Indian Journal of Pure & Applied Physics
Vol. 61, June 2023, pp. 416-422
DOI: 10.56042/ijpap.v61i6.2411

Assessment of Natural Radionuclides Content and Radon Exhalation of Clay Pulverized Fly Ash Bricks

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Received 20 February 2023; accepted 23 May 2023

The bulk of bricks made worldwide are burnt clay bricks, and this volume of manufacture requires 230 - 242 million m^3 of agricultural land. This corresponds to 25,500 hectares of rich farmland at a depth of 1 meter. This type of exploitation will have a substantial detrimental impact on national food security. According to the present need, efficient use of industrial waste, such as fly ash, is necessary. So it becomes very important to limit the removal of top soil to build bricks and to encourage the use of pulverized fly ash (PFA) instead. So it is essential to examine the levels of radioactivity from natural sources and the amount at which radon is exhaled from building materials to manage the exposure of occupants to natural radiation. The radioactivity concentration of nuclides is determined using a high-resolution sodium iodide with a thallium activator gamma spectrometer. Radon exhalation rates were examined in the exhalation chamber using a hermetically shaped container and observing the activity over time. The focus of this research is to examine the radioactive concentration levels of radium, thorium, and potassium, as well as radon exhalation rates of Clay-Pulverized Fly Ash bricks mixed in different proportions. All radiation hazard factors linked with the radioactive nuclide and radon exhalation rates of clay pulverized fly ash bricks are examined and compared to the recommended limits from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) data.

Keywords: NaI(Tl) scintillator; Fly ash; Radioactivity; Radon exhalation; Natural radiation exposure

1 Introduction

The study of radiation is essential for our everyday lives as it makes us aware of the harmful effects of naturally occurring radiation sources. In actuality, radioactivity may be found in our bodies, the soil we walk on, and the air we breathe. For environmental protection, radioactive material monitoring is crucial. It is important to use quick and precise radioactivity measurement techniques[1]. Radon and its progeny produce alpha particles[2]. Radon is a highly radioactive inert gas, the isotope of the daughter nuclei of uranium ^{238}U. It has a physical half-life of 3.825 days, sufficient to permeate from construction buildings to the inner atmosphere of dwellings [3]. Gamma radiations are emitted by radioisotopes that occur naturally, like ^{40}K, and by their progeny from the ^{238}U and ^{232}Th families. Sufficient information on the levels and distribution patterns of radionuclides in the surroundings should be available, which helps to understand human exposure to radiation from natural and man-made sources for developing radiation protection policies[4]. Every year, India makes over 170 billion bricks, utilizing approximately 440 million tonnes of soil. The bulk of bricks made worldwide are burnt clay bricks, and this volume of manufacture requires 230 - 242 million m^3 of agricultural land. This corresponds to 25,500 hectares of rich farmland at a depth of 1 meter. This type of exploitation will have a substantial detrimental impact on national food security[5]. On the other hand, fly ash is a waste product derived from thermal power plants using coal as fuel. According to a survey conducted in 2016, 1.2 billion tonnes of coal ash were produced globally[6,7]. Approximately 226.13 million tonnes of fly ash were produced in India in the year 2019–20, and by the year 2025, it is expected to reach around 300–400 million tonnes[8,9]. The volume of waste generated by human activities and industry by products has been steadily rising over the past few decades, which has impacted disposal prices and escalated environmental concerns. In addition, the availability of geo-resources as virgin raw materials is a growing problem for our society. As a result, the industry is looking for alternate secondary raw materials, such as recycling wastes and by products. This trend is being driven in part by a growing awareness of the importance of green

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strategies[10-12]. So it becomes very important to limit the removal of top soil for the purpose of building bricks and to encourage the use of pulverized fly ash (PFA) instead. However, before it is used, it is necessary to understand the radiological aspect of it, which is crucial for human dwellings. Clay, sand, gravel, and soils, which are made into building materials from rocks and sediments, include naturally radioactive elements at wildly varying concentrations and can therefore be considered Naturally Occurring Radioactive Materials (NORM)[10,13-15]. The inherent radioactivity of building materials impacts the population's average yearly radiation dosage in terms of radiation that is emitted outside from the material and inside via exposure to radon exhalation[10,16,17]. Radium, thorium, and potassium activity concentrations are used to measure the gamma radiation which is emitted directly from external sources. The gaseous radon from the building materials, especially from holes and cracks, is a significant factor in exposure to internal radiation[10,18-20]. The focus of this study is to examine the radioactive concentration levels of radium, thorium, and potassium, as well as the radon exhalation rates of Clay-Pulverized Fly Ash bricks mixed in different proportions. All radiation hazard factors linked with the radioactive nuclide and radon exhalation rates of clay pulverized fly ash bricks are examined and compared to UNSCEAR approved limits.

2Materials and Methods

2.1 Collection and preparation of samples

Fly ash of Class C exhibit pozzolanic and cementing capabilities and was added to six locally sourced clay soil combinations. These mixtures were to be representative of those often found in the state of Kurukshetra, Haryana. Each sample has a predetermined total mass of 310 g with the variation of fly ash in proportion from 0 to 50%, with an increase of 10% by mass in each sample. The samples were precisely weighed with an electronic balance. Initially, the specimens were pulverized, then the powder was strained using a scientific sieve with a 150 mm mesh size. After that, the specimens were desiccated in the oven at 110 °C for 120 minutes to eliminate any leftover moisture content. Thereafter, all the samples were mixed properly, then sealed in a hermetic shaped container, and stored for six weeks to achieve an equilibrium state between the progenies of radon and thoron. The collected fly ash and clay samples were combined in various ratios to generate pulverized fly ash bricks in order to discover the strongest and most cost-effective combination. When the quantity of fly ash in bricks exceeded 50% by weight, casting the bricks became problematic[21]. The clay fly ash bricks were prepared by varying the fly ash contents in proportion from 0 to 50% with an increase of 10% by mass in each sample; they were denoted by FA0, FA10, FA20, FA30, FA40, and FA50, respectively as shown in Fig. 1.The previously made mixture was poured into the cubic clean casting mold by adding the necessary proportion of distilled water to thoroughly mix them and applying a hydrolytic pressure of 20 tons and allowed to dry for three days in the open under the sun. The residual moisture was then eliminated by drying it in an oven for two hours at 100 °C.

2.2 Determination of radionuclide content in samples

NaI(Tl) scintillator was used to measure the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in each brick sample. It is the perfect tool for radionuclide analysis because it is quick, non-destructive, and extremely effective. When the secular equilibrium of radionuclides was achieved in the samples in six

Fig. 1 — Different compositions of clay fly ash bricks.

weeks, the content of radium, potassium, and thorium in clay-fly ash mixtures was studied using sodium iodide with thallium activator gamma spectrometer which was attached to a multi-channel analyzer via a pre-amplifier. The instrument which is used for measuring the activity concentration was Atomtex's AT1315 gamma spectrometer and a detailed illustration of the device is mentioned in previously reported work[22]. The gamma rays that emerged from the materials were identified with the instrument and analyzed by an electronic system. The spectrum generated from the software SPTRATC (AT1315) is shown in Fig. 2, where gamma-ray energy is shown by a horizontal line and activity at particular energy by a vertical line. The relative limits of detection by the detector are 3,33 and 303Bq/kg for radium, thorium, and potassium, respectively. The instrument has a measuring inaccuracy of about ±20%.

2.3 Measurement of the radon exhalation rate

The quantity of radon activity concentration released per unit surface area or mass per unit time is the ^{222}Rn exhalation rate. The brick samples were enclosed in an accretion chamber with a known volume, and radon concentration inside the chamber was monitored through Smart Radon Monitor. The description of Smart Radon Monitor has been prescribed elsewhere[23]. To eliminate the moisture, the test samples were dried in an oven at 100 °C for a whole night. The hardened steel cylindrical chamber has an interior altitude of 190 mm and a diameter of 280 mm, in which the upper connection point for a photomultiplier tube is connected to a Lucas cell, as shown in Fig. 3. To preserve air tightness, a metallic cover with screws and a circular rubber gasket of size

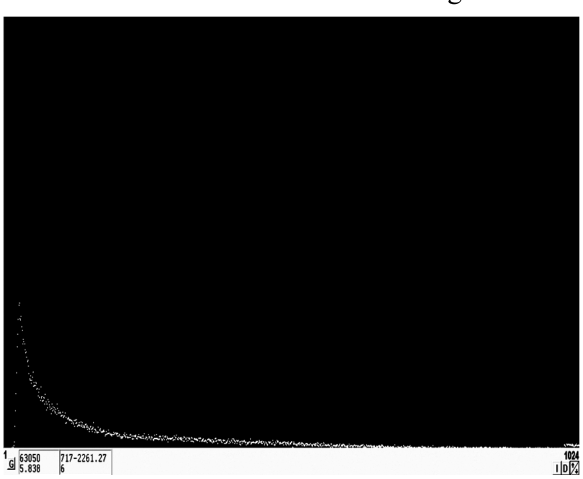

Fig. 2 — Analysis of Gamma spectrum of 10% fly ash with 90% Clay specimen.

280 mm was used to close the accumulator. The assessment cycle for each material was performed in diffusion mode for 180 minutes. Lucas cell-based SRM was used to measure the increase in radon concentration over time inside the container. To determine the appropriate brick that would have the maximum brick exhalation, all radon exhalation measurements were performed on various samples of bricks. The radon growth in the accumulator[24] is given by:

$$C = \frac{J_s S}{V\lambda_e}\left(1 - e^{-\lambda_e t}\right) + C_i e^{-\lambda_e t} \quad \ldots (1)$$

Where the symbols have their standard notation.

2.4 Estimation of the radiation hazard values

2.4.1 Radium equivalent activity (R_{eq})

To compare the activity concentration of samples containing various radionuclides (*i.e.*, radium, thorium, and potassium), the term "radium equivalent activity," or "Raeq," has been devised[25]. The production of 370, 259 and 4810Bq/kg of radium, thorium, and potassium, respectively, is anticipated to result in the same gamma ray dosage rate. The following equation is used to calculate $Ra_{eq.}$[26,27]:

$$Ra_{eq.} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad \ldots (1)$$

where the symbols have their standard notation.

2.4.2 Hazard index(H) : Internal H_{in} and External H_{ex}

The quantity of exposure to internal and external radiation caused by construction materials can be calculated by using the following hazard indices[28] equations:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad \ldots (2)$$

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad \ldots (3)$$

H_{in} depicts the radiological threat caused by radioactive radon to lung tissue. The fluctuation in

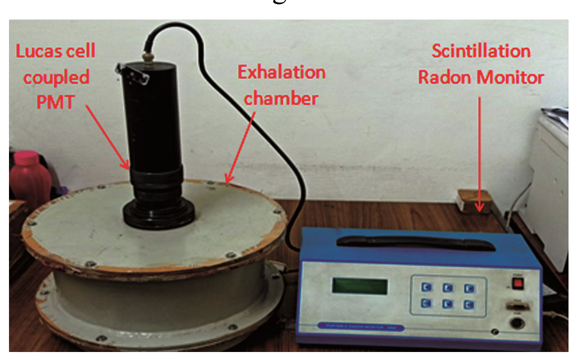

Fig. 3— Scintillation Radon Monitor with Exhalation chamber.

radon concentration in a dwelling was assumed to be compatible with the air-flow dynamics, nature, type, and permeability of the building material. The maximum level of $Ra_{eq.}$ (370Bq/kg) is assigned the greatest value Hex corresponding to unity. As a result, the value indices must be smaller than one in order to limit the influence of radiological hazards.

2.4.3 GammaIndex (Iɣ)

The method which is used to study the radioactive content in the building material which is originated from minerals is done by activity concentration index. Further, it gives information regarding the evaluation of concrete. The non-dimensional value of Iɣ, determined using equation, which accounts for the combined effect of the three primary natural radionuclides[28], is the requirement for satisfying the standard equation (4):

$$I\gamma = \frac{C_{Ra}}{370} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \leq 1 \quad \text{... (4)}$$

When evaluating the dosage criteria of $1mSva^{-1}$, the value of Iɣ should not be more than one.

2.4.4 Annual Gonadal Dose Equivalent (AGDE)

The effects of dose equivalents on the bone marrow and bone surface cells have been studied by UNSCEAR. The following equation (5) was employed for the calculation of specific activity of ^{226}Ra, ^{232}Th, and ^{40}K to study AGDE[29]:

$$AGDE = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_K \quad \text{... (5)}$$

2.4.5 Indoor air absorbed dose rate

The EC 1999 study report states that the indoor air absorbed dose rate (D)[30] from naturally existing radionuclides in building materials like gypsum, cement, bricks *etc*., emits gamma rays is calculated using the following equation (6) :

$$D(nGyh^{-1}) = 0.92A_{Ra} + 1.1A_{Th} + 0.08A_K \quad \text{... (6)}$$

Where the activity (Bq kg^{-1}) of ^{226}Ra is denoted by A_{Ra}, ^{232}Th is denoted by $A_{Th,}$ and ^{40}K is denoted by A_K.

2.4.6 Alpha index for radon concentration

The radon gas exhaled out from building materials, consumed excessively, caused a high level of alpha radiations, which is measured by alpha radiation index (Iα)[31], which is specified by the equation below (7) :

$$I_{\propto} = \frac{A_{Ra}}{200\ Bq\ Kg^{-1}} \quad \text{... (7)}$$

where the symbols have their standard notation, the suggested limit for the radon concentration for the indoor environment is $200Bq/m^3$.if the alpha radiation index surpasses unity, radon exhalation from the material may result in an indoor radon concentration[30] more than $200Bq/m^3$.

2.4.7 Annual effective dose (AED)

AED factor[32] for indoor dwellings due to gamma rays coming out from the fly ash mixes can be calculated by using the conversion factor of 0.7Sv/Gy. This factor is globally 0.8 for those who spent 80 % of their time inside the dwellings:

$$AED = D_{in}(\ nGyh^{-1}) \times\ 8760\ h\ \times\ 0.7\ Sv\ Gy^{-1} \times 0.8 \times 10^{-6} \quad \text{... (8)}$$

The permitted annually effective dosage for the residents should not exceed $1mSvy^{-1}$ as advised by the International Commission on Radiological Protection (ICRP), except pure fly ash[33].

3 Results and Discussion

3.1 Specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K

Table 1 provides an overview of the spectrometric analysis findings for the six samples (every sample repeated twice) of clay soil with varying fly ash amounts. With an average value of 13.87± 2.62, the concentration of ^{226}Ra spanned from 11.22±2.40 Bq/kg to 15.09±2.77 Bq/kg for a pure Clay soil sample to a pure fly ash sample. The ^{232}Th concentration varied similarly, with an average value of 6.32 ±1.9 and values ranging from 6.40 ±1.83 Bq/kg in a sample of 100% clay soil to 7.21±2.03 Bq/kg in a sample of 50% fly ash.

Table 1 — Radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Clay–fly ash mixtures.

Sample	Composition of the sample by mass		Radionuclide concentrations (Bq/kg)		
	CLAY%	Fly ash%	C_{Ra} (Bq/kg)	C_{Th} (Bq/kg)	C_K (Bq/kg)
FA 0	100	0	11.22±2.40	6.40± 1.83	80.55±13.16
FA 10	90	10	12.79±2.51	6.78±1.86	74.02±12.63
FA 20	80	20	12.70±2.53	5.28±1.87	58.17±11.47
FA 30	70	30	13.37±2.59	7.19±1.89	58.02±11.47
FA 40	60	40	18.04±2.92	5.09±1.92	67.72±12.34
FA 50	50	50	19.03±2.65	7.56±1.90	64.29±11.73

Figure 4 compares the specific activity of ^{226}Ra, ^{232}Th, and ^{40}K with varying fly ash percentage composition in clay soil. According to a 1999 report by the European Commission[16], the average global values of radium, thorium, and potassium are 40, 30, and 400 Bq/kg, respectively. The findings demonstrate that the mean activity concentrations of radium, thorium, and potassium are below the average values for the entire world.

3.2 Radiological hazard assessment results

All radiation hazard factors linked with the radioactive nuclide were used to assess the radiological hazard of natural radionuclides to residents in the study building materials. Table 2 displays the computed outcomes for the radiological hazard assessment of frequently used building materials. All of the samples' $R_{eq.}$ values were discovered to be lesser than the 370 Bq/kg limit advised for construction materials[16]. According to Table 2, the value of $Ra_{eq.}$ was lowest for pure clay soil (26.57 Bq/kg) and highest for 50% fly ash samples (34.79Bq/ kg). Due to a rise in the radionuclide content of radium and thorium, Ra_{eq}'s value increased as fly ash concentration in the mixture increased. The highest limit of Ra_{eq} is 370 Bq kg^{-1} which corresponds to the most significant value of H_{ex} *i.e*. Unity. These indices' values must be less than unity to maintain a negligible radiation risk. The safe limit of 1 was determined to be met for all samples' exterior and internal hazard indices, as indicated in Table 2. Given the dosage criteria of 1 mSva^{-1}, the value of I_{α} should not be greater than unity for materials used in bulk amounts for construction. As indicated in Table 2, all sample values were found to be under the safe limit of 1. All samples' AGDE values were below the suggested upper dosage limit of 1 mSvy^{-1}. In six samples under analysis, the value of AGDE ranged from 0.087 mSvy^{-1} to 0.110 mSvy^{-1} with an average value of 0.093 mSvy^{-1}. According to Table 2, the estimated indoor air absorbed dose rate (D) values vary from 23.81 nGyh^{-1} to 30.96 nGyh^{-1} for pure clay soil to 50% fly ash samples, respectively. The average D values of every building material under investigation were lower than the 84 nGy h^{-1} global population-weighted average indoor absorbed gamma radiation rate[16]. As indicated in Table 2, the value for the alpha index for radon concentration was discovered to be within the acceptable range of 1. The International Commission on Radiological Protection's recommended acceptable threshold of 1mSvy^{-1} value of annual effective dose. This threshold limit was not reached by any of the samples[33].

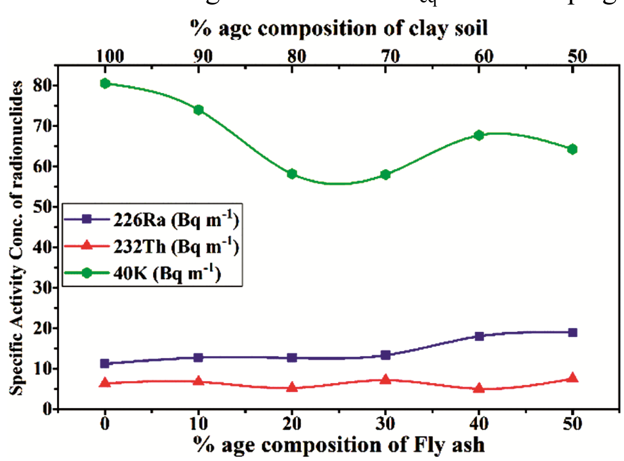

Fig. 4 — Graph showing activity concentration of radionuclides for different percentage compositions (fly ash in clay soil).

3.3 Radon surface exhalation rates results

The measurement of radon surface exhalation rates for all the samples was conducted using active methods via SRM and exhalation chamber (as described above) listed in Table 3.

The rise of radon was measured using a scintillation radon monitor within a closed chamber with building materials of a predefined mass or dimension. The average radon surface exhalation rate was 1.97±0.38 Bq/m^2/hr, ranging from 3.9± 0.85 Bq/m^2/hr in a pure clay soil sample to 0.85± 0.05 Bq/m^2/hr in a pure fly ash sample. Upon analyzing the results, it can be concluded that there is a significant drop in radon surface exhalation as the fly ash level in

Table 2 — Hazard indices and radiological dose parameters in Clay–fly ash mixtures.

Samples	R_{eq} (Bq/Kg)	H_{ex}	H_{in}	gamma index ($I\gamma$)	AGDE (Sv y^{-1})	D (nGy h^{-1})	($I\alpha$)	AED (mSv y^{-1})
FA 0	26.57	0.072	0.103	0.090	86.71	23.81	0.056	0.12
FA 10	28.14	0.075	0.110	0.093	91.10	25.15	0.064	0.13
FA 20	24.72	0.066	0.100	0.080	79.58	22.14	0.063	0.11
FA 30	28.12	0.076	0.110	0.091	89.58	24.58	0.067	0.12
FA 40	30.53	0.083	0.130	0.096	98.28	27.61	0.090	0.14
FA 50	34.79	0.094	0.145	0.110	110.59	30.96	0.095	0.15

Table 3 — Radon surface exhalation rate of clay pulverized fly ash bricks.

Samples	Mass of bricks (in Kg)	Radon Surface Exhalation Rates (in $Bq/m^2/hr$)
FA 0	3.545	3.9 ± 0.85
FA 10	3.220	2.85 ± 0.65
FA 20	2.238	1.0 ± 0.10
FA 30	3.244	1.75 ± 0.35
FA 40	3.302	1.5 ± 0.30
FA 50	3.315	0.85 ± 0.05

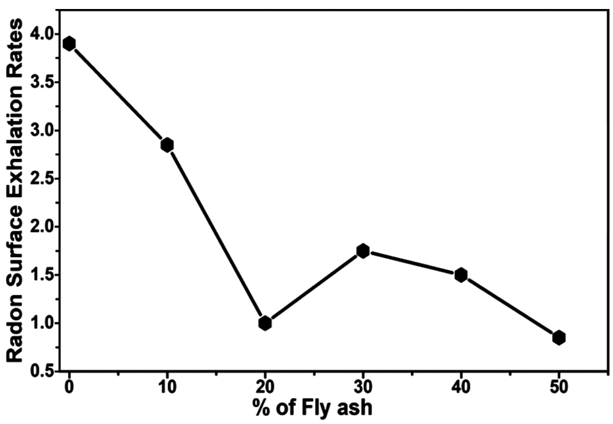

Fig. 5— Variation of radon surface exhalation rate with different amount composition of fly ash in clay soil.

the clay soil brick increases. The reason behind it may be due to the low porosity, increase in packing density, and presence of heavy elements like Pb, Sr, Zn, Rb, Co, etc. in fly ash, which may hinder the flow of radon when it is added up with clay soil. Fig. 5 shows the radon surface exhalation rate variation with different amounts of composition of fly ash in clay soil.

4 Conclusions

Fly ash has a wide range of uses in building materials, so it is essential to measure risk to the habitants from concentrations of ^{226}Ra, ^{232}Th and ^{40}K activity and radon exhalation rates when clay-pulverized fly ash brick is intended for use as a building material. The activity concentrations of radium, thorium, and potassium and radon exhalation rates of Clay-Pulverized Fly Ash bricks mixed in different proportions are within the safe limit corresponding to worldwide average values. So it shows that by the excess usage of fly ash in the soil, there is no risk of radiation hazard to human life, and it will stop the exploitation of fertile farmland and will limit the removal of top soil for the purpose of building bricks and encourage the use of pulverized fly ash (PFA).

Acknowledgment

The author would like to thank to Dr. Rohit Mehra, and N.I.T Jalandhar for providing the laboratory facilities and technical assistance for gamma spectroscopy.

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