New cementitious binders for immobilization of radioactive waste

EURECA-PRO

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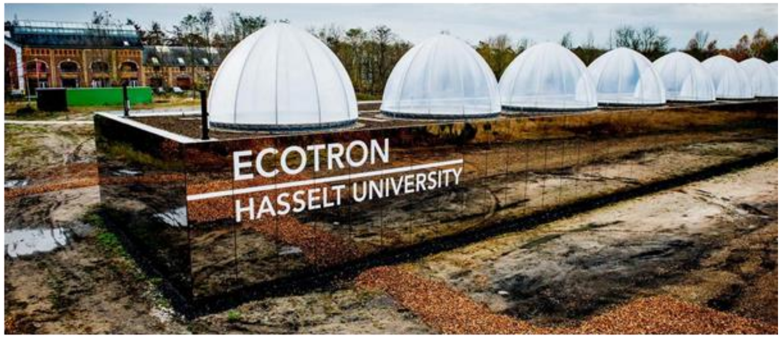
NuTeC
Nucleair Technologisch Centrum

Centre for Environmental Science (CMK)

CMK conducts fundamental and applied research:

1. To understand **influences of the environment** on organisms

2. To develop and assess sustainable **clean technologies** to mitigate influences of the environment on organisms



3. To monitor, value and **optimize biodiversity and ecosystem services** under different stress conditions, including climate change

CMK
CENTRE FOR
ENVIRONMENTAL SCIENCES
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Nuclear Technology (NuTeC) research group is embedded in the CMK

NuTeC

NuTeC – Research in environmental technology

- **Nuclear decommissioning** and immobilization of **nuclear waste**

Characterization Treatment Encapsulation -immobilization

- **Recycling** of **naturally occurring radioactive materials (NORM)**

Characterization Treatment Recycling in construction

- **Adsorption & desorption** of **(radiological) pollutants**

Pyrolysis technology

Adsorbent Biochar, AC, zeolite, …

Adsorption/desorption of pollutants in water, air, soil

Outline

1. **Introduction – current situation**

2. New cementitious binders
    a) Immobilization
    b) Effects of ionizing radiation on the encapsulation matrix
    c) Treatment/management of liquid radioactive waste

3. Lessons learned

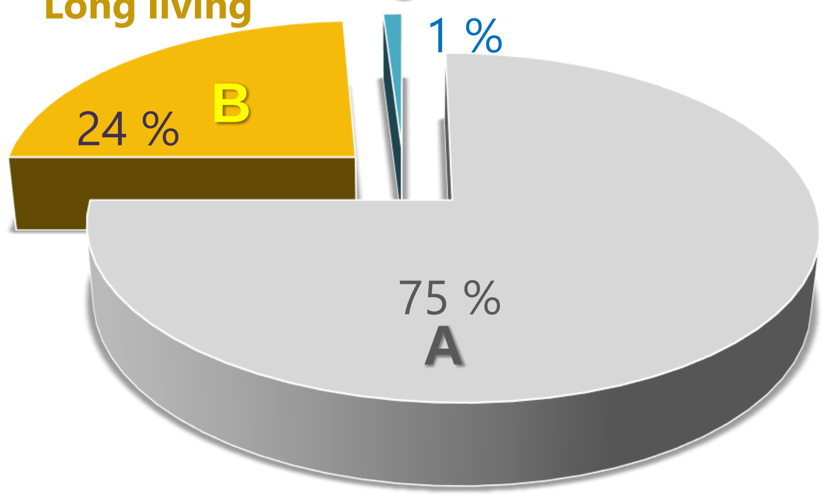
Classification radioactive waste (Belgium)

- Category A – B – C waste

	Low activity [< 5 mSv/h]	Intermediate activity	High Activity [> 2 Sv/h]
Short half life [< **30 year**]	A	A	C
Long half life [> **30 year**]	B	B	C

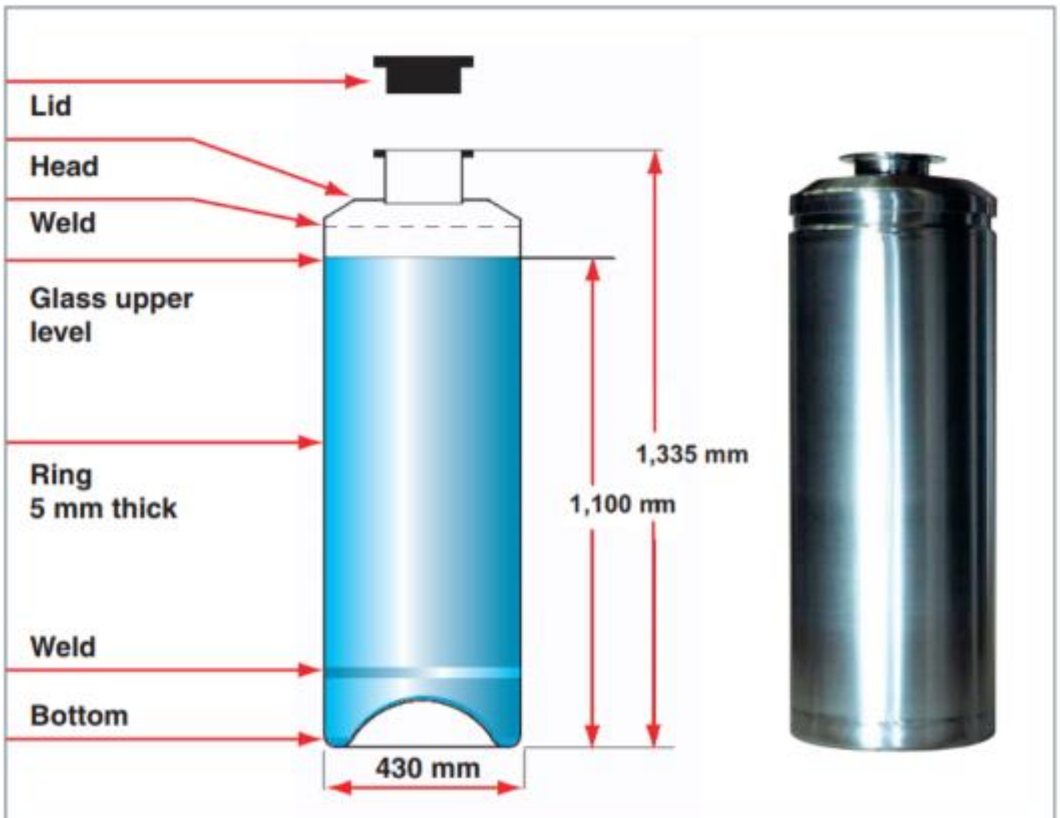
Classification radioactive waste (Belgium)

- Category A – B – C waste



High activity waste: vitrification

- Liquid waste added to **ceramic- or glass**-forming chemicals, mixed and thermally treated to immobilize in solid waste-forms.
- **Effective** but very **expensive** → less suitable for larger volumes



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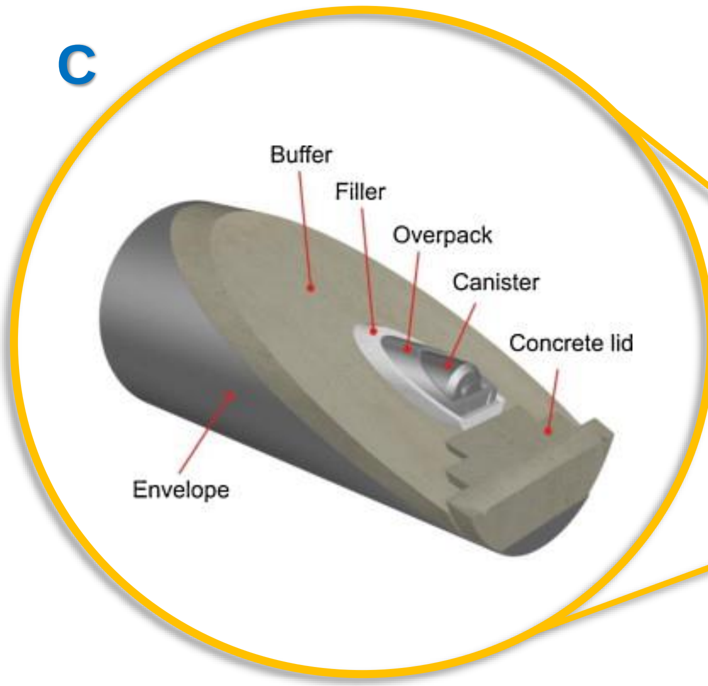
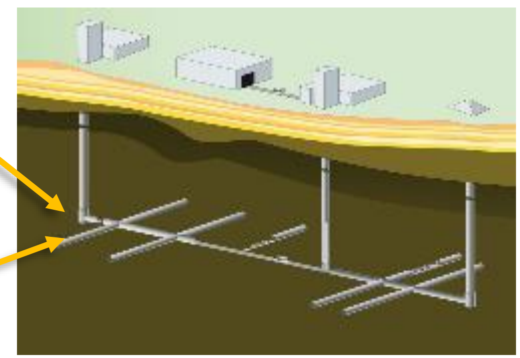
[a, b] CEA, http://www.materials.cea.fr/en/PDF/MonographiesDEN/Nuclear%20waste%20conditioning_CEA-en.pdf, , accessed 2018
[c] Belgoprocess, https://www.belgoprocess.be/activiteiten/tussentijdse-opslag-van-geconditioneerd-belgisch-radioactief-afval, accessed 2018

High-level waste: Deep geological disposal

C



B & C



Low and Intermediate Level Waste: cementation

- Widely used: compaction or incineration, followed by **cementation** as a cost-efficient and proven technology

Short living waste (< 30 year)

Low and Intermediate Level Waste: near surface disposal

NIRAS, https://www.niras.be/oppervlaktebergingsinstallatie-dessel-0, accessed 07/09/2018

Low and Intermediate Level Waste: cementation

Current methods

Cementation using ordinairy Portland cement (OPC)

Advantages OPC

Availability

Relative **low-cost**

Easily modified to fit specific purpose

Robust and tolerant to a wide variety of wastes

Relatively easily **processed remotely**

…

Low and Intermediate Level Waste: cementation

Current methods

Cementation using ordinairy Portland cement (OPC)

Disadvantages OPC [a]

***High temperature increase** resulting from the heat of hydration, giving rise to thermal stresses and possible cracking.

*The **weak immobilization potential** for monovalent cations such as Cs^+.

***Moderate durability** (e.g. leaching through alkali-silica reaction (ASR) with certain types of RAW under certain conditions)

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Similar materials, different uses

Construction

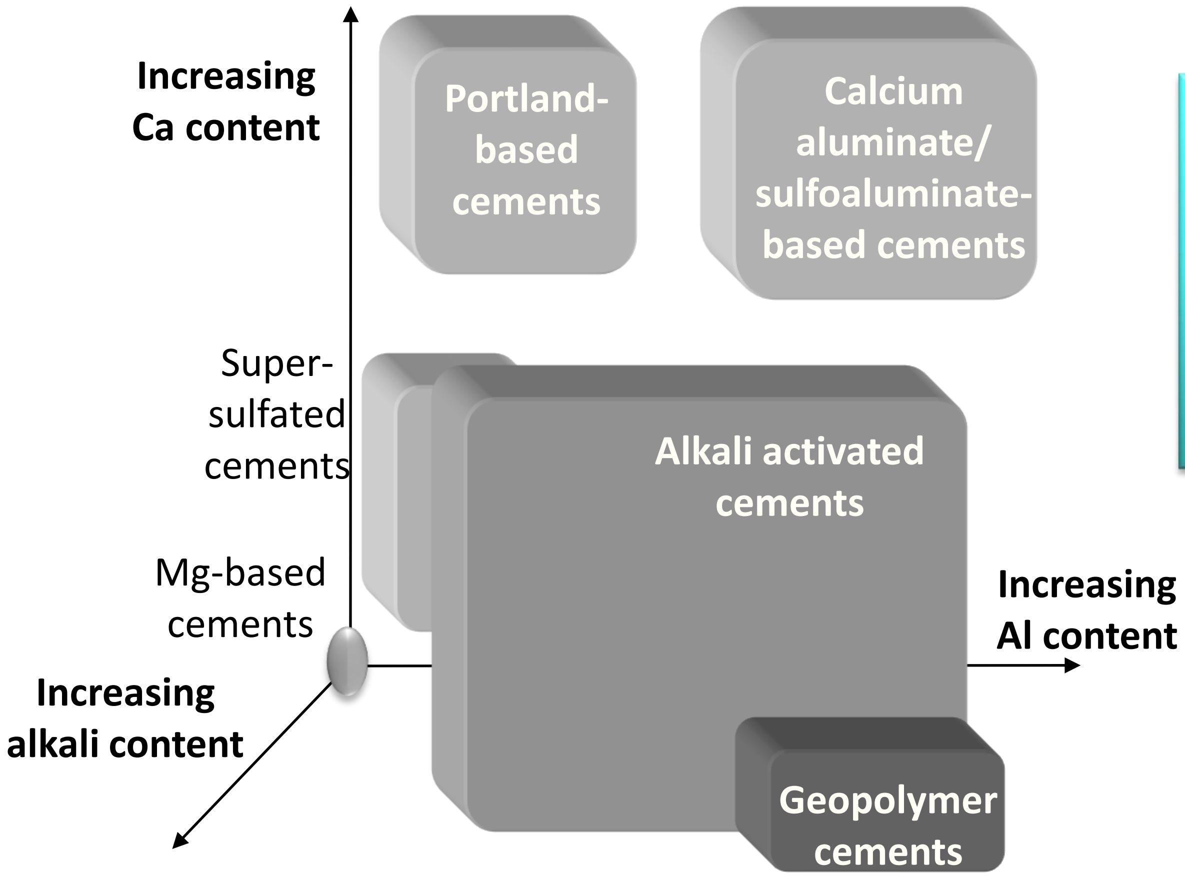
- High strength
- Fast strength development
- Control of flow properties with organic admixtures
- Durability for 50-200 years service life
- Passivation of mild steel
- Low cost

Waste immobilisation

- **Durability** for 100,000 – 1,000,000 years service life
- **Binding of radionuclides**
- **Low heat evolution**
- **Dimensional stability**
- Controlled **corrosion** of reactive metals
- Stability under **irradiation**

Can we realistically expect all this from a single material?

Traditional and non-traditional cements



Moving from a single universal cement to an array of cement types

Most important: Designing materials that are fit for purpose!

J.L. Provis & J.S.J. van Deventer (eds.), State of the Art Report of RILEM TC 224-AAM, 2014

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Production Alkali activated materials (AAMs)

Solid **aluminosilicate** source + **Alkali** silicate/hydroxide **activating** solution

Dissolution

Oligomerization

Polymerization

Precursor

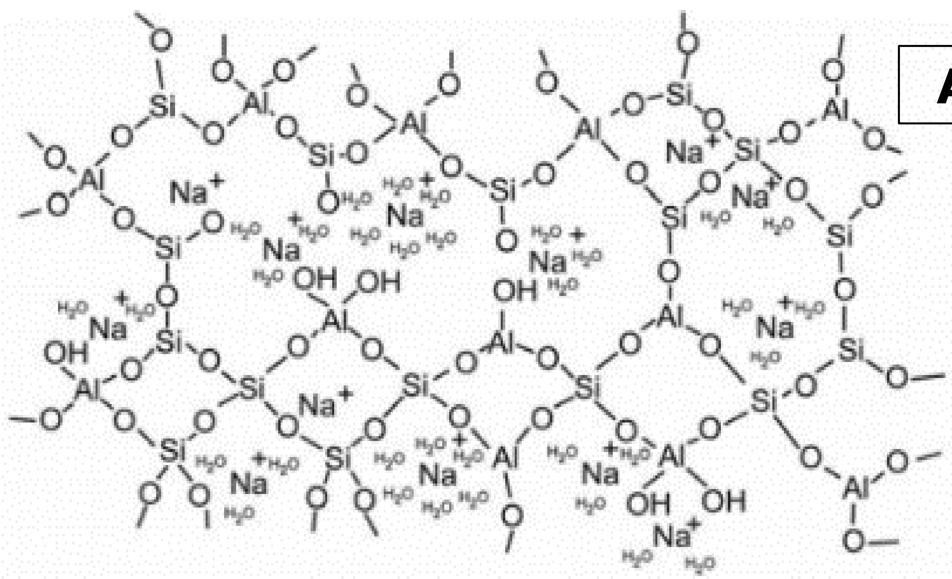
Activator

Synthesis parameters

Aluminosilicate polymer

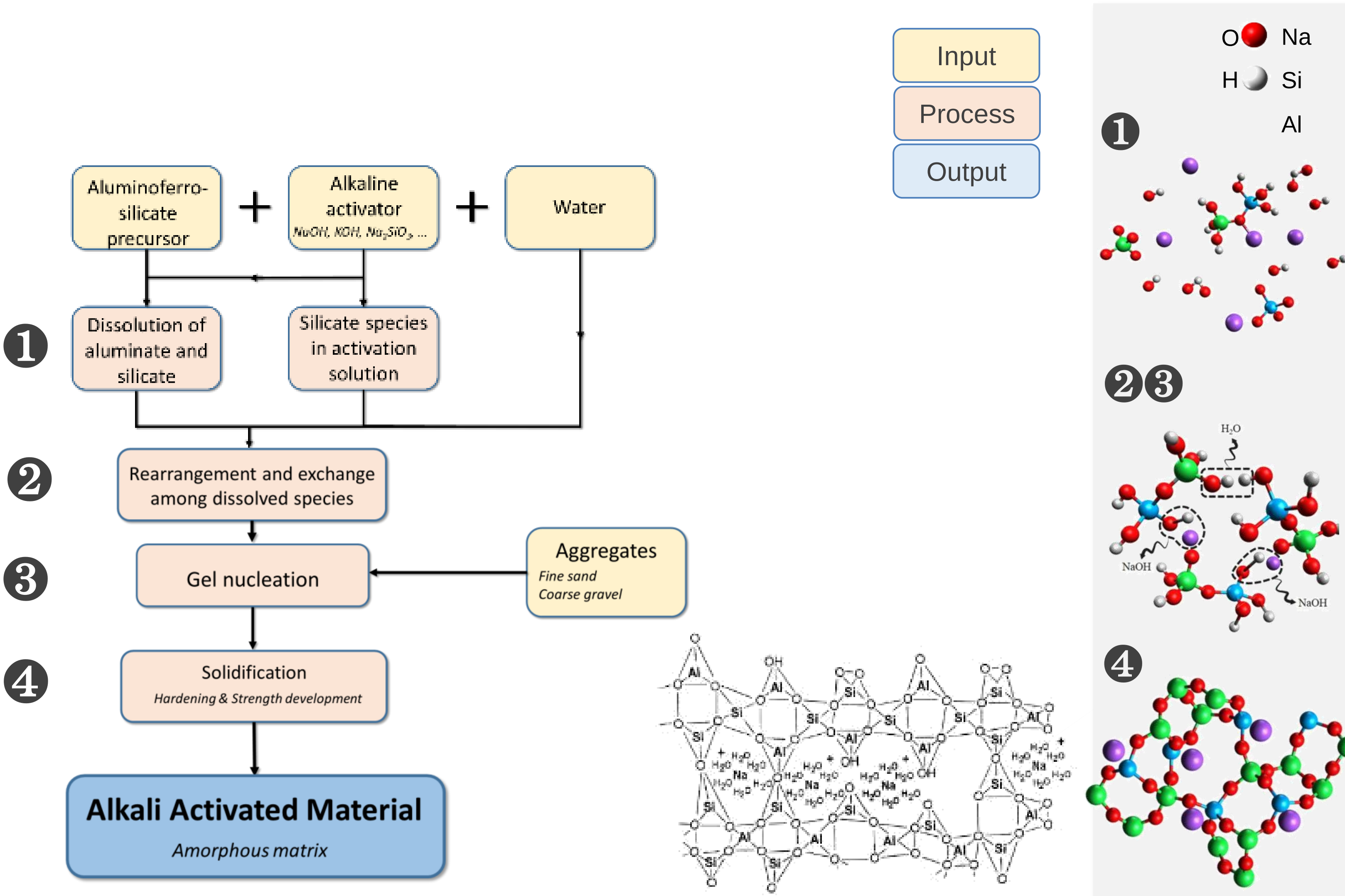
Adapted from Deventer (2007)

AAM



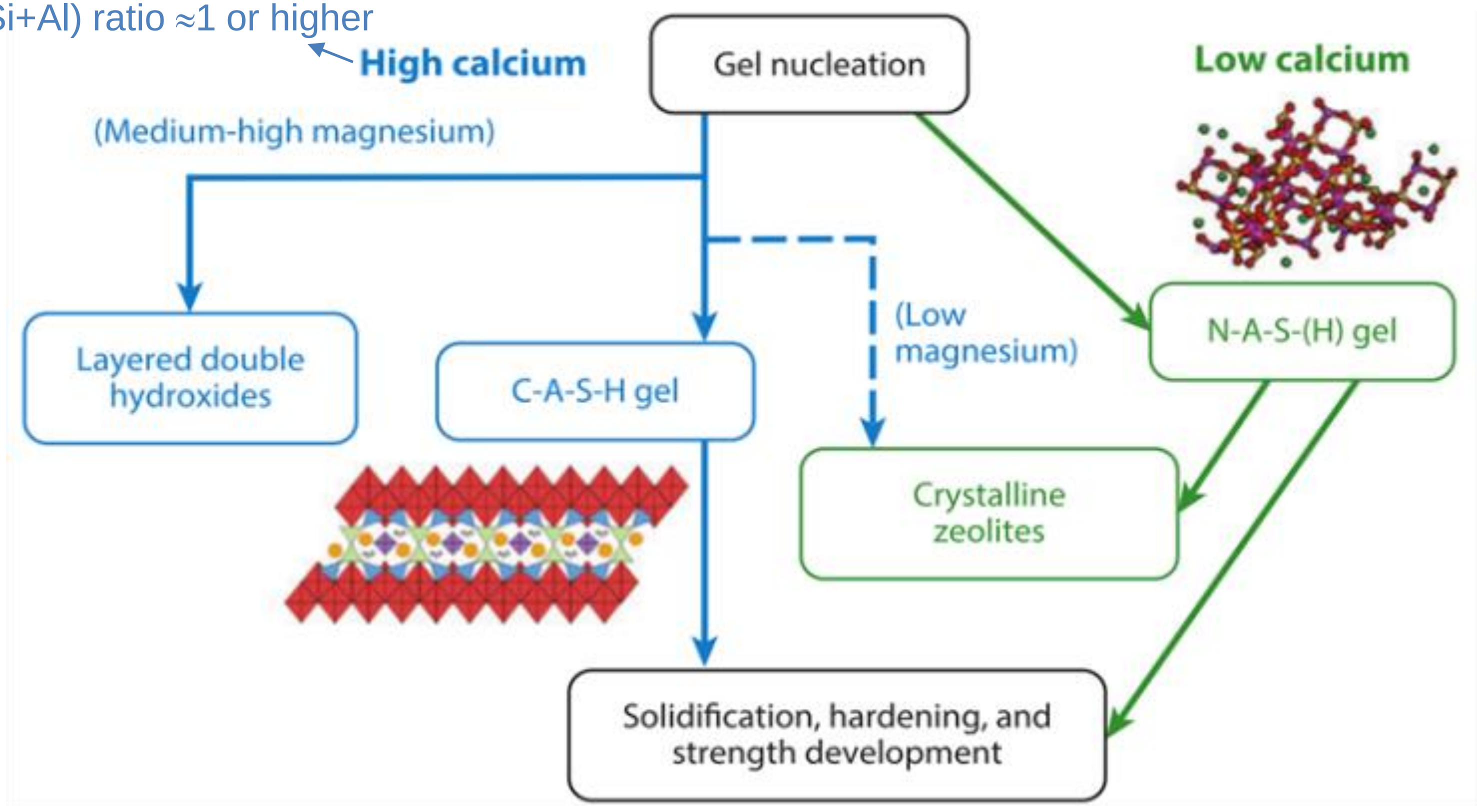
Adapted from Rowles (2008)

Alkali activated materials (AAMs) - chemistry



Alkali activated materials (AAMs) - chemistry

Ca/(Si+Al) ratio ≈1 or higher



Calcium (sodium) aluminosilicate hydrate (C-(N)-A-S-H)-type gel.

Alkali (sodium/potassium) aluminosilicate hydrate (N-A-S-H or K-A-S-H)

Low permeability

Excellent chemical and thermal resistance

Blended binder systems: can offer benefits of two gels

Provis, J.L., Bernal, S.A., 2014. Geopolymers and related alkali-activated materials. Annu. Rev. Mater. Res 44, 299–327

Immobilisation of Cs and Sr in alkali activated materials

ASTM C1220-98 (*Static monolithic leaching test)*

"Clean" precursor development	Start from pure Al, Si and Ca oxides
	Mix and melt at high temperature (1630 °C)
	Quench in water
Model AAM-composition	Different compositions
	2 M NaOH
	1 wt% Cs ; 0.1 wt% Sr
	Leaching **28 d** at 90 °C

- [PhD Niels Vandevenne, 2019]

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Immobilisation of Cs and Sr in alkali activated materials

ASTM C1220-98 (*Static monolithic leaching test)*

Cs

92 % immobilisation 28 d leaching 90 °C

Dependent on the **Si/Al** and **Ca/(Si+Al)** ratios of the precursor

Combination of initial **wash-off**, **diffusion** and **depletion** of an **easily-leachable fraction**

Sr

99 % immobilisation 28 d leaching 90 °C

Dependent on the **Ca/(Si+Al)** ratio

Limited to a **small fraction** present on or near the **surface**

- [PhD Niels Vandevenne, 2019]

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Immobilisation of Cs and Sr in alkali activated materials

- **Control** of the matrix?
  - Residues <-> model compounds
  - Variations in the (trace) element concentration?
  - Impact of the **waste-loading**?
  - Using **lower alkaline** activating solutions
  - **Durability** studies for use in long-term waste management

Effect of waste-loading on matrix properties

Cs^{+} addition: Homogeneous incorporation in AAM
No significant effects on
Early reaction kinetics

Sr^{2+} addition: Sr-precipitation ($Sr(OH)_2$)
Significant effects on
Early reaction kinetics

- [PhD Niels Vandevenne, 2019]

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Effects of ionizing radiation on concrete

Dehydration	Radiation-induced carbonation

Radiolysis
Decomposition of water and formation of H_2 gas

Heating
Evaporation of water

Radiolysis
Formation of H_2O_2

→ $CaO_2.H_2O$
→ Ca(OH)
→ $CaCO_3$

Drying & Shrinkage

Decomposition

Cracking & Loss of strength

- [PhD Bram Mast, 2020]

Radiation-induced mechanical degradation in AAMs

IP composite	wt.%
SiO_2	34.0
Fe_xO_y *	24.1
CaO	21.1
Al_2O_3	11.9
Na_2O	6.3
Other	2.1

* 57% FeO and 43% Fe_2O_3

- Design4application
  - **Radioactive waste encapsulation**
    - Less $Ca(OH)_2$
    - Reduced H_2O content: less damage as a result of dehydration
    - High immobilization capacity
  - **Good Shielding properties**

- [PhD Bram Mast, 2020]

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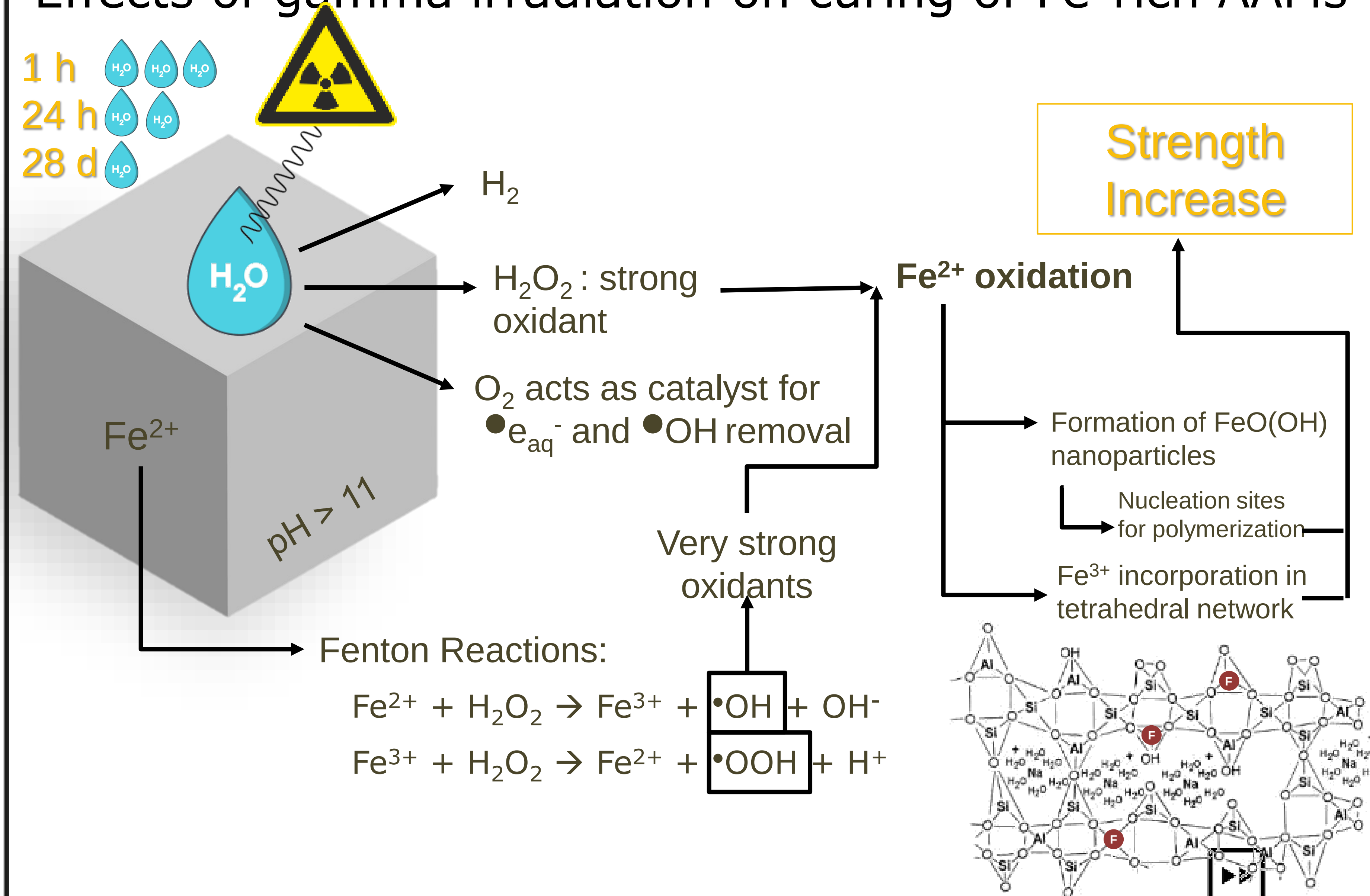
Effects of gamma irradiation on curing of Fe-rich AAMs

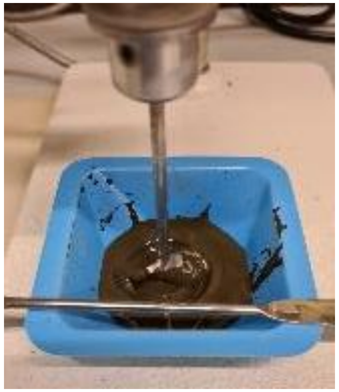


- [PhD Bram Mast, 2020]

Immobilisation of liquid nuclear waste

1. Adsorption on **nanoparticles**

Cerium Oxide Nanoparticles (CeO_2 NPs)



2. Final immobilisation with **new cementitious binders**

Collaborative Doctoral Partnership with JRC (Joint Research Centre)

- [PhD Angela Mooren]

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3. **Lessons learned**

Lessons learned

- AAMs have the potential to outperform OPC but a lot of extra studies are required.

- **Availability** and **variations** in elemental composition of input materials
  → issues for **controlled** nuclear waste encapsulation

- **Presence of iron**: additional effects of gamma irradiation on curing of Fe-rich AAMs

- Not the best idea to use (long lived) NORM residues for immobilization of (short lived) Cat A waste