NRG

contact
P. de Jong
P.O.Box 9034
6800 ES Arnhem
The Netherlands

phone
+31 26 356 85 69

fax
+31 26 356 85 39

e-mail
p.dejong@nrg.eu

Dr. Omri Lulav
National Coal Ash Board
20 Lincoln street
TEL AVIV 67134
ISRAEL

Arnhem, 20 October 2010

our reference : K5098/10.104560 RE/PdJ/VL
your reference :

subject : Analysis of radon exhalation rate and activity concentrations

Dear Mr. Lulav,

Please find attached the results of the analysis of the radon exhalation rate and the activity concentrations of six concrete mixtures.

If you have any questions on the report, please do not hesitate to contact me.

Sincerely yours,

Peter de Jong
NRG Radiation & Environment

Annex: 1

NRG Petten
T +31 (0)224 56 4950
F +31 (0)224 56 8912
Westerduinweg 3
P.O. Box 25
1755 ZG Petten
The Netherlands

NRG Arnhem
T +31 (0)26 356 8524
F +31 (0)26 356 8536
Utrechtseweg 310
P.O. Box 9034
6800 ES Arnhem
The Netherlands

Trade register
37082135

www.nrg.eu
info@nrg.eu

ANNEX: RESULTS ON RADIOLOGICAL ANALYSIS

1 Test samples

On August 17, 2010 the following concrete test samples were received:

a. Three test samples sized 10x10x20 cm^3 coded AP-4 to AP-6
b. Three test samples sized 10x10x20 cm^3 coded BP-10 to BP-12
c. Three test samples sized 10x10x10 cm^3 coded AQ-1 to AQ-3
d. Three test samples sized 10x10x10 cm^3 coded BQ-7 to BQ-9

The test samples mentioned under a. and b. were analysed together to determine the radon exhalation rate. The samples under c. and d. were analysed individually for gamma-ray emitting radionuclides.

2 Sample pre-treatment

The test samples for the ^{222}Rn exhalation rate measurements were conditioned at a temperature of 20 ± 2 °C and a relative humidity of 50 ± 5%. According to the applied standard method the conditioning is continued until the decrease in moisture content of the material is less than 0.07% measured over a period of seven days. Prior to the gamma-ray measurements, the samples are broken to pieces with a size smaller than 0.2 cm.

3 Applied methods

The radon exhalation rate was determined according to the standard method NEN 5699 [1]. In this method the samples are enclosed in a container that is purged with nitrogen gas with a relative humidity of 50%. The exhaled radon, carried along with the nitrogen gas, is trapped on silica gel at -190 °C. After sufficient radon has been trapped, the nitrogen stream is stopped and the trapping agent analysed by liquid scintillation counting. The results are expressed as mass exhalation rates by normalizing the amount of exhaled radon per kg material.
The natural radioactivity concentrations are determined according to a standard method published as NEN 5697 [2]. 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 1 litre, weighed, 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 a HPGe detector in a low-background facility. The samples of broken building 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.

The ^{222}Rn release factor of a sample, F_{Rn}, is defined as the ratio between the amount of ^{222}Rn gas released to the environment and the amount that is generated within the material. In formula:

$$F_{Rn} = \frac{E}{\lambda_{Rn}\, a_1} 100\%,$$

where E is the ^{222}Rn exhalation rate in Bq/kg/s, λ_{Rn} is the decay constant of ^{222}Rn (2.1×10^{-6} s^{-1}) and a_1 is the activity concentration of ^{226}Ra (Bq/kg). This factor is calculated for each of the test series.

4 Results

The results of the radon measurements are given in Table 1. The standard deviation in this table refers to a triplicate analysis of the same test sample.

Table 1: Sample weight and exhalation rate expressed as μBq/s and μBq/kg/s.

Samples	Weight (kg)	Exhalation rate				
		(μBq/s)		(μBq/kg/s)		
		x	SD	x	SD	SD%
AP-4 to AP-6	14.20	149	3	10.5	0.2	2.2
BP-1 to BP-12	13.58	74	6	5.4	0.5	8.3

Table 2 presents the results of the gamma-ray analysis. The counting time of each sample was 70,000 s. The standard deviation refers to counting statistics.

Sample	Weight (kg)	Activity concentration (Bq/kg)							
		^{226}Ra		^{228}Th		^{228}Ra		^{40}K	
		x	SD	x	SD	x	SD	x	SD
AQ-1	1.669	29.5	0.2	9.2	0.2	8.7	0.2	60.2	1.0
AQ-2	1.667	29.6	0.2	9.0	0.2	9.6	0.3	57.1	0.9
AQ-3	1.643	29.2	0.2	7.6	0.2	7.7	0.2	50.9	0.8
BQ-7	1.586	38.3	0.3	17.8	0.3	18.6	0.4	64.8	1.0
BQ-8	1.634	38.9	0.3	18.6	0.3	17.6	0.3	57.1	0.9
BQ-9	1.605	36.6	0.3	18.0	0.3	18.1	0.3	60.7	1.0

From the three separate analyses the average ^{226}Ra concentration is calculated at 29.4 ± 0.2 Bq/kg for the A series of samples and 37.9 ± 1.2 Bq/kg for the B series.

The radon release factor, calculated according to the above mentioned formula is then calculated at:

A series: (17.0 ± 0.4) %
B series: (6.8 ± 0.6) %

References

[1] Radioactivity measurements – Determination method of the rate of the radon exhalation of dense building materials. NEN 5699(en). Nederlands Normalisatie-instituut, Delft, the Netherlands.

[2] Radioactivity measurements – Determination of the natural radioactivity in stony building materials by means of semiconductor gamma ray spectrometry. NEN 5697(en). Nederlands Normalisatie-instituut, Delft, the Netherlands.

Technical note

From	:	Govert de With	(NRG)
To	:	Omri Lulav / Kosta Kovler	(NCAB)
Subject	:	Dose assessment from radon progeny in a concrete based room using CFD calculation	
Datum	:	17-1-2012	
Referentie	:	912559/12.112181 RE/GdW/VL	

1 Radon simulation and boundary conditions

Geometry, material and ventilation conditions
Based on information provided by the customer the following conditions of the room have been agreed:

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

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

^{222}Rn source
According to exhalation measurements carried out by NRG[1] the ^{222}Rn exhalation rate is determined as $4.8 \cdot 10^{-6}$ and $6.8 \cdot 10^{-6}$ $Bq \cdot kg^{-1} \cdot s^{-1}$ for the concrete samples with and without fly ash, respectively. 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). In this way the ^{222}Rn source in the room is calculated at $7.4 \cdot 10^{-2}$ $Bq \cdot s^{-1}$ for the room constructed with concrete to which no fly ash is added and $5.2 \cdot 10^{-2}$ $Bq \cdot s^{-1}$ for the concrete with fly ash.

These values, however, have to 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.[2] have introduced a formalism to calculate the one-dimensional exhalation rate from the experimentally determined results. Therefore, they defined an equivalent sample geometry with a reduced thickness to compensate for the ^{222}Rn diffusion to the planes other than the front and backside. With the x-axis directed towards the room, the shortest mean distance is given by:

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

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:

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

$E_m^{(1)}$ 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.[3] 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 in the considered room comes to $5.9 \cdot 10^{-2}$ Bq·s^{-1} (no fly ash added) and $4.1 \cdot 10^{-2}$ Bq·s^{-1} (with fly ash), respectively. Since these values are supposed to be more realistic, these are applied in the model calculations.

Radon and radon progeny modelling

The modelling 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 De With and De Jong[4]. In this work, a Computational Fluid Dynamics (CFD) model is used to simulate the concentration of thoron and thoron 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 1 Concentrations radon and radon progeny.

	^{222}Rn (Bq·m^{-3})	^{218}Po (Bq·m^{-3})	^{214}Pb (Bq·m^{-3})	^{214}Bi (Bq·m^{-3})
Concrete without fly ash	14.3	12.3	5.6	3.0
Concrete with fly ash	13.0	11.3	5.2	2.8

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

2 Dose assessment

Calculation of 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:

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

in which C_1, C_2 and C_3 are the activity concentrations in the indoor environment of ^{218}Po, ^{214}Pb and ^{214}Bi ($Bq \cdot m^{-3}$), respectively, as found by the 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 $nSv \cdot h^{-1}$ per $Bq \cdot m^{-3}$ is applied[5]. The time spent indoors is taken as 7000 h per year (80% of the total time).

Based on the findings reported in Table 1, the above conversion factor and the hours spent indoors the internal dose is as follows:

Table 2 Annual internal dose from radon exposure.

	Internal dose ($mSv \cdot a^{-1}$)
Concrete without fly ash	0.34
Concrete with fly ash	0.31

Calculation of 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.[6,7]. 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:

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

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:

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

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 to the residents due to external radiation the absorbed dose as found from equation (5) is multiplied by a conversion factor of 0.7 $Sv \cdot Gy^{-1}$ and the annual number of hours spent indoors, taken as 7000. The required activities for each of the three radionuclides are based on measurements performed by NRG[1]. The mean activity concentrations for both types of concrete are as follows:

Table 3 Activity concentrations.

	^{226}Ra ($Bq \cdot kg^{-1}$)	^{232}Th ($Bq \cdot kg^{-1}$)	^{40}K ($Bq \cdot kg^{-1}$)
Concrete without fly ash	29.4	8.6	56.1
Concrete with fly ash	37.9	18.1	60.9

Based on those experimental results the annual external dose for is as follows:

Table 4 Annual external dose from primordial radionuclides.

	Internal dose ($mSv \cdot a^{-1}$)
Concrete without fly ash	0.20
Concrete with fly ash	0.29

Calculation of total dose

Based on the calculated internal and external dose as presented in Table 2 and Table 4 the total annual dose per year is:

Table 5 Annual total dose from primordial radionuclides.

	Total dose ($mSv \cdot a^{-1}$)
Concrete without fly ash	0.54
Concrete with fly ash	0.60

3 References

[1] De Jong, P. (2010)
Analysis of radon exhalation rate and activity concentrations. Letter K5098/10.104560 RE/PdJ/VL, Nuclear Research and consultancy Group.

[2] Berkvens, P., Kerkhove, E. and Vanmarcke, H. (1988)
Three-dimensional treatment of steady-state ^{222}Rn diffusion in building materials: introducing a practical modified one-dimensional approach. Health Phys. 55:793-799.

[3] Kovler, K., Perevalov, A., Steiner, V. and Rabkin, E. (2004)
Determination of the radon diffusion length in building materials using electrets and activated carbon. Health Phys. 86:505-516.

[4] De With G. and De Jong P. (2011).
CFD modelling of thoron and thoron progeny in the indoor environment. Rad. Prot. Dosim., 145 (2-3): 127-132.

[5] UNSCEAR (2000).
Sources and effects of ionizing radiation. UNSCEAR 2000 report to the General Assembly, with scientific annexes. Volume 1: Sources. United Nations Scientific Committee on the Effects of Atomic Radiations. United Nations, New York.

[6] De Jong, P. and Van Dijk, W. (2008)
Modeling gamma radiation in dwellings due to building materials. Health Phys. 94: 33-42.

[7] De Jong, P. and Van Dijk, J.W.E. (2008)
Calculation of the indoor gamma dose rate distribution due to building materials in the Netherlands. Radiat. Prot. Dosim. 132:381-389.

[8] Koblinger, L. (1978)
Calculation of exposure rates from gamma sources in walls of dwelling rooms. Health Phys. 34: 459-463.

[9] De Jong, P. (2010)
PhD Thesis Exposure to natural radioactivity in the Netherlands: the impact of building materials, Arnhem, The Netherlands.