EXHALATION OF RADON AND THORON FROM PHOSPHOGYPSUM USED AS BUILDING MATERIAL

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ABSTRACT

The radioactive properties of two types of phosphogypsum, were determined. Gypsum plates with different thickness were produced. The ^{226}Ra and ^{232}Th concentrations were measured by means of high resolution gamma spectrometry. The results are for type 1 ^{226}Ra : 75 Bq/kg and ^{232}Th 230 Bq/kg and for type 2 ^{226}Ra : 155 Bq/kg and ^{232}Th : 160 Bq/kg. The radon (^{222}Rn) exhalation rate was evaluated by closing the plates in airtight barrels and measuring the radon concentration. The exhalation rate of type 1 is 1.2 10^{-5} Bq/(kg s) and type 2 4.7 10^{-5} Bq/(kg s). In combination with the ^{226}Ra concentration an emanating fraction of respectively 7.6% and 14% is obtained.

The ^{220}Rn (thoron) exhalation of the plates was determined by measuring the concentration of the decay products in a chamber of 1 m^3 with normal aerosol concentrations. The exhalation rate was found to be independent of the thickness of the plates, as expected from the short half-life of ^{220}Rn. Covering the entire surface of the plates with two layers of a common Latex paint decreased the thoron exhalation by a factor of 10 to 20.

The laboratory results for the radon and thoron exhalation were converted using realistic assumptions for a room. The contribution of phosphogypsum to the average radon concentration in a room is found to be about 1 Bq/m^3 for type 1 and 4 Bq/m^3 for type 2 resulting in an annual effective dose of the order of 0.1 mSv/year. The contribution to the effective dose from the thoron exhalation is much greater, namely, 1.8 mSv/year for type 1 and 0.9 mSv/year for type 2. Painting the gypsum lowers the thoron dose by a factor of 10 to 20 making the thoron dose comparable to that of radon.

INTRODUCTION

Gypsum is widely used in Belgium as a building material. The walls in most dwellings are covered with a layer of 1 to 2 cm of gypsum before they are painted or wallpapered. A considerable fraction of the gypsum applied in Belgian dwellings comes from the phosphate industry. The reaction of phosphate ore with sulfuric acid yields a variety of phosphate products and calcium-sulfate (gypsum). On average 4 kg of gypsum are produced for each kg of phosphoric acid (1). The phosphate ore has generally a high content of ^{238}U and/or ^{232}Th in radioactive equilibium with its daughter products. Most of this radioactivity will be found in the gypsum waste which is called phosphogypsum.

The paper consists of two parts. In the first part the radioactive properties of two types of phosphogypsum are determined. Results are given concerning the ^{226}Ra and ^{232}Th concentration, the radon (^{222}Rn) exhalation rate and the thoron (^{220}Rn) exhalation with and without painting. The radiological impact of using these types of phosphogypsum in home construction are discussed in the second part.

METHODS AND RESULTS

From each of the two types of phosphogypsum 60 plates with a surface of 0.295 x 0.210 m were made. Half of the plates was produced with a thickness of 0.5 cm and the other half with a thickness of 2 cm.

The ^{226}Ra and ^{232}Th concentrations of the two types of phosphogypsum were measured by means of high resolution gamma spectrometry. The detector of the measurement system is a 20% efficient germanium cristal with a resolution of 1.70 keV (fwhm). The detector is calibrated in the same geometry with a ^{134}Cs source traceable to NIST. The results for the two types are:

Type 1 : ^{226}Ra : 75 Bq/kg ^{232}Th : 230 Bq/kg

Type 2 : ^{226}Ra : 155 Bq/kg ^{232}Th : 160 Bq/kg

The radium concentrations are lower and the thorium concentrations are higher than those found in phosphogypsum from Moroccan origin (1).

The radon (^{222}Rn) exhalation rate was measured by closing some of the plates in an airtight barrel flushed previously with radon free air. One week or more later, the radon concentration in the barrel was determined by means of the Lucas technique. From the dimensions of the barrel, the weight of the sample and the radon concentration the exhalation rate was calculated:

Type 1 : $1.2\ 10^{-5}$ Bq/(kg s)

Type 2 : $4.7\ 10^{-5}$ Bq/(kg s)

In combination with the ^{226}Ra concentration an emanating fraction of 7.6% for type 1 and 14% for type 2 is obtained.

The thoron exhalation (^{220}Rn) of the plates was evaluated by means of its decay products because of the short half-life of thoron (55.6 s). The plates were brought into a 1 m^3 chamber which was ventilated continuously at a rate between 0.3 to 0.4 h^{-1} with outside air containing natural aerosol particles. The outside air was taken at a height of 10 m where the thoron and thoron decay product concentrations are very low. Each sample was measured 3 to 4 times. The reproducibility of the equilibrium equivalent thoron concentration was about 25% due to fluctuations in the aerosol concentration in the chamber. The equilibrium equivalent thoron concentration (EEC_{Th}) is calculated from the ^{212}Pb and ^{212}Bi concentrations in the following way:

$$EEC_{Th} = 0.91\ C\ (^{212}Pb) + 0.09\ C\ (^{212}Bi)$$

The results given in table 1 are normalised to an exhalation surface of 4 m^2 and corrected for the influence of humidity on the exhalation of the freshly produced gypsum plates.

TABLE 1. Equilibrium equivalent thoron concentration of the phosphogypsum plates with and without painting in the experimental chamber

Type	Thickness cm	Not painted EEC_{Th} Bq/m^3	Painted EEC_{Th} Bq/m^3	Ratio Not painted / Painted
1	0.5	35	2.2	16
1	2.0	25	2.3	11
2	0.5	14	0.7	20
2	2.0	12	1.0	12

The thoron exhalation is, as expected from the short half-life of ^{220}Rn, independed of the thickness of the samples. The exhalation of type 1 is about two times higher than that of type 2. This is more or less in line with the measured ^{232}Th concentrations.

The effect of covering the entire surface of the plates with two layers of a common Latex paint was also investigated. The set-up was identical to the measurements of the un-painted plates. The measured equilibrium equivalent thoron concentrations are shown in table 1. The reduction in thoron exhalation by painting the gypsum plates is a factor of 10 to 20.

DISCUSSION

In order to evaluate the radon and thoron exhalation of the phosphogypsum plates, the laboratory results were converted using realistic assumptions for a room:

- size of the room : 4 x 5 x 3 m (60 m^3);
- ventilation rate : 0.5 h^{-1} ;
- walls and ceiling covered with type 1 or type 2 phosphogypsum (74 m^2);
- residence time : 80%.

Covering the walls and the ceiling of the room with 1 cm of phosphogypsum increases the radon concentration of the room with 1 Bq/m^3 for type 1 and 4 Bq/m^3 for type 2 resulting in a contribution to the effective dose of the order of 0.1 mSv per year.

In the calculation of the effective dose from the thoron exhalation the conversion factor between the equilibrium equivalent thoron concentration and the effective dose from ICRP publication 50 (2) is taken: 3.9 10^{-5} mSv/(Bq h/m^3). The effective dose from the thoron exhalation of the uncovered phosphogypsum is much greater than the one from the radon exhalation, namely, 1.8 mSv/year for type 1 and 0.9 mSv/year for type 2. Painting the gypsum reduces the thoron exhalation by a factor of 10 to 20 making the thoron dose comparable to that of radon

REFERENCES

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