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Radon immobilization potential of alkali-activated materials containing ground granulated blast furnace slag and phosphogypsum



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HIGHLIGHTS

- Alkali-activated materials developed from ground granulated blast furnace slag.
- Incorporation of 10 wt% phosphogypsum.
- Study of the ²²²Rn immobilization potential dependent on alkali activator.
- ²²²Rn release successfully reduced by physical encapsulation.

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ABSTRACT

This study evaluates the immobilization potential of ²²²Rn in alkali-activated materials (AAMs) based on ground granulated blast furnace slag (GGBFS). As radon source, 10 wt% phosphogypsum (PG) was added to evaluate PG reuse via AAMs. Matrix properties were modified by varying alkali activator's SiO₂/Na₂O and H₂O/Na₂O ratios. During hardening of AAMs, radon release occurs in three stages resulting, after 30 h, to 0.26 Bq/(kg*h). By varying the alkali-activator, massic exhalation for hardened samples decreased down to 0.01 Bq/(kg*h). Radon release was successfully reduced mainly by physical encapsulation, with best results being achieved by adjusting the alkali-activator to SiO₂/Na₂O = 0.75 and H₂O/Na₂O = 20.

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1. Introduction

Phosphogypsum (PG) is a residue from the phosphate production. PG consists mainly of CaSO₄·2H₂O, but can also include a relatively high level of fluoride and some heavy metals (such as Cd or Cr). Depending on the ore and the production process, PG typically contains high levels of ²²⁶Ra, owing to similar chemical properties of radium and calcium, and low levels of ²³²Th and ⁴⁰K [1,2]. The ²²⁶Ra concentration in PG can range from 200 to 3000 Bq/kg [3]. For this reason, PG is classified as naturally occurring radioactive material (NORM). Worldwide, 170 million tons of PG are disposed annually [2] and the recycling rate is still very low, i.e. less than 5%.

It is estimated that the total amount of PG stored will reach 7 to 8 billion tons by 2025 [2]. At the same time, recent evolutions in the phosphate industry, together with rapidly developing economies, are leading to a major increase in the production of PG [2,4], requiring a large disposal area with an elaborated management strategy to avoid soil, water and air pollution. To transform this practice into a more sustainable one, the more effective use of PG needs to be researched. Only the construction industry, with its very large production volumes, is able to process such large amounts. Despite, no effective technologies are known for PG processing and utilization in the construction industry since efficient purification techniques for the removal of radium are lacking [5]. Some studies report the recycling of PG in traditional cementitious [6–9] or alkali-activated materials (AAMs) [10], but the radiological impact was never considered. In order to achieve reuse of PG in construction materials several issues need to be tackled: (1) the presence of potentially hazardous elements in PG (e.g. Cd, naturally

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occurring radionuclides) should be controlled by selecting and optimizing the industrial process; (2) commercially interesting techniques need to be developed for the purification of PG and (3) the resulting construction materials need to be able to immobilize or reduce the impact of the remaining hazardous elements.

The focus of this study is on the immobilization of hazardous elements from PG in AAMs based on ground granulated blast furnace slag (GGBFS). GGBFS is the main by-product originating from the blast furnaces during iron production and is produced by water quenching of molten blast furnace slag. GGBFS is an excellent precursor for alkali-activation because of rapid setting, fast and high strength development, low porosity, and high resistance to chemical attack [11]. Moreover, it represents a viable and sustainable alternative to Ordinary Portland Cement (OPC) because of its advantage to allow the incorporation of larger quantities of residues from other industrial manufacturing processes which are less suitable as precursor [12,13]. GGBFS contains relatively low levels of radionuclides from the ^{238}U and ^{232}Th series, in most cases meaning that activity concentrations are below or around 200 Bq/kg [14].

The incorporation of industrial NORM residues in construction, and more specifically in building materials, requires a cautious attitude because it increases the external gamma dose rate [15] and any porous material containing radium becomes a permanent radon source. It has to be noted that there exist three naturally occurring isotopes of radon (^{222}Rn , ^{220}Rn and ^{219}Rn), their prevalence is significantly different and from the radiological protection point of view, the primary focus is on ^{222}Rn mainly owing to the relatively long half-life (3.82 days for ^{222}Rn ; 55.6 s for ^{220}Rn ; 3.96 s for ^{219}Rn). When ^{226}Ra decays into ^{222}Rn , an α -particle of energy 4.78 MeV is emitted and by recoil, the ^{222}Rn nucleus can get removed from the solid phase into the pore space, provided it is close enough to the boundary, and becomes free to migrate (further called ‘emanation’). Followed by diffusion and advective flow through the pore network, ^{222}Rn atoms can get exhaled to the atmosphere (further called ‘exhalation’) [16]. The amount of radon that can exhale from a building material is dependent on numerous factors like temperature, air pressure, humidity fluctuations, total porosity, pore size distribution and pore type (open or closed) [17]. The alkali-activation mix design is the most important factor affecting the physical structure of AAMs because the chemical reactions occurring during synthesis are dependent on the availability of chemical species. Consequently, it is expected that the type and concentration of alkali-activator can have a large influence on radon exhalation.

Since 2013, a unified regulation regarding radiological protection was defined (EU-BSS, Council directive 2013/59/EURATOM, European Basic Safety Standards) which sets radiological restrictions related to the use of residues in building materials. The reference level applying to indoor external exposure to gamma radiation emitted by building materials, in addition to outdoor external exposure, is set at 1 mSv/y. Concerning radon in workplaces and indoor exposure to radon, the EU-BSS requires that the reference level for annual average activity concentration in air, shall not be higher than 300 Bq/m³ [18].

The study herein investigates the ^{222}Rn immobilization potential of GGBFS-based AAMs when incorporating 10 wt% PG, as a function of type and concentration of alkali-activator. To the best of the authors knowledge, this is the first study that considers valorization of PG in GGBFS-based AAMs and their radon exhalation behavior.

2. Materials and methods

2.1. Sample preparation

GGBFS and PG were used as precursors. GGBFS was provided by a Belgian steel company and PG constitutes an International

Table 1
Sample's mix design.

Sample	wt% GGBFS	wt% PG	Alkali-activator/ Precursor	SiO ₂ /Na ₂ O	H ₂ O/Na ₂ O
SS1	90	10	0.6	0.75	20
SH2	90	10	0.6	0	27.8
SH3	90	10	0.6	0	18.5

Atomic Energy Agency (IAEA) reference material (IAEA 434). Prior to sample preparation, the GGBFS was dried and milled to a Blaine fineness of 4050 ± 200 cm²/g, determined according to EN 196-6 [19]. The density was measured according to ASTM C204 [20] (Quantachrome Multipycnometer MVP-6DC) and found to be 2.9 g/cm³. The chemical composition of GGBFS was determined by X-ray fluorescence analysis (Philips, PW 1830) and consisted of (in wt%): 36.2 ± 0.2 SiO₂, 40.3 ± 0.5 CaO, 11.4 ± 0.2 Al₂O₃, 8.2 ± 0.1 MgO, 1.1 ± 0.1 S, 0.8 ± 0.1 TiO₂, 0.8 ± 0.1 Na₂O and 0.5 ± 0.1 K₂O. PG was used as received and its matrix composition, provided by the supplier, is (in wt%): 96 CaSO₄·2H₂O, 1–2 P₂O₅, 1.2 F, 1 SiO₂ and 0.2 Al₂O₃. Three different alkali-activators (see Table 1) were prepared by mixing together, sodium hydroxide pellets (grade 99%, supplied by Chem-Lab), sodium silicate (molar ratio SiO₂/Na₂O = 3.3 and 65% water, supplied by ABCR GmbH) and distilled water (ASTM type II). The alkali-activators were prepared 1 day prior to sample preparation. Paste samples were produced by using the precursors at a GGBFS/PG mass ratio of 9/1. The ratio of alkali-activator/precursor was kept constant at 0.6. Precursor and alkali-activator were mixed together for 3 min.

2.2. Radiological analysis

2.2.1. Gamma spectroscopy

The natural radionuclide content of GGBFS was determined by gamma spectroscopy. A sample of 356.3 g of GGBFS was put into a sealed cylindrical polystyrene container of 55 mm diameter and 105 mm height, weighed and enclosed for 30 days to reach the secular equilibrium with the ^{226}Ra progeny. Thereafter, the GGBFS sample was measured on top of a High-Purity Germanium (HPGe) detector (Mirion Technologies (Canberra) model BE5075-7500SI) of 48% relative efficiency (FWHM: 0.346 at 5.9 keV, 0.587 at 122 keV and 1.768 at 1332.5 keV). The shielding was composed of 0.2 cm copper and 10 cm lead. Energy calibration was performed with the same procedure as in [21] using the PG reference material 434 with known activity concentrations (95% confidence level) for radionuclides from the ^{238}U decay chain provided by the IAEA. The PG was transferred into the standard counting geometry enclosed for 30 days to reach the secular equilibrium with the ^{226}Ra progeny prior to measurement. The geometry dimensions, mass, shape, material composition and detector configuration and position were loaded into Canberra Laboratory Sourceless Calibration software (LabSOCS) to determine the counting efficiency for each gamma peak (ϵ) of the measurement setup, which is fitted by a fifth order polynomial function (Eq. (1)):

$$\ln(\epsilon) = -5.410^{-3}(\ln E)^5 + 1.210^{-1}(\ln E)^4 - 8.610^{-1}(\ln E)^3 + 9.310^{-1}(\ln E)^2 + 11.1(\ln E) - 27.6 \quad (1)$$

The sample measuring time was 20 h. The data and spectra were recorded by a Lynx MCA. The determination of the GGBFS massic activity was carried out by the software program Genie 2000 from Canberra. To determine the background of the detector, an empty sealed beaker was counted with the same procedure as the GGBFS sample. The background spectrum was used to correct the net peak area of measured gamma rays from GGBFS. The ^{226}Ra activity was estimated from the 609.3 keV (46.1%),

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