Use of phosphogypsum in alkali-activated binders: radiological and leaching assessment

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NuTeC
Nucleair Technologisch Centrum

Outline

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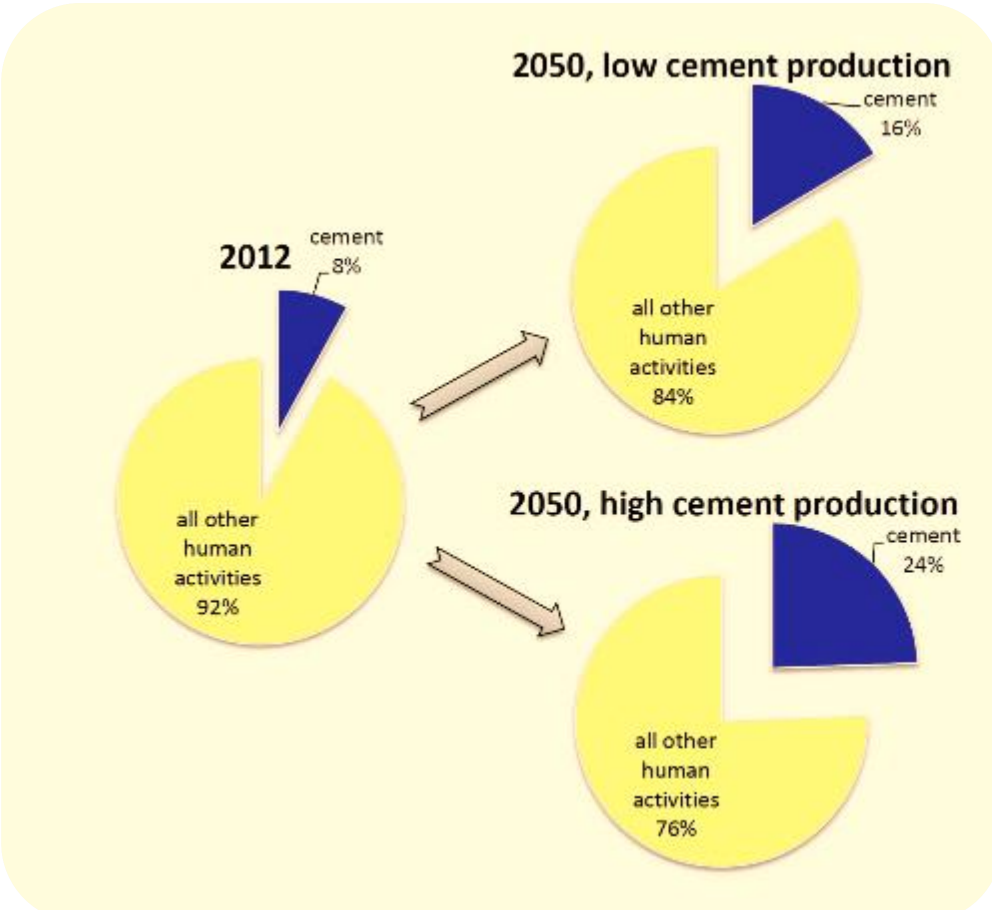
Terranova phosphogypsum deposit (DEME, Zelzate, Belgium)

https://www.deme-group.com/news/terranova-solar-largest-solar-park-low-countries?lang=nl

CO_2 emissions from Ordinary Portland Cement (OPC)



- **Alternative? Alkali Activated Materials (AAMs)**:
  - Reduce CO_2 emissions by up to 80%
  - Comparable technical performance in many aspects
  - Allow incorporation and recycling of several types of industrial residues.

Production Alkali activated materials (AAMs)

Solid aluminosilicate source + Alkali silicate/hydroxide activating solution

NORM precursor

Dissolution

Oligomerization

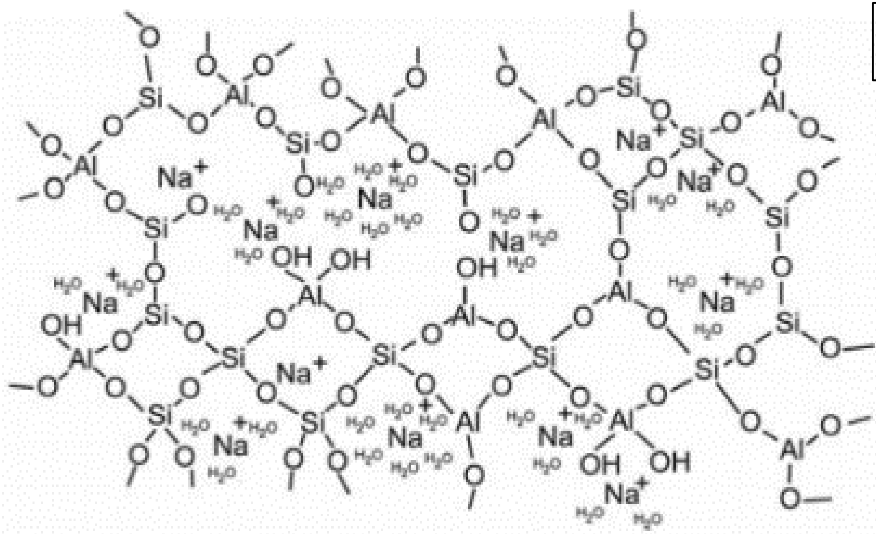
Polymerization

Activator

Synthesis parameters

Aluminosilicate polymer

Adapted from Deventer (2007)



AAM

Adapted from Rowles (2008)

Research @ NuTeC (Uhasselt)- MTM (KULeuven)

CMK CENTRUM VOOR MILIEUKUNDE UHASSELT

By-products

Fly ash

Red mud

Phosphogypsum

Alkali activation

Use in publicly accessible environment

Use in nuclear safety applications

- Gamma dose evaluation

- Radon exhalation
- Leaching

- Leaching

- Durability prediction in gamma radiation field

Tom Croymans

Katrijn Gijbels

Zoltan Sas

Niels Vandevenne

Bram Mast

Use of by-products in Alkali Activated Materials

Industrial by-products

Activating solution

NaOH
KOH
Na_2SiO_3/NaOH
...

T, t

Curing

AAMs

Research questions:

- Do the AAMs provide a **suitable incorporation matrix** for **recycling of phosphogypsum**?
- Can we **control/limit leaching & radon emissions** from the AAMs?

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3. Results

4. Conclusion & outlook

Sample mix design

Phosphogypsum (PG): IAEA reference material n° 434

96 wt% $CaSO_4.2H_2O$, 1-2 wt% P_2O_5,
1.2 wt% F, 1 wt% SiO_2 and 0.2 wt% Al_2O_3

Ground Granulated Blast Furnace Slag (GGBFS) from Belgian steel producing company

40.3 wt% CaO, 11.4 wt% Al_2O_3, 8.2 wt% MgO, 1.1 wt% SO_3,
0.8 wt% TiO_2, 0.8 wt% Na_2O, 0.5 wt% K_2O, 0.3 wt% Fe_2O_3

Alkali activators:

- sodium silicate
- sodium hydroxide

Sample	wt% GGBFS	wt% PG	SiO_2/Na_2O	H_2O/Na_2O
SS1	90	10	0.75	20.0
SH2	90	10	0	27.8
SH3	90	10	0	18.5

*Note: alkali activator/precursor ratio of 0.6 was chosen due to a decline in the workability upon PG incorporation (high specific surface area)

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Leaching & radon exhalation tests

Type of Radiation	Nuclide	Half-life
α	Uranium-238	4.5 x 10^9 years
β	Thorium-234	24.5 days
β	Protactinium-234	1.14 minutes
α	Uranium-234	42.33 x 10^5 years
α	Thorium-230	8.3 x 10^4 years
α	Radium-226	1590 years
α	Radon-222	3.825 days
α	Polonium-218	3.05 minutes
β	Lead-214	26.8 minutes
β	Bismuth-214	19.7 minutes
α	Polonium-214	1.5 x 10^-4 seconds
β	Lead-210	22 years
β	Bismuth-210	5 days
α	Polonium-210	140 days
	Lead-206	stable

Leaching test: up-flow percolation CEN/TS 16637-3.

Radon exhalation tests (SARAD RadonScout)

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Experimental methods

- Chemical & radiological analysis:
  - ^{232}Th and ^{238}U:
    - Thermal and epithermal **neutron activation analysis (NAA)**
  - (Long-living) radionuclides:
    - **HPGe**
  - Non-radiological elements:
    - Quantitative analysis via **ICP-OES and ion chromatography**
- Microstructural analysis:
  - N_2 sorption measurements
  - Mercury intrusion porosimetry (MIP)
  - Scanning electron microscopy (SEM)

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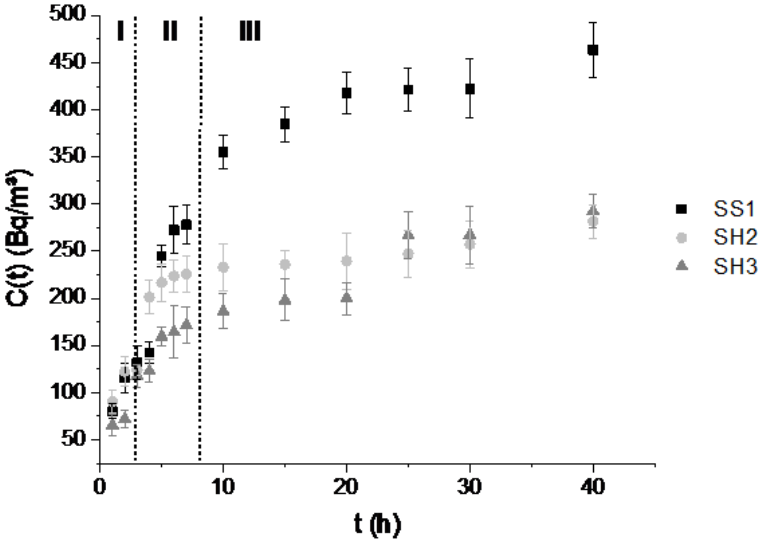
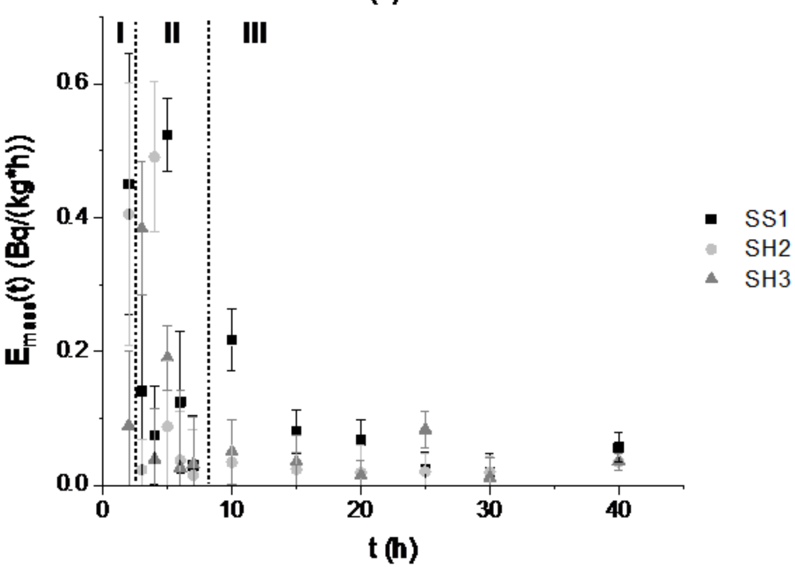
Radon concentration & exhalation rate in accumulation chamber during **hardening of fresh pastes**



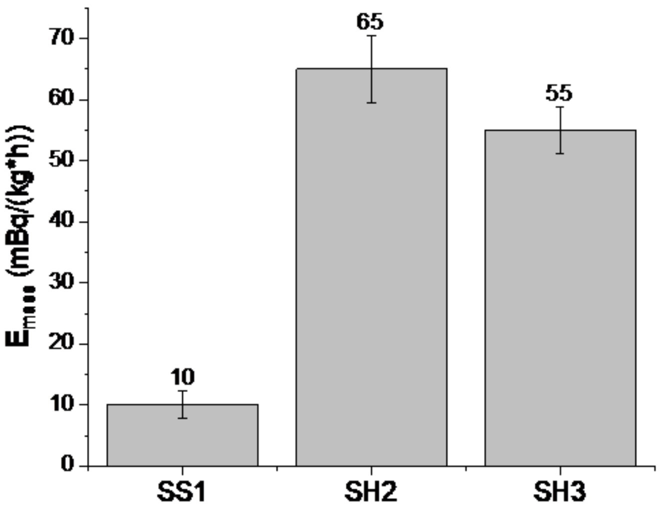


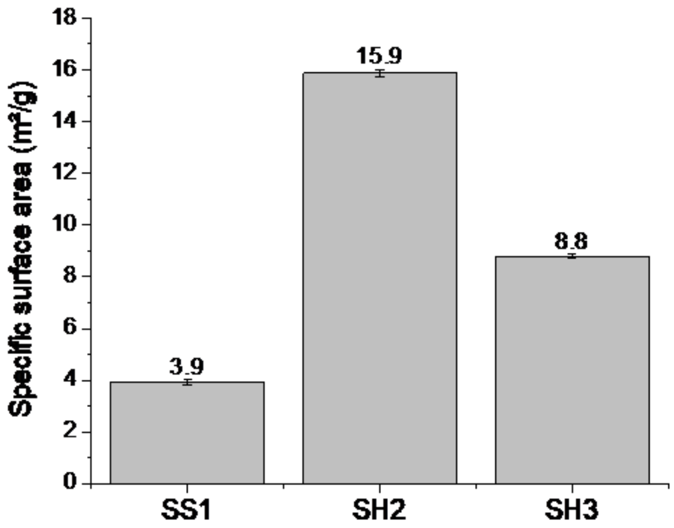
I Dissolution of precursors

II Densification and drying of the sample

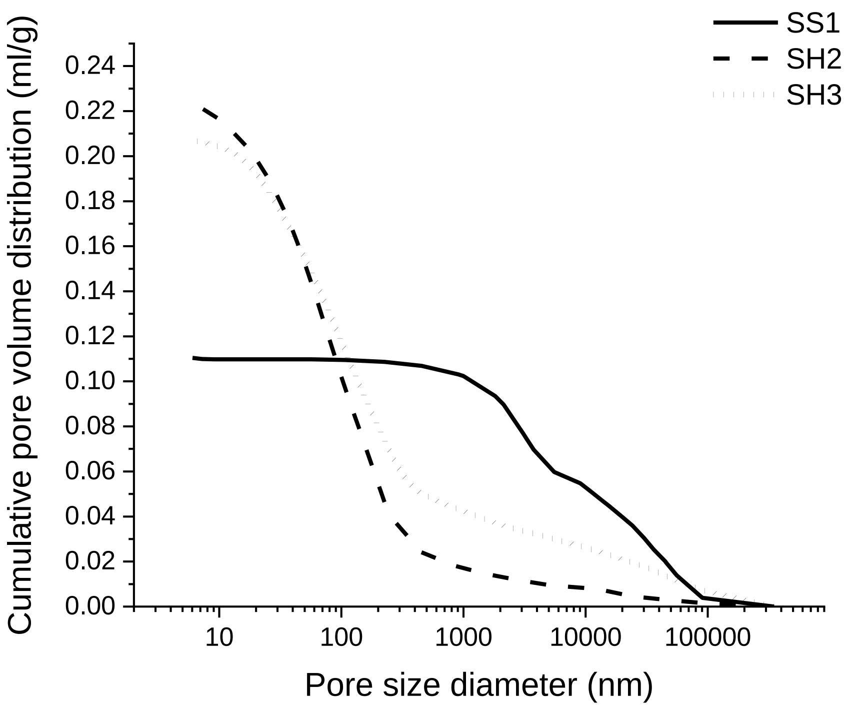
III Further densification and decreasing porosity

Exhalation rate of samples in dry condition after 28 days curing





- Greatly dependents on the used alkali activator
- Availability of **silicate species** in the alkali activator for SS1 resulted in **lowest exhalation rate**



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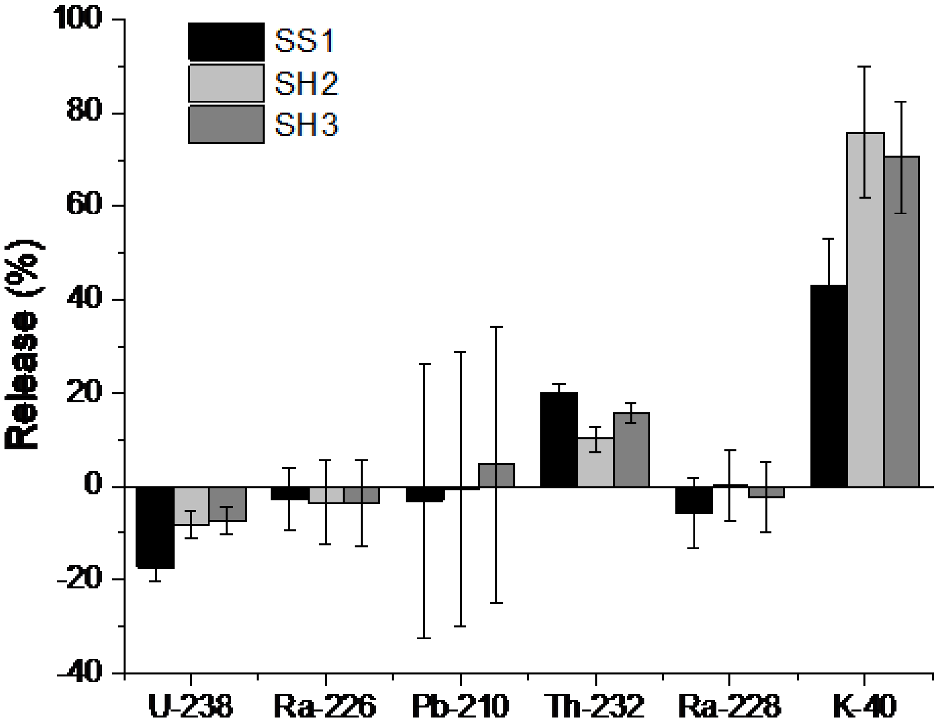
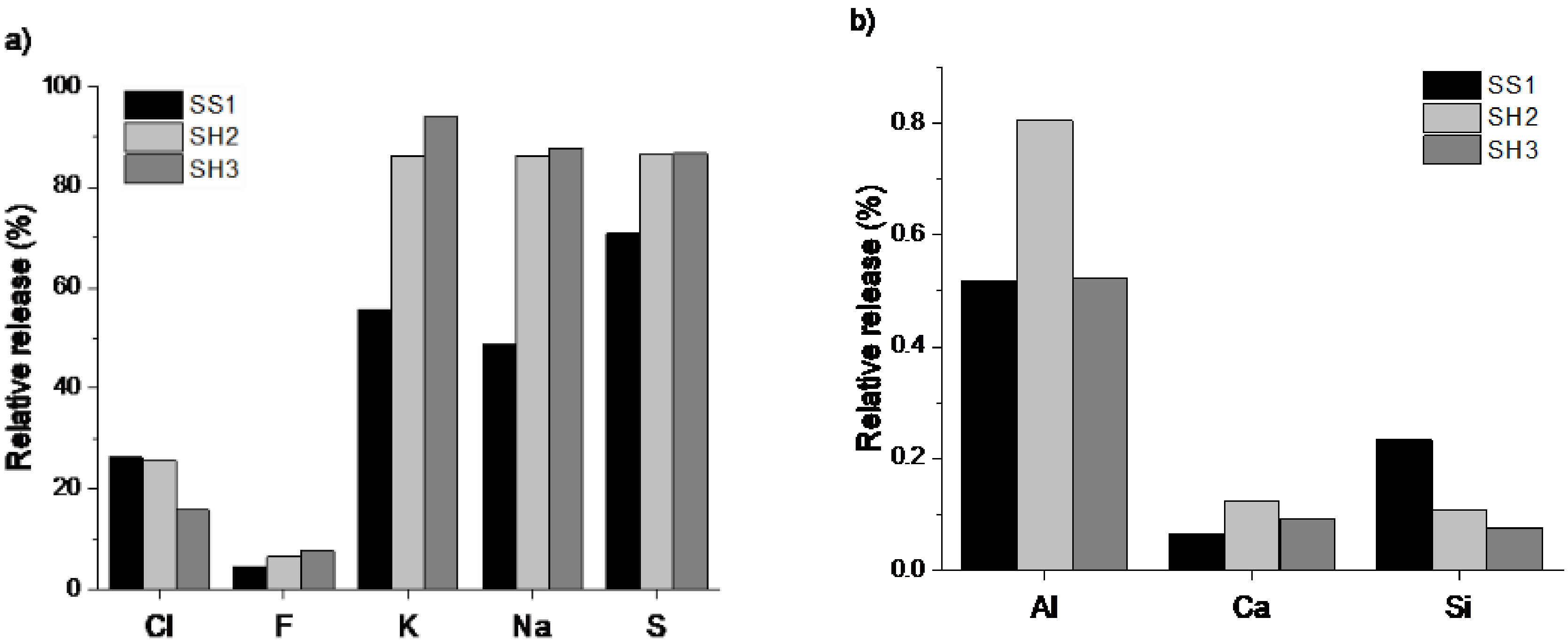
Release of naturally occurring radionuclides upon leaching



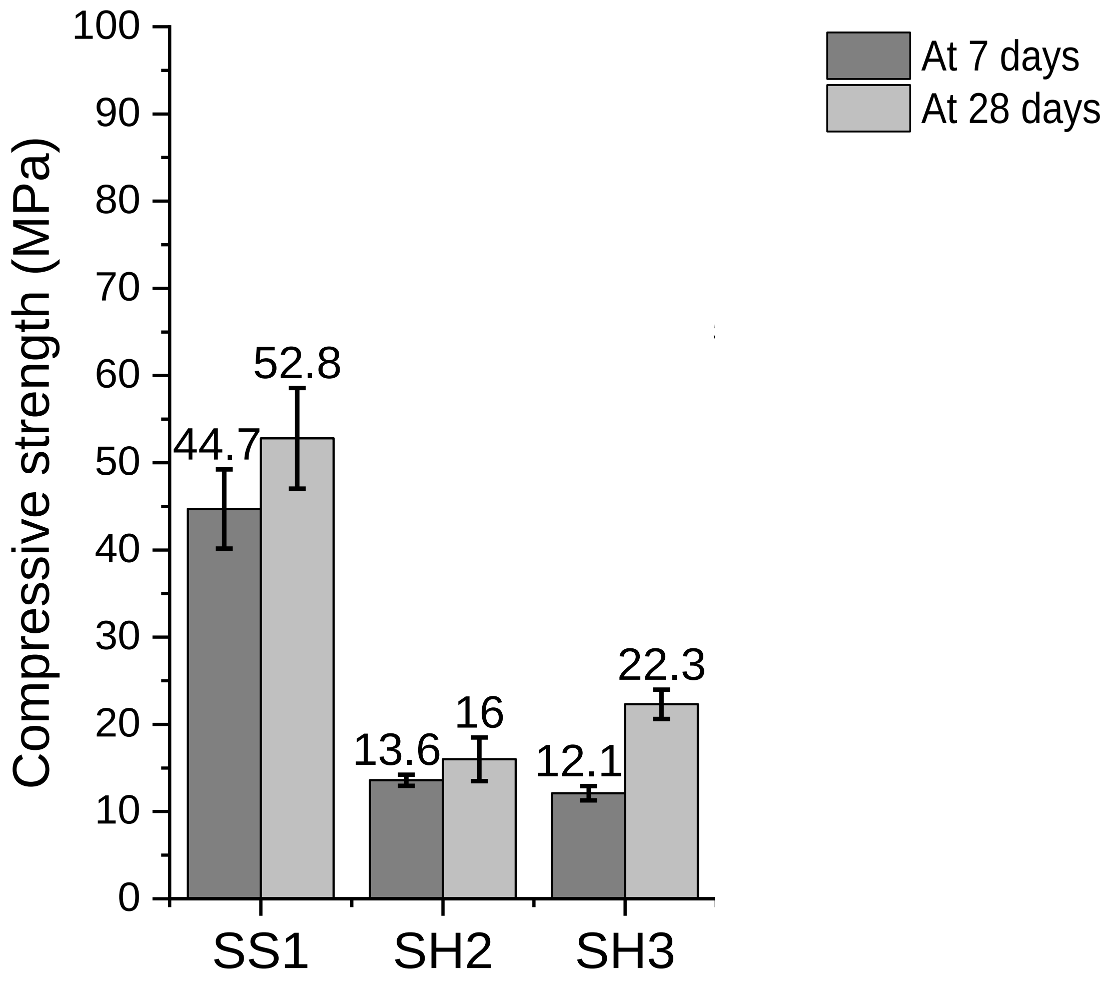
- Alkaline environment:
  - Potassium: large release [charge balancing ion]
  - Thorium (Th^{4+}): leachable and non-leachable complexes [hydroxide & hydroxo-carbonate complexes]
    - ***Na_2SiO_3 activated***: more leachable thorium complexes, (compared to NaOH)
  - Uranium (U^{6+}): retained [in calcium-silicate-hydrate structure or absorbed on silicate surfaces]
  - Radium: retained [similar to Ca]
  - Lead: retained [precipitation $Pb(OH)_2$]

Relative release of non-radiological elements



- From phosphogypsum: F and Ca are well retained in AAM structure (in contradiction to S)

Compressive strength



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Conclusion & outlook

- AAMs: by selecting an appropriate activator, it is possible to control:
  - **Porosity**
  - **Radon emissions**
  - **Leaching behavior**

- Note: Alkali activator/precursor ratio of 0.6 was chosen because of a decline in the workability upon PG incorporation

- In **follow-up studies**:
  - We are trying to **reduce the alkalinity** of the solutions
  - Increase the PG content, while simultaneously achieving good mechanical properties and immobilization of hazardous (radiological) elements

Would like to know more?

- Gijbels K., Iacobescu R.I., Pontikes Y., Vandevenne N., Schreurs S., Schroeyers W. (2018). Radon immobilization potential of alkali-activated materials containing ground granulated blast furnace slag and phosphogypsum. https://doi.org/10.1016/j.conbuildmat.2018.06.162

- Gijbels K., Landsberger S., Samyn P., Iacobescu R.I., Pontikes Y., Schreurs S., Schroeyers W. (2019). Radiological and non-radiological leaching assessment of alkali-activated materials containing ground granulated blast furnace slag and phosphogypsum. https://doi.org/10.1016/j.scitotenv.2019.01.089

- Gijbels K., Iacobescu R.I., Pontikes Y., Schreurs S., Schroeyers W. (2019). Alkali-activated materials containing ground granulated blast furnace slag and phosphogypsum. https://doi.org/10.1016/j.conbuildmat.2019.04.194

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