ASSESSMENT OF HEALTH RISK DUE TO VARIATION IN CONCENTRATION OF FLYASH IN CEMENT

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ABSTRACT. Fly ash is widely used in the production of cement and concrete, and improves durability and strength of the concrete. As for all materials of mineral origin, fly ash contains mainly natural radio nuclides such as ^{226}Ra, ^{232}Th and ^{40}K. By the determination of the radioactivity level in building materials, the indoor radiological hazard to human health could be assessed. The aim of this study is to investigate the variation of natural radio nuclides (^{226}Ra, ^{232}Th and ^{40}K) in cement due to variation in the concentration of fly ash. For this purpose, mixtures with different concentration of fly ash are experimentally tested with the help of NaI(Tl) gamma spectrometer. To evaluate the potential radiological risk to individuals associated with these building materials, various radiological hazard indicators were calculated. From the measured gamma-ray spectra, radium equivalent, external and internal hazards, annual gonadal equivalent dose are determined for ^{226}Ra, ^{232}Th and ^{40}K. The results demonstrate that as the concentration of fly ash increased in the cement, concentration of ^{226}Ra and ^{232}Th also increased whereas ^{40}K concentration remained similar for all practical purposes.

Keywords: Natural radio nuclides, Radiation hazards, AGED, Emanation coefficient, Alpha dose equivalent

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INTRODUCTION

Most building materials of natural origin contain small amounts of Naturally Occurring Radioactive Materials (NORM), mainly radio nuclides from the ^{238}U and ^{232}Th decay chains, and ^{40}K. The worldwide average indoor effective dose due to gamma rays from building materials is estimated to be about 0.4 mSv per year [1]. Cement is a commonly used building construction material, and India is the world's second largest cement producer after China, surpassing the developed nations such as the USA and Japan. The natural radioactivity in cement gives rise to internal and external radiation exposure. However, the internal radiation exposure mainly affecting the respiratory tract is caused by the short-lived daughter products of radon, which are emitted from the construction material including the cement into the room air [2]. During respiration radon progeny deposit in the lungs and irradiate the tissue, thereby damaging the cells, and may cause lung cancer [3]. However, elevated levels of natural radio nuclides causing annual doses of several mSv were identified in some regions around the world, e.g. in Brazil, France, India, Nigeria, Iran. The materials that contain high concentration of NORM, have the potential to be carcinogenic when exposed to them. Therefore, monitoring of the radioactive materials in environmental samples is of primary importance from the view point of radiation protection of the environment.

It is well known that the most important source of external and internal exposure in building materials of dwellings is caused by gamma-rays and alpha-particles emitted from radio nuclides of the uranium (^{238}U) and thorium (^{232}Th) decay series as well as from ^{40}K [4-7]. Radon and thoron are both generated from radium isotopes decay in the solid grains. Both emanated to pore gases or fluids and then migrated to a significant distance from the site of generation in rock, soil and/or building materials into the atmosphere (exhalation) before undergoing radioactive decay. This can be explained as an exhalation process and plays an important role [8-10] for outdoor and indoor radon concentration. In order to assess the radiological hazards to human health in environment, it is required to measure the radioactivity levels in those building materials. The commercially available cements in the market are called PPC (Pozzolana Portland Cement) which contain fly-ash content in the range of 15%-35%. Out of various types of cements like Portland cement, high alumina cement and fly ash based cement (Portland Pozzolana Cement i.e. PPC), about 70% of cement produced worldwide is ordinary Portland cement (OPC) i.e without fly ash. The analyzed samples are mainly of Ordinary Portland Cement (OPC) and Portland Pozzolana Cement (PPC) collected from their manufacturers being manufactured by various factories.

The present study is dedicated to achieve the following objectives:-

1. Study of variation of natural radio nuclides (Ra-226, Th-232 and K-40) in cement due to variation in the concentration of fly ash.
2. Assessment of health risks, if any, associated with the usage of cement mixed with fly ash as building material.
3. Calculation of external and internal hazard indices for each mixture of cement and fly ash.

EXPERIMENTAL PROCEDURE

Pre-processing of samples

To evaluate a variety of cement and fly ash combinations, seven mixtures of Type I Portland cements and Class C fly ash were chosen to be representative of those commonly available in the state of Punjab. The total mass of each sample was fixed at 150g. The percentage composition of fly ash in the mixture was varied as 0, 10, 20, 30, 40, 50, and 100 percent by mass.

- The mixture was grinded, sieved by using scientific sieve of mesh size 150 µm and then, dried in open for 24 hours.
- In order to remove any trace of moisture, each sample was heated in an oven for 2 hours at 110°C.
- After heating, each sample was stored and sealed in plastic containers for 6 weeks to attain secular equilibrium.
- The samples were analyzed for radio nuclide content using NaI(TI) Gamma beta spectrometer. The pre-processed and packed samples are shown in Figure 1

Figure 1   Samples of different concentration of Fly Ash with Portland cement

Gamma ray spectrometry using NaI(Tl) detector

The concentration of natural radio nuclides is measured in prepared samples of cement and fly ash using a NaI(TI) gamma radiation detector as shown in Figure 2. If a gamma spectrometer is used for identifying samples of unknown composition, its energy scale must be calibrated first. Calibration is performed by using the peaks of a known source, such as caesium-137 or cobalt-60. Because the channel number is proportional to energy, the channel scale can then be converted to an energy scale. If the size of the detector crystal is known, one can also perform an intensity calibration, so that not only the energies but also the intensities of an unknown source—or the amount of a certain isotope in the source—can be determined. Because some radioactivity is present everywhere (i.e., background radiation), the spectrum should be analyzed when no source is present. The background radiation must then be subtracted from the actual measurement. Lead absorbers can be placed around the measurement apparatus to reduce background radiation.

The samples are counted for a period of 3 hours and are analyzed of the photo peak of ^{226}Ra, ^{232}Th and ^{40}K. The activity of 226Ra, 232Th and 40K is evaluated from 1764 KeV, 2610

KeV and 1460 KeV photo peaks. This spectral analysis is done with the help of computer software SPTR-ATC (AT-1315). The detection limits of the radio nuclides, ^{226}Ra, ^{232}Th and ^{40}K are 3 Bq kg^{-1}, 3 Bq kg^{-1} and 30 Bq kg^{-1} respectively.

Figure 2 NaI (Tl) detector

RADIUM EQUIVALENT ACTIVITY (R_{eq})

The distribution of natural radioactivity in the soil is non-uniform. Uniformity with respect to exposure to radiation is defined in terms of radium equivalent activity (Ra_{eq}) in Bq kg^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. The gamma transitions of energy 609 KeV or 1760 KeV (due to ^{214}Bi) is used to determine the concentration of ^{226}Ra. The criterion for this model considers the external hazard due to gamma rays corresponds to maximum Ra_{eq} of 370 Bq Kg^{-1} for the building material. This Ra_{eq} is calculated through the following relation [11]*:*

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.07A_K \qquad (1)$$

Where A_{Ra}, A_{Th} and A_K are the specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

EXTERNAL (H_{ex}) AND INTERNAL (H_{in}) HAZARD INDICES

The external and internal hazard index can be calculated by the following equation [12]:

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

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

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq Kg^{-1}, respectively. The values of this index must be less than unity in order to keep the radiation hazard significant. The radiation exposure due to the radioactivity from a construction material is limited to 1.5 mGy y^{-1}. The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} (370 Bq).

ANNUAL GONADAL DOSE EQUIVALENT (AGDE)

UNSCEAR considered the activity of bone marrow and bone surface cells as the organ of interest when estimating dose equivalent. Therefore, AGDE due to the specific activities of ^{226}Ra, ^{232}Th and ^{40}K was calculated using the following relation:

$$AGDE = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \quad (4)$$

GAMMA LEVEL INDEX (Iγ)

In specific samples, the gamma level index (Iγ) is used to estimate the level of γ-radiation hazard associated with the natural radionuclides. This index is also used to correlate the annual dose rate due to excess external gamma radiation caused by superficial materials. The gamma level index (Iγ) in the soil samples has been calculated using equation 5 given by European Commission [13]:

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \quad (5)$$

RESULTS AND DISCUSSION

The concentration of natural radio nuclides namely, ^{226}Ra, ^{232}Th and ^{40}K in cement – fly ash mixtures with varying percentage composition is summarized in Table 1. The concentration of ^{226}Ra ranges from in pure Portland cement sample to in pure fly ash sample. Similarly, the ^{232}Th content varies form in 100% cement sample to in 100% fly ash sample. ^{40}K does not vary much over the entire range of samples with varying fly ash concentration in cement. Thus, it can be concluded that as the content of fly ash increases in the Portland cement, the concentration of ^{226}Ra and ^{232}Th also increases.

The radio nuclides (^{226}Ra, ^{232}Th and ^{40}k) are not homogeneously distributed in the cement. It has been assumed that 370 Bq kg^{-1} of ^{226}Ra or ^{259} Bq kg^{-1} of ^{232}Th or 4810 Bq kg^{-1} of ^{40}K produce the same gamma dose rate. The acceptable limit for Ra_{eq} as recommended by the Organization for Economic Cooperation and Development [14] is 370 Bq kg^{-1}. As we can see in the table, the radium equivalent activity values for all samples were found to be lower than the recommended limit for building materials of 370Bq kg^{-1}. Ra_{eq} value is minimum for pure

OPC (195.6 Bq kg^{-1}) and maximum for pure fly ash sample (322.89 Bq kg^{-1}). The value of Ra_{eq} increases with increasing concentration of fly ash in mixture due to increase in radionuclide content of ^{226}Ra and ^{232}Th.

The external radiation exposure is usually associated with the gamma radiations emitted by ^{226}Ra, ^{232}Th and ^{40}K. For most of the samples, external and internal hazard indices are found to be within the safe limit of 1. H_{ex} for pure fly ash sample however, exceeds unity.

The Annual Gonadal Dose Equivalent (AGDE) for most samples (i.e. the mixtures with upto 30% fly ash in Portland cement) fell below the recommended upper dose limit of 1 mSv y^{-1}. The value of AGDE varies from 0.79mSv y^{-1} to 1.16mSv y^{-1} in 7 samples under investigation with mean of 0.95 mSv y^{-1}. But as the fly ash concentration becomes ≥40%, the values of annual dose effect are found to be higher than 1 mSv y^{-1}, which is harmful for residents in indoor dwellings.

Table 1 Concentration of ^{226}Ra, ^{232}Th and ^{40}K in cement-fly ash sample mixtures

%AGE COMPOSITION		A_{Ra}	A_{Th}	A_K	A_{eff}	Ra_{eq}	I_{yr}	H_{ex}	H_{in}	AGDE
OPC	Fly ash	(Bq/kg)	(Bq/kg)	(Bq/kg)	(Bq/kg)	(Bq/kg)				(mSv/y)
100	0	26.79 ±6.97	20.31 ±6.00	1996.03 ±256.18	273.7	195.6	1.7	0.5	0.6	0.79
90	10	32.52 ±7.70	25.67 ±6.59	1982.59 ±254.54	291.4	208.0	1.8	0.6	0.6	0.82
80	20	55.22 ±9.11	41.98 ±7.64	1804.31 ±238.43	342.8	241.6	2.0	0.6	0.8	0.91
70	30	57.49 ±9.33	44.22 ±7.80	1889.38 ±247.07	358.3	252.9	2.1	0.7	0.8	0.95
60	40	63.11 ±9.80	55.30 ±8.71	1829.16 ±242.77	379.3	270.2	2.2	0.7	0.9	1.00
50	50	65.20 ±10.00	55.49 ±8.75	1849.66 ±245.20	385.8	274.0	2.2	0.7	0.9	1.01
0	100	81.79 ±11.36	76.54 ±10.52	1880.80 ±248.83	429.3	322.8	2.5	0.9	1.1	1.16

CONCLUDING REMARKS

From the measured gamma-ray spectra, radium equivalent, external and internal hazards, annual gonadal equivalent dose are determined for ^{226}Ra, ^{232}Th and ^{40}K. The radium equivalent for ^{226}Ra, ^{232}Th and ^{40}K was found to be varying in the range of 195.6 Bq Kg^{-1} to 322.89 Bq Kg^{-1} with mean value of 252.188 Bq Kg^{-1}. The external and internal hazards was found to be varying in the range of 0.57 to 0.91 Bq Kg^{-1} and 0.64 to 1.13 Bq Kg^{-1} respectively with mean value of 0.71 and 0.86 Bq Kg^{-1} respectively. The annual gonadal equivalent dose was found to be varying in the range of 0.79mSv y^{-1} to 1.16mSv y^{-1} with the mean value of 0.95mSv y^{-1}.

From the present study, it can be concluded that:

- Concentration of ^{226}Ra and ^{232}Th increases with the increase in the concentration of Fly ash in the Portland cement.
- Results match with the considerable limit of the concentration of Fly ash in cement in the range of 15% to 25% which is confirmed by external hazard values as reported by OCED, 1979.
- In general, the average values of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the samples are lower than the world figures as reported in UNSCEAR, 2000 [1].
- The calculated value of H_{ex} for the samples is less than unity. It is inferred that for all cement samples, the Ra_{eq} value is well within the permissible limit (370Bq kg^{-1}).
- No considerable change in ^{40}K as the concentration of fly ash increases in Portland cement.

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