



Measurement of radon exhalation and emanation in synthesized coal fly ash-based geopolymer paste for building applications

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ABSTRACT

This study examines the radon exhalation potential of CFA and CFA-based GPP products, focusing on their elemental composition, structural properties, and radon containment capabilities. The particle size analysis result revealed that a substantial portion of CFA particles are fine, contributing to a greater surface area, which increased the potential for radon emanation. SEM analysis highlighted microstructural differences, with denser structures in the geopolymer products. XRF result revealed reductions in potassium (⁴⁰K), uranium (²³⁸U, ²³⁵U), and thorium (²³²Th) progenitors in CFA-based GPP products compared to CFA, and resulted in lower radioactivity due to the dilution effect of fillers and additives. XRD analysis showed the presence of crystalline phases such as quartz, mullite and amorphous phases in CFA and in the CFA-based GPP. The amorphous glassy phase in CFA, being rich in SiO₂ and Al₂O₃ precursors was critical for forming the aluminosilicate geopolymer network, that reduced porosity and trapped radon progenitors. Variations in CaO content and the formation of Ca containing mineral phases enhanced radon retention in CFA-base GPP products. Density measurements indicated that higher-density products exhibited lower permeability, reducing radon exhalation. Radioactivity measurements showed that CFA emitted 684 Bq/kg, above the world average of 420 Bq/kg and CFA-based GPP emitted between 408 and 459 Bq/kg. These findings underscored the need for controlling formulations and curing conditions to mitigate radon emanations of CFA-based products such as geopolymers.

Introduction

Waste management has become a critical issue for the world as economic growth is coupled with urban densification, which leads to a considerable rise in the quantity and variety of wastes [Elagroudy et al., 2016; Soni et al., Jun. 2022]. Due to cement production being responsible for 5–7 % of global CO₂ emissions, it is necessary to find alternatives to cement. Utilizing waste material as filler in concrete or as a geopolymer-based concrete substitute is becoming a key component of reducing carbon emissions and final production costs. Waste materials have been used as replacement fillers or cement substitutes, being available at lower cost [Allwood et al., Mar. 2010; Friedlingstein et al., 2010; Kalombe et al., Dec. 2020; McLellan et al., Jun. 2011]. Coal power generation is still the world's primary energy source, providing about 38

% of the total energy demand. As a result of coal burning, coal fly ash (CFA) is generated in quantities of nearly 1 billion tons annually globally as a by-product (fly ash—85 % and bottom ash—15 %) [Sultana et al., 2021]. For instance, in South Africa only, coal is responsible for about 72.1 to 77 % of the country's primary energy needs, and coal-based power generation uses about 224 million tons of marketable coal annually. This production results in approximately 40 million tons of CFA waste being produced annually, of which only about 7 % is reused while the remainder is predominantly disposed of in open dumps [Ige and Olanrewaju, Sep. 2023; Reynolds-Clausen and Singh, 2019]. CFA is rich in inorganic constituents such as oxides of silicon, aluminium, iron, and calcium, etc., and can replace cement as a binder, ranging from 10 to 80 % in concrete [Fucic, Jan. 2012]. Since 2010, the percentage of CFA used in cement and concrete products ranged from 19 % to 27 %, which

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has improved the mechanical and durability properties of concrete mixes and reduced greenhouse gas emissions [Gursel et al., Jan. 2016]. The use of CFA waste as a construction material has been encouraged as a way of managing waste. Due to its physical and chemical characteristics, CFA waste has become acceptable as a zero-cost raw feedstock for construction rather than requiring special management [Fucic, Jan. 2012; Nisnevich et al., Jul. 2008]. The safe utilization of coal-burning power plant wastes and byproducts depends on acceptable radioactivity levels and other environmental factors since they contain toxic metals and radionuclides. Generally, it is stated that CFA radioactivity levels are indistinguishable from other natural materials [Baykal and Saygili, Apr. 2011]. Developing sustainable buildings is a significant step forward from both an economic and an ecological perspective. However, the microenvironment created in such living environments could present a complex radiochemical environment that can pose a significant risk to its occupants' health [Fucic, Jan. 2012]. It is imperative to understand the carcinogenic properties of radon-emitting building materials. Houses are primarily affected by radon emitted from building materials [Abu-Jarad et al., 1980]. The World Health Organization (WHO) reports that radioactive radon gas in buildings is the second leading cause of lung cancer after smoking, with an excess relative risk of 16 % per 100 Bq m⁻³ [Fucic et al., Oct. 2010; Jiránek and Kačmaříková, Nov. 2019; WHO 2009].

Radon is a natural atomic element, a radioactive noble gas in the ²³⁸U decay series that can easily move out of its host material since it is a gas [NRC 2006]. It is an invisible, odourless, tasteless, colourless, inert, and radioactive gas. Radon generally occurs from the natural breakdown of uranium in the Earth's crust. It usually travels through soil and thus enters homes, schools, and other buildings. Levels of radionuclides such as uranium (²³⁵U, ²³⁸U), radium (²²⁶Ra), thorium (²³²Th), lead (²¹⁰Pb), polonium (²¹⁰Po), and potassium (⁴⁰K) in CFA as a by-product of thermal plants may vary by a factor of 20 to 25 from the original peat [10]. A typical illustration of the radioactive decay chain of ²³⁸U is presented in supplementary figure SF1. Tracing the occurrence of Radon (Ra) is of considerable importance since it is the primary health risk associated with chronic indoor radon exposure and increases the lifetime risk of lung cancer [NRC 2006]. Understanding radium's decay pathways is crucial to managing potential health and environmental hazards associated with exposure to radioactive substances. Radium occurs naturally in the crust of the Earth. Gamma rays are penetrating ionizing electromagnetic radiation that is emitted by most materials from naturally occurring long-lived radionuclides (⁴⁰K and members of the ²³⁸U decay series). Radon emits alpha particles as part of one of the decay pathways, resulting in lead (Pb) and polonium (Po). It decays relatively quickly compared to other radioactive elements, where half-lives are longer. Ra-226, for instance, has a half-life of around 1600 years. As it decays, alpha particles are emitted, transforming into radon-222 (Rn-222) noble gas. Furthermore, radon-222 itself undergoes radioactive decay, producing polonium-218 (Po-218) and lead-214 (Pb-214) [Rajkhowa et al., Jan. 2021; Seiler and Wiemels, 2012; Strumińska-Parulska and Falandysz, Nov. 01, 2020]. The level of radon in outdoor air is usually so low that it poses very little risk. The mechanism that increases the potential for developing lung cancer is the emission of an alpha particle from radon decay products Po-218, Po-214, and Pb-214, causing physical and chemical damage to cells and DNA [Grzywa-Celińska et al., Dec. 2020; Lorenzo-González et al., Sep. 2019]. The mechanisms of DNA damage leading to lung cancer are shown in supplementary figure SF2.

When radon enters or emanates from a building, it and its decay products can accumulate at high concentrations. Radon exhalation from materials depends on many factors, such as grain size and pore space. As radon decays, it produces radioactive products. As one breathes, radon decay products, which are solids, can be trapped in the lungs. Further decay of these decay products releases alpha particles that damage lung tissue and lead to lung cancer [Ereos and Yener, 1999]. Even though concrete emits radionuclides as it ages, studies on the decay products of these radionuclides have not been extensively conducted. This is

especially of concern in the case where about 70 % of cement is replaced by CFA [Chambers et al., Nov. 2011; Fucic, Jan. 2012; Ramachandran and Sahoo, 2009]. A study by Kovler [Kovler et al., Jan. 2005] found that despite the higher ²²⁶Ra content in CFA, >3 times higher than of Portland cement, the radon emanation in CFA is only 0.52 %, which is significantly lower than the 7.62 % obtained in cement. By replacing only 25 % of the volume of cement with FA, the ²²⁶Ra concentration was increased by 1.5. Yet, despite the higher ²²⁶Ra content in the concrete, they reported that the rate of radon exhalation was only slightly increased or even decreased. Radon exhalation measurements from a sample of concrete must be standardized based on humidity and age. According to the investigations, adding CFA to concrete can reduce radon exhalation under certain conditions [Kovler et al., Jan. 2005; Roelofs and Scholten, 1994]. Specific radioactivity activity associated with ²³²Th for construction materials was listed as 44–860, 42–918, 50–200, and 10–30 Bq/kg for cement, concrete, red brick, and lime brick respectively with CFA at 100–300 Bq/kg in the specific study [EC 1999; Harrison, 2004; Ramachandran and Sahoo, 2009; IAEA 1994]. It seems that there are two reasons for such a “paradoxical” behavior. CFA particles may have a low emanation coefficient. Additionally, the pozzolanic reaction between CFA with water and calcium hydroxide that forms because of Portland cement hydration densifies the concrete structure over time [Kovler et al., Jan. 2005]. This study covers the investigation of the potential indoor radon concentrations and gamma radiation from CFA-based geopolymer paste (CFA-based GPP) formulations cured at room temperature and intended to be used for building applications in which cement is replaced wholly by CFA.

Materials and methods

Materials

Low calcium class F DURAPOZZ coal fly ash (CFA) obtained from Lafarge Industries South Africa (Pty) Limited, Private Bag X26, Gallo Manor 2052, Republic of South Africa was used as a solid precursor of CFA. Sodium hydroxide pellet (98 %) and Shutter oil were acquired from Kimix Airport Industrial, Cape Town, South Africa. Sodium silicate with the following composition 30 % SiO₃, 9.2 % Na₂O, and 60.8 % H₂O, was purchased from Protea chemicals, Cape Town, South Africa. NaOH and sodium silicate were used as alkaline activators for the synthesis of CFA-based GPP.

Formulation and CFA-based geopolymer product specifications

Different CFA-Based GPP formulations were purely composed of CFA and alkaline activators. Fine and coarse aggregates (sand and stone) were excluded from all formulations to better understand radon measurement associated with using CFA alone as a construction material. The products were cast and cured at room temperature for 28 days whilst covered with a plastic film. The sample nomenclature and material mass per cent of each formulation cast are shown in Table 1.

Sample characterization

Different characterization techniques, including particle size analysis

Table 1
Mass composition (%) of CFA-base GPP formulations used for radioactivity measurement.

Formulation code	Composition, w/w, %				
	CFA	Dye oxide pigment	Alkaline activator		
			Na ₂ SiO ₃	NaOH, 12M	Additional H ₂ O
GPP SM 1A	74.05	0	18.55	7.4	0
GPP MR1	68.97	0	17.24	6.9	6.9
GPP MR2	67.25	1.72	17.24	6.9	6.9

and XRF, were used to identify the properties of the solid precursor, CFA. This was to understand its impact on formulation design as well as note any changes when exposed to environmental conditions. The particle size analysis of CFA was measured using the Malvern Panalytical Mastersizer 2000 analyzer with a small volume dispersive unit (Hydro 2000SM) and a stirring speed of 1500 rpm. Three scans per sample were performed for accuracy. Propan-2-ol, also known as isopropyl alcohol (IPA), was used as a dispersant (particle refractive index = 1.73 and assay of 99.5 %) in a well-coated slurry at a ratio of 1:3 (CFA: IPA, using a spatula). This method provides detailed volume and cumulative particle size distribution, essential for evaluating the surface area and its influence on radon emanation.

To complement particle size analysis scanning electron microscopy (SEM) was used to observe the microstructure of coal fly ash (CFA) and synthesized CFA-based geopolymer products (GPP SM1A = geopolymer paste sample mix 1 phase A, GPP MR1 = geopolymer paste mix reference 1, and GPP MR2 = geopolymer paste mix reference 2). The Back-scattered Electron (BSE) imaging technique was selected. To commence imaging, the high voltage (HV) function was activated, and the desired mode for analysis was selected based on the imaging testing area requirements. A beam intensity of 10 kV was considered for SEM imaging to capture high-resolution surface details. The working distance (WD) was set at about 20 mm to optimize focus and clarity during imaging.

X-ray fluorescence (XRF) techniques were used to determine the presence of major oxides and minor elements based on the weight per cent (wt.%) from fused disks made with ignited powder and concentration (ppm) methods, on a Panalytical Axios wavelength dispersive X-ray spectrometer for CFA and CFA-based geopolymer optimized products. This measurement was specifically focused on potassium uranium (U), thorium (Th), and potassium (K) content to evaluate changes in radioactivity after geopolymerization. X-ray diffraction (XRD) was employed to analyze the crystalline phases in CFA as well as in CFA-based geopolymer products.

CFA and product radon exhalation and emanation characterization techniques

Radon exhalation and emanation measurements were performed using RAD7 to investigate the potential radioactivity issue related to the use of South African Lethabo low coal fly ash as a binder in building materials in the form of CFA-based GPP. A 100 mm cubic hardened block of each of the Lethabo low calcium CFA-based GPP was used for measurements. Two issues were considered.

- The gamma radiation from the blocks. Gamma rays are penetrating ionizing electromagnetic radiation that are emitted by most materials from naturally occurring long-lived radioactive nuclides (^{40}K and members of the ^{238}U and ^{232}Th decay series).
- The radon emanation from the block. Radon exhalation from materials depends on many factors, such as grain size and pore space.

Gamma radiation measurement. For the gamma radiation, a CFA-based GPP 100 mm cubic block was placed in a lead castle gamma chamber (to minimize background gamma radiation) with a Sodium Iodide NaI (TI) detector. This detector does not have as good a resolution as an HPGe (High Purity Germanium) detector, but it was perfectly suited to measure the activity of the radioactive materials in the blocks [Pilakouta et al., Sep. 2018]. The radium content (and therefore uranium if the decay series is in secular equilibrium) and thorium concentrations were obtained using this detector in a very similar way to the methods described in a previous study [Pilakouta et al., Sep. 2018]. The efficiency of the detector needed for the block's geometry was tested. To do this, a control sample of known activity was used. A sample of soil from a mine dump was used as a control for the measurements described in this report. This mine dump sample had a high activity of ^{226}Ra (290 Bq/kg)

and 40 K (230 Bq/kg), so it could be used to calculate the ^{226}Ra and ^{40}K concentrations in the CFA-based GPP blocks.

It is acknowledged that there were some limitations in the design of radon and gamma concentration measurements. Firstly, CFA-based geopolymer products were not exposed to normal environmental conditions such as temperature and sunlight, which could also affect radioactivity. Secondly, these results may not be like those of an existing building envelope, as concentrations may differ. Thorium concentrations were measured as shown in Fig. 1.

Radon measurement. For radon measurement, a 100 mm CFA-based GPP product in the form of a block was placed in an air-tight vacuum chamber that was connected to a radon detector namely a RAD7 radon detector (Available at <https://durridge.com/products/rad7-radon-detector/>, Accessed on 08th August 2022) and a computer system as shown in Fig. 2. Following the setting of the parameters in RAD7 software on the computer, measurements were conducted. The radon exhaled from the CFA-based GPP block causes the radon concentration to increase in the chamber.

According to Amin (2015), the radon exhaled from the block causes the radon concentration $C(t)$ to increase in the chamber as shown in Eq. (1).

$$C(t) = \frac{C_{Ra}}{V} (1 - e^{-\lambda t}) \quad (1)$$

Where: V is the volume of the chamber and the detector = 14 liters, $C(t)$ is the radon concentration in the chamber, in Becquerels per cubic meter (Bq.m^{-3}), C_{Ra} is the effective strength of the radium source in the CFA and CFA-based GPP blocks that cause radon release into the chamber in Bq, t is the time in seconds (s) and λ is the decay constant of Radon 2.1×10^{-6} , in s^{-1}

It was assumed that for a particular sample, the radon increased in the chamber (as read by the RAD7) at a rate of $x \text{ Bq.m}^{-3}$ per hour for the first few hours. In this period, Eq. (2) can be approximated to Eq. (3) since $e^{-\lambda t} \sim 1 - \lambda t$, and λt is very small. The slope, m , is then defined as shown in Eq. (3) and the effective strength C_{Ra} can be obtained.

$$C(t) = \frac{C_{Ra}\lambda t}{V} \quad (2)$$

$$m = \frac{C_{Ra}\lambda}{V} \text{ or } C_{Ra} = \frac{Vm}{\lambda} \quad (3)$$

CFA-based geopolymer product compression strength

SANS method 5863:2006 (concrete tests - compressive strength of hardened concrete) was used as the standard to conduct this test using a fully automatic 1800 kN concrete cube press complete with a digital controller having a load capacity of 1800 kN and load rate of 18 kN/min.

Three 100 mm cube specimens at a specific curing age were tested of the cured CFA-based geopolymer paste products, for their average mortar and concrete compression strength. The machine compression loading rate was set up at 180 kN/mm. The crushing load endpoint was recorded, and the compression strength of the specimen was calculated according to the formula in Eq. (4).

$$f_c = \frac{P}{A} \quad (4)$$

Where: f_c is the characteristic strength of a specimen in mega-pascal (MPa)

- P is the crushing load obtained from the machine in Newton (N)
- A is the load applied surface area of the specimen in millimetre square (mm^2).

The average density of GPP was recorded, where individual density was calculated based on the density standard equation of mass and volume. Based on density measurements, the assumption was that

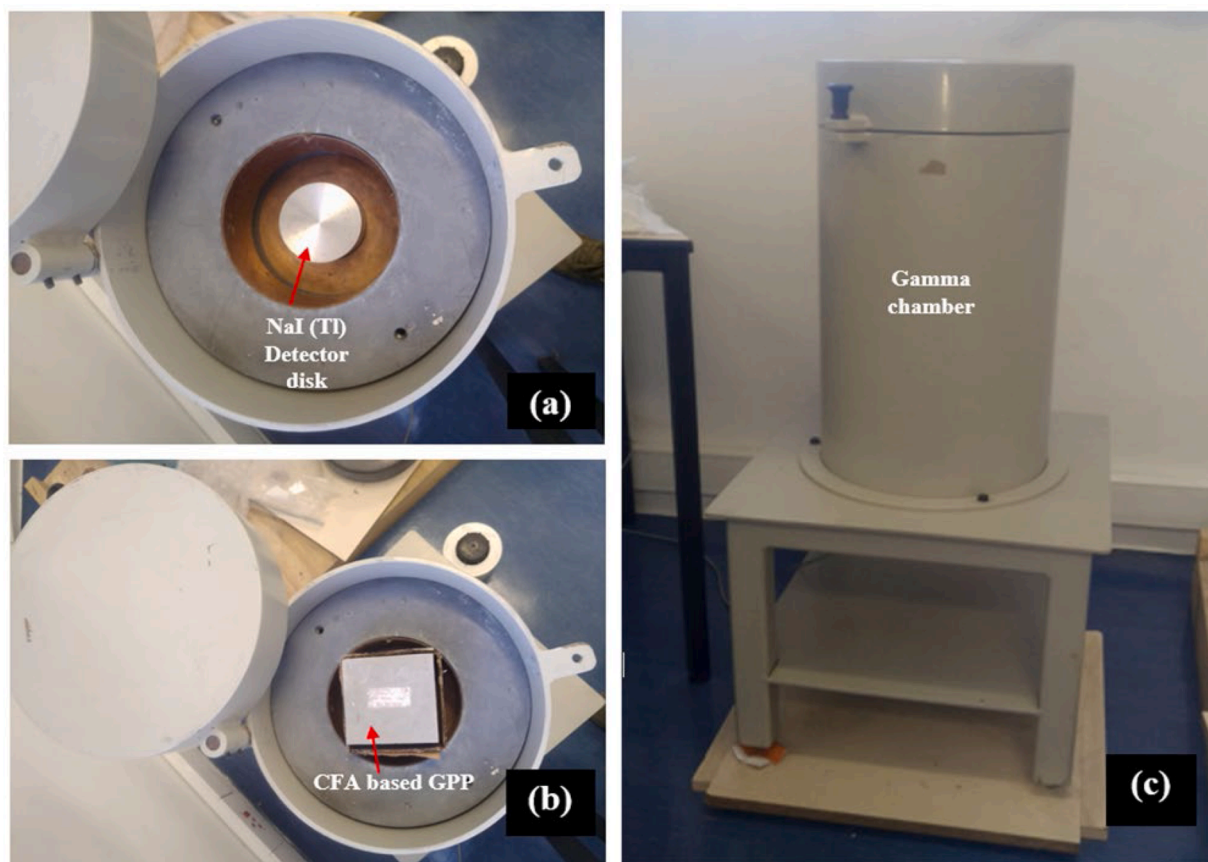


Fig. 1. Process involved in thorium concentrations measurement; (a) Chamber showing the detector, (b) CFA-based GPP specimen in a chamber and (c) chamber closed for testing.

higher-density products were expected to exhibit lower permeability, thus limiting radon exhalation. A simplified experimental protocol covered in this work is summarised and exhibited in Fig. 3.

Results and discussion

Particle size analysis

CFA particle size, cumulative and volume distributions of CFA were analyzed and are shown in Fig. 4. These results are in the range of 0.3–0.25 μm , similar to previous investigations [Kumar Vadapalli et al., 2007; Li et al., 2014; Van Der Merwe et al., Oct. 2014]. The PSD plot reveals a volume distribution with a peak at around 10–100 μm , indicating that a significant portion of the particles fall within this size range. The cumulative distribution (red line) reaches close to 100 % at around 1000 μm (with about 24 % CFA particle size >10 μm and about 86 % <100 μm , suggesting that some particles are large, but most are below this threshold.

As part of this study, particle size might be a critical factor affecting radon emanation and exhalation as smaller particles, particularly in the micrometer (μm) range, tend to have a higher surface area, which can enhance radon release because radon gas, formed by the decay of ^{238}U and ^{232}Th within the material, has more accessible pathways to escape from the surface. This is particularly important in CFA, where radon progenitors, ^{238}U and ^{232}Th , are present. The presence of fine particles in this CFA may increase the potential for radon exhalation when their surfaces remain openly disposed of as these particles provide more points of access for radon to diffuse into the surrounding environment. Larger particles in the distribution, on the other hand, contribute less to radon exhalation due to their lower surface area-to-volume ratio.

Scanning electron microscopy and synthesized CFA-based geopolymer products

Fig. 5 shows the SEM analysis of class F DURAPOZZ CFA and synthesized CFA-based GPP. The SEM images reveal distinct microstructural differences between raw CFA and GPP SM1A, GPP MR1, and GPP MR2. CFA exhibits smooth, well-defined, spherical particles with significant interparticular void spaces, which may lead to higher radon emanation due to insufficient encapsulation of radon progenitors. In contrast, GPP SM1A, GPP MR1, and GPP MR2 products display rougher, more irregular morphologies with variable agglomeration, indicating structural transformation through geopolymerization. This densification may aid in encapsulating ^{238}U and ^{232}Th and their decay products, such as ^{226}Ra , potentially reducing radon release... These results may reveal the impact of geopolymer microstructure on radon mitigation.

X-ray fluorescence major and minor elements of CFA and CFA-based geopolymer products

XRF analysis of class F DURAPOZZ CFA and synthesized CFA-based GPP were analyzed for major, minor, and trace elements using the XRF method described in Section 2.2.1. The analytical results for major and minor elements are shown in Tables 2 and Table 3, respectively and are reported in % dry weight. As displayed in Table 2, XRF results reveal the major elemental composition of CFA and synthesized geopolymers, highlighting major components like SiO_2 , Al_2O_3 , and CaO , along with trace elements such as Zn, Sr, and Th. The SiO_2 and Al_2O_3 contents are essential for forming the aluminosilicate network, which could play a significant role in radon containment by reducing the porosity and providing a stable structure for trapping radon progenitors. Geopolymers GPP SM1A, MR1, and MR2 show increased SiO_2 levels due to

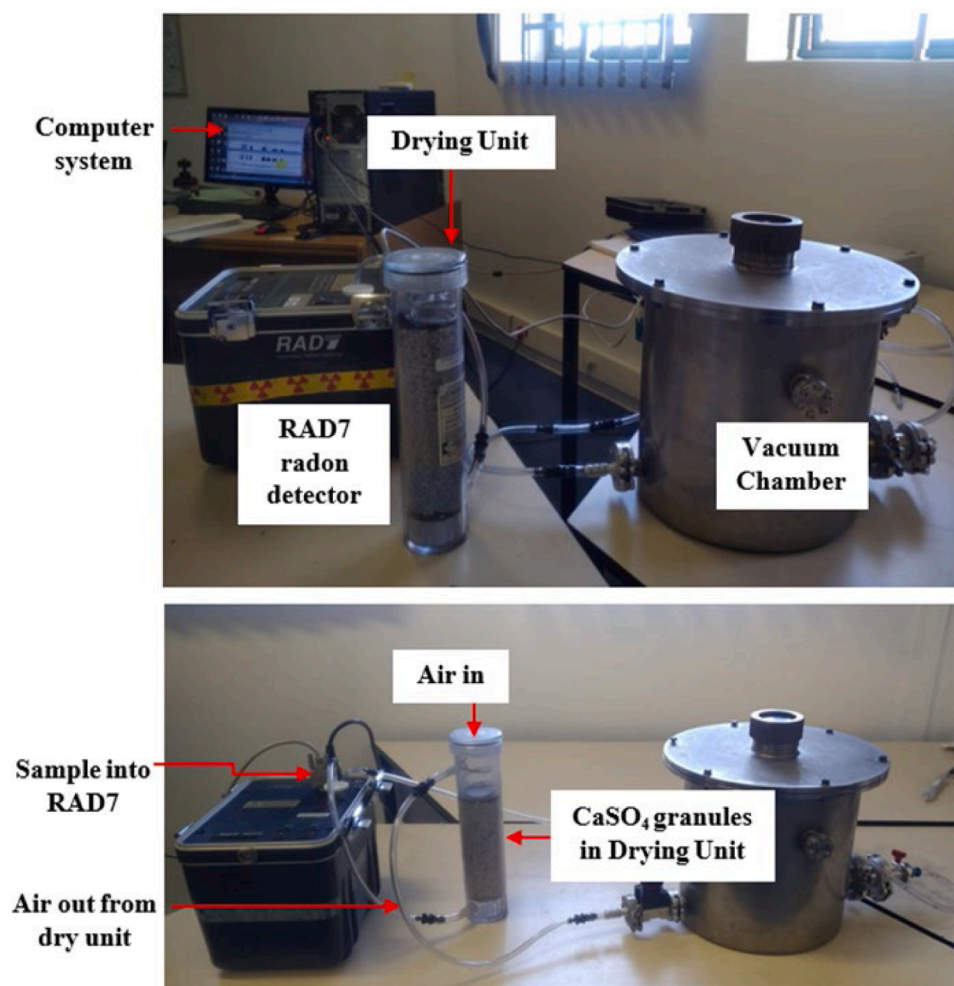


Fig. 2. Radon monitor set-up and CFA-based GPP block measurements in progress.

the additional sodium silicate added in the formulations compared to CFA, thus the lower Al_2O_3 can be attributed to the dilution effect of the added sodium silicate. The sodium silicate could be a source of SiO_2 species during geopolymerization. The elevated CaO content in the synthesized geopolymers products compared to the initial content in CFA suggests the formation of calcium-rich phases. Calcium phases can either strengthen the matrix or introduce additional microstructures that may affect permeability. This higher CaO content in GPP products might influence radon emanation by altering the geopolymer's internal structure, enhancing its radon containment properties under certain conditions. Based on minor elemental results, trace elements like U and Th present in CFA were found to be present in lower quantities in synthesized CFA-based geopolymer products. U was 9 ppm in CFA and lower than 5 ppm in GPP SM1A, GPP MR1, and GPP. Additionally, Th was 44 ppm in CFA and 26–27 ppm in GPP SM1A, GPP MR1 and GPP. The reduction in U and Th concentration could be ascribed to the additional components added to the geopolymer having a dilution effect and indicates that in the future, adding fillers to the geopolymer formulations could be beneficial.

X-ray diffraction of CFA and CFA-based geopolymer products

The XRD patterns of class F DURAPOZZ CFA and synthesized CFA-based GPP are shown in Fig. 6. For CFA, crystalline phases like quartz and mullite were identified by dominant peaks at 2θ positions 16.4° , 25.9° (not present in GPP products), 26.2° , 33.2° , and 40.8° (quartz) and 20.8° , 26.6° , 36.6° , 39.4° 50.1° (except in GPP MR1) (mullite),

respectively as well as hematite at 2θ 20.8° , 26.6° (strongest and most intense peak), 36.6° , 39.4° 50.1° (except in GPP MR1) which represent typical mineral components of fly ash. However, a distinct peak at approximately 11.6° (suggested to be mullite) observed in the CFA sample was found to be eliminated in all GPP products, indicating the transformation or dissolution of this phase during geopolymerization. Similarly, peaks present in raw CFA starting at about 50° to 70° were found to be greatly reduced or absent in synthesized GPP SM1A, GPP MR1, and GPP MR2 products further reflecting a reduction in the short-range order and crystallinity of the refractive quartz and mullite mineral phases, during geopolymerization. CFA had an enhanced amorphous glassy phase, indicated by the increased hump between 20 and 30° 2θ , which was less pronounced in the geopolymer matrices, indicating the availability of the glassy phase in CFA to provide the precursor species necessary for the geopolymerization process to proceed. This indicated that the glassy aluminosilicate matrix in CFA may contribute to the formation of stable amorphous phases in geopolymers, potentially allowing better encapsulation of the ^{238}U and ^{232}Th and their decay products, such as ^{226}Ra which directly produces radon. Low mobility of radon gas in a stable geopolymer matrix could reduce radon release. The XRD patterns of raw CFA and investigated geopolymer products GPP SM1A, GPP MR1, and GPP MR2 provide some insights into their structural changes and potential for radon containment. These mineralogical changes contribute to the creation of an amorphous aluminosilicate framework, which could enhance radon containment by reducing surface area. In addition, apart from the retained peaks for quartz and mullite, the GPP SM1A and GPP MR2 display new phases such as

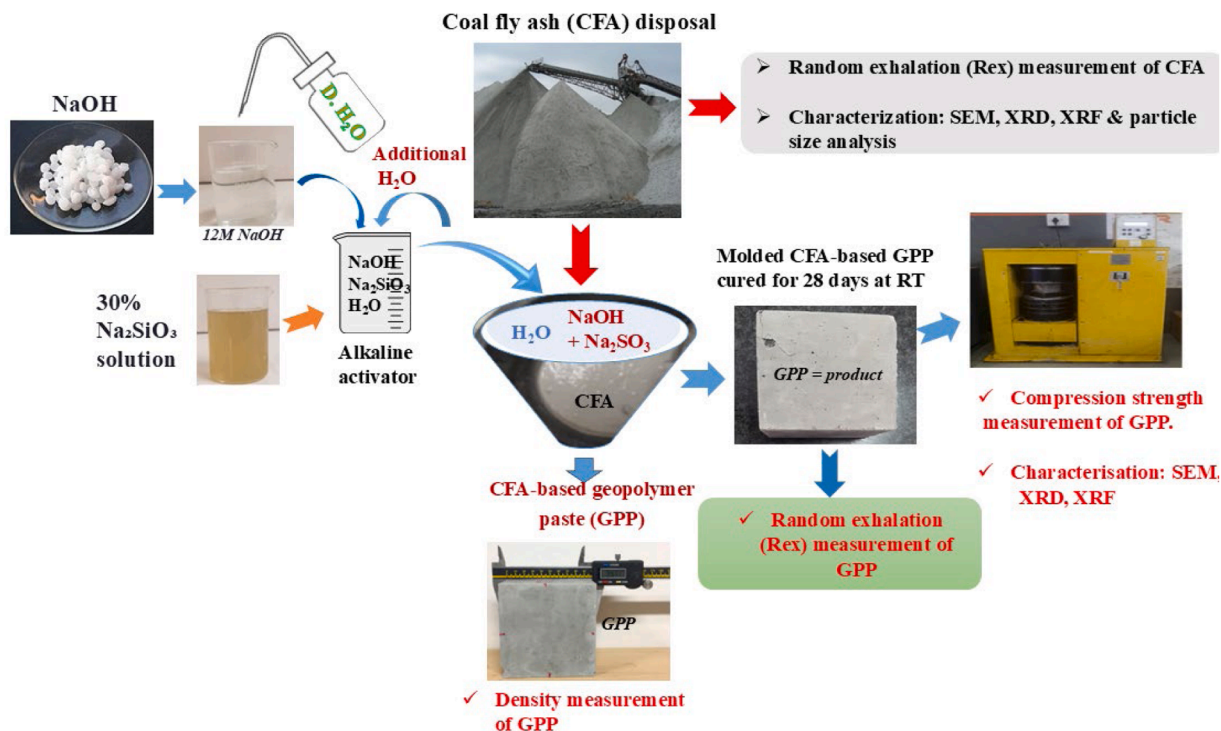


Fig. 3. Experimental schematic steps for synthesizing CFA-based geopolymer (GPP).

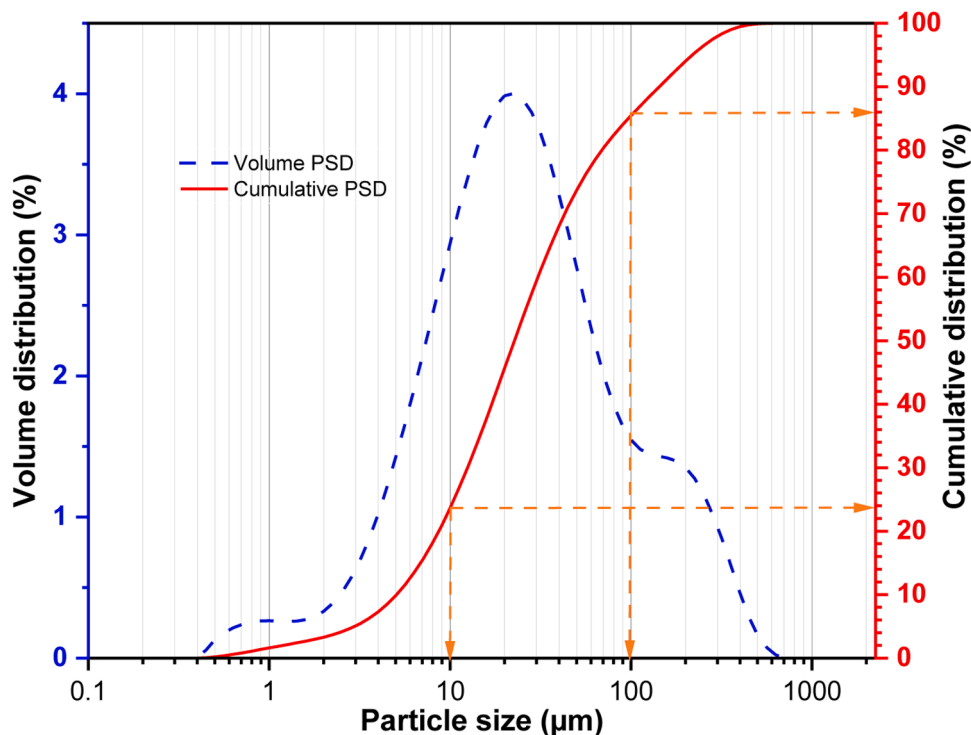


Fig. 4. Particle size cumulative and volume distribution CFA.

calcium sulphate (CaSO₄) or calcium carbonate (CaCO₃), visible at 20 positions near 29.4°. These calcium-rich phases may improve the microstructural stability of the geopolymer matrix, reducing pathways for radon migration and enhancing their effectiveness in radon containment compared to raw CFA.

Radon exhalation and emanation

Radioactivity content and gamma radiation

Table 4 shows the radioactivity of radium (Ra), thorium (Th), and potassium-40 (⁴⁰K) in CFA and synthesized CFA-based GPP products, as determined by gamma radiation activity, with results indicating that raw CFA generally exhibits higher gamma radiation activity than the

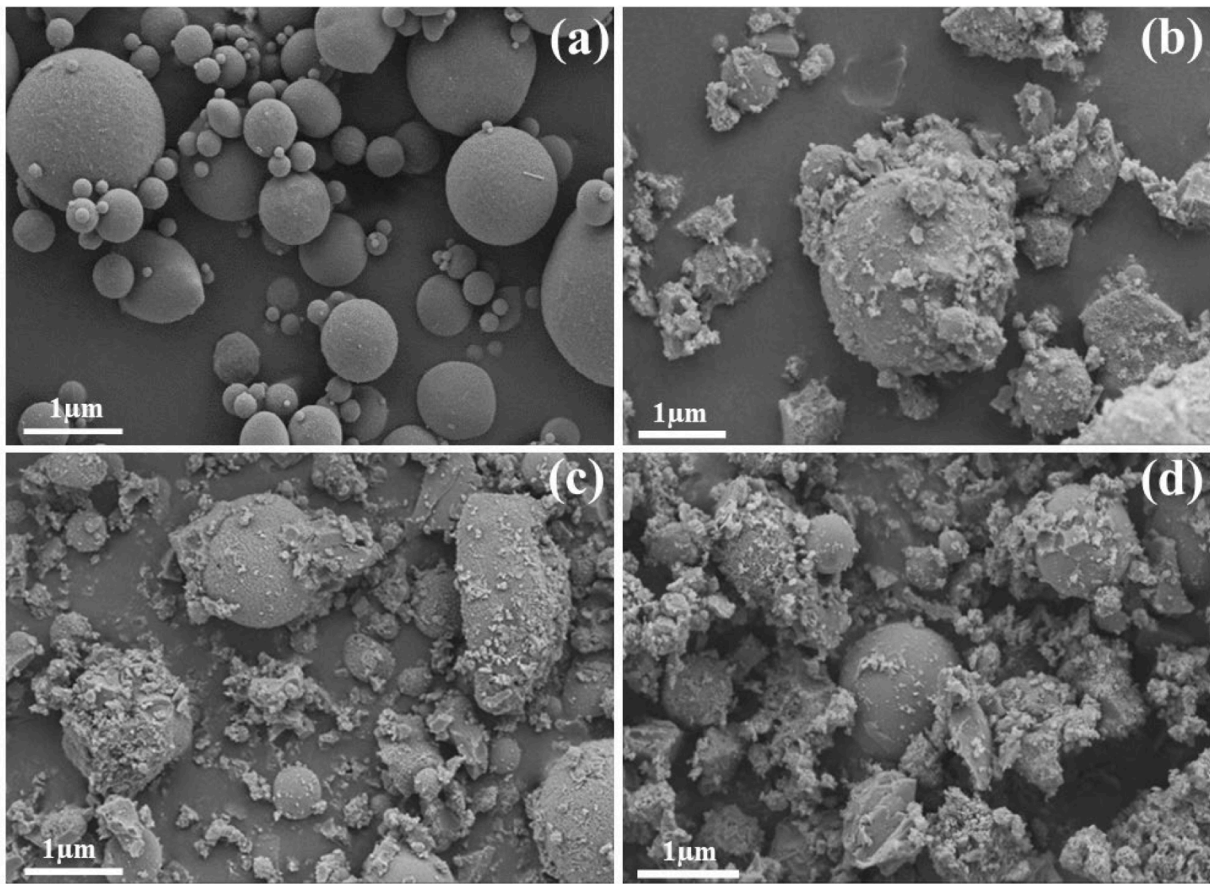


Fig. 5. SEM Micrographs of Raw CFA (a) and Synthesized CFA-Based Geopolymer Products GPP SM1A (b), GPP MR1 (c), and GPP MR2 (d) showing microstructural differences.

Table 2
Elemental analysis of CFA and synthesized CFA-based geopolymer (wt. %) for major and minor elements by XRF.

Major element	CFA	GPP SM1A	GPP MR1	GPP MR2
SiO ₂	55.27	62.15	62.64	62.72
Al ₂ O ₃	33.03	19.58	19.09	18.87
CaO	4.17	11.14	10.54	10.99
Fe ₂ O ₃	3.14	1.89	1.88	1.83
TiO ₂	1.70	0.97	0.94	0.93
MgO	1.06	0.75	0.74	0.69
K ₂ O	0.71	0.51	0.52	0.48
P ₂ O ₅	0.46	0.28	0.29	0.29
SO ₃	0.23	0.16	0.25	0.28
Na ₂ O	0.15	2.53	3.06	2.88
Cr ₂ O ₃	0.04	0.02	0.02	0.02
MnO	0.03	0.02	0.02	0.02

GPP SM1A, GPP MR1, and GPP MR2 products. Specifically, Ra and Th values for CFA were 263 Bq/kg and 288 Bq/kg, respectively, with a high average total activity of 684 Bq/kg. Additionally, the ⁴⁰K was low at <100 Bq.kg⁻¹ in geopolymer products compared to 133 Bq.kg⁻¹ in raw CFA. Synthesized GPP products exhibited lower gamma radiation activity levels, with Th activity ranging from 181–209 Bq/kg, radium from 159–194 Bq/kg, and average total activity levels falling to between 408 and 459 Bq/kg. These reductions in radioactivity for GPP products may be attributed to the dilution effect from additional materials such as sodium silicate in the geopolymer formation, lowering the overall concentration of radionuclides.

A study conducted by Estokova et al. [Estokova et al., Oct. 2022] on radioactivity in a historical building in Slovakia reported a world

Table 3
Determination of CFA and CFA-based synthesized geopolymer (ppm) trace elements by XRF.

Minor element	CFA	GPP SM1A	GPP MR1	GPP MR2
Zn	76	60	63	124
Cu	53	30	31	31
Ni	67	43	45	42
Mo	<5	<5	<5	<5
Nb	48	28	27	26
Zr	389	206	220	225
Y	64	32	32	32
Sr	905	816	740	745
Rb	40	27	27	26
U	9	5	<5	<5
Th	44	27	27	26
Pb	81	62	64	62
Co	10	<5	<5	5
Mn	261	204	227	207
Cr	290	227	239	237
V	194	130	153	155
F	543	4378	2608	3059
S	1481	1173	1415	1625
Cl	36	69	59	93
Sc	25	21	19	20
Ba	934	547	568	562

average value of 594.0 Bq.kg⁻¹, which was deemed acceptable for residential use, as it was not anticipated to pose any significant health risks.

Until now, the impact of gamma radiation from building materials on the inhabitants of residential spaces remains uncertain, as it depends on factors such as whether the material is used solely for walls, walls and floors, or walls, floors, and ceilings. Additionally, the duration of oc-

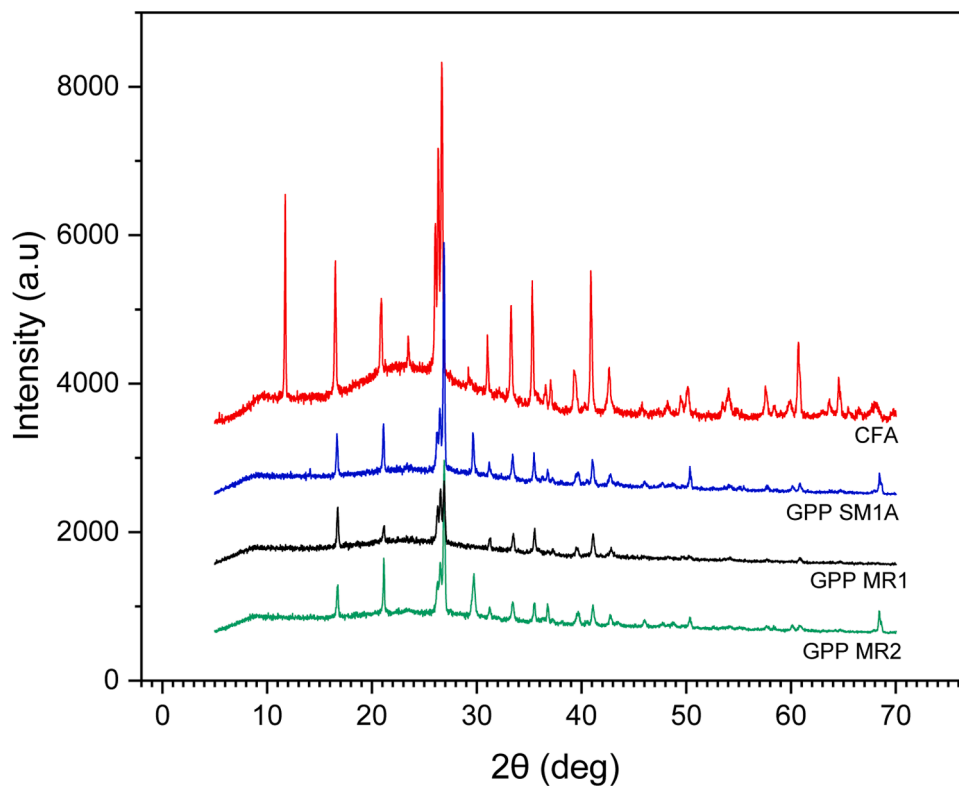


Fig. 6. XRD Analysis of phase composition in CFA and CFA-Based Geopolymers.

Table 4

The gamma radiation activity of uranium and thorium decay series and the ^{40}K in different synthesized geopolymer products.

Products ID	Radioactivity concentration (Bq/kg)			
	Ra	Th	K	Average total activity
CFA	263	288	133	684
GPP SM 1A	159	200	79	438
GPP MR1	139	187	82	408
GPP MR2	209	181	69	459

cupancy within the dwelling is also an unknown variable that complicates the assessment. The European Commission or EC (1999) gives an indicative rule of an activity concentration index (I), providing an idea of the concentrations in building materials that would lead to a dose of 1 mSv per annum (millisievert per annum). This is the accepted maximum annual dose that members of the public can be exposed to. Eq. (5) provides a formula for calculating the Activity Concentration Index (I), which is used to evaluate whether the radioactivity levels in building materials meet the EC standards for safety. This index considers the weighted contributions of ^{226}Ra , ^{232}Th , and ^{40}K gamma radiation activity and establishes a maximum annual dose of 1 mSv per person as an acceptable threshold. For CFA-based materials, this calculation is crucial to determining if the geopolymer products can be used safely in residential construction applications.

$$I = \frac{C_{\text{Radium}}}{300 \text{ Bq/kg}} + \frac{C_{\text{Th}}}{200 \text{ Bq/kg}} + \frac{C_{\text{K}}}{3000 \text{ Bq/kg}} \quad (5)$$

It is important to note that the weighting factors in the denominators relate to the number of gamma rays and their energies in the decay series. ^{232}Th has the most gamma rays, ^{238}U has fewer and there is only one in the case of ^{40}K . An activity Concentration Index of 1 corresponds to an annual dose of 1 mSv. However, this does depend strongly on the structure of the building as explained by the EC standard of 1999. As

stated in the EC of 1999, clay bricks usually do not contribute zero doses. On the contrary, the radioactivity values are about half of that of coal fly ash for ^{232}Th .

Radon exhalation

Radon exhalation in synthesized CFA-based geopolymer blocks was calculated using Eq. (3). Fig. 7 illustrates radon exhalations over time for three geopolymer products labelled GPP MR2, GPP MR1, and GPP SM1A measured by the RAD7 detector. Each sample's radon exhalation increases linearly over 24 h, indicating a consistent exhalation rate. Among the products, GPP MR1 showed the highest radon release rate with a slope of 21.0278 Bq/m³.hr, followed closely by GPP MR2 (20.2818 Bq/m³.hr), while GPP SM1A exhibits the lowest rate at 9.9813 Bq/m³.hr. These linear trends are further supported by high R² values for each sample, suggesting strong fits and consistent behavior throughout the experiment.

By observing formulations listed in Table 1, CFA content may not be the only factor influencing radon exhalations, nor the incorporation of sodium silicate or dye oxide pigments in specimens. However, the difference may be primarily attributed to variations in each sample's density because of other factors such as curing conditions or the level of filler content in the case of CFA-based geopolymer augmented with activators such as sodium silicate, etc. GPP SM1A's emanations with a lower slope may indicate a more compact matrix that limits radon diffusion. The higher exhalation rates for GPP MR1 and GPP MR2 than GPP SM1A could result from increased porosity in those materials, allowing radon to escape more freely. The main difference between these three geopolymer products was that GPP SM1A had no additional water added to the formulation during blending, thus geopolymerization took place in a more concentrated environment, promoting the formation of a denser matrix (Section 3.6).

Using Eq. (2) the effective strength C_{Ra} of the radium source in the block that exhausts into the chamber in Bq was found to be 18 Bq. This means that the CFA-based GPP blocks act as a radium source that causes 18 radon atoms to enter the chamber every second. This can be used to

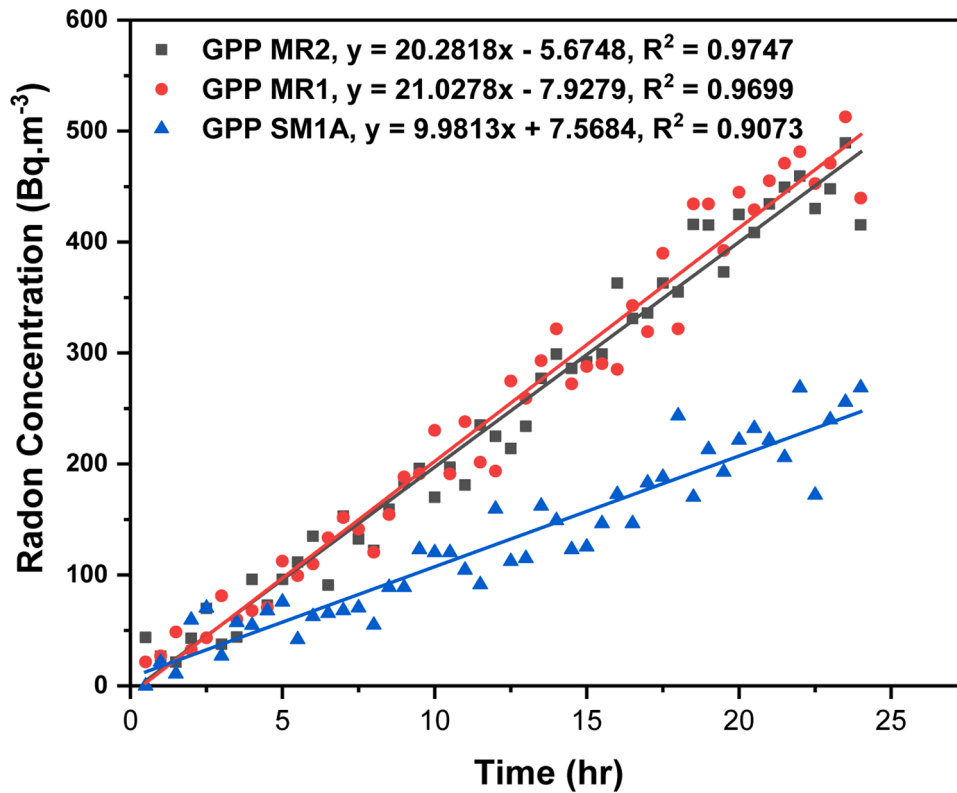


Fig. 7. Trend of radon build up in 14 L chamber as measured with RAD7 detector for synthesized CFA-based GPP products.

calculate the radon exhalation rate from the block.

The exhalation of radon, or radon flux, is usually given in terms of the radon activity concentration that is released from 1 m² of the material per second or per hour. Taking this per hour as in Amin [Amin, Oct. 2015] the radon flux can be considered as E_A and determined according to Eq. (6).

$$E_A = \frac{\lambda C_{Ra}}{Area} \times 3600 \quad (6)$$

Where $C_{Ra}\lambda$ is the radon activity concentration released into the air, $Area$ is the area of the brick and 3600 converts from flux per second to flux per hour. For CFA-based GPP SM1A, this instead leads to 2.4 Bq.m⁻².hr⁻¹. These results can also be written as 2 400 mBq. m⁻².h⁻¹ if this is the subject of comparison with results obtained from Amin's [Amin, Oct. 2015] study. This value is higher than any of the values reported in that paper. The radon flux of the different CFA-based GPPs products GPP SM1A, GPP MR1, and GPP MR2 was based on the slope of radon buildup, calculated radon concentration (C_{Ra}) and flux values are presented in Table 5. These results emphasize differences in radon exhalation potential across the products, with GPP MR1 showing the highest radon flux of 4.92 Bq.m⁻².h⁻¹, followed by 4.67 Bq.m⁻².h⁻¹ from GPP MR2 with GPP SM1A exhibiting the lowest flux of 2.38 Bq.m⁻².h⁻¹. These trends are consistent with the respective slopes of radon buildup, where GPP MR1 has the steepest slope of 21.10 Bq/m³.h and GPP SM1A the lowest, at

10.20 Bq.m⁻².h⁻¹.

It has been reported that granite countertops may cause radiation damage, but realistic calculations suggest this is unlikely to be a serious concern [Llope, Jun. 2011]. In general, the measured radon escape values are typical for building materials and do not seem to imply any specific radiation protection issue.

CFA base GPP formulations compression strength and density

CFA-based GPP formulations used for radon and thorium measurement were also tested for compression strength and monitored. The compression strength and density at 28 days of maturity age, cured under ambient conditions, are shown in Fig. 8. Fig. 8 also illustrates the relationship between the compression strength and density of different CFA-based GPP formulations, specifically GPP SM1A, GPP MR1, and GPP MR2. The results obtained show that the compression strength of GPP SM1A and GPP MR1 was higher than the 25 to 35 MPa required for normal and structural applications (Mindess et al., 2003). GPP SM1A shows the highest compression strength at 45.0 MPa, followed by GPP MR1 at 38.7 MPa, and GPP MR2 at 30.2 MPa. These GPP formulations also resulted in density in the range of 1700 kg /m³ as obtained in other studies, which were used for low-density applications [Baziak et al., Oct. 2021; Nguyen et al., Jan. 2022]. Despite having excellent compression strength, the use of these formulations for structural application may be determined by the characterization of other properties [Ntsa, 2023]. The variation in compression strength and density could have implications for radon exhalation rates, as denser and stronger materials generally exhibit lower permeability, potentially reducing radon escape. The substantially lower radon flux observed in GPP SM1A (Fig. 7) may thus be related to higher structural density due to the low water formulation used, which might have reduced void spaces and effectively impeded radon migration. Conversely, higher flux values in GPP MR1 and GPP MR2 compared to GPP SM1A may be attributed to their relatively lower density which in return corresponds to greater porosity, enhancing the

Table 5
Radon flux of different CFA-based geopolymer paste mix tested.

Products	Slope of radon build-up, Bq. m ⁻³ .hr ⁻¹	C_{Ra}	radon flux ($C_{Ra} \lambda / Area$) $\times$ 3600 Bq.m ⁻² .h ⁻¹
GPP SM 1A	10.20	18.89	2.38
GPP MR1	21.10	39.07	4.92
GPP MR2	20.00	37.04	4.67

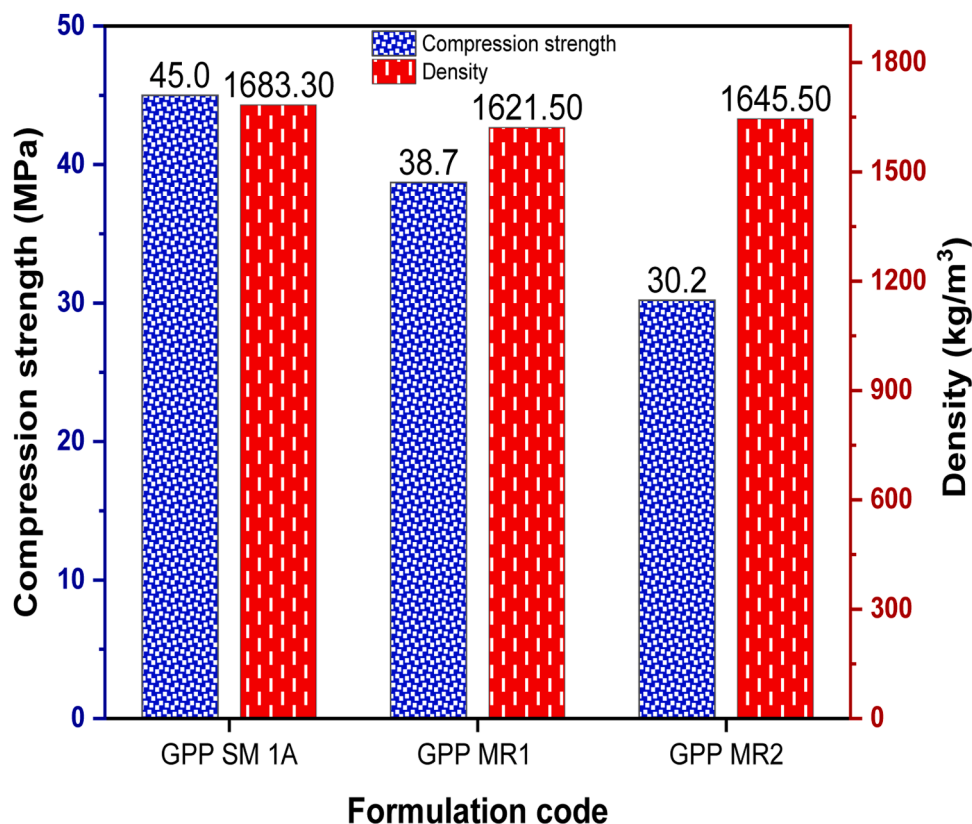


Fig. 8. Compression strength of CFA-based GPP formulations that were tested for radon measurement.

potential for radon emanation. Higher compression strength in GPP SM1A may indicate a more compact and stable matrix that could limit radon diffusion compared to less dense and weaker matrices that resulted from slight differences in the formulations used, as in the case of GPP MR2. The denser structures of GPP SM1A (1683.3 kg/m³) and GPP MR2 (1645.5 kg/m³) relative to GPP MR1 (1621.5 kg/m³) might thus restrict radon mobility, correlating with lower radon exhalation rates observed in the denser geopolymer sample. However, radon exhalation also depends on microstructure, porosity, and specific radionuclide content, so while high compression strength and density suggest a reduction in radon release, they may not be the sole factors influencing it.

Conclusions and prospects

The findings of this study underscore the potential of CFA-based GPP as an effective radon mitigation material by revealing critical interrelationships between formulations, activator content, elemental composition, structural properties, particle size distribution, and radon containment capabilities. Radioactivity measurements indicate that CFA has an average total activity of 680 Bq/kg, exceeding the accepted limit of 420 Bq/kg. In contrast, geopolymer products synthesized from it had lower activity values ranging from 408 to 459 Bq/kg. Additionally, the reduction of U and Th levels in GPP SM1A, GPP MR1, and GPP MR2, relative to raw CFA, contributed to decreased radioactivity and lower radon emanation. This showed that the dilution of CFA by the additional components in the formulations used in forming the geopolymer effectively reduced radionuclide content. Using CFA in its raw form or as an additive to concrete may lead to higher radon emissions due to the uranium and thorium content. In contrast, synthesizing CFA into GPP helps to encapsulate these elements to a greater extent, significantly reducing radon exhalation. This reduction is attributed to the GPP's increased density, which minimizes voids and restricts radon

emanation, as well as the dilution effect from additives in the formulation. Subtle differences in the formulations applied, such as water content, affected the aluminosilicate framework, primarily formed by SiO₂ and Al₂O₃, and show the critical role of the formulations used for geopolymer formation in creating a dense, stable matrix, demonstrated by increased compressive strength that restricts radon diffusion.

SEM analysis results highlight the microstructural compactness of GPP, like GPP SM1A, enhancing radon containment, while XRD results confirm the changes in mineralogical phases that may have aided in stabilizing the matrix. Compression strength and density analysis indicate that higher-density GPP, such as GPP SM1A, exhibited greater density and strength, most probably from the reduced water content used in the formulation. The optimized compositional adjustments in GPP SM1A resulted in the lowest radon exhalation rate among the products, underscoring the importance of precise material formulation, processing, and structural control in enhancing radon containment. Since it is uncertain what the effect of gamma radiation in building materials will be on inhabitants of houses, CFA-based GPP, with activities below 684 Bq/kg, may particularly be suitable for non-habitable construction applications, such as highway infrastructure, coastal protection, and articulated concrete block mat for erosion control, etc. However, for housing purposes, monitoring is recommended to ensure the radon emanations remain below safe levels. Future research should expand on these findings by investigating radiation risks within a full building envelope of CFA-based geopolymer formulations, providing a comprehensive assessment aligned with radiation protection guidelines.

CRediT authorship contribution statement

Evral Ntse: Writing – original draft, Data curation, Conceptualization. **Robert Lindsay:** Writing – review & editing, Data curation. **Tunde Victor Ojumu:** Writing – review & editing, Supervision, Resources. **Emile Salomon Massima Mouele:** Writing – review & editing. **Jean**

Luc Mukaba: Writing – original draft. **Chuks Paul Eze:** Supervision. **John T. Kevern:** Writing – review & editing. **Leslie F. Petrik:** Writing – review & editing, Visualization, Supervision, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

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Data availability

Data will be made available on request.

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