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Alpha radioactivity in Indian cement samples

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Background: The essential constituents of cements like lime, silica and alumina are derived from earth's crust in which radioactive elements like uranium, thorium etc are also present in varying amounts almost everywhere. These two elements are considered as the parent elements of uranium and thorium radioactive decay series in which radon and thoron are produced respectively as decay products. In the present study the samples of ordinary Portland cement (OPC), Portland pozzolana cement (PPC) and some other cementious finishing materials like white cement, Plaster of Paris (POP), cement putty etc were collected and analysed for radium and radon concentrations along with radon exhalation rates. **Materials and Methods:** Alpha sensitive LR-115 Type II plastic track detectors commonly known as "Solid State Nuclear Track Detectors" were used to measure the radium and radon concentration. The alpha particles emitted from the radon causes the radiation damaged tracks. The Chemical etching in NaOH at 60 °C for about 90 minutes was done to reveal these latent tracks, which were then scanned and counted by an optical microscope of suitable magnification. By calculating the track density of registered tracks, the radon and radium concentrations along with exhalation rate of radon, were determined using required formulae. **Results:** The radon and radium concentration in various brands of cements found to vary from 333 ±9.9 to 506 ±13.3 Bq m-3 and from 3.7 ± 0.1 to 5.6 ± 0.2 Bq kg-1 while in various cementious finishing materials used in the construction, these were found to vary from 378 ±19.7 to 550 ± 9.8 Bq m-3 and from 4.2 ± 0.2 to 6.1 ± 0.1 Bq Kg-1, respectively. Based on the data the mass and surface exhalation rates were also calculated. **Conclusion:** The measurements indicate that there is marginal variation of the concentration of radium and radon in various brands of cements in India with lower levels in the cement samples having red oxide and higher levels in fly ash based cement samples but overall concentration levels of radon and radium are lower than that of average global values. **Iran. J. Radiat. Res., 2006; 3 (4): 171-176**

Keywords: *Portland cement, radioactivity, radon concentration, nuclear tracks, exhalation rate.*

INTRODUCTION

Radiation plays an important role in our every day life as the world is naturally radioactive and each of us is exposed to naturally occurring quantities of radiation. In fact, radioactivity can be in the air we breathe, the soil on which we walk the dwellings in which we live and even within our bodies (1). The present study deals with radon exhalation in Portland cement in which radon gas with a half life of 3.8 days is emanated in air as a product of ^{238}U that occur as a trace element almost in all the naturally occurring materials including the cementious materials used in the construction of cement grouts, mortars and concrete. The direct predecessor of radon in the decay series of ^{238}U is ^{226}Ra is often incorporated into the solid matrix of the cement. Before the radon can enter the pore space of the material and become available for transport to the indoor environment, it has to escape from the solid matrix of the particle. Radon is released from the mineral grain via alpha recoil. If no structural changes and chemical reactions takes place in the material containing the radium then the emanation of radon in the pore space is controlled by two mechanisms i.e. recoil and diffusion through the solid matrix (2).

Cement is a commonly used building construction material and India is the world's second largest cement producer (6% share of world cement production) after China (44%), surpassing the developed nations like US and Japan. The natural level of radioactivity in cement gives rise to internal and external indoor exposure. The external exposure is caused by gamma radiation originating from the members of the uranium (^{238}U) and thorium (^{232}Th) decay series and also from potassium (^{40}K) (3). However, the internal

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radiation exposure mainly affecting the respiratory tract is caused by the short-lived daughter products of radon, which are exhaled from the construction material including the cements into the room air (4). The hazard of radon comes from its radioactive progeny, which use their physical properties to spread or attach in the same way as the aerosols do, trapped in the lung and deposit their alpha particle energies in the tissue, beta particles and the gamma radiations (5). Lung cancer, skin cancer and kidney diseases are the hazards by the inhalation of radon decay products (6). The radiological impact caused by nuclides is due to radiation exposure of the body by the gamma rays and irradiation of the lung tissues from inhalation of radon and its progeny (7). Therefore, keeping in view the natural risk, it is necessary to know the dose limits of public exposure (8) and to measure the natural environmental radiation level in dwellings in which cement and other cement based finishing materials are used as construction materials as most of the individuals spend 80% of their time indoors. Due to this fact great attention was given to make assessment of the concentration of radionuclides in the various brands of cements and other cementious finishing materials like white cement and plaster of Paris.

Out of various types of cements like Portland cement, high alumina cement and fly ash based cement (called Portland Pozzolona Cement i.e. PPC), about 70% of cement produced world wide is ordinary Portland cement (OPC). But in India, the production of fly ash based cement called PPC, has increased dramatically over the last two years. Portland cement is made by heating the correct mixture of lime stone ($CaCO_3$) with sand (SiO_2) and clay (alumina silicate) at a temperature of about 1450-1600°C in a rotary kiln. When mixed with water, the Portland cement sets to give concrete which is a hard insoluble solid similar in appearance Portland rock which is a famous building stone of England (9). It is a grayish heavy powder containing calcium aluminates and silicates. About 2-5% of natural gypsum ($CaSO_4.2H_2O$) is added to slowdown the setting as the slow setting greatly increases its strength. High alumina cement is made by fusing limestone and bauxite (an ore of aluminum) with small amounts of SiO_2 and TiO_2 at 1400-1500 °C. These are more expensive than the Portland cement but it sets quickly and develops high strength within one day. It is used to make beams for bridges and buildings.

In the present study the analyzed samples are mainly of ordinary Portland cement (OPC), Portland pozzolana cement (PPC) and some other cementious finishing materials like white cement, plaster of Paris (POP), cement putty etc. collected from their manufactures being manufactured by various factories located in the states of Rajasthan, Madhya Pradesh and Himachal Pradesh in India keeping in view the availability of raw material required for the manufacturing of above cements and finishing materials. The state of Madhya Pradesh is situated in central part of India and Rajasthan belongs to western region both forming the part of Aravali range of hills while Himachal Pradesh is having its location in northern India in sub Himalayan terrain and a part of Shivalik range of hills.

This paper outlines the methodology used for the assessment of concentration of radium and radon and hence the radon exhalation rates from various brands of cements and cementious finishing materials used in the construction of buildings. The potential radiological hazard of the different samples is estimated by measuring the radon concentration as well as radon exhalation rate using 'Solid State Nuclear Track Detector (SSNTDs)'.

MATERIALS AND METHODS

For the measurement of radon concentration along with radium concentration etc we used 'CAN' technique (10). Three samples of each brand of cements and other cementious finishing materials used as building construction material like plaster of Paris

(POP) and white cements were collected from the manufacturers along with their detailed literature having technical details etc. Known amounts (0.01 kg) of each of the samples (Oven dried and filtered through a sieve) were taken in plastic cans. Alpha sensitive LR-115 Type III plastic track detectors were fixed on the bottom of the lid of each 'Can' with cello tape such that the sensitive side of the detector faced the specimen. The cans were tightly closed from the top and sealed. The geometrical parameters of the detector and container were: LR-115 Type II plastic track detectors (1 cm × 1 cm), plastic cans with internal diameter 7.66 cm, height 9.9 cm, surface area = 46 cm^2, volume = 454 cm^3.

The exposure time of detectors was taken as 90 days after which these were removed and subjected to a chemical etching process in 2.5 NaOH solutions at 60 °C for one and half hour. The detectors were washed and dried. The tracks produced by alpha particles were observed and counted under an optical microscope of suitable magnification. The measured track density (track/cm^2/day) was converted into radon concentration in Bq m-3 using calibration factor for the given LR-115 type II detector as 0.021 track cm^{-2} day^{-1}=1 Bq m^{-3} as used by other workers (11-13). Radium concentration has been calculated using the following relation (14):

$C_{Ra} = \rho\, h\, A / K\, T_e\, M$

Where ρ = Background corrected alpha track density due to radon (Track cm^{-2})

h = Distance between the detector and top of the sample (m)

A = Surface area of sample (m^2)

K = Sensitivity factor (Tracks cm^{-2} day^{-1} per Bq m^{-3})

M = Mass of sample (kg)

Te = effective time of exposure (days) where $T_e = [T-\lambda^{-1}(1-e^{-\lambda T})]$ in which λ is decay constant for radon given by $\lambda = 0.6931/T_{1/2}$. Here $T_{1/2}$ is half-life of Radon and it is 3.82 days.

Exhalation rates (Ex) were calculated using the following equations (15-16):

For mass exhalation rate:

$$E_x = \frac{CV\lambda / M}{T + 1/\lambda(e^{-\lambda t} - 1)} (BqKg^{-1}h^{-1}) \qquad (1)$$

and for surface exhalation rate :

$$E_x = \frac{CV\lambda / A}{T + 1/\lambda(e^{-\lambda t} - 1)} (BqKg^{-1}h^{-1}) \qquad (2)$$

Where: C = Integrated radon exposure (Bq m^{-3} h^{-1})

V = Volume of air in the CAN (m^3)

T = Time of exposure (hrs)

λ = Decay constant for radon (h^{-1})

M = Mass of sample (kg)

A = Surface area of the sample (m^2)

RESULTS AND DISCUSSION

The calculated values of radium concentration, radon concentration, and mass and surface exhalation rates of radon for various brands of cements in India are presented in table 1 while for various cementious finishing materials like white cement, white cement putty and Plaster of Paris (POP) are shown in table 2. It can be seen from the results that the radium and radon concentration vary appreciably from sample to sample in both the cases. It may be due to variation in the location of raw materials from where these are derived and the percentage of constituents used by various manufacturers in the manufacturer of elements as these parameters depends upon the distribution of naturally occurring radionuclides present in soil and rocks in the earth's crust. Since these radionuclides are not uniformly distributed, the knowledge of their distribution plays an important role in radiation protection and measurement (17).

The radon concentration and radium concentration in various brands of cements varies from 333 ± 9.9 to 506 ±13.3 Bq m^{-3} and from 3.7 ± 0.1 to 5.6 ± 0.2 Bq kg^{-1} respectively while the mass and surface exhalation rates found to change from 12.1 ± 0.4 to 18.3 ± 0.5 m Bq kg^{-1} hr^{-1} and from 263 ± 7.5 to 402 ± 10.6 m Bq m^{-2} hr^{-1} respectively.

Similarly, the radon concentration and radium concentration in various cementious finishing materials used in the construction of buildings found to vary from 378 + 19.7 to 550 + 9.8 Bq m^{-3} and from 4.2 ± 0.2 to 6.1 ±

Table 1. Radium concentration and radon exhalation rates in some cement samples of various brands in India.

S. N.	Sample names	Samle codes	Radon concentration (Bq m^{-3})	Radium concentration (Bq kg-)	Mass exhalation rates (mBq kg^{-1} hr-1)	Surface exhalation rates (mBq m^{-2} hr^{-1})
1	Cement OPC-RJ 43 grade	1C-1 1C-2 1C-3	326 365 307	3.6 4.0 3.4	11.9 13.3 11.2	258 288 244
AM ± SE*			333 ± 9.9	3.7±0.1	12.1±0.4	26.3±7.5
2	Cement PPC-RJ 53 grade	2C-1 2C-2 2C-3	518 461 538	5.8 5.1 6.0	18.9 16.8 19.6	412 366 427
AM ± SE*			506±13.3	5.6±0.2	18.4±0.5	402±10.6
3	Cement OPC-RJ 43 grade	3C-1 3C-2 3C-3	403 422 480	4.5 4.7 5.3	14.7 15.4 17.5	320 336 381
AM ± SE*			435 ± 13.3	4.8 ± 0.1	15.9 ± 0.5	346 ± 10.5
4	Cement OPC-RJ 43 grade	4C-1 4C-2 4C-3	442 403 480	4.9 4.5 5.3	16.1 14.7 17.5	351 320 381
AM ± SE*			442 ± 12.8	4.9 ± 0.1	16.1 ± 0.5	350 ± 10.2
5	Cement OPC-RJ 53 grade	5C-1 5C-2 5C-3	461 384 422	5.1 4.3 4.7	16.8 14.0 15.4	366 305 336
AM ± SE*			422 ± 12.8	4.7 ± 0.1	15.4 ± 0.5	336 ± 10.2
6	Cement PPC-RJ 43 grade	6C-1 6C-2 6C-3	480 518 442	5.3 5.8 4.9	17.5 19.0 16.1	381 412 351
AM ± SE*			480 ± 12.7	5.3 ± 0.2	17.5 ± 0.5	381 ± 10.2
7	Cement OPC-MP 53 grade	7C-1 7C-2 7C-3	442 403 461	4.9 4.5 5.1	16.1 14.7 16.8	351 320 366
AM ± SE*			435 ± 9.9	4.8 ± 0.1	15.9 ± 0.4	346 ± 7.8
8	Cement OPC-MP 43 grade	8C-1 8C-2 8C-3	422 365 403	4.7 4.0 4.5	15.4 13.2 14.7	336 288 320
AM ± SE*			398 ± 9.7	4.4 ± 0.1	14.4 ± 0.4	315 ± 8.1
9	Cement OPC-HP 53 grade	9C-1 9C-2 9C-3	480 499 442	5.3 5.6 4.9	17.4 18.2 16.1	381 396 351
AM ± SE*			474 ± 9.7	5.3 ± 0.1	17.2 ± 0.4	376 ± 7.6
10	Cement OPC-HP 43 grade	10C-1 10C-2 10C-3	461 480 422	5.1 5.3 4.7	16.8 17.5 15.4	366 381 336
AM ± SE*			454 ± 9.9	5.0 ± 0.1	16.6 ± 04	361 ± 7.6

Table 2. Radium concentration and radon exhalation rates in some cementious finishing materials of various brands in India.

S. N.	Sample names	Samle codes	Radon concentration (Bq m^{-3})	Radium concentration (Bq kg-)	Mass exhalation rates (mBq kg^{-1} hr^{-1})	Surface exhalation rates (mBq m^{-2} hr^{-1})
1	White Cement RJ	1WC-1 1WC-2 1WC-3	365 326 442	4.0 3.6 4.9	13.3 11.9 16.1	288 258 351
AM ± SE*			378 ± 19.7	402 ± 0.2	13.8 ± 0.7	299 ± 15.8
2	White Cement MP	2WC-1 2WC-2 2WC-3	556 518 576	6.2 5.8 6.4	20.3 18.9 21.0	442 412 458
AM ± SE*			550 ± 9.8	6.1 ± 0.1	20.1 ± 0.4	437 ± 7.8
3	Wall Putty MP	WP-1 WP-2 WP-3	518 576 480	5.8 6.4 5.3	18.9 21.0 17.5	412 458 381
AM ± SE*			525 ± 16.1	5.8 ± 0.2	19.1 ± 0.6	417 ± 12.9
4	Plaster of Paris-HR	1PP-1 1PP-2 1PP-3	556 480 422	6.2 5.3 4.7	20.3 17.5 15.4	442 381 336
AM ± SE*			486 ± 22.4	5.4 ± 0.3	17.7 ± 0.8	386 ± 17.7
5	Plaster of Paris-RJ	2PP-1 2PP-2 2PP-3	518 422 461	5.8 4.7 5.1	18.9 15.4 16.8	412 336 366
AM ± SE*			467 ± 16.1	5.2 ± 0.2	17.0 ± 0.6	371 ± 12.8
6	Plaster of Paris-RJ	3PP-1 3PP-2 3PP-3	442 499 537	4.9 5.6 6.0	16.1 18.2 19.6	351 396 427
AM ± SE*			493 ± 15.9	5.5 ± 0.2	18.0 ± 0.6	391 ± 12.7

"SE (standard error) = S /ÖN, Where S is SD (standard deviation) and N is the no of observations

OPC - RJ	(Ordinary Portland cement from Rajasthan State)
OPC - MP	(Ordinary Portland cement from Madhya Pradesh State)
PPC - RJ	(Portland Polozona cement from Rajasthan State)
OPC - HP	(Ordinary Portland cement from Himachal Pradesh State)
RJ	Rajasthan State
MP	Madhya Pradesh State
HR	Haryana State.

0.1 Bq Kg^{-1} respectively while the mass and surface exhalation rates are varying from 13.8± 0.7 to 20.1± 0.4 m Bq kg^{-1} hr^{-1} and from299 ± 15.8 to 437± 7.8 m Bq m^{-2} hr^{-1}.

The measurements indicate higher levels of radon and radium concentrations in fly ash based cement (called Portland pozolona cement-PPC) as compared to ordinary Portland cement (OPC) as compared to ordinary Portland cement (OPC), although these are lower than the global average value (5).

Similarly, the radon and radium concentrations in other cementious finishing materials used in the construction of buildings are within safe limits. From the present study it is concluded that the fly ash based cement (PPC) is found to have radon concentration and radium content below the average global value and its production should be encouraged as compared to ordinary Portland cement as it leads to high savings in terms of energy and environmental pollution. If 15% of cement is replaced by supplementary cementing materials including fly ash then there would be reduction of 227 million tons of CO_2 worldwide due to reduction in the use of lime stone which will save the environment from being polluted. Per ton of fly ash used in cement manufacturing saves 1.40 ton of limestone with a saving of 3.306 Joule thermal energy and reduction in 1.0 ton of CO_2, a pollutant green house gas (18)

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