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A study of the natural radioactivity and radon exhalation rate in some cements used in India and its radiological significance

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ABSTRACT

The presence of natural radioactivity and radon exhalation from building materials contribute to the radiation dose received by human. So, it is essential to evaluate the activity levels of the primordial radionuclides (^{238}U, ^{226}Ra, ^{232}Th and ^{40}K) present in the building materials for the assessment of natural radiation dose. Cement is one of the major component of the building materials and is used on a large scale. In the present study, the commercially available cement samples of fifteen different brands were used to study the radon exhalation rate and activity concentration of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K nuclides. The radon exhalation rate was measured using the can technique, while the concentration of radionuclides content was determined by using gamma ray spectroscopy. The radon exhalation rate from different brands of cements was found in the range from 1.56 to 13.1 mBqkg^{-1}h^{-1} with a mean value of 5.27 mBqkg^{-1}h^{-1}. The specific activity of uranium was found in the range 45.3–218.9 Bqkg^{-1} with a mean value of 111.2 Bqkg^{-1}; ^{226}Ra from 20.3 to 60.1 Bqkg^{-1} with a mean value of 35.8 Bqkg^{-1}; ^{232}Th from 18.8 to 60.1 Bqkg^{-1} with a mean value of 33.2 Bqkg^{-1} and ^{40}K varied from 160.9 to 248.1 Bqkg^{-1} with a mean value of 199.1 Bqkg^{-1}. The radiological parameters – radium equivalent activity, absorbed dose rate, annual effective dose, external hazard index, internal hazard index, gamma activity index and alpha index were also evaluated to assess the potential radiological hazard associated with these cement samples. Correlation coefficients for the different radionuclides have been evaluated and studied.

1. Introduction

Most building materials of natural origin contain small amounts of naturally occurring radioactivity materials (NORM), mainly radio nuclides from the ^{238}U and ^{232} Th decay chains and ^{40}K. The radionuclides in the series headed by ^{235}U are relatively less important from a dosimeteric point of view (UNSCEAR, 1992). The origin of these materials is the Earth's crust, but they find their way into building materials, air,

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water, food and the human body itself. The world wide average indoor effective dose due to gamma rays from building materials is estimated to be about 0.4 mSv per year (UNSCEAR, 2000). In many parts of the world, building materials containing radioactive nuclides have been used for generations. As individuals spend more than 80% of their time indoors, the internal and external radiation exposure from building materials creates prolonged exposure situations (ICRP, 1999). The external radiation exposure is caused by the gamma emitting radionuclides, which in the uranium series mainly belongs to the decay chain segment starting with ^{226}Ra. The internal (inhalation) radiation exposure is due to ^{222}Rn, and marginally to ^{220}Rn, and their short lived decay products, exhaled from building materials into the room air. A study carried out by Papastefanou, Stoulos, and Manolopoulou (2005) in Greece on building materials has shown that out of the investigated building materials, granite and phosphogypsum are the highly radioactive materials for which the absorbed dose rate in indoor air becomes upto five times higher than the dose criterion. However, a study carried out by Lu, Li, Yang, and Zhao (2013) on building materials collected from Yan'an, China has shown that the investigated building materials may be used safely as construction materials. All this depends upon the level of radioactivity in the soil, rock and industrial by-products from where these building materials are derived. So, knowledge of the level of natural radioactivity in building materials is then important to assess the possible radiological hazards to human health and to develop standards and guidelines for the use and management of these materials.

Portland cement (chemically known as calcium aluminosilicate) is an important construction material for houses and other buildings. It is used for making blocks and concrete, and for plastering the buildings made of bricks. The essential raw materials of the Portland cement are: Limestone ($CaCO_3$), latrite (Fe_2O_3), slate ($Al_2O_3SiO_2$) and gypsum ($CaSO_4 \cdot 2H_2O$) (Shreve, 1972). The cement industry is considered one of the basic industries which play an important role in the national economy of developing countries (El-Bahi, 2004). It is one of the lowest-cost materials widely used in construction. White cement is made from specially selected raw materials, usually pure chalk and white clay (Kaolin) containing very small quantities of iron oxides and manganese oxides. The specific levels of gamma radiation in cement depend upon the content of radium, thorium and potassium in the raw products, used in the manufacture of cement.

Recent studies of people exposed to radon have confirmed that radon in homes represents a serious health hazard (WHO, 2009). Out of the average annual dose of 2.4 mSv from natural sources, 1.2 mSv is through inhalation, mainly from radon (UNSCEAR, 2000). The main health risk associated with long term, elevated exposure to radon is an increased risk of developing lung cancer, which depends on the radon concentration and the length of exposure. It is estimated that radon causes between 3% and 14% of all lung cancers, depending on the average radon level in a country. An increased rate of lung cancer was first seen in uranium mines exposed to high concentrations of radon. In addition, studies in Europe, North America and China have confirmed that even low concentrations of radon such as those found in homes also confer health risks and constitute significantly to the occurrence of lung cancers worldwide (WHO, 2014). Radon gas enters the indoors from different sources, such as soil or rock under or surrounding the buildings, building materials, water supplies, natural gas and outdoor air. As in soil and rock, radon gas (^{222}Rn) is formed inside building materials by decay of the parent nuclide ^{226}Ra. However it is not possible to determine the radon exhalation rate simply from the activity concentration of ^{226}Ra (Jing, Navreen, & Ibrahim, 2010). The radon exhalation rate may vary from one building material to the other having the same ^{226}Ra activity levels.

The objective of the present work was to measure the natural activity concentration due to radionuclide contents and radon exhalation rates in different brands of cement used in India and to assess the associated potential radiological hazards by the use of these cements. Out of the 15 samples studied, 10 samples belong to different manufacturing companies of India and 5 are the imported ones. Earlier Kumar, Ramachandran, and Prasad (1999) and Ramola, Manjulata, and Gusain (2014) have carried out study on different building materials and soil in India.

2. Materials and methods

2.1. *Measurement of radioactivity by gamma ray spectrometry*

Cement samples of various brands were collected from the suppliers and processed as per the procedure described in IAEA TRS-295 (IAEA, 1989). The processed samples were filled in the cylindrical plastic containers of dimensions 7 cm (diameter) × 6.5 cm (height), sealed completely to make them air tight and kept for about one month so as to ensure secular equilibrium between radium and radon progenies of both uranium and thorium series.

The samples were analysed by using high resolution gamma spectrometry system. It consists of coaxial High Purity Germanium (HPGe) detector having 50% relative efficiency with respect to 7.62 cm × 7.62 cm NaI (Tl), its energy resolution measured in terms of full width at half maximum (FWHM) is 2.1 keV at 1332.5 keV of ^{60}Co gamma energy at 25 cm from the top of the detector. The gamma spectra were analyed using an 8 K MCA (PHAST, Peak Shape Analysis Software developed by Electronics Division, Bhaba Atomic Research Centre, India) along with other electronic accessories coupled with the HPGe detector. The detector is shielded with 7.5 cm lead to reduce the background contribution of the surrounding. The certified reference materials IAEA RGU-I and RGTh-I have been used for the energy and efficiency calibration of the system in the energy range of 46.53–2614.53 keV. The spectra were acquired for 100000 s and the photo peaks were evaluated by using the MCA emulation software. The minimum detectable activity (MDA) of the system has been estimated with 95% confidence level (Currie, 1968). The combined standard uncertainty was calculated by considering the error associated with counting, gamma emission probability and efficiency calibration. ^{238}U emits very very weak gamma rays; hence, its progeny ^{234}Th gamma line such as 63.25 keV and 92.59 keV can be used to assess it. Among the two, the former is free from major