Radiation exposure from four different concrete mixtures

Measurement of activity concentrations, radon exhalation and dose assessment

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Summary

As part of the here presented work a total of four different concrete mixtures have been studied to determine the activity concentrations and the radon exhalation as well as an assessment of the radiation exposure when these concrete mixtures are used in regular dwellings. Experiments were carried out in compliance with the Dutch standards NEN 5697 and NEN 5699.

Based on these experimental findings, a dose assessment for both external and internal radiation exposure is performed. The results indicate an estimated external dose between 0.19 and 0.27 mSv per year and an internal dose between 0.62 and 0.68 mSv per year. Based on those estimates the maximum total dose for these concrete mixtures is about 0.91 mSv per year.

Introduction

Four different concrete mixtures have been studied to determine the activity concentrations and the radon exhalation as well as the radiological dose when these materials are used in regular dwellings.

Gamma-spectrometry measurements are performed to determine the radioactivity concentrations of the gamma-ray emitting radionuclides in three samples of each of the four concrete mixtures, according to the Dutch standard NEN 5697[1]. For each concrete mixture the received three samples were analysed together according to NEN 5697[1]. This analysis was performed in triplicate from which results the average ^{222}Rn exhalation rate was calculated.

Subsequently, a dose assessment for both internal and external radiation exposure is performed for each of the four concrete mixtures. For the internal dose assessment the radon and radon progeny concentrations are computed in a predefined dwelling, using advanced numerical calculation. For the external radiation exposure the methodology proposed by De Jong et al.[3] is used. Finally, the work is completed with a summary of the total exposure levels.

1 Experimental testing

1.1 Gamma-ray measurement

1.1.1 Principles of the method

The natural radioactivity concentrations are determined according to a standard method published under NEN 5697[1]. According to this method the density dependent photo peak efficiencies are determined for the gamma-ray energies 352 keV (^{214}Pb), 583 keV (^{208}Tl), 911 keV (^{228}Ac) and 1,461 keV (^{40}K). Four calibration standards are assembled with increasing densities. The materials used are stearic acid, starch, gypsum and quartz sand, homogeneously mixed with certified amounts of ^{238}U and ^{232}Th, in equilibrium with their daughter nuclides, and ^{40}K. The standards are placed into Marinelli beakers with a volume of about 0.7 litre, weighted and closed radon-tight. To obtain secular equilibrium, a waiting time of at least three weeks is taken into account before counting the samples. All samples are counted using an HPGe detector in a low-background facility. The samples of the material are analysed in an identical way as the calibration standards with respect to geometry, waiting time and radon-tightness of the beaker. The photo-peak efficiencies of the samples are deduced from the efficiency curves of the standard samples by interpolation. The results are expressed per unit of dry weight.

1.1.2 Sample pre-treatment

The concrete samples are crushed and placed into a Marinelli beaker. To reach a secular equilibrium a waiting time of at least three weeks is applied before the samples are counted in an HPGe detector.

1.1.3 Results

Table 1 shows the results from the gamma-ray measurements. The counting time of each sample is 57,600 s. The standard deviation refers to the counting statistics.

Table 1 Sample specifications, activity concentration and standard deviation expressed in Bq·kg^{-1}.

Sample code	Weight (kg)	Activity concentration (Bq·kg^{-1})							
		^{226}Ra		^{228}Th		^{228}Ra		^{40}K	
		x	SD	x	SD	x	SD	x	SD
2872-A	1.129	34	2	4.2	0.4	5	1	43	2
2872-B	1.119	33	3	5	1	5	1	42	4
2872-C	1.150	32	1	5.1	0.3	5.5	0.4	42	2
5850-A	1.135	39	3	15	1	15	1	43	5
5850-B	1.129	39	3	14	1	15	1	50	3
5850-C	1.138	40	3	15	1	16	1	48	3
5852-A	1.110	36	3	8	1	8	1	62	4
5852-B	1.113	35	1	8.4	0.4	8	1	58	2
5852-C	1.118	35	3	8	1	8	1	51	6
7699-A	1.146	36	3	8	1	8	1	54	2
7699-B	1.147	36	3	8	1	8	1	61	6
7699-C	1.127	37	3	7.4	0.4	8	1	53	3

1.2 Radon exhalation measurement

1.2.1 Principles of the method

The radon exhalation rate is determined according to the standard method NEN 5699[2]. In this method the samples are enclosed in a container, which is purged with nitrogen gas with a relative humidity of 50%. The three concrete prisms are placed in the container. The exhaled radon, carried along with the nitrogen gas, is trapped on silica gel at -190ºC. After sufficient radon has been trapped, the nitrogen stream is stopped and the trapping agent is analysed by liquid scintillation counting. The results are expressed as mass exhalation rates by normalizing the amount of exhaled radon per kg material.

1.2.2 Sample pre-treatment

The test samples for the ^{222}Rn exhalation rate measurements are conditioned at a temperature of 20ºC and a relative humidity of 50%.

1.2.3 Results

The results from the radon exhalation measurements are given in Table 2. The standard deviation (SD) in this table refers to a triplicate analysis of the same test sample.

Table 2 Sample specifications and exhalation rate expressed as $\mu Bq \cdot s^{-1}$ and $\mu Bq \cdot kg^{-1} \cdot s^{-1}$.

Sample code		**Exhalation rate**				
	Weight	**($\mu Bq \cdot s^{-1}$)**		**($\mu Bq \cdot kg^{-1} \cdot s^{-1}$)**		**(%)**
	(kg)	**x**	**SD**	**x**	**SD**	**SD**
2872	14.58	107	6	7.3	0.4	5
5850	14.39	121	14	8	1	12
5852	14.39	104	9	7	1	9
7699	14.55	121	14	8	1	11

2 Radon simulation and boundary conditions

For the computation of the radon concentrations in the room a three dimensional numerical based software is used. For this purpose the details of the room are specified and the radon source terms for all four concrete mixtures are determined.

2.1 Geometry and boundary conditions

Based on information provided by the customer the following conditions of the room have been agreed:

2.1.1 Geometry and material features of the room

Dimensions	: length 3 m; width 3 m; height 2.7 m
Inner area	: 50.4 m^2
Volume	: 24.3 m^3
Window	: 1.2 x 1.2 m^2 (1.4 m^2)
Door	: 2.0 x 0.8 m^2 (1.6 m^2)
Area concrete	: 47.4 m^2
Concrete thickness	: 0.2 m
Concrete density	: 2300 $kg\cdot m^{-3}$
Furniture	: Unfurnished room

2.1.2 Flow conditions and ^{222}Rn background

Air exchange rate	: 0.5 h^{-1}
^{222}Rn background concentration	: 10 $Bq\cdot m^{-3}$
^{222}Rn background equilibrium factor	: 0.4

2.2 ^{222}Rn source

In building materials the diffusion of ^{222}Rn is often described as a one-dimensional process, i.e. directed to the living space and its opposite side. The ^{222}Rn source for the considered room follows from the multiplication of the exhalation rates by the total area in the room taken by concrete and the surface density, the product of the thickness of the building parts and their density. In formula:

$$S = E_m \; A \, \rho \, L \tag{1}$$

In this equation S represents the ^{222}Rn source in the room ($Bq{\cdot}s^{-1}$); E_m the ^{222}Rn exhalation rate ($Bq{\cdot}kg^{-1}{\cdot}s^{-1}$); A the area in the room taken by concrete (m^2); ρ the density of concrete ($kg{\cdot}m^{-3}$); and L the half-thickness of the construction parts (m).

This method, however, has to be considered as a maximum. The exhalation from the test specimen takes place from all sides instead of one-dimensional as in a practical situation. This may lead to an overestimation of the ^{222}Rn source in the room under investigation. Berkvens et al.[4] have introduced a formular to calculate the one-dimensional exhalation rate from the experimentally determined three-dimensional results. Therefore, they defined an equivalent sample geometry with a reduced thickness to compensate for the ^{222}Rn diffusion to the surfaces other than the front and backside. With the x-axis directed towards the room, the shortest mean distance is given by:

$$\frac{L'_x}{2} = \frac{L_x}{2} - \frac{1}{6}\left(\frac{L_x{}^2}{L_y} + \frac{L_x{}^2}{L_z} - \frac{1}{2}\frac{L_x{}^3}{L_y L_z}\right) \tag{2}$$

In this expression L_x, L_y and L_z represent half of the sample thickness of the test specimen in the x-, y- and z-direction. As the dimensions of the present test samples are 0.10 x 0.10 x 0.20 m^3, L'_x is calculated at 0.33 m. Subsequently the exhalation rate from a wall in practice is calculated from the measured exhalation rate according to:

$$\frac{E_m^{(1)}}{E_m} = \frac{L'_x}{L_x} \frac{\tanh\,(L_x/l)}{\tanh\,(L'_x/l)} \tag{3}$$

$E_m^{(1)}$ is this equation is the one-dimensional exhalation rate; E_m the experimental determined exhalation rate; and l the diffusion length of ^{222}Rn in concrete. As reported by Kovler et al.[5] the literature data on the diffusion length in concrete ranges from 0.06 m for heavy concretes to 0.30 m for light-weight species. If for the present concretes a diffusion coefficient of 0.10 m is assumed, a correction factor of 0.79 can be calculated from equation (3). With that the ^{222}Rn source for the four concrete mixtures is shown in the table below.

Table 3 Radon source in Bq/s for the four different concrete mixtures.

Sample code	Radon source (mBq·s^{-1})
2872	64
5850	72
5852	62
7699	72

2.3 Radon and radon progeny modelling

The computation of the ^{222}Rn concentration and the concentrations of the short-lived ^{222}Rn progeny nuclides ^{218}Po, ^{214}Pb and ^{214}Bi follows the method as described by De With and De Jong[6]. In this work, a Computational Fluid Dynamics (CFD) model is used to simulate the concentration of radon and radon progeny products in a typical Dutch living room. The dispersion is computed using the fundamental flow equations for gas and aerosols, which enables detailed simulation of the three-dimensional flow structures from ventilation and buoyancy. Extra algorithms are developed and coupled with the CFD model to take account of all relevant physical processes. These include the formation and attachment of the progeny products to aerosol particles as well as their dispersion and deposition.

The computed concentrations for radon and its progeny are as follows:

Table 4 Concentrations radon and radon progeny.

Sample code	^{222}Rn (Bq·m^{-3})	^{218}Po (Bq·m^{-3})	^{214}Pb (Bq·m^{-3})	^{214}Bi (Bq·m^{-3})
2872	29.0	24.7	10.4	5.4
5850	31.7	26.9	11.3	5.8
5852	28.5	24.2	10.3	5.3
7699	31.6	26.8	11.3	5.8

These concentration levels also include the radon background concentration of 10 Bq·m^{-3} and its background progeny.

3 Dose assessment

3.1 Calculation of the external dose

The external dose due to gamma radiation from the building materials in the room is calculated according to the method described by De Jong et al.[3,7]. The standard geometry as defined by Koblinger[8] is taken as starting point for our calculation model. This geometry has dimensions of 5 x 4 m^2 and 2.8 m in height, with each construction part (i.e. floor, walls and ceiling) made of 20 cm thick concrete and no doors or windows. Correction factors have been deduced for alternative situations. The absorbed dose rate in air (unit: $Gy \cdot h^{-1}$) in a particular room is than calculated according to:

$$\dot{D}_{air} = \left\{ \sum_{i=1}^{6} \left[F_{dose} \cdot F_1 \cdot F_2 \cdot F_3 \cdot \cdots \cdot F_n \right]_i \right\} F_{zoning} \cdot F_{adjac} \tag{5}$$

in which i is the index for a construction part, F_1 to F_n are the correction factors for construction part i, F_{zoning} is a correction factor which takes internal zoning of the construction into account, and F_{adjac} is the contribution from adjacent floors and dwellings. F_{dose} is the so-called dose factor, defined as:

$$F_{dose,i} = k_1 a_{1,i} + k_2 a_{2,i} + k_3 a_{3,i} \tag{6}$$

In this equation k_1, k_2 and k_3 represent the specific absorbed dose rates, defined as the absorbed dose rate in air due to an activity concentration of 1 $Bq \cdot kg^{-1}$ of each of the primordial radionuclides in equilibrium with its decay products ($Gy \cdot h^{-1}$ per $Bq \cdot kg^{-1}$); and $a_{1,i}$ to $a_{3,i}$ the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K of construction part i ($Bq \cdot kg^{-1}$), respectively. The values of the specific absorbed dose rate depend amongst others on the thickness, density and dimensions of the various construction parts.

Several researchers have determined the specific absorbed dose rate for the standard Koblinger-construction, using various codes. Averaged over all data the specific absorbed dose rates are[9]:

^{226}Ra 0.90 $nGy \cdot h^{-1}$ per $Bq \cdot kg^{-1}$
^{232}Th 1.10 $nGy \cdot h^{-1}$ per $Bq \cdot kg^{-1}$
^{40}K 0.08 $nGy \cdot h^{-1}$ per $Bq \cdot kg^{-1}$

For the calculation of the annual indoor effective dose, the absorbed dose is determined by equation (5) and is multiplied with a conversion factor of 0.7 $Sv \cdot Gy^{-1}$. The annual number of hours spent indoors is taken as 7000 (80% of the total time). The mean activity concentrations for the four concrete mixtures are as follows:

Table 5 Activity concentrations.

Sample code	^{226}Ra (Bq·kg^{-1})	^{232}Th (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})
2872	33	5	42
5850	39	15	47
5852	35	8	57
7699	36	8	56

It is important to stress that the above ^{232}Th concentration is based on the average concentration of ^{228}Th and ^{228}Ra. Based on those experimental results the annual external dose is as follows:

Table 6 Annual external dose from primordial radionuclides.

Sample code	External dose (mSv·a^{-1})
2872	0.19
5850	0.27
5852	0.22
7699	0.22

3.2 Calculation of the internal dose

The effective dose to residents due to inhalation of the short-lived ^{222}Rn progeny is based on the so-called equilibrium equivalent ^{222}Rn concentration. This concentration is calculated according to the following expression:

$$C_{eq}^{222} = 0.105\, C_1 + 0.515\, C_2 + 0.380\, C_3 \qquad (4)$$

where C_1, C_2 and C_3 are the activity concentrations in the indoor environment of ^{218}Po, ^{214}Pb and ^{214}Bi (Bq·m^{-3}), respectively. These concentrations are obtained using the CFD model computations. The constants in the equation are the relative contributions of each decay product to the total potential alpha energy from the decay of a unit ^{222}Rn gas. To convert the equilibrium equivalent concentration into an effective dose rate, a conversion factor of 9 nSv·h^{-1} per Bq·m^{-3} is applied[10]. The time spent indoors is taken as 7000 h per year (80% of the total time).

Based on the findings reported in Table 4, the above conversion factor and the hours spent indoors the internal dose is as follows:

Table 7 Annual internal dose from radon exposure.

Sample code	**Internal dose (mSv·a^{-1})**
2872	0.63
5850	0.68
5852	0.62
7699	0.68

3.3 Calculation of the total dose

Based on the calculated internal and external dose as presented in Table 6 and Table 7 the total annual dose is:

Table 8 Annual total dose from primordial radionuclides.

Sample code	**Total dose (mSv·a^{-1})**
2872	0.82
5850	0.96
5852	0.84
7699	0.91

4 Conclusions

The findings of this work are as follows:

1. The activity concentrations and radon exhalation have been experimentally investigated for four different concrete mixtures.
2. The results indicate that the ^{226}Ra concentration is between 32 and 40 $Bg \cdot kg^{-1}$, and broadly similar for all concrete mixtures. The ^{228}Th and ^{228}Ra concentration varies between 4.2 and 16 $Bg \cdot kg^{-1}$, while the concentration ^{40}K varies between 42 and 62 $Bg \cdot kg^{-1}$.
3. The radon exhalation varies between 7.3 and 8 $\mu Bq \cdot kg^{-1} \cdot s^{-1}$.
4. Based on computational modelling the radon concentration for a room of 3×3×2.7 m^3 and an air exchange rate of 0.5 h^{-1} is estimated between 29.0 and 31.6 $Bq \cdot m^{-3}$. This includes a background concentration of 10 $Bq \cdot m^{-3}$.
5. Computation of the external and internal dose for those concrete mixtures results in an estimated yearly external dose of up to 0.27 mSv and an internal dose of up to 0.68 mSv.

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