→ induced changes in transport properties
- Accelerated test results might be extrapolated to the natural environments
→ accelerated experiments + modelling
- Systematic approach: multiple processes at multiple scales



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STUDIECENTRUM VOOR KERNENERGIE
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Modelling of concrete structures: the role of cracks

21st Exchange Meeting ESV EURIDICE GIE

Janez Perko

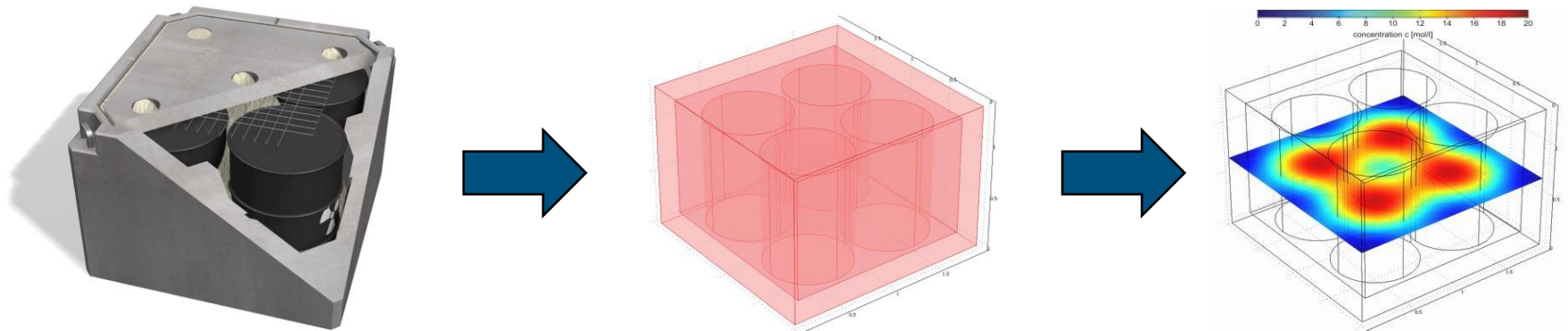
Engineered and Geosystem Analysis, Expert Group Waste & Disposal

janez.perko@sckcen.be



STUDIECENTRUM VOOR KERNENERGIE
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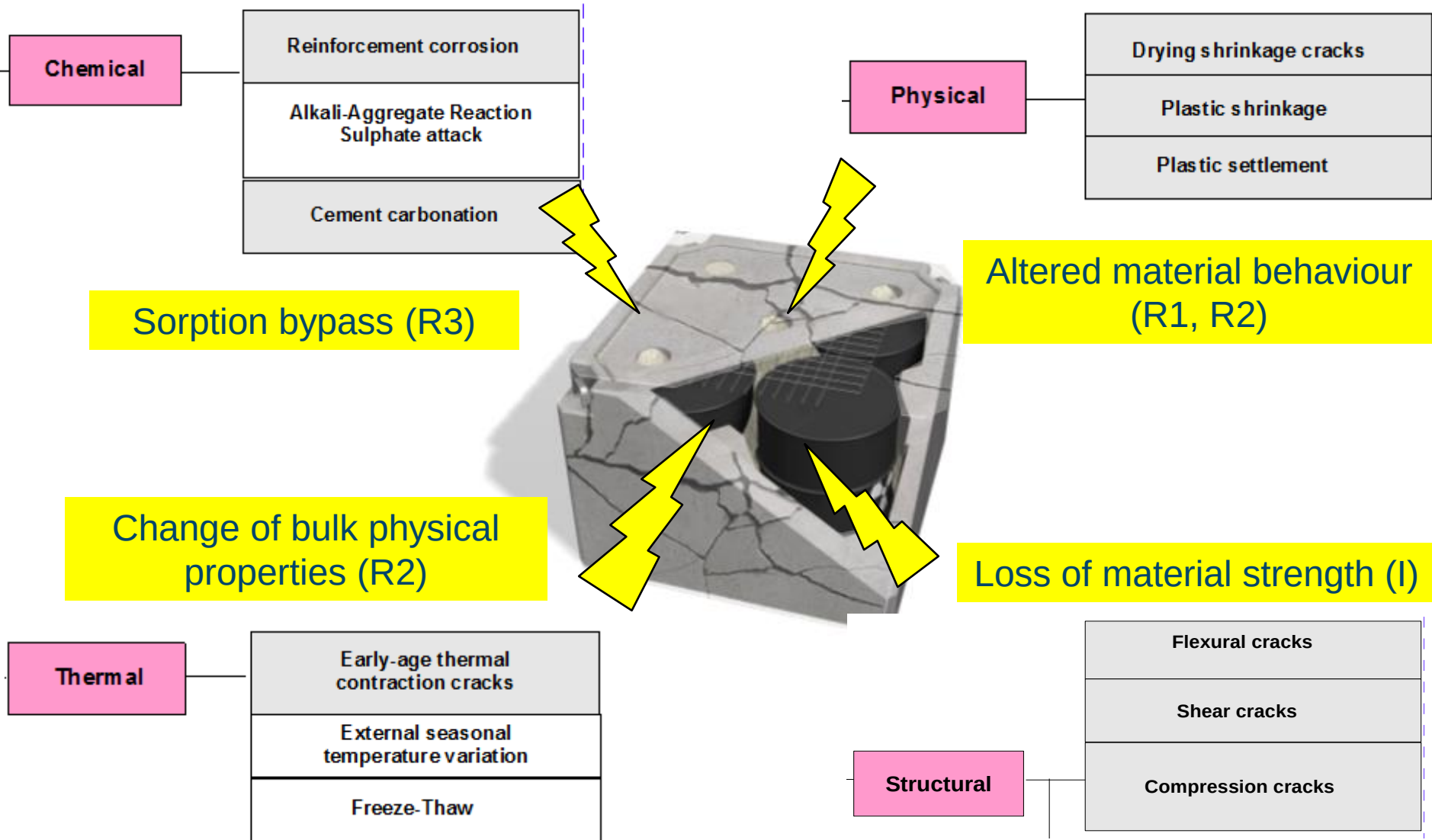
- Long term safety of disposal facility is demonstrated by performance assessment (PA)
 - PA = modelling = conceptualisation



- Conceptualisation => ability to evaluate uncertainties

● Uncertainties

Motivation



● Conceptualisation

Complexity

● Equivalent effective medium (upscaling)

2.2.8 HS14-060: Benadering voor de modellering van **scheuren**, spleten en openingen

Het volgende wordt vermeld in OD-108 ([4], p.235): “*The effective hydraulic conductivity is defined here as the hydraulic conductivity of concrete with initial macro-cracks ($>100\ \mu\text{m}$)*”

● Explicit fractures, saturated

2.2.1 HS14-053: Verwachte evolutie(s) en distributie van de saturatiegraad

De voorspelling van de tijdsevolutie van de verzadigingsgraad van de verschillende onderdelen van de bergingsinstallatie (spleten en **scheuren** inbegrepen) is essentieel bij het identificeren en

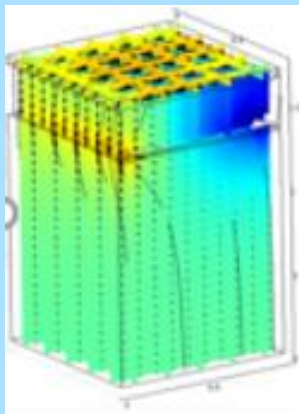
● Explicit fractures, partially saturated

2.2.10 HS14-062: Hypothesen i.v.m. de modellering van **scheuren**, spleten en openingen

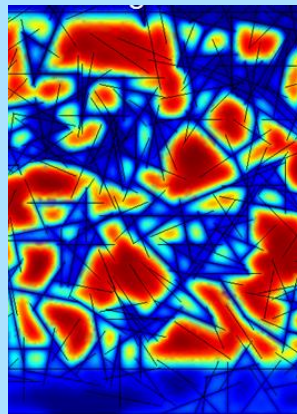
In HS14-061 wordt gevraagd om de verwachte evolutie(s) en initiële toestand qua **scheuren**, spleten en andere openingen te beschrijven in het hoofdstuk 14. Het is verder ook soms

● Explicit fractures, partially saturated, dynamic

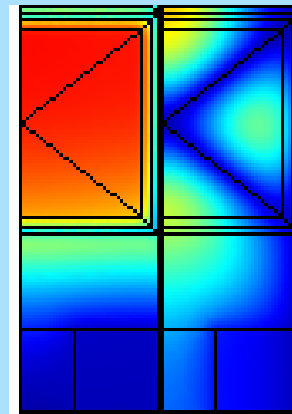
Cracks & water flow



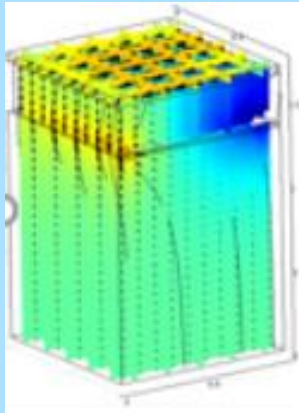
Cracks & migration



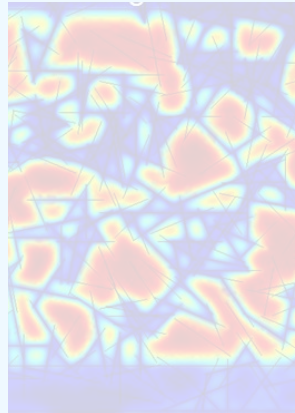
Conclusions & Outlook



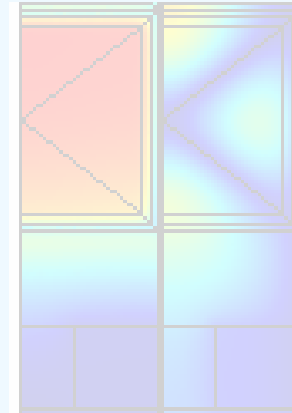
Cracks & water flow



Cracks & migration



Conclusions & Outlook



- Water flow in saturated conditions

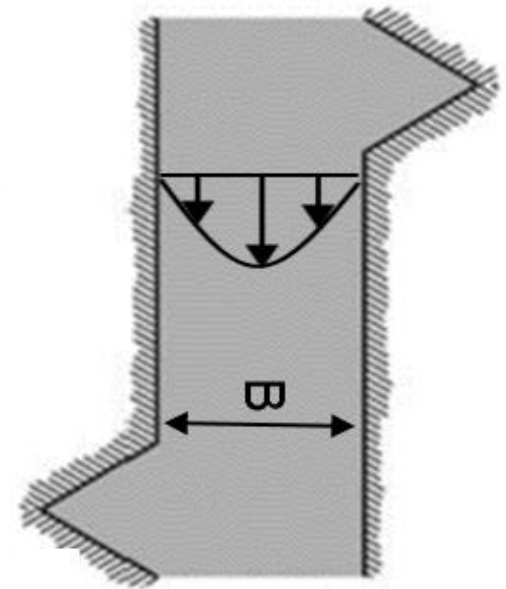
- Darcy's equation



- Parabolic velocity profile

- Parameters defining water flux

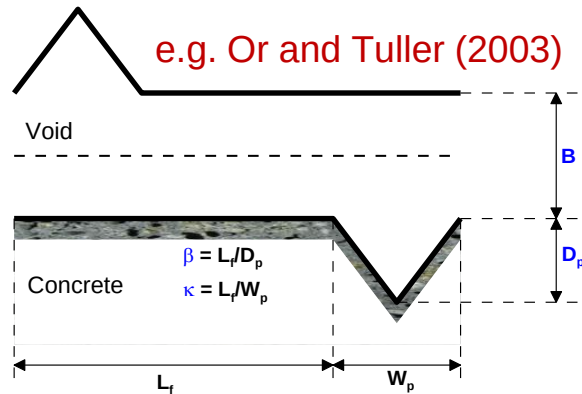
- Crack aperture
 - Crack strength (number of cracks per meter)
 - Boundary conditions



Water flow in partially saturated cracks

● Water flow in partially saturated cracks

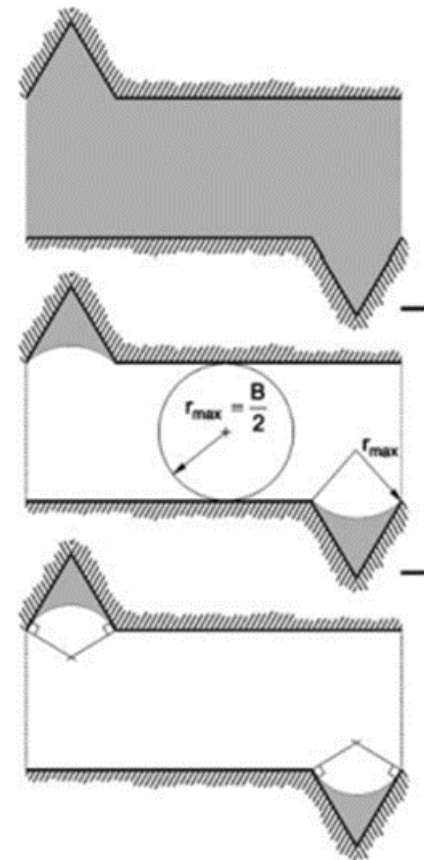
- New relative permeability calculation



- Parameters defining water flux

- Crack aperture
- Crack strength (number of cracks per meter)
- Boundary conditions
- Explicit crack roughness
- Water-air and water surface interactions

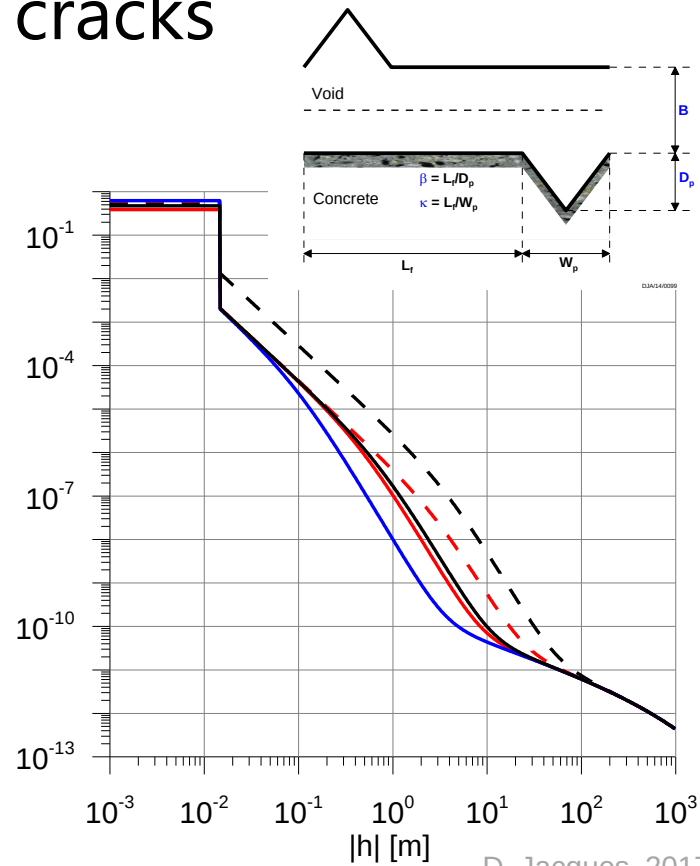
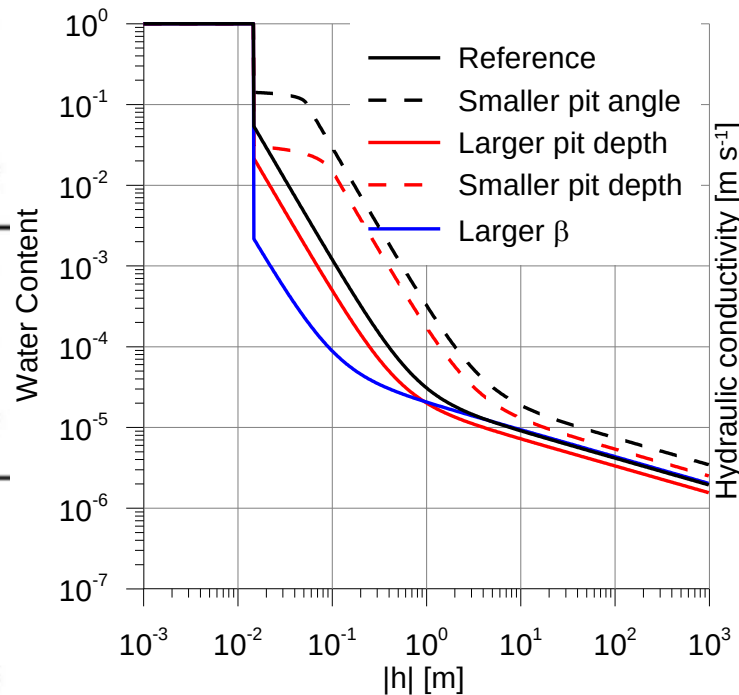
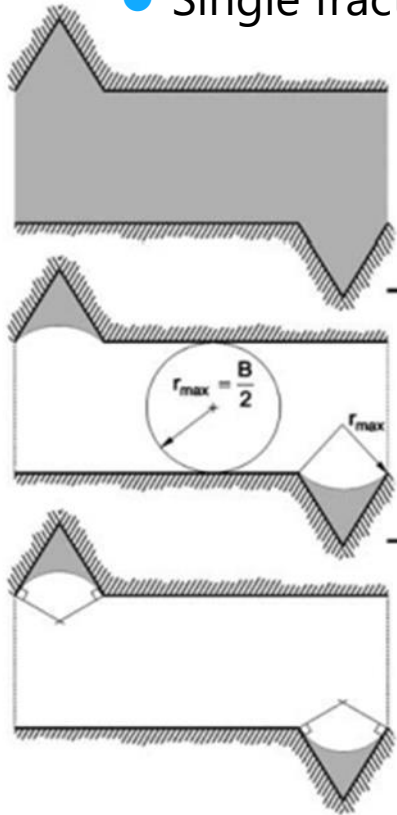
Different possible saturation regimes



Water flow in partially saturated cracks

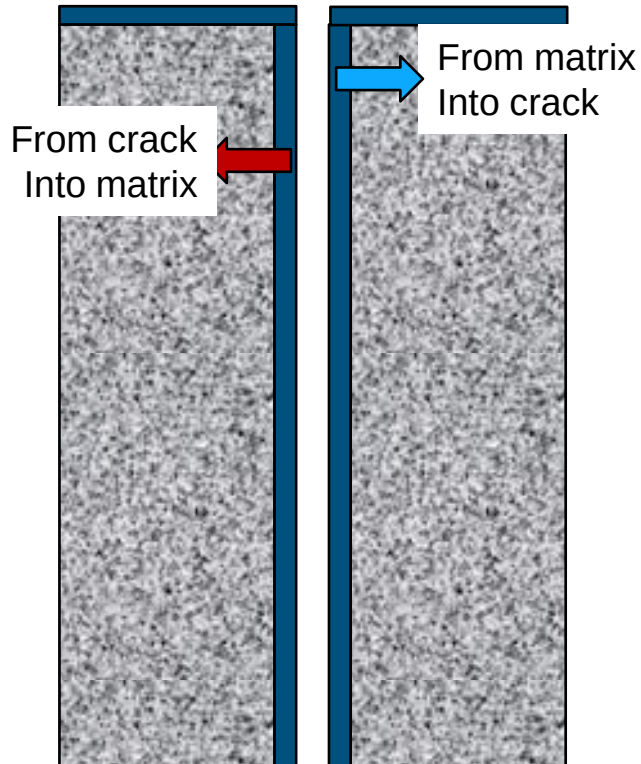
● Water flow in partially saturated cracks

● Single fracture



D. Jacques, 2017

Water flow in partially saturated cracks/matrix interaction



- Many possible conditions

- Matrix

- Intact
 - Physically degraded

- Crack

- Narrow/wide (distribution)
 - Smooth/rough (evolution)

- Boundary conditions

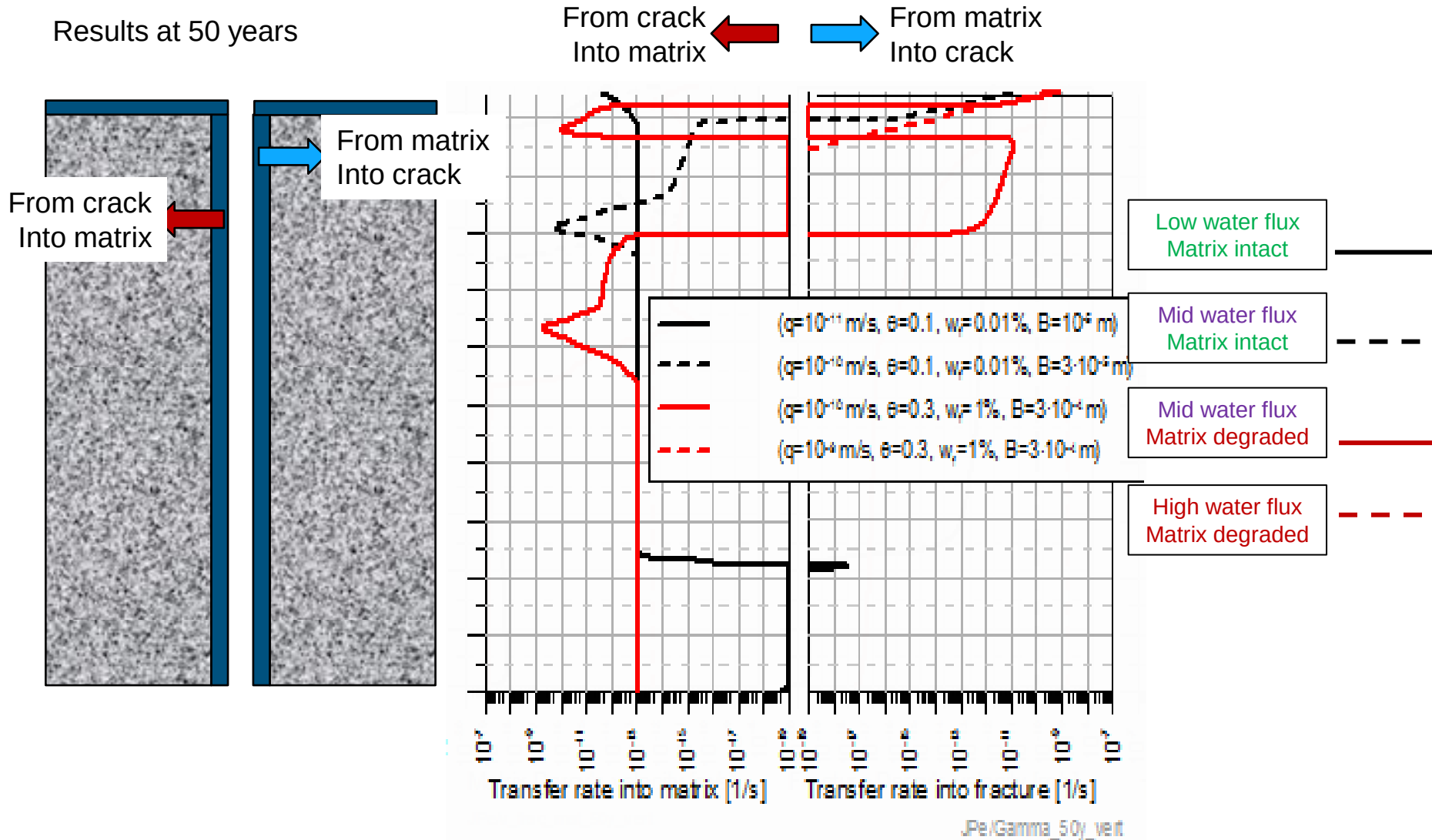
- Amount of available water

- Type of problem

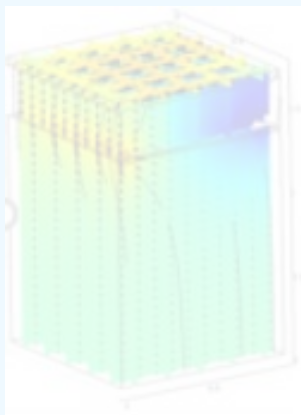
- Initial value (e.g. monolith)
 - Boundary value (e.g. module base)

Water flow in partially saturated cracks/matrix interaction

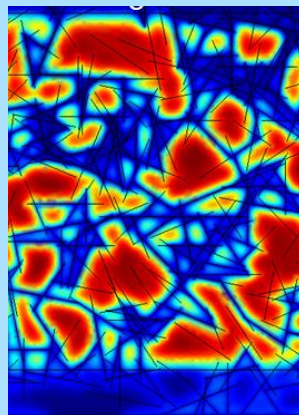
Results at 50 years



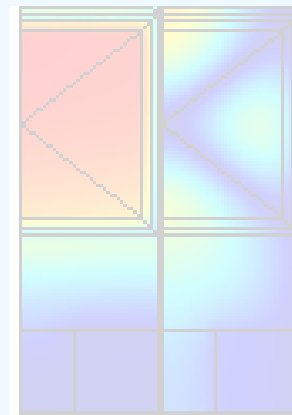
Cracks & water flow



Cracks & migration



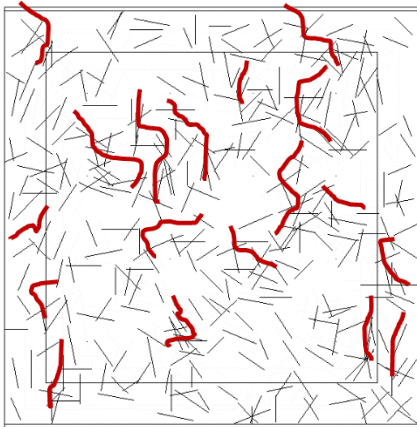
Conclusions & Outlook



Transport in fractured porous media

- Explicit approach
- Concrete block 1m×1m
 - Explicit cracks (based on the initial conditions)
 - Random crack position, length and orientation
 - Same pattern used for all systems, only length increases

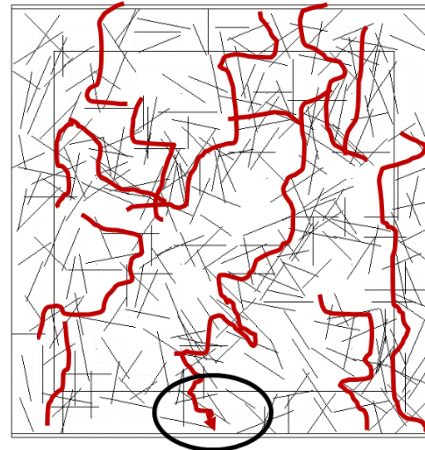
configuration 1



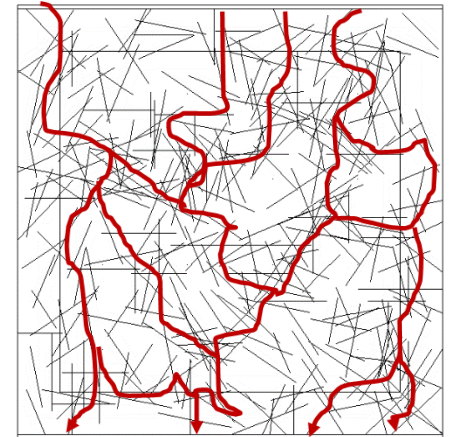
configuration 2



configuration 2.5

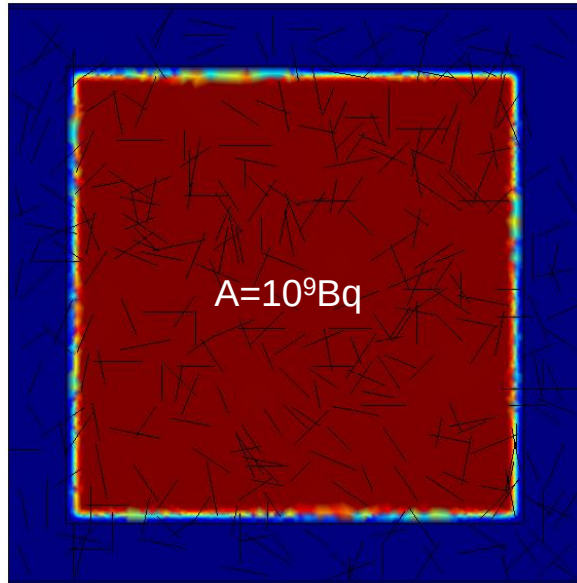


configuration 3



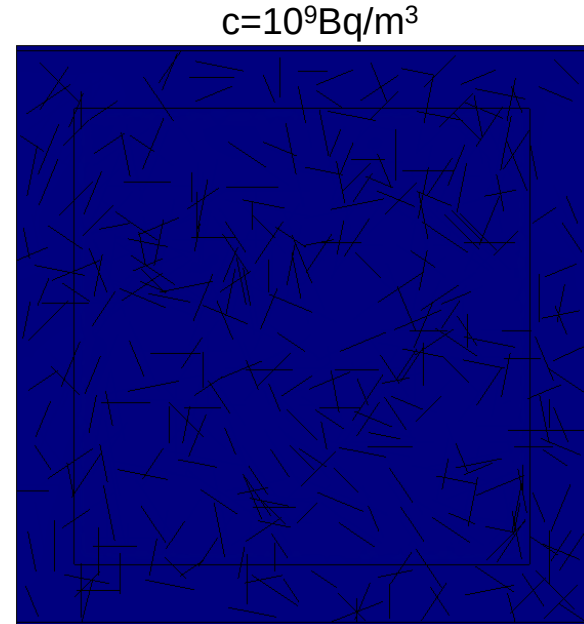
Transport in fractured porous media

- Initial value problem and boundary value problem



representing monolith

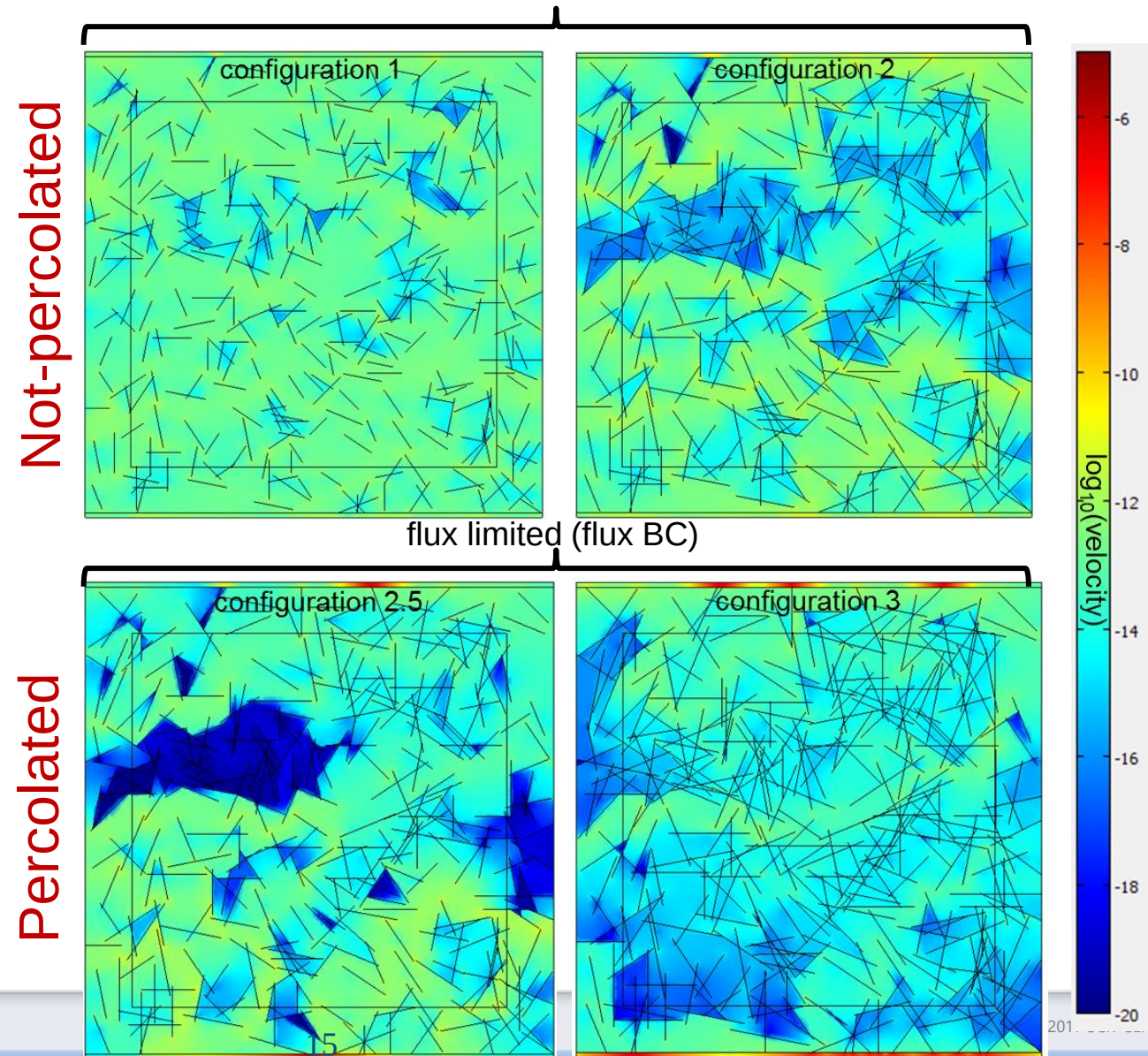
$$K_s = 2 \cdot 10^{-14} \text{ m/s}$$
$$K_s = 1 \cdot 10^{-11} \text{ m/s}$$



representing module base

Transport in fractured porous media

- Flow calculations
 - More connected cracks - Lower matrix velocity



Transport in fractured porous media

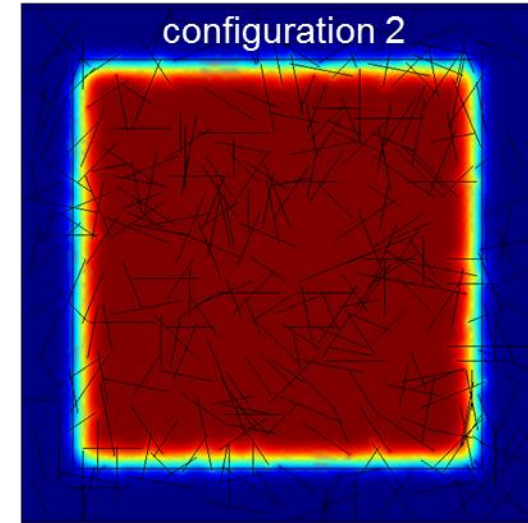
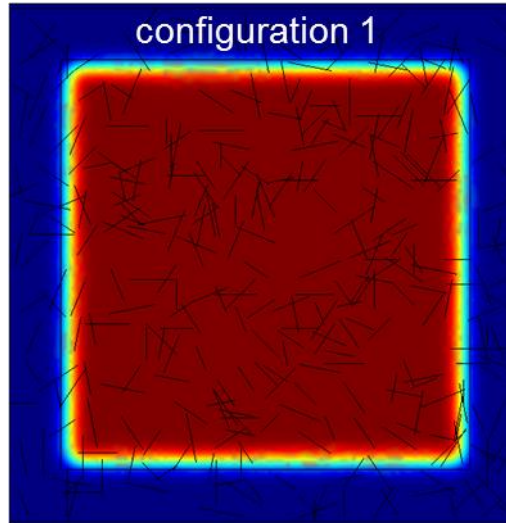
- Initial value problem

- homogeneous like process



- Effective properties

Not-percolated

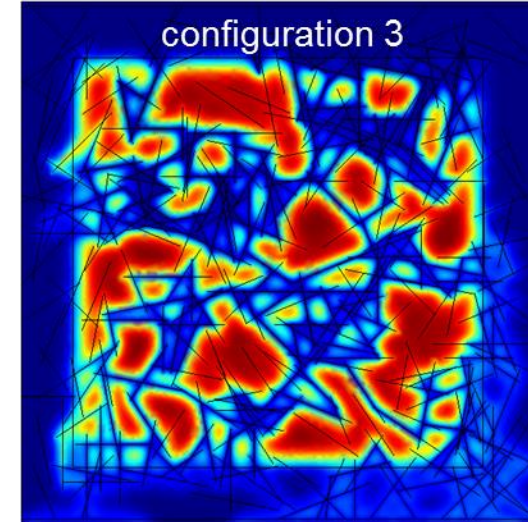
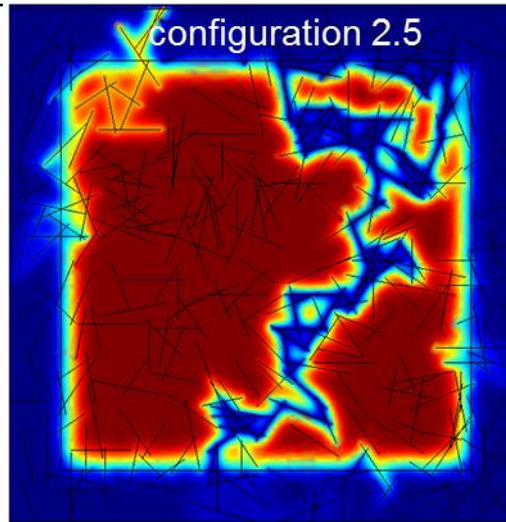


- Heterogeneous transport



- Mobile (instant leaching)
- Immobile (diffusive release)

Percolated



Breakthrough curve © 2017 SCK•CEN

Transport in fractured porous media

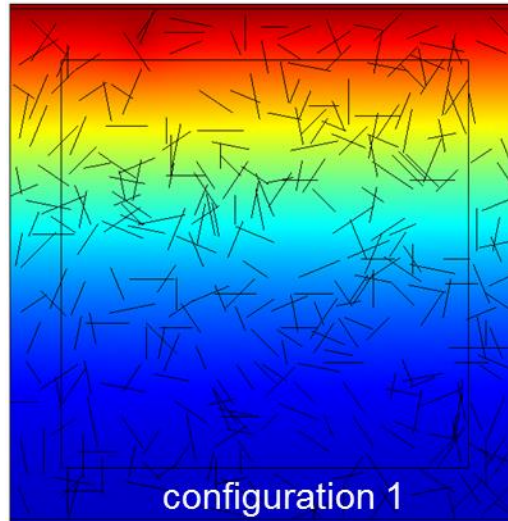
- Boundary value problem

- homogeneous like process

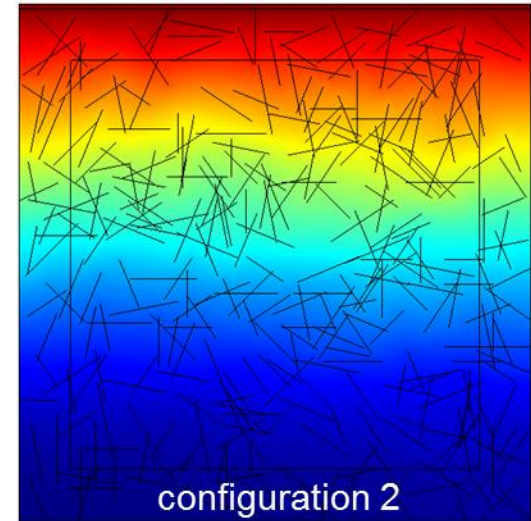


- Effective properties

Not-percolated



configuration 1



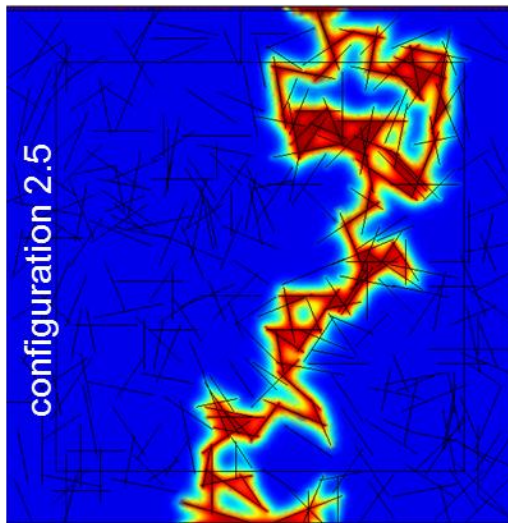
configuration 2

- Heterogeneous transport

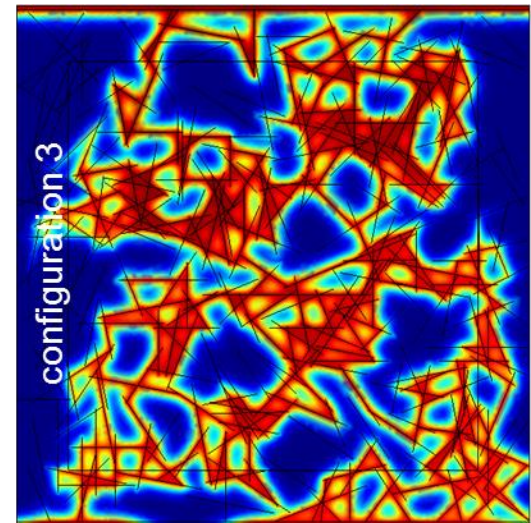


- Mobile (bypass)
- Immobile (back diffusion)

Percolated



configuration 2.5

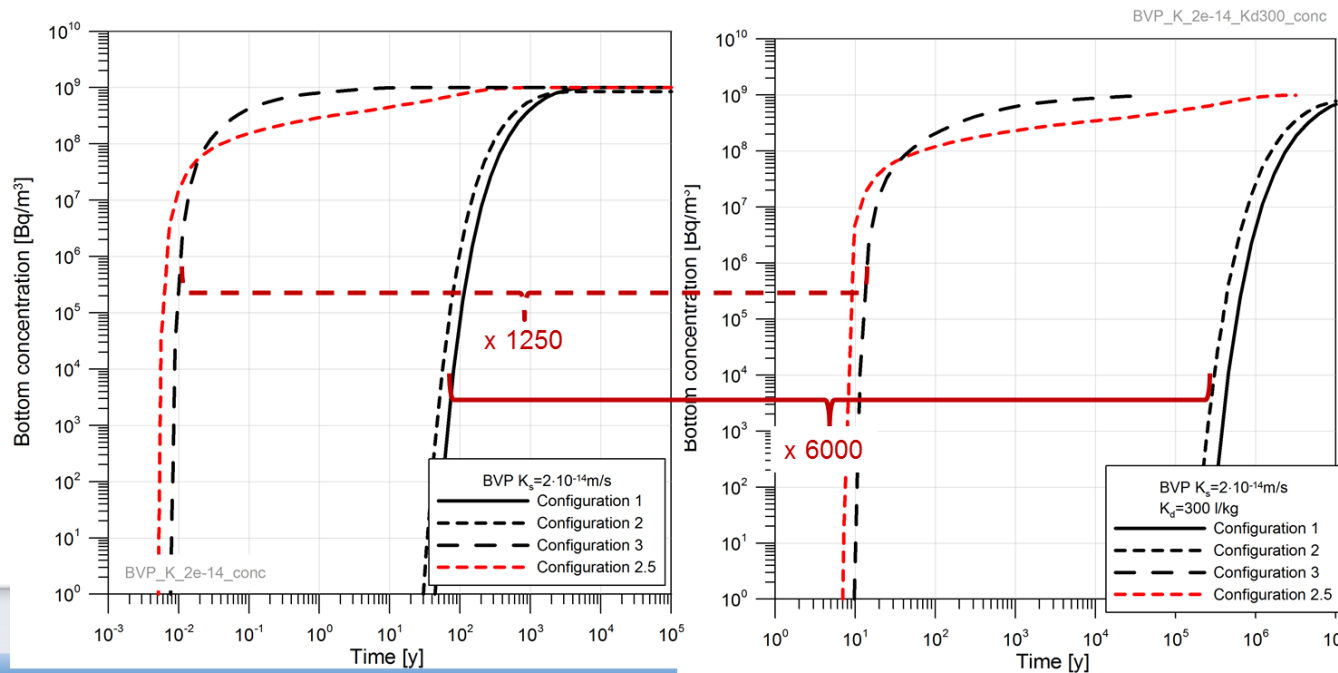


configuration 3

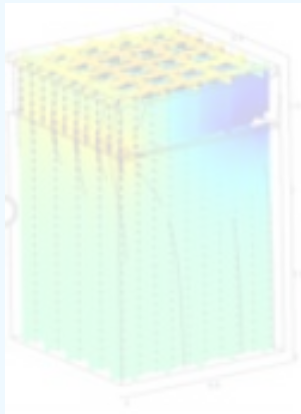
Transport in fractured porous media

Selected outcomes

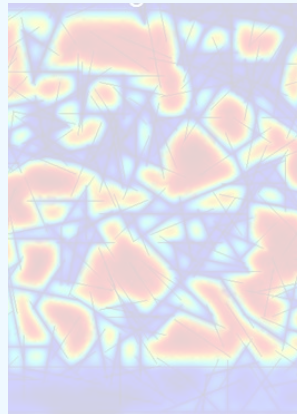
- Smaller matrix diffusion $\Rightarrow$ more spread (IVP)
- Smaller matrix diffusion $\Rightarrow$ less spread (BVP)
- Reduction in sorption in terms of peak magnitude and time is only valid when the system is fully percolated.
- Sorption is reduced only for fully percolated systems
- Upscaling is more effective for diffusion driven transport.



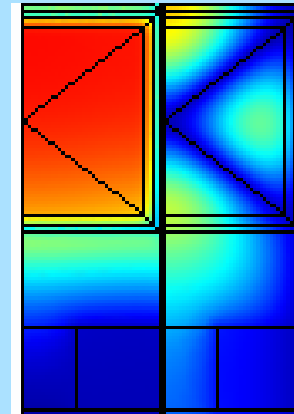
***Cracks
&
water flow***



***Cracks
&
migration***



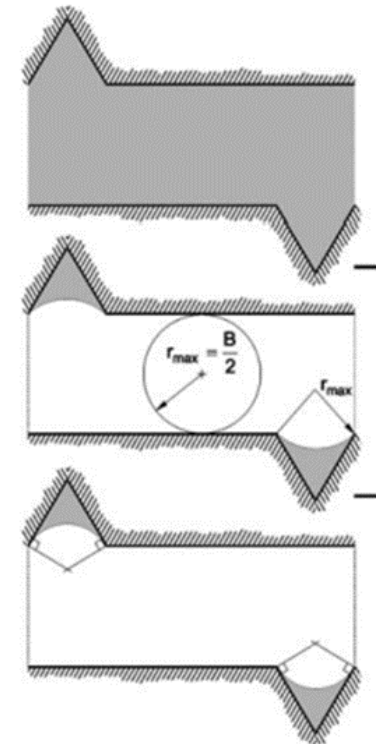
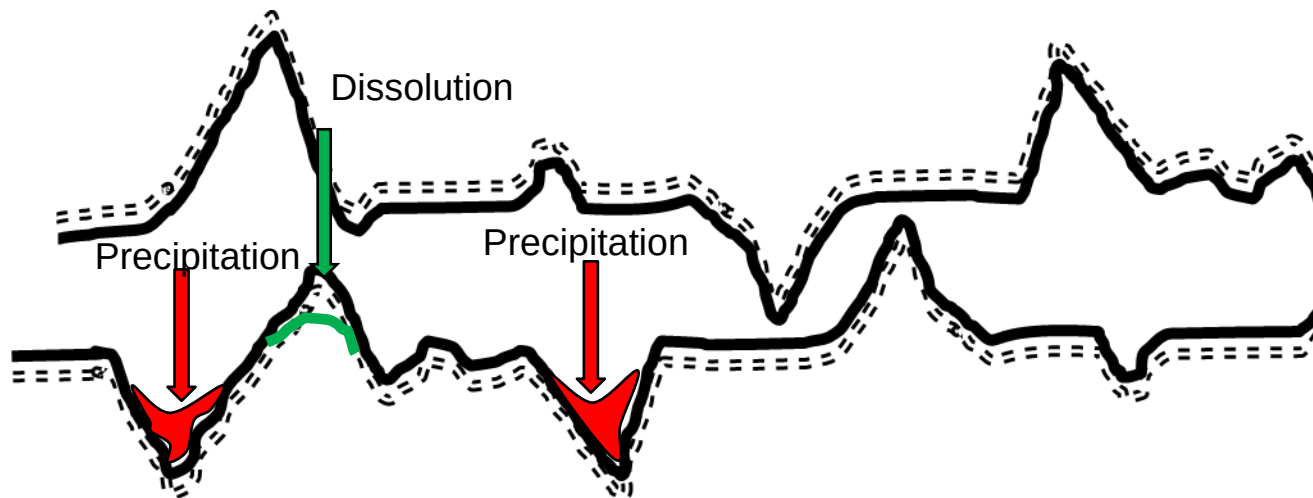
***Conclusions
&
Outlook***



Reactive transport in fractured porous media

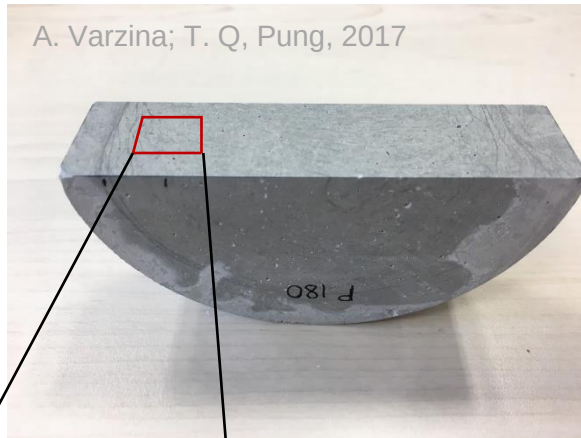
- Depending on environmental conditions fractures can evolve differently with time
 - Precipitation (e.g. carbonation)
 - Dissolution (e.g. Ca leaching)
 - Combined precipitation-dissolution

PhD programme

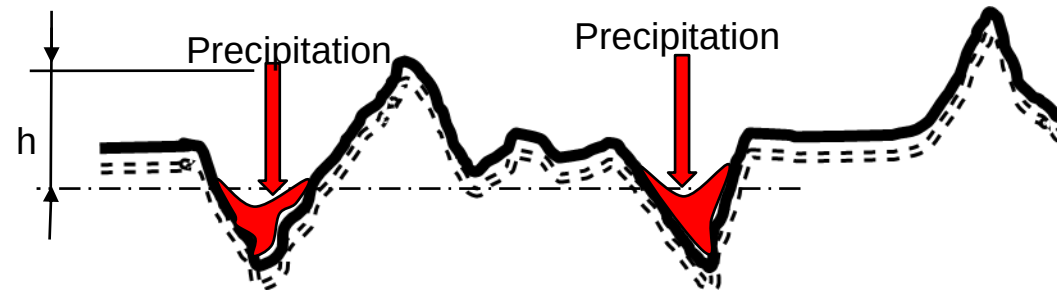


Reactive transport in fractured porous media

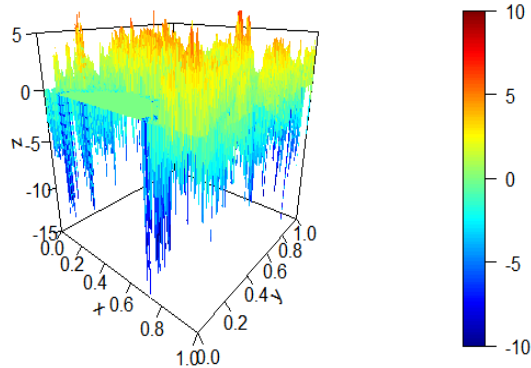
● Experimental evidences



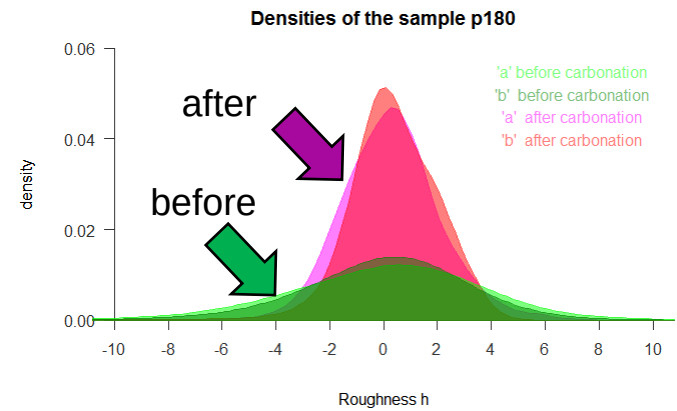
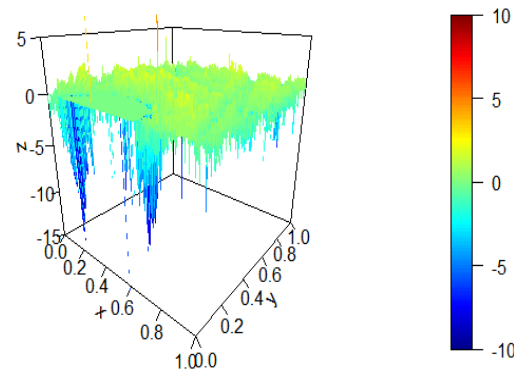
Coarse polished sample



Area (a) before carbonation



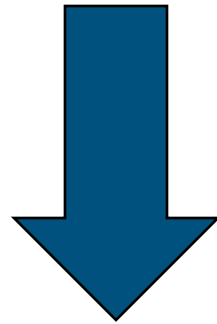
Area (a) after carbonation



● To be complemented by modelling

- Mechanistic understanding of
 - Water retention in cementitious materials
 - Matrix and cracks
 - Chemical retention
 - Crack surface
 - Crack/matrix interaction
 - Time evolution
 - Crack properties
 - Up/down-scaling of processes

Fractures are the “reliable” long-term features of the disposal system



Understanding fractures =
understanding long term safety

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ONDRAF/NIRAS

RD&D priorities on cement within the Belgian national programme

Xavier Sillen

Erik Coppens

Maarten Van Geet

21st Exchange Meeting ESV EURIDICE GIE

29/09/2025

Belgian Agency for Radioactive Waste and Enriched Fissile Materials



RD&D priorities on cement within the Belgian national programme

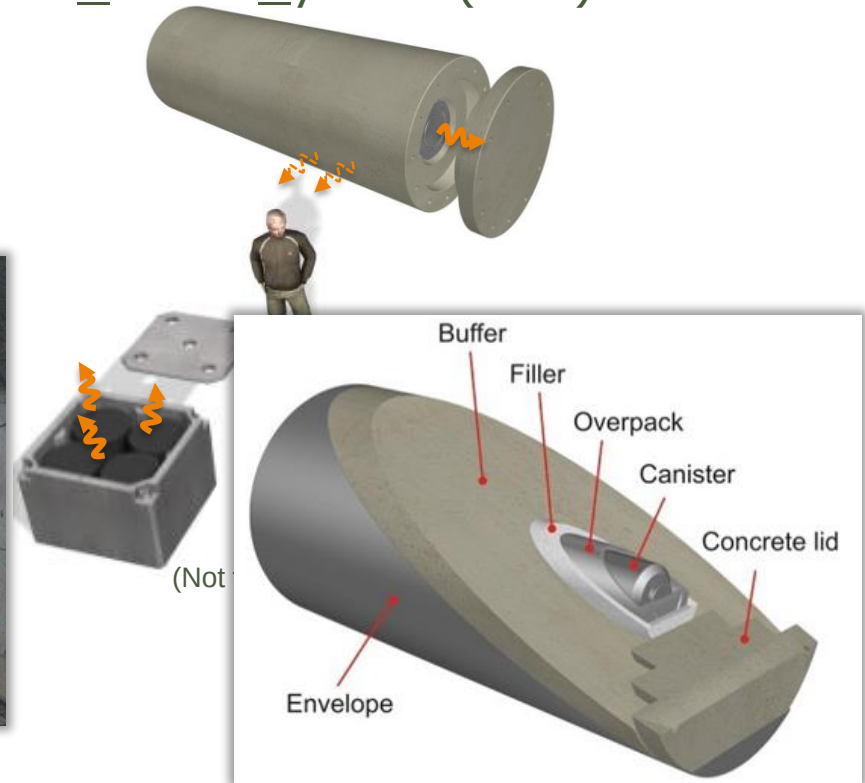
- **Cementitious materials in the long term management of radioactive waste**
- **Research, Development and Demonstration (RD&D) on cementitious materials in that context**
- **Priorities in the Belgian national programme**
- **Conclusions**

RD&D priorities on cement within the Belgian national programme

- **Cementitious materials in the long term management of radioactive waste**
- **Research, Development and Demonstration (RD&D) on cementitious materials in that context**
- **Priorities in the Belgian national programme**
- **Conclusions**

Cementitious materials in long term management of radioactive waste

- Cementitious materials are **ubiquitous** in long term management of radioactive waste
 - **Many uses:** waste conditioning, structural elements, shielding, key component of the Engineered Barrier System (EBS)



Cementitious materials in long term management of radioactive waste

- **Cementitious materials are ubiquitous in long term management of radioactive waste**
 - **Many uses:** waste conditioning, structural elements, shielding, key component of the Engineered Barrier System (EBS)
- **Engineered materials**
 - One can freely **select** and to some extent **design** the material to meet certain **requirements** (see 2nd presentation – Erik Coppens)
- **Evolving materials**
 - in the short term (see 1st presentation – Erik Coppens)
 - and the long term (see 3rd presentation – Quoc Tri Phung)

RD&D priorities on cement within the Belgian national programme

- Cementitious materials in the long term management of radioactive waste
- **Research, Development and Demonstration (RD&D) on cementitious materials in that context**
- Priorities in the Belgian national programme
- Conclusions

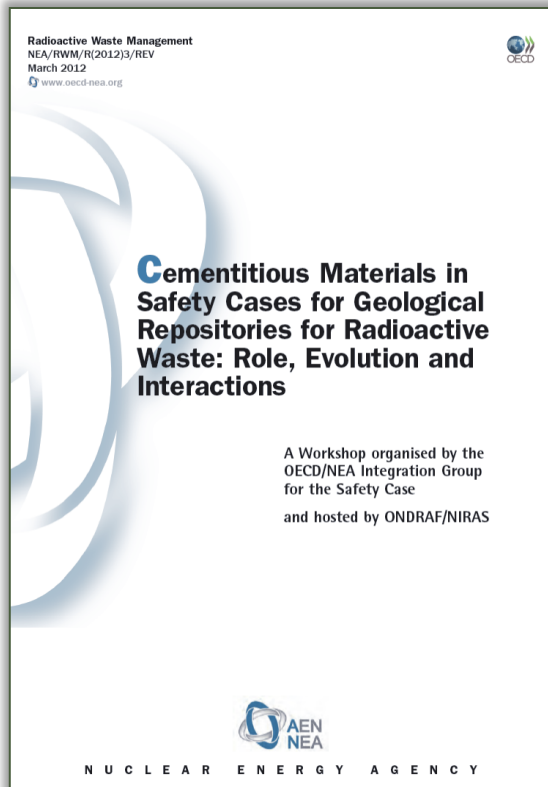
RD&D on cementitious materials for the long term management of radwaste

- Many uses
 - + many choices
 - + evolution } → plenty of possible RD&D topics!

- RD&D activities generally organised along the lines of
 1. **Material selection/design** (w.r.t. requirements)
↓
 2. **Characterisation** of key properties that will affect the evolution of the **component** and the **system** of which it is a part
↓
 3. **Assessment** of the expected **evolution**
 - will the requirements be met over the necessary time frames?
 - compatibility with other components of the system?

RD&D on cementitious materials for the long term management of radwaste

www.oecd-nea.org/rwm/docs/2012/rwm-r2012-3.pdf



<http://goo.gl/hVMWJw>

A good reference: NEA WS proceedings

- Workshop held in Brussels in 2012
- Discusses roles, evolution and interactions of cementitious materials in the context of geological disposal and on the related RD&D

Notable evolution (~ early 2000's)

- cementitious materials considered as "**necessary evil**" w.r.t long term performance of geological repositories; focus on detrimental effects, perturbations



- **taking advantage** of key properties of cementitious materials to **enhance** the long term performance of the EBS

RD&D priorities on cement within the Belgian national programme

- Cementitious materials in the long term management of radioactive waste
- Research, Development and Demonstration (RD&D) on cementitious materials in that context
- **Priorities in the Belgian national programme**
- Conclusions

Priorities in the Belgian national programme: elements of context

- **In Belgium O/N is responsible, by law, of the management of the radioactive waste (& associated RD&D programme)**
 - RD&D on cementitious materials follows the chain of this management from treatment/conditioning to long-term storage & disposal, with the aim to develop **applicable solutions** for each step: strong focus on **applied science**
- **RD&D commensurate with programme stage**
 - **Cemented wastes:** existing / future
 - RD&D can be pro-active but sometimes must be reactive (e.g. "gelvaten")
 - **cAt:** towards licensing for construction
 - RD&D should address legitimate concerns of licensing authority & stakeholders
 - **B&C:** assuming geological disposal but **no decision-in-principle yet!**
 - More generic RD&D as some boundary conditions remain uncertain
 - SFC-1: from exploratory to increasingly targeted RD&D

Priorities in the Belgian national programme: setting RD&D priorities

- **For each RD&D topic (question) under consideration, check**
 - **Urgency** (e.g. is answer required for current programme stage?)
 - **Impact** (e.g. could result strongly affect the programme, or can we accommodate any outcome by minor adaptations?)
 - **Effort** involved (e.g. can we do it ourselves, did others face the same issue, can we "design around" the issue?)
- **Make the best use of available programme resources**
 - We live in a small country, but the challenges we face in long term management of radioactive waste are not smaller
 - For R&D topics/issues that are less application-specific (e.g. structural performance, concrete pathologies,...), leverage expertise from others domains (e.g. civil engineering use of cementitious materials)
 - Seek return of experience from sister programmes (F, E, GB, CH...), European projects (currently CEBAMA, upcoming EJP)
- **Remain open-minded (look for the unexpected)**

Priorities in the Belgian national programme: **RD&D on cemented wastes**

- **compatibility of cemented waste forms with disposal**
 - For **existing** cemented wastes
 - for **known problematic wastes**, characterisation efforts to understand involved processes → assess consequences → propose remediation measures (e.g. "gelvaten")
 - In general, risk studies + development of new conditioning
 - For **future** cemented waste matrices (e.g. concrete)
 - E.g. concrete
 - Return



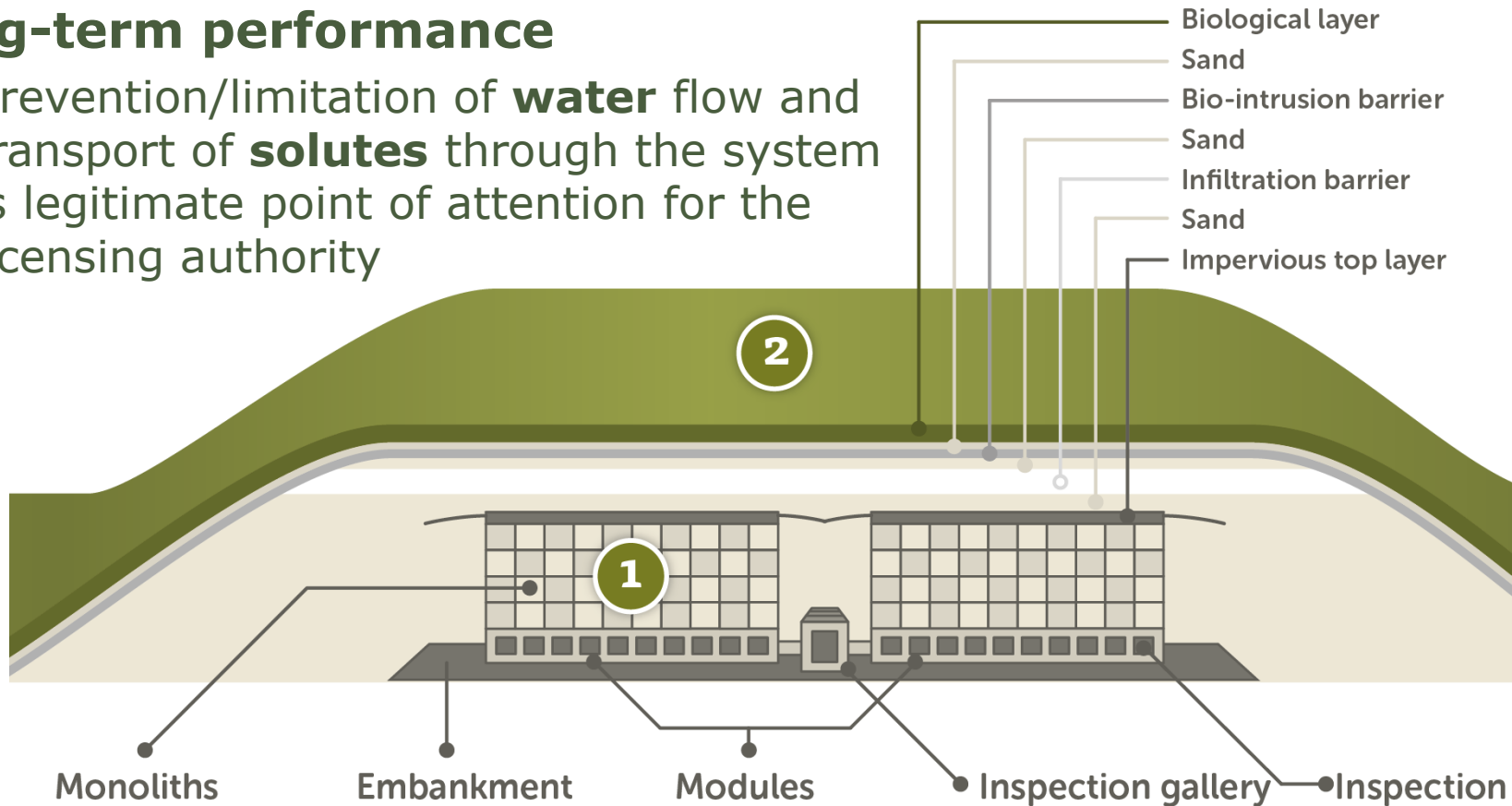
Priorities in the Belgian national programme: **cAt RD&D**

▪ Nearing implementation → Engineering aspects

- Detailed engineering aspects
- QA measures (towards: as built = as designed)

▪ Long-term performance

- prevention/limitation of **water** flow and transport of **solutes** through the system is legitimate point of attention for the licensing authority



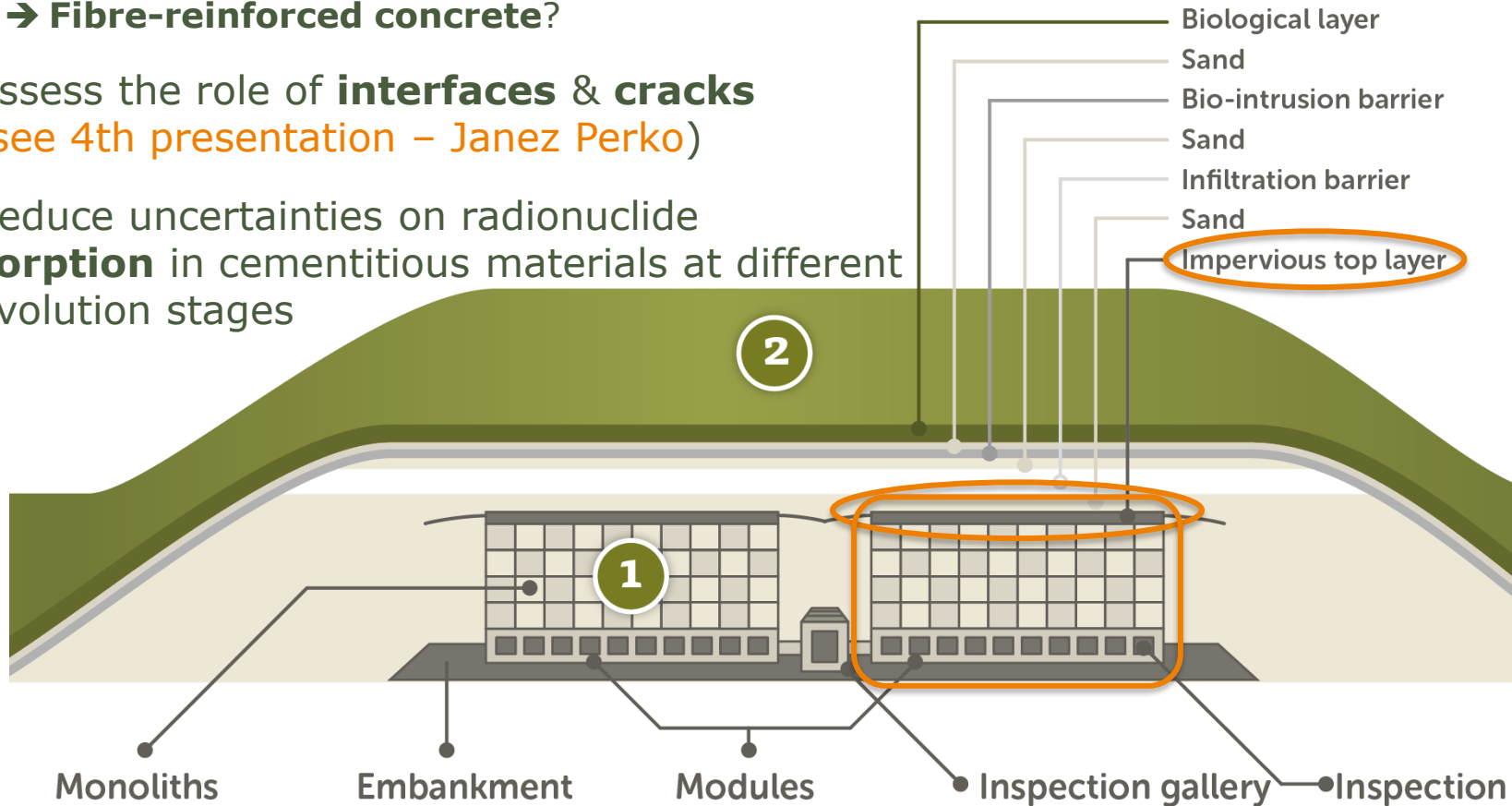
Priorities in the Belgian national programme: cAt RD&D

- **Long-term performance, w.r.t. water and solute transport**

- Finalize the development of the **Impervious Top Slab**
 - Mechanical resistance should be robust w.r.t. chemical degradation
 - Post-failure behaviour favouring diffuse rather than localized damage

→ **Fibre-reinforced concrete?**

- Assess the role of **interfaces** & **cracks**
(see 4th presentation – Janez Perko)
- Reduce uncertainties on radionuclide **sorption** in cementitious materials at different evolution stages



Priorities in the Belgian national programme: RD&D on geological disposal (B&C wastes)

- **Preliminary repository design & costing exercise → Engineering aspects**
 - Structural elements (gallery lining): high performance concrete?
 - Choice of non-metallic fibres as reinforcement in concrete elements (e.g. in cat. B monoliths) to limit generation of gas from metal corrosion?
 - Backfilling mortars: flowability, strength to be limited for retrievability, QA aspects (e.g. homogeneous backfilling)
- **Thermo-hydro-mechanical and chemical (THMC) behaviour of the **supercontainer** for category C wastes**
 - "Short" term **structural & shielding** function
 - Long term **buffer** function

} See next slides

Priorities in the Belgian national programme: "short" term evolution of the supercontainer

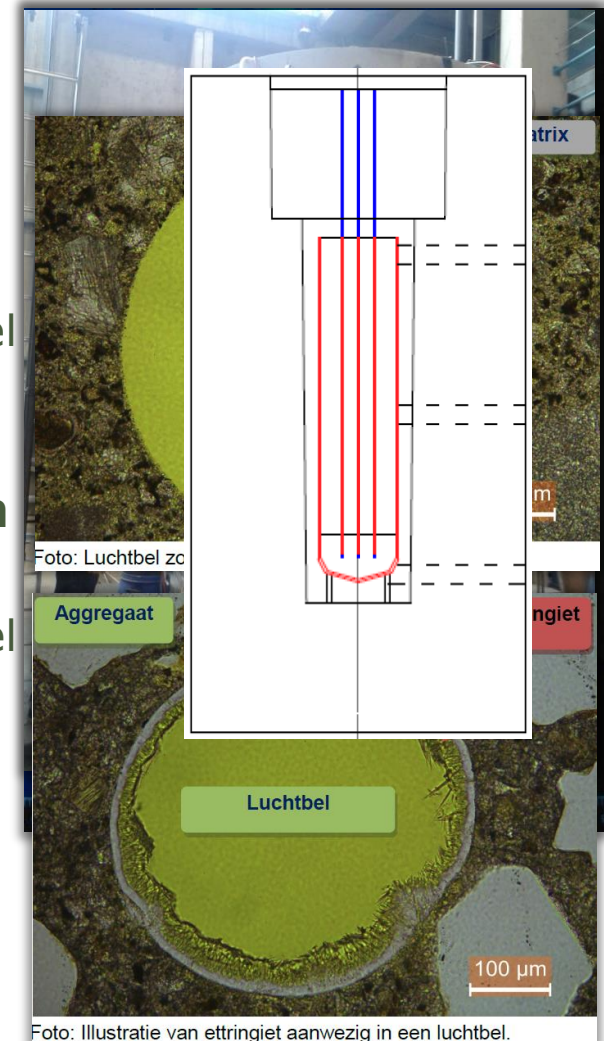
- Supercontainer for C-wastes (vitrified high level waste and spent fuel)
 - Made on surface and to be **transported** into the repository
 - Provides **shielding** during the operational and retrievability phase



- Can **structural integrity** be relied upon at least during operational and retrievability phase?

Priorities in the Belgian national programme: **"short" term evolution of the supercontainer**

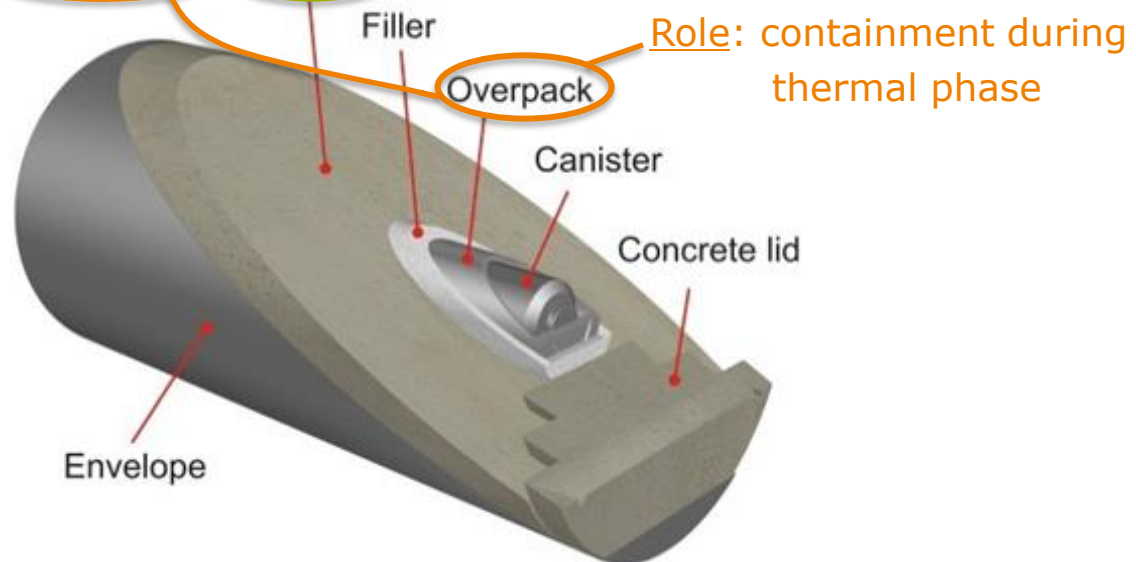
- **"Half-Scale Tests" (HST) programme**
 - Demonstrate feasibility of construction: 2 tests have been performed (HST-1, HST-2) of the construction of an (instrumented) buffer
- **Phase 1: hardening after casting**
 - Check temperature, deformations against model predictions
 - Verify absence of early-age thermal cracking
- **Phase 2: dummy waste canister insertion**
 - Casting of filler under thermal load
 - Check temperature, deformations against model predictions
 - Assess risk of cracking under thermal load
- **Phase 3: cooling**
 - Verify absence of Delayed Ettringite Formation (see presentation 1) after high temperature excursion



Priorities in the Belgian national programme: long term evolution of the supercontainer

▪ Contained environment concept

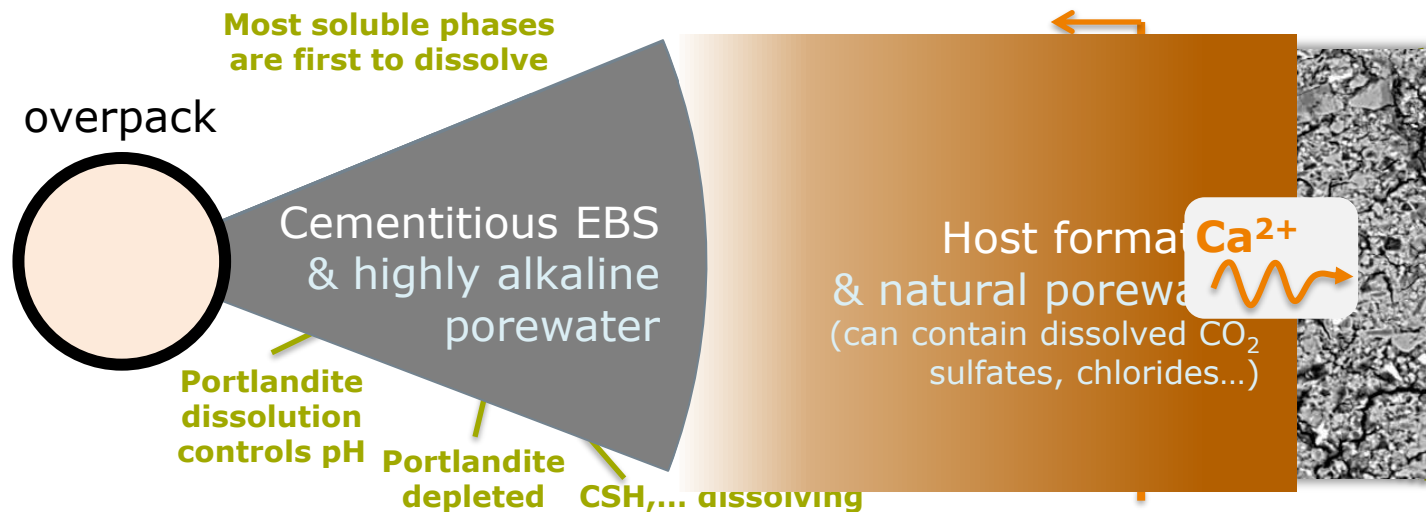
- The cementitious **buffer** provides a **high pH** environment to the carbon steel **overpack**



- Under high pH, the passive layer formed on carbon steel ensures **uniform corrosion** with **very low corrosion rates**
→ **how long** can the cementitious buffer ensure the high pH conditions at the buffer/overpack interface?

Priorities in the Belgian national programme: long term evolution of the supercontainer

- Time scales involved → stepwise approach to bound the evolution, by modelling
 - **Ca-leaching** (as in presentation 3), but **diffusion**-controlled



Priorities in the Belgian national programme: long term evolution of the supercontainer

- **Example of Ca-leaching model results (Diffu-Ca model, CEA)**
 - Model with simplified chemistry
 - Except for time, distance, all parameters are expressed as functions of the concentration of calcium in solution
 - Provides profiles of calcium in solution and solid, flux of leached calcium, pH, porosity and diffusion coefficient

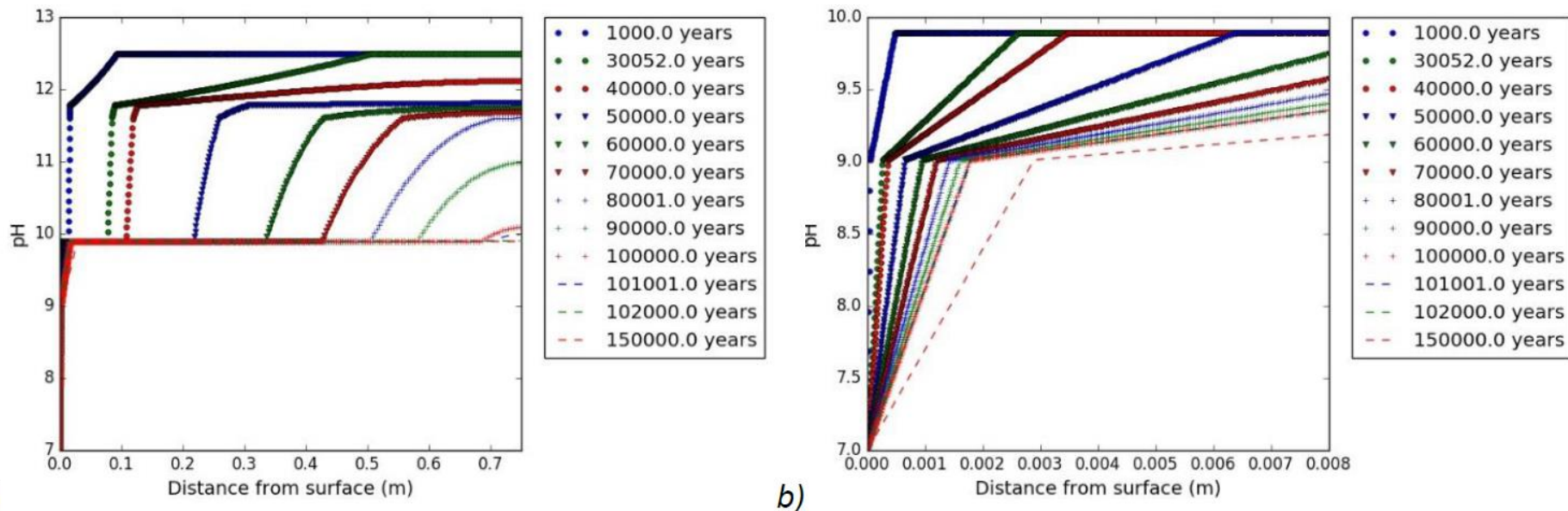
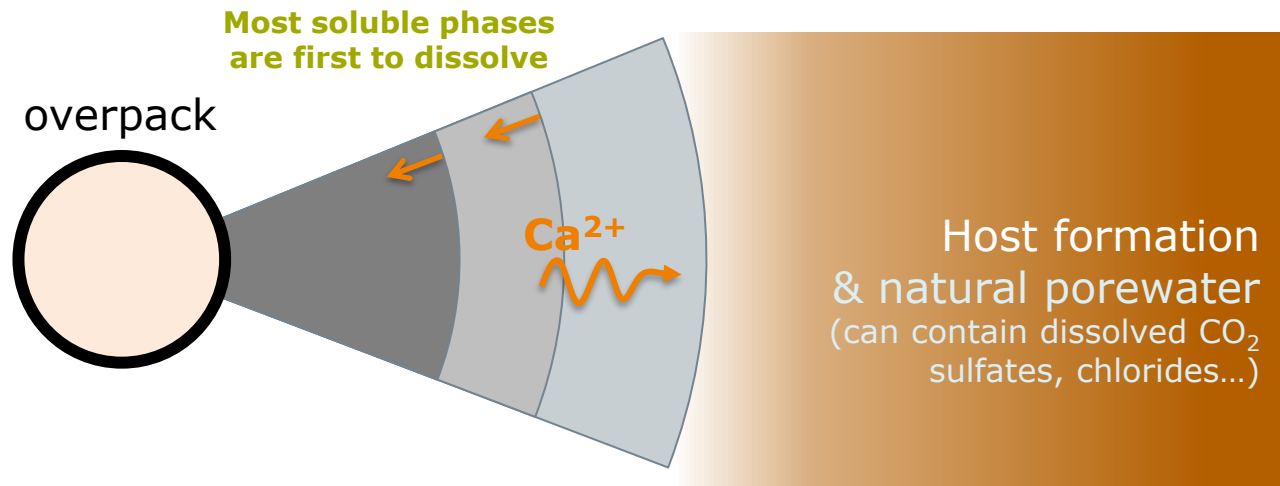


Figure 16 : Evolution of pH within concrete buffer at different duration (a) and a zoom on the few first millimeters from the interface between aggressive solution and concrete buffer (b) for a discretization with 1000 meshes.

Priorities in the Belgian national programme: long term evolution of the supercontainer

- Time scales involved → stepwise approach to bound the evolution, by modelling
 - Ca-leaching (as in presentation 3), but **diffusion**-controlled



- Carbonation: what happens at the interfaces? (→CEBAMA)
 - Effect of sulfates,... on buffer? (→resistance to chemical attacks?)
 - What if the porosity is heterogeneous (cracks)?
- With more "realism" come more parameters & associated uncertainties!

RD&D priorities on cement within the Belgian national programme

- Cementitious materials in the long term management of radioactive waste
- Research, Development and Demonstration (RD&D) on cementitious materials in that context
- Priorities in the Belgian national programme
- **Conclusions**

Conclusions

- **Cementitious materials are ubiquitous in long term management of radioactive wastes**
- **As uses are multiple, possible RD&D topics abound**
- **R&D priorities strongly linked to programme stage and boundary conditions**
 - Focus first on application specific, urgent & potentially high impact topics (for the latter, compare to potential cost of non-R&D in terms of later design change)
 - Leverage generic expertise from "classical" uses of cementitious materials + benefit from return of expertise from sister programmes
 - Some level of long-term, more fundamental research is desirable in a spirit of confidence building, evaluation of performance reserves and openness for the unexpected

Conclusions

▪ **Current priorities for ONDRAF:**

- compatibility of **cemented waste forms** with disposal
 - Screening for problematic behaviour, characterisation efforts
 - Formulations of waste immobilization matrices
 - Strong link with waste acceptance process, criteria
- For **cAt**,
 - Impervious top slab development
 - Role of discontinuities in flow and transport through modules
 - Reduce uncertainties on radionuclide sorption in cementitious materials at different evolution stages
- For **deep disposal**, focus is on performance of supercontainer
 - THM evolution of the buffer: can structural integrity be relied upon at least during operational and retrievability phase?
 - Long term chemical evolution of the buffer, starting with Ca-leaching: how long could high pH conditions prevail at the overpack?
 - Other long-term processes that could affect the role of the buffer

Thank you for your attention

▪ Questions?

- Thanks to colleagues S. Ben Hadj Hassine, W. Wacquier, S. Brassinnes, D. Léonard, H. Van Baelen and R. Gens for valuable input to this presentation

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The CEBAMA project

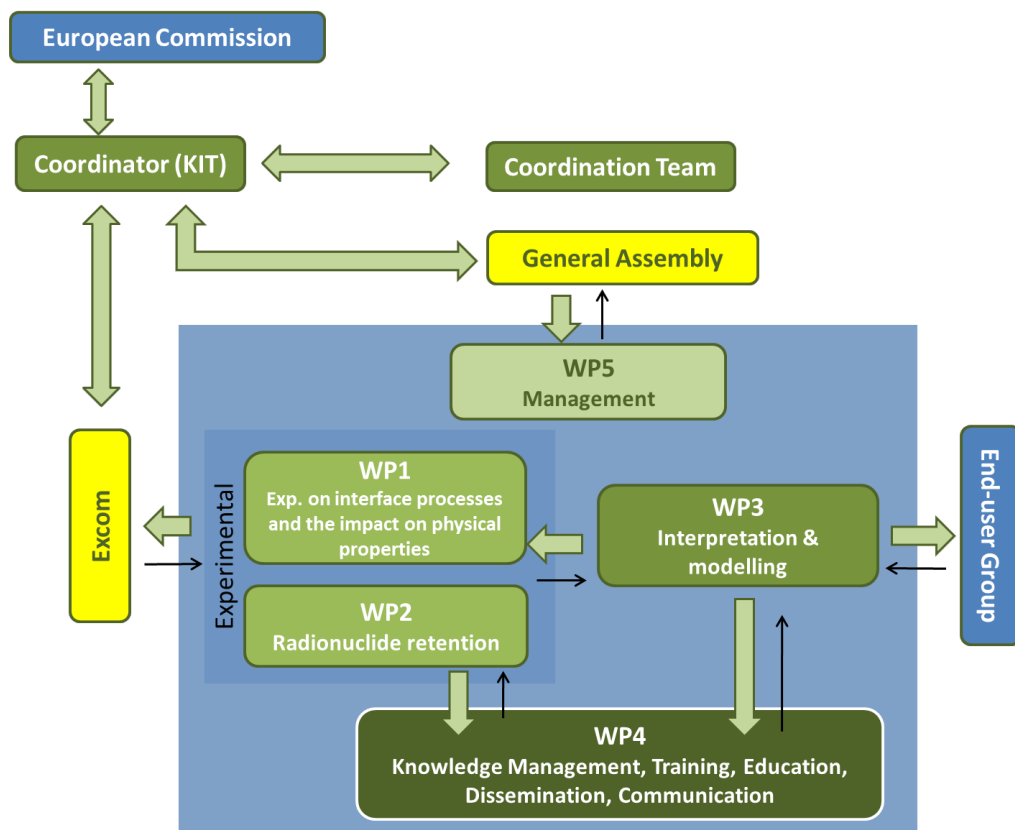


■ What is CEBAMA?

- A collaborative project related to the use of CEment-BASed MAterials in nuclear waste disposal applications
- Funded by the EC (Horizon 2020)
- 4 years started in June 1st 2015
- 27 Beneficiaries (SCK•CEN included), O/N part of the End-Users



The CEBAMA project



- **WP1:** Experiments on interface processes between the host rock and the concrete materials and the impact on physical properties
- **WP2:** Radionuclide retention in high pH concrete environments (Be, C, Cl, Ca, Se, Mo, I and Ra)
- **WP3:** Interpretation and modelling of transport processes in cement-based materials and interfaces with host rocks

Urgency – impact – effort

