

## **Moisture dependence of radon transport in concrete: measurements and modelling**

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### **Abstract**

The moisture dependence of the radon-release rate of concrete was measured under well controlled conditions. It was found that the radon-release rate almost linearly increases upto moisture contents of 50 to 60%. At 70 to 80% a maximum was found and for higher moisture contents the radon-release rate decreases very steeply. It is demonstrated that this dependence can be succesfully modelled on basis of the multi-phase radon-transport equation in which values for various input parameters (porosity, diffusion coefficient, emanation factor, etc.) were obtained from independent measurements. Furthermore, a concrete structure development model was used to predict at any moment in time the values of input parameters that depend on the evolution of the concrete microstructure. Information on the concrete manufacturing recipe and curing conditions (temperature, relative humidity) was used as input for the concrete structure model. The combined radon transport and concrete structure model supplied sufficient information to assess the influence of relative humidity on the radon source and barrier aspects of concrete.



## Introduction

As in most instances radon originating from the soil makes up the largest fraction of the radon concentration indoors, a fair amount of research has been undertaken to construct and validate transport models that describe radon generation and migration in soils.

Rogers and Nielson (1991b) developed a very general description for radon generation and transport in a porous medium in which both diffusion and advection were considered. Furthermore, dynamical transfer of radon between three phases (air, liquid, and adsorbed to the solid phase) is modelled and condensed in a single equation with the radon concentration in the air-filled pore space as the observable parameter. This model has been extensively tested in the laboratory for sand (as a simple porous medium) in both dry, and moistened conditions; in a cylindrical sand column (Van der Graaf et al. 1992; Van der Spoel et al. 1997, 1998a, 1998b, 1999) and in a rectangular soil chamber (Mosley et al. 1996). In these laboratory tests a good correspondence between measurements and modelling is observed. It should be remarked that application of similar models to *in situ* situations like real houses (Nielsen et al. 1994) or small test structures (Garbesi et al. 1993; Andersen et al. 1994) often results in less favourable correspondence. Nielsen et al. (1994) found deviations of a factor of two in comparing with real house data. Garbesi et al. (1993) and Andersen et al. (1994) reported comparable differences between experiments and model calculations for small test structures. These discrepancies were attributed to incomplete or inaccurate knowledge of the input parameters, especially house data, air permeability, and moisture content of the soil.

In most countries building materials are, compared to soil, a relatively small source of radon. Notable exceptions are Italy (Bochicchio et al. 1996; Risica et al. 2001) and the Netherlands (Lembrechts et al. 2001). In the latter country, in new dwellings, radon from building materials constitutes 70% of the average radon concentration ( $28 \text{ Bq m}^{-3}$ ). In the Netherlands concrete provides the largest source of radon amongst the five types (concrete, aerated concrete, brick, gypsum, sandy lime-stone) of normally used stony building materials (De Jong and Van Gellecum 2001). In most other countries concrete is seen as a useful barrier against soil gas radon e.g., in the case of a concrete ground floor slab (Gadd and Borak 1995; Renken and Rosenberg 1995; Klink et al. 1999; Keller et al. 2001).

Both the radon generation and transport in concrete will depend on the amount of moisture inside the pores. Radon generation depends on moisture through the moisture dependence of the radon emanation factor. Radon transport is affected because both, the diffusion coefficient and the air permeability are likely to decrease with increasing moisture filling of the pores. This implies that both radon source strength and the barrier function of concrete are moisture dependent.

In this paper the moisture dependence of radon release from concrete test cubes is measured. The results are then taken as a starting point to describe radon generation and (diffusive) transport in concrete using a similar modelling approach as has been done previously for soil. After validation, this model is applied to study the effect of changing moisture contents on the radon source and barrier aspects of concrete.

## Model description

The model for radon transport is based on the multi-phase radon transport equation (Rogers and Nielson 1991b; Van der Spoel, 1998). Under the experimental conditions of our radon release experiments (steady-state, no pressure differences) this equation (taking the radon concentration in air as a reference) simplifies to,

$$\nabla \cdot (D\nabla C) - \beta\lambda C + S = 0, \quad (1)$$

where

- $D$  = the bulk radon diffusion coefficient ( $\text{m}^2 \text{s}^{-1}$ );
- $C$  = the radon concentration in the air-filled pore space ( $\text{Bq m}^{-3}$ );
- $\beta$  = the partition-corrected porosity;
- $\lambda$  = the radon decay constant ( $2.1 \times 10^{-6} \text{ s}^{-1}$ ); and
- $S$  = the radon production rate per unit bulk volume ( $\text{Bq m}^{-3} \text{ s}^{-1}$ ).

The partition-corrected porosity  $\beta$  and the bulk radon production rate  $S$  are given by,

$$\beta = \epsilon(1 - m + Lm) + \rho_b k_a, \quad (2)$$

and

$$S = \eta \rho_b \lambda C_{Ra}, \quad (3)$$

with

- $\epsilon$  = the porosity of concrete;
- $m$  = the fraction of water saturation of the pores ( $\text{m}^3 \text{m}^{-3}$ );
- $L$  = the Ostwald coefficient (0.26 at 293 K);
- $\rho_b$  = the bulk dry density of concrete ( $\text{kg m}^{-3}$ );
- $k_a$  = the radon surface-adsorption coefficient ( $\text{m}^3 \text{kg}^{-1}$ );
- $\eta$  = the radon emanation factor; and
- $C_{Ra}$  = the radium ( $^{226}\text{Ra}$ ) content on basis of dry mass ( $\text{Bq kg}^{-1}$ ).

Eqn (??) defines the partition-corrected porosity (a term introduced by Andersen (1992)) that incorporates the distribution of radon between the three phases (air, water and adsorbed to the solid surface). This approach assumes that the Ostwald coefficient  $L$  (ratio of the radon concentrations in the water- and air phase) and the surface-adsorption coefficient  $k_a$  (ratio of concentrations in air and adsorbed to the solid surface) are constant. This corresponds to an equilibrium assumption and is generally used in studies of radon transport in soil. A discussion about its validity for soil can be found in Van der Spoel (1999) and Nazaroff (1992). The equilibrium assumption is satisfied if typical time scales involved in transfer between phases are small with respect to other transport processes. As the average pore diameter in concrete is considerably smaller than in soils, typical transport paths for exchange between phases are correspondingly shorter. Consequently, time scales for transport between phases will be smaller for concrete than for soil and thus the equilibrium assumption will also be better satisfied for concrete than for soil.

Eqn (??) has been solved numerically (control-volume method, Patankar (1980)) for the radon concentration  $C$  using a grid of 30x30x30 control volumes for a 15x15x15 cm<sup>3</sup> cube with the NGD-KVI radon-transport code (for more details about this code we refer to Van der Spoel (1998) and Van der Spoel et al. (1998a)).

In the radon-release rate experiments, zero radon concentration around the concrete test cubes was enforced and, correspondingly, in solving eqn (??), boundary conditions of a zero concentration at the concrete surfaces were applied. The radon-release rate  $R$  (Bq s<sup>-1</sup>) was then deduced from the calculated concentration profiles in the cube by using the fact that the amount of radon released per second from the surfaces of the cube is the difference of the amount produced into the pore volume and the amount that decays in the pore volume per unit time. This radon-transport model (1D and 2D versions) and its implementation have been extensively validated for both steady state and transient situations against experiments with dry and wet sand (Van der Spoel et al. 1997, 1998a, 1999) and against analytical solutions (Van der Spoel 1998) and other numerical codes (Andersen et al. 1999).

## Material Properties and Experimental Techniques

### Concrete test cubes

The concrete test cubes (side: 15 cm) were provided by the concrete laboratory of a Dutch cement manufacturer<sup>1</sup> and were made according to a standard recipe. On mass basis, the composition of the cubes was, blast furnace cement: 14.4%; sand: 28.4%, gravel: 50.5% and water: 6.7%. A total of 15 cubes was produced. All cubes were conditioned under water for 21 days. Thereafter six cubes were used for strength tests by the concrete laboratory (measured strengths approximately 50 N mm<sup>-2</sup>, typical for this type of concrete), and nine were used in this research. These nine cubes were allowed to cure for at least six months before being used in the experiments. The initial masses (before curing) of the nine cubes ranged between 8.1 and 8.2 kg with a mean value of 8.14±0.06 kg. The detailed composition of a cube is given in Table ?? and is considered as being representative for the most widely used concrete in the Dutch building practice.

### Radon-release rate measurements

Radon-release rates ( $R$  in Bq s<sup>-1</sup>), were measured, under controlled conditions, with the NGD-KVI radon-release rate setup (Cozmuta and Van der Graaf 2001) consisting of a stainless-steel box (80 litres) inside which a test sample (BM) is placed (Fig. ??). A nitrogen flow of 400 ml min<sup>-1</sup> passes through this box. This flow is conditioned to a preset relative humidity by combining a flow wetted by a humidifier (H) and a dry flow. Flow controllers (FC) regulate both flows and a relative humidity sensor (RH<sub>1</sub>) monitors the combined flow. The flow is then split and directed to two inlets, in the upper and lower part of the box, to provide a uniform and homogeneous flow pattern. The nitrogen flow leaves the box through two outlets that join each other before passing to a flow meter (FM) and a second relative humidity sensor (RH<sub>2</sub>) yielding information about a possible change in humidity of the sample. Supplementary, a sensor (T) inserted via the

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<sup>1</sup>ENCI, Eerste Nederlandse Cement Industry, IJmuiden, The Netherlands.

Table 1: *Detailed composition of a concrete cube.*

Component	Type	Size (mm)	Mass (kg)	Fraction (%)
Cement	Blast furnace	<0.063	1.17	14.4
Sand	Fine	0.1-0.3	0.26	3.2
	Medium	0.2-0.5	0.58	7.1
	Coarse	0.5-1.0	0.96	11.8
	Very coarse	1.0-1.8	0.52	6.4
	Gravel	Granules	2.0-4.0	0.45
Gravel	Pebbles I	4.0-8.0	0.83	10.2
	Pebbles II	8-16	1.35	16.6
	Pebbles III	16-25	1.47	18.1
Water	water/cement=0.48		0.56	6.7
Total			8.15	100

lid monitors the temperature variations during the measurement. The pressure inside the setup is kept constant to a preset value of 1060 Pa by a feedback system (PCU) to prevent fluctuations due to atmospheric pressure variations.

Figure 1: *Schematic presentation of the NGD-KVI radon-release setup. FC) flow controller; H) humidifier; RH<sub>1,2,3</sub>) relative humidity sensors; T) temperature sensor; FM) flow meter; BM) building material; D<sub>1</sub>) pre-dryer (silica gel); D<sub>2</sub>) dryer (water trap); ACC) activated charcoal; LN) liquid nitrogen; V) valve; PCU) pressure control unit; DP) differential pressure transducer.*

After being pre-dried in a column of silicagel (D1) and freeze-dried via a vial (D2) cooled to liquid-nitrogen temperature (LN), the flow passes through a second vial that contains a column of 17.1 g of activated charcoal (ACC). The radon exhaled by the sample and transported with the nitrogen flow is adsorbed by the charcoal (kept at liquid-nitrogen temperature). The exhalation volume with sample is pre-flushed for at least 17 hours before the adsorber is connected to obtain a steady-state condition. After a certain adsorption time  $t_a$  ( $s^{-1}$ ) the charcoal is transferred into a cylindrical pill box (diameter 50 mm) and placed in a calibrated geometry on one of the NGD-KVI

low-background high-purity germanium spectrometers<sup>2</sup> for a gamma-ray measurement. The count rate is determined in the energy range of 50 keV to the upper energy limit of the spectrometer. The spectrometers have a higher energy resolution than actually needed, spectroscopic information was used to ensure that only radon (<sup>222</sup>Rn) was adsorbed (and not thoron, <sup>220</sup>Rn). The count rate due to adsorbed radon originating from the test sample,  $N_{bm}$  (s<sup>-1</sup>), is related to the total count rate,  $N_{tot}$  (s<sup>-1</sup>), in the energy range of the spectrometer. Corrections for the contribution of the charcoal,  $N_{bg}$  (s<sup>-1</sup>), for radon decay during the measurement time,  $t_m$ , of the gamma-ray determination, and for the time interval between the end of the adsorption and the beginning of the gamma-measurement (waiting time  $t_w$ ), are applied. The contribution of the charcoal was determined from a spectrum recorded with a standard amount of activated charcoal in the same geometry. The waiting time  $t_w$  was always longer than three hours to approximate equilibrium between radon and its short-lived decay products. The radon-release rate  $R$  (Bq s<sup>-1</sup>), of the test sample is then calculated from,

$$R = \frac{N_{bm}\lambda(\lambda + \lambda_v)}{\omega\lambda_v(1 - \exp(-\lambda t_a))} - R_{setup} \quad (4)$$

where,

$$N_{bm} = (N_{tot} - N_{bg}) \frac{\lambda t_m \exp(\lambda t_w)}{1 - \exp(-\lambda t_m)} \quad (5)$$

and

- $\lambda_v$  = the ventilation rate (s<sup>-1</sup>) of the setup (ratio between the flow rate and the volume of the box);
- $\omega$  = the efficiency of the method, the product of the fraction of radon adsorbed on the charcoal and the efficiency of the gamma detection; and
- $R_{setup}$  = the contribution (Bq s<sup>-1</sup>) of silica gel in the pre-dryer of the setup. This contribution  $(2.9 \pm 0.4) \times 10^{-6}$  Bq s<sup>-1</sup> is small compared to release rates of concrete test cubes.

The efficiencies of the spectrometers were determined by conducting release rate experiments with a 20.2 kBq certified radon-emanating source<sup>3</sup>. Typically, these efficiencies are in the range 0.16 to 0.26.

This method to determine radon-release rates conforms to the Dutch standard for these measurements (NEN 2001). The method as it is implemented at NGD-KVI has been involved in national (De Vries 2000) and international (Petropoulos et al. 2001) intercomparisons with excellent results.

After the curing period of six months one of the cubes was immersed in water until saturation (constant mass) and then dried stepwise in a ventilated oven at 200°C. After each step the cube was equilibrated to room temperature and thereafter its mass and radon-release rate were measured. From the mass determinations an average value ( $\bar{m}$ ) of the water saturation of the pores after each drying step was calculated as  $\bar{m} = (M - M_{dry}) / (M_{wet} - M_{dry})$ , with  $M$ : mass after a drying step;  $M_{dry}$ : dry mass and  $M_{wet}$  the mass in saturated condition of the cube. Consequently,  $\bar{m} = 0$  for the dry

<sup>2</sup>EG&G ORTEC LO-AX Series HPGe coaxial low-energy photon spectrometer and EG&G Ortec GEM Series HPGe coaxial detector system. 100 Midland road, Oak Ridge, TN 37830.

<sup>3</sup>RN-2000A, serial no. 169, Pylon Electronic Development Company, Ltd. Ottawa, Canada.

cube and  $\bar{m} = 1$  for the water-saturated cube. To minimize gain or loss of moisture during the radon-release rate measurement the relative humidity of the nitrogen flow was conditioned at either 100% ( $\bar{m} > 0.7$ ), 0% ( $\bar{m} < 0.2$ ) or 50% (all other  $\bar{m}$ -values). Mass gain or loss during the radon-release measurements was never more than a few grams.

### Input parameters

To solve eqn (??) and obtain quantitative results, values for a multitude of parameters that occur in the equation have to be determined. These parameters can be ordered in two groups namely those that do not depend on the fraction of water saturation in the pores,  $m$ , and those that are  $m$ -dependent. It should be noted that  $m$  may vary with spatial position in the cube, whereas  $\bar{m}$  is the volume average of  $m$  over the entire volume of the cube.

We start with estimates of the values of the  $m$ -independent parameters, radium content, porosity, bulk dry density and radon-surface adsorption coefficient. Thereafter the  $m$ -dependence of the radon diffusion coefficient and the emanation factor will be studied.

**Radium content.** The activity concentration ( $C_{Ra}$ ) of  $^{226}\text{Ra}$  was determined via gamma-ray spectrometry. One cube was crushed to parts of size smaller than 2 cm. These parts were dried to constant mass at  $105^\circ\text{C}$  and thereafter left sealed in a 1 litre radon-tight Marinelli beaker for three weeks to establish approximate equilibrium between  $^{226}\text{Ra}$ ,  $^{222}\text{Rn}$  and short-lived radon decay products. The  $^{226}\text{Ra}$  content was then determined using gamma-rays from  $^{214}\text{Bi}$  (609, 1120 and 1764 keV) and from  $^{214}\text{Pb}$  (295 and 352 keV). The geometry was calibrated using nuclide cocktails with certified activities. Corrections for self absorption and background radiation of the setup have been applied.

**Porosity.** Porosity ( $\epsilon$ ) of samples was determined by the water-immersion method. In this method the mass of a sample when completely water-saturated and after oven drying ( $150^\circ\text{C}$ ) is measured. The porosity is then calculated by taking the difference and dividing by the density of water and the bulk volume of the specimen.

**Bulk dry density.** Bulk dry density ( $\rho_b$ ) was calculated from the dry mass and the dimensions of the samples.

**Radon surface-adsorption coefficient** The radon surface-adsorption coefficient  $k_a$  was not measured. For wet surfaces this adsorption is usually assumed to be negligible (Rogers and Nielson 1991b; Van der Spoel 1998). In case of room dry sand only a negligible amount of radon is adsorbed (Van der Spoel et al. 1999). Considering its composition most likely the same holds true for gravel. Consequently, for the materials comprising the bulk of the concrete, surface adsorption is likely a small effect. For dry surfaces of cement the value of the coefficient might be non-zero. For dry concrete ( $k_a=2.1\times 10^{-4}\text{ m}^3\text{ kg}^{-1}$ , Schery and Lopez 1991) and for dry aerated concrete ( $k_a=10.7\times 10^{-4}\text{ m}^3\text{ kg}^{-1}$ , Van der Pal and Van der Spoel 2001) non-zero values were reported. In both these studies it was also found that  $k_a$  decreased very fast with

increasing moisture content. Lacking any experimental information for the type of concrete used in this study,  $k_a$  was set to zero at all values of the moisture content used in the modelling. Possible effects of this assumption are assessed in the discussion.

The values of the  $m$ -independent parameters used throughout the modelling are presented in Table ??.

Table 2: *Values of  $m$ -independent parameters used in the modelling.*

Parameter	Value
$C_{Ra}$ (Bq kg <sup>-1</sup> )	21.7±0.4
$\epsilon$	0.115±0.005
$\rho_b$ (kg m <sup>-3</sup> )	2260±30
$k_a$ (m <sup>3</sup> kg <sup>-1</sup> )	set to zero

**Radon-diffusion coefficient.** The radon-diffusion coefficient in water,  $D_w=1.2\times 10^{-9}\text{m}^2\text{s}^{-1}$  (Broecker and Peng 1974) is four orders of magnitude smaller than the coefficient for diffusion in air,  $D_a=1.1\times 10^{-5}\text{m}^2\text{s}^{-1}$  (Rogers and Nielson 1991a). This implies that with increasing water saturation of the concrete the bulk radon-diffusion coefficient will decrease. To assess the functional dependence of  $D$  on  $m$  two hollow concrete cylinders (height  $h=15$  cm, inner and outer radius  $a=2.5$  and  $b=6.7$  cm, respectively) were machined out of two concrete cubes. These cylinders were then immersed in water and thereafter various values of the water saturation of the concrete were obtained by drying the cylinders step by step. At each set value of water saturation the radon-diffusion coefficient was determined by enclosing a 20.2 kBq radon-emanation source inside the cylinder and sealing the top and bottom surface using a rubber disc and grease (Fig. ??). After three weeks (to approximate a steady-state situation) a radon-release rate measurement was conducted on the hollow cylinder with enclosed source. The bulk radon-diffusion coefficient can then be calculated from the measured value of the radon-release rate using an analytical solution on basis of the radon-transport equation.

The effectiveness of the sealing of the radon source inside the hollow cylinder was tested by using the same procedure on a aluminum dummy. The-radon release rate measured for the situation with the source in the aluminium dummy was less than approximately 1% of the strength of the radon source ( $4.24\pm 0.16$ ) $\times 10^{-2}$  Bqs<sup>-1</sup>) showing that the sealing is almost completely radon tight. As the strength of the radon source is more than three orders of magnitude larger than the radon production of the hollow cylinder itself (approximately  $2\times 10^{-5}$  Bqs<sup>-1</sup>), the latter can be neglected in the transport equation. Consequently, using this hollow cylindrical geometry, the radon-transport equation reduces to a radial 1-dimensional form (due to the effective sealing no radon flux from top and bottom surface occurs) which can be analytically solved (see Appendix A). In this way an expression is obtained for the radon-release rate of the hollow cylinder with radon source enclosed (Appendix A, eqn (??)) with  $D$ ,  $\beta$  and the dimensions of the cylinder as parameters. Using values for the dimensions of the cylinders that were measured with a calliper and  $\beta$  that was derived from the porosity

and moisture content of the hollow cylinder a value of  $D$  was calculated from eqn (??) for each set value of the moisture content of the cylinders.

Figure 2: *Concrete or aluminium hollow cylinder with a radon source enclosed.*

The results of these measurements (Fig. ??) show indeed that the bulk radon-diffusion coefficient decreases as a function of increasing  $m$ . To obtain a description of the data a relation, eqn (??), that was earlier used for sand (Van der Spoel et al., 1999) was fitted (least-squares method) to the data points,

$$D(m) = D(0) \exp(-a(m + m^5)). \quad (6)$$

This relation ( $D(0) = (7.1 \pm 0.7) \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$ ;  $a = 2.76 \pm 0.09$ ; dotted line in Fig. ??) describes the data quite well. The curve is more or less similar to that for sand (open circles and dashed line in Fig. ??, Van der Spoel et al. (1999)) with the exception that the decrease at high  $m$ -values is less steep. In fact, the ratio  $D(1)/D(0)$  for concrete that follows from our measurements is only  $6.3 \times 10^{-2}$ . On the other hand, from first principles, we can write,

$$\frac{D(1)}{D(0)} = \frac{\epsilon \tau D_w}{\epsilon \tau D_a} = 1.1 \times 10^{-4} \quad (7)$$

where  $\tau$  denotes the tortuosity. That the ratio above is not satisfied by eqn (??) is probably related to the lack of data for  $D$  at high  $m$ -values. Attempts to obtain data at higher moisture contents were not succesful because during the period of three weeks to approximate steady-state the sample lost some water (even if kept at 100% RH). To

Figure 3: Bulk radon-diffusion coefficient  $D$  as a function of  $m$  for concrete (filled circles: hollow cylinder 1; filled squares hollow cylinder 2) and for sand (open circles). The dotted and solid line are least-squares fits of eqns (??) and (??), respectively, to the data for concrete. The dashed line is a least-squares fit of eqn (??) to the data for sand (data from Van der Spoel et al. (1999)).

enforce the ratio given by eqn (??) this equation was used as a constraint of a fit to the data of the following three-parameter function describing the  $m$ -dependency of  $D$ ,

$$D(m) = D(0) \exp(-a(m + bm^5)). \quad (8)$$

This function ( $D(0) = (1.77 \pm 0.06) \times 10^{-8} \text{m}^2 \text{s}^{-1}$ ;  $a = 2.57 \pm 0.08$ ;  $b = 2.55 \pm 0.08$ ; solid line in Fig. ??) seems to describe the data reasonably well and has the desired feature that the diffusion coefficient decreases more rapid at higher  $m$ -values.

**Radon-emanation coefficient.** It is well-known that the radon-emanation factor depends on the moisture content of the porous medium. For soil-like materials (Sun and Furbisch 1995; Menetrez et al. 1996), the emanation factor seems to increase with increasing moisture content  $m$  upto 0.1-0.5 at which point the emanation factor reaches a constant maximum value for higher moisture contents.

The moisture dependence of the radon-emanation factor of concrete was measured on a batch of crushed parts (dimensions 1-2 cm; dry mass of the batch  $M_{dry} = 1.531 \text{ kg}$ ) produced from one of the concrete cubes. The size of the parts was chosen such that on the one hand, they are so small that even in completely saturated conditions almost all radon that is produced into the pore space is expected to be released from the parts and on the other hand, the parts are so large that their pore structure is still representative for intact concrete. Radon-release rates were measured of this batch for various values of the moisture content (Fig. ??). The amount of water ( $M_{sat}$ ) needed for saturation ( $m = 1$ ) was estimated from  $M_{sat} = \epsilon \rho_w M_{dry} / \rho_b$  with  $\rho_w$  the density of water. It is realised that the porosity of the crushed parts is not a well-defined quantity and is probably somewhat larger than the porosity of the concrete cube. To be sure

that full saturation is obtained, approximately four times the amount of water needed for saturation was added and the radon-release rate of this supersaturated sample was measured ( $m = 1$  values in Fig. ??). Thereafter the sample was dried in steps and the  $m$ -value was calculated by dividing the mass of water in the sample by  $M_{sat}$ . With this procedure the values of  $m$  in the range  $0 < m < 1$  are most likely overestimates (because  $\epsilon$  is likely estimated too small also  $M_{sat}$  is estimated to small) of the actual  $m$ -values. However, lacking definite information on the porosity of the crushed parts, no other, more accurate estimate could be made.

From the radon-release rates and the radium content of the concrete, the emanation factor was then calculated under the assumption that all radon produced into the pore space also was released (Fig. ??). The emanation factor of concrete shows an almost linear increase over the entire range  $0 < m < 1$  which is in contrast to the behaviour of e.g. sand (open circles, Fig. ??) that shows a fast increase at low  $m$ -values and thereafter obtains an constant value. A least-squares fit of a linear relation ( $\eta(m) = a + bm$ ;  $a=0.010\pm 0.008$ ,  $b=0.270\pm 0.016$ ) to the data shows indeed that such a relation is in very good agreement with the data.

Figure 4: Radon-emanation factor  $\eta$  of concrete (filled circles) and sand (open circles, Van der Spoel et al. (1999)) as a function of  $m$ . The solid line is a least-squares fit of a linear relation to all data points. The dotted line is a fit to only the data points at  $m = 0$  and  $m = 1$  with the  $m = 1$  data adjusted for diffusion (filled squares).

## Moisture distribution

The actual spatial distribution of the moisture inside the concrete test cubes during the radon-release measurements is unknown. In the modelling three distributions were compared, namely 1) a homogeneous moisture distribution (constant  $m$ -value for at all

positions inside the cube), 2) a distribution with zero moisture content at the surfaces and increasing  $m$ -values towards the centre of the cube (Fig. ??) and 3) a distribution with non-zero moisture at the surfaces (Fig. ??) and increasing  $m$ -values towards the centre. The last two distributions were constructed from parametrized integrable functions  $m(x, y, z)$  of which the volume-averaged moisture content of the concrete cube (side length:  $2L$ ) could be calculated analytically from

$$\bar{m} = \frac{1}{(2L)^3} \int_0^{2L} \int_0^{2L} \int_0^{2L} m(x, y, z) dx dy dz. \quad (9)$$

Details on the construction of these distributions can be found in the Appendix B.

Figure 5: *Moisture distribution in a concrete cube at  $x, y=7.5$  cm. Situation with zero moisture at the surfaces. Shown are curves with  $\bar{m}$  of 0.1, 0.2, ..., 0.9. Curves generated with procedure from Appendix B.*

## Concrete structure modelling

To aid the interpretation of the results also a model for concrete structure development was used. This model was developed at the University of Tokyo and for details we refer to Maekawa and Kishi (1999).

This model is based on a description of the hydration process using a multicomponent heat of hydration formalism for blended cements. This provides the concrete heat-generation rate that is used as the source term in the equation of thermodynamic energy conservation. By solving this equation for the space and time domain of interest, the temperature distribution, degree of hydration of concrete and the amount of chemically combined water are calculated. These parameters are further used to compute the micro-aspects of pore-structure development of the hydrated cement paste. The dynamic coupling of the hydration, development of microstructure and moisture

Figure 6: *Moisture distribution in a concrete cube at  $x, y=7.5$  cm. Situation with non-zero moisture at the surfaces. Shown are curves with  $\bar{m}$  of 0.1, 0.2, ..., 0.9. Curves generated with procedure from Appendix B.*

transport is achieved by integrating all these contributions in the mass conservation equation written in terms of pore liquid-water pressure. The sink term of the moisture mass conservation reflects changes in the bulk porosity and its distribution, the rate of moisture loss due to hydration as well as the change in the degree of saturation of various types of pores.

The concrete model as described above has been implemented in the 3D-DuCOM code (version 5.1)<sup>4</sup>. The code is self-contained and it has a much broader applicability than used in this work. DuCOM was designed to provide a quantitative evaluation of concrete performance, necessary for serviceability, durability and safety evaluation of concrete structures. The computational system was tested by applying it to sets of experimental data related to various physical phenomena. The effect of various water-cement ratios, mix proportions, curing conditions and specimen geometry on the microstructure and strength development were investigated. Under arbitrary drying-wetting conditions the code was applied to predict the moisture loss and gain behaviour. Water loss together with hydration also generate volumetric deformation of concrete composite such that an estimation of the degree of shrinkage can be derived. Preliminary verifications have shown reasonable agreement with experiments especially considering that the overall combined system uses only a few empirical parameters (Maekawa and Kishi 1999).

The input information required by the model is: geometrical shape and dimensions of the structure; nodes, elements and surfaces for the finite element analysis; initial

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<sup>4</sup>The numerical code is implemented in FORTRAN programming language using the Microsoft Project Developer, Fortran Power Station 4.0 compiler. The DuCOM code is a product of the Concrete Laboratory, Department of Civil Engineering, University of Tokyo and is developed by K. Maekawa (program frame developer and project leader), CR Chaube (hygro-physical system), CT Kishi (hydration heat modelling) and CT Ishida (ion migration and corrosion).

temperature of concrete; initial liquid pressure in the pore space; environmental boundary conditions (temperature, relative humidity) to which the structure is exposed at each subsequent time step; mix proportion of concrete (water-powder ratio, unit mass of concrete components) and mineral composition of cement/powder material characteristics (specific densities). A numerical code was written at NGD-KVI that automatically generates the input files for a rectangular/cubical sample geometry as required by the DuCOM code. This is especially useful for the proper implementation of the finite-element geometry (nodes, elements and surfaces) and for long simulation times. Besides the solution for the two primary variables, temperature and liquid pressure, other internal variables can also be calculated at each step, in both space and time domain. Of interest for this research are: micropore structure of concrete as expressed by pore-size distribution; degree of hydration of cement in concrete and pore-water content and saturation of various concrete phases.

Concrete structure calculations were performed for a cube of 15 cm sides. Including the two end points, on each edge six nodes were defined. With a total of 216 nodes, the three-dimensional structure is then divided in 125 elements (eight neighbouring points define an element) and 150 boundary surfaces through which transport may take place. The initial temperature of concrete at placement is defined at each node as 20° C. Initially the pore space is completely filled with water, thus the liquid pressure equals atmospheric pressure and the initial liquid pressure at every node is set at 1000 mbar. A time step (in days) is then specified and for each step the environmental conditions (temperature and relative humidity) are set so that they reflect the curing conditions of the samples investigated.

Inside the cube, aggregates are assumed to be a smeared dispersed suspension. This assumption holds because the size of the samples is significantly larger than the size of aggregates.

The fractional densities of the components (defined as the ratio of the mass of the component divided by the total volume of the sample) were set at values derived from Table ???. Mass percentages of the cement powder components were set at Portland cement 40% and blastfurnace slag 60%<sup>5</sup>. The percentages of mineral compounds of the Portland cement were set at 49.7% for tricalcium silicate, 23.9% for dicalcium silicate, 8.8% for tricalcium aluminate and 9.4% for tetracalcium aluminoferrite.

Blaine values (powder surface area) of cement powder components were set at: 3520 m<sup>2</sup> kg<sup>-1</sup> for Portland cement and 3300 m<sup>2</sup> kg<sup>-1</sup> for blastfurnace slag. Specific densities were taken as 2720 kg m<sup>-3</sup> for gravel, 2580 kg m<sup>-3</sup> for sand, 3150 kg m<sup>-3</sup> for Portland cement and 2900 kg m<sup>-3</sup> for blastfurnace slag.

The time evolution of the porosity in concrete was calculated over a time period of 160 days. In Fig. ?? this evolution is shown for the relative contribution to the total porosity of the capillary pores and the gel pores. The capillary porosity constitutes the total pore space at the time the concrete is newly mixed. Thereafter, part of this is filled by the hydration of grains of cement that expand in the process. These hydration products form the cement gel. This is a cohesive mass of hydrated cement, including so-called gel pores that are interconnected interstitial spaces between the gel particles. As

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<sup>5</sup>Actually, according to the manufacturer the mass percentage of the blastfurnace slag is 75%. However, DuCOM was designed for a maximum slag dosage in the cement powder of 60%. To obtain the best approximation of the actual situation the maximum value allowed by DuCOM was used.

hydration products can not precipitate in pores having a diameter smaller than 2 nm this is the diameter of a stable gel pore (Neville 1995). At the end of the simulation period the total porosity was 0.09 (of which approximately 35% capillary and 65% gel pores).

Figure 7: *Time evolution of the relative contributions of capillary (dotted line) and gel (solid line) porosity in concrete. DuCOM simulation for 15 cm cube.*

The time-evolution of the pore-size distribution can be also simulated for capillary pores (Fig. ??). A significant decrease in the average pore size mainly occurs in the early stages of hydration. This result is consistent with the evolution of the capillary and gel porosities shown in Fig. ?. Fig. ? shows the gel-pore size distribution. Actually this does not change in time because in the model it is assumed that gel pores are formed according to the same distribution function at all times.

At the end of the simulation time, the sizes of the capillary pores are distributed around a value of about  $10^{-8}$  m. This value is characteristic for pores in well-hydrated, relatively low water-cement ratio pastes but is an order of magnitude larger than 2 nm, the diameter of a stable gel pore. It should be pointed out that the gel and the capillary pores have a distribution spanning two orders of magnitude and thus, these pore distributions partially overlap.

Moisture profiles depicting the variation of the degree of water-saturation of pores with position over a sample were also simulated for different times after freshly mixing the concrete. Both, the fraction of water in capillary and gel pores,  $m_c$  and  $m_g$  were calculated. Moisture profiles are presented as diagonal cross sections of the three-dimensional curve at three different heights (close to the surface, at  $z_1=0.75$  cm; at an intermediate position,  $z_2 =3.25$  cm and through the centre at  $z_3=7.5$  cm). At each height, the time evolution of the calculated moisture-distribution profiles is shown (Fig. ??).

If the cube is underwater, both types of pores are fully saturated with water ( $m_c$  and  $m_g$  equal unity) and thus during this period (first 21 days), the moisture profiles are

horizontal. Next, the sample is placed at room conditions (20°C, 50% RH) and starts to dry. The profiles show that at first water is removed from the capillary pores, starting with those located at the surface. At the centre of the cube a small dip occurs during the first 25 days. This dip is due to the desiccation of the sample related to the heat production in the hydration process<sup>6</sup>.

Similarly, the water fraction of the gel pores decreases, however, remaining at a higher level than in capillary pores. For both types of pores the water loss is largest near the surface and decreases towards the centre. Except for the magnitude, the patterns are quite similar.

Figure 8: *DuCOM simulation of the time evolution (shown are results for 0.01, 1, 5 21 and 160 days) of the pore-size distribution of capillary pores ( $dv/d(\ln r)$ ), where  $v$  is the fraction of pore volume made up of pores with radius less than  $r$ .*

## Model-measurements comparisons

The results of the measurements of the radon-release rate ( $R$ ) of the concrete cube at various average moisture contents ( $\bar{m}$ ) are presented in Fig. ?? (filled circles). The radon-release rate increases almost linearly with increasing  $\bar{m}$  up to  $\bar{m}=0.5-0.6$ . After a maximum is obtained at  $\bar{m}=0.7-0.8$  the radon-release rate decreases rapidly for higher  $\bar{m}$ -values. In the following the earlier described model for radon release will be used on basis of the values measured for the input parameters to explain this behaviour.

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<sup>6</sup>Even for underwater curing, after cement and water are mixed, the rate of heat liberated in the hydration process is larger than the rate at which water diffuses. For a short time, the central region of the sample is more strongly affected by desiccation and thus, the moisture profiles exhibit a concavity. With water surrounding the sample, the ulterior process of equilibration may be described as the movement of the two maxima initially located close to the surface towards the middle, ending with a fully saturated sample.

Figure 9: *Pore-size distribution of gel pores ( $dv/d(\ln r)$ ) used in DuCOM simulation.*

### Initial modelling

The radon-release rates were initially calculated for a homogeneous moisture distribution on basis of the input parameters listed in Table ??, the linear dependence for the emanation factor (solid line, Fig. ??) and for both fits of the diffusion coefficient (eqns (??) and (??)). Although the model calculations using eqn (??) for  $D(m)$  (Fig. ??, dotted line) show some of the features of the experimental data (linear increase, occurrence of a maximum), the position of the maximum is not at the correct position and, more seriously, the modelled decrease of  $R$  at high  $\bar{m}$ -values is not nearly steep enough. The calculations based on eqn (??) (Fig. ??, solid line) do show a maximum at the correct position and moreover the steep decrease of  $R$  after the occurrence of the maximum is correctly modelled. This indicates that eqn (??) most likely gives a more consistent description of the  $m$ -dependence of  $D$  than eqn (??). Consequently, eqn (??) will be used in further modelling.

### Influence of moisture distribution

The model calculations presented up to this point assumed a homogeneous moisture distribution. However, during the drying procedure of the concrete it is likely that a moisture gradient develops with low  $m$ -values (possibly almost zero) at the surfaces and increasing  $m$ -values towards the centre of the cube. To assess the influence of such alternative  $m$ -distributions model calculations were conducted on basis of the  $m$ -distributions shown in Figs. ?? and ??. It is found that the  $m$ -distribution with zero-moisture at the surfaces results in a shift of the position of the maximum towards lower  $m$ -values which is not in agreement with the experimental data (Fig. ??, dashed line). The  $m$ -distribution with non-zero moisture at the boundaries gives results that are approximately similar to the case with the homogeneous distribution (Fig. ??, dashed

Figure 10: *DuCOM simulation of the time evolution of capillary ( $m_c$ ) and gel ( $m_g$ ) fraction of pore-water saturation.*

dotted line). It is concluded that the actual moisture distribution in the concrete during the radon-release rate measurements most likely resembled the distribution of Fig. ?? (of which the homogeneous distribution is a special case). This is corroborated by the calculations with the concrete structure code DuCOM (Fig. ??) for a drying concrete cube with identical composition as the test cube that also resulted in  $m$ -distributions that are in agreement with Fig. ?. Moreover, also NMR measurements during drying of mortar (Pel, 1995) show drying profiles similar to the curves in Fig. ?.

### Reassessment of the emanation factor

Although the model calculations are consistent with the experimental data with respect to the shape of the radon-release rate curve, the absolute values are underestimated with approximately 25%. This is probably related to the earlier mentioned systematic overestimate of the  $m$ -values in the range  $0 < m < 1$  on the horizontal axis of Fig. ?. Shifting the data points for  $m$  in this range to lower  $m$ -values would increase in the slope of the linear relation for  $\eta(m)$  and consequently result in more radon production in the model. Another possible cause could be that the emanation factor measured for high  $m$ -values is underestimated because part of the radon that emanates into the pore space of the concrete parts is not released due to the small diffusion coefficient at these high  $m$ -values. To estimate this effect, release rates were calculated for concrete cubes with sides of 1.5 cm (the approximate size of the crushed parts) using eqn (??) for  $D(m)$ . It was found that at  $m = 1$  only 80% and at  $m=0.9$  already 99.7% of the emanated radon was also released. The data points at  $m=1$  in Fig. ? were corrected for this effect (filled squares, Fig. ?). Using only the data at  $m=0$  and  $m=1$  (corrected values) an alternative linear relation ( $\eta(m) = a + bm$ ;  $a=0.010\pm 0.010$ ,  $b=0.35\pm 0.02$ )

Figure 11: Radon-release rate  $R$  (filled circles) as a function of average moisture content  $\bar{m}$ . Also shown are model calculations using a homogeneous  $m$ -distribution with  $D(m)$  on basis of eqn (??) (dotted line) and eqn (??) (solid line).

was constructed (dotted line, Fig. ??) and model calculations on basis of this relation were conducted for a homogeneous moisture distribution. These calculations (solid line, Fig. ??) are in good agreement with the experimental data. Especially in the range  $0 < \bar{m} < 0.8$  the correspondence is excellent. In the range  $0.8 < \bar{m} < 1.0$  the calculations slightly overestimate the data.

### Sensitivity analysis

To assess how sensitive the model calculations are for variations in the input parameters the model was used with values of  $C_{Ra}$ ,  $\epsilon$  and  $\rho_b$  in a range of 5%, 20% and 5% respectively, around the values that were used as standard input (values in Table ??) and using  $D(m)$  from eqn. (??) and the alternative linear relation for  $\eta(m)$ . The ranges were selected as being tentatively representative for the maximum differences in the input parameters between different test cubes. The results (for a homogeneous  $m$ -distribution) showed that these variations do not change the calculated values with more than approximately 10%. Thereafter, using values for the  $m$ -independent parameters from Table ??, the sensitivity of the calculations for  $D(m)$  and  $\eta(m)$  was assessed. For both relations the parameters  $a$  and  $b$  were increased and decreased with one standard deviation and calculations were performed for a homogeneous  $m$ -distribution. Fig. ?? shows that these variations may alter the modelled radon-release rate with approximately 20%. In the  $0 < \bar{m} < 0.8$  part of the figure all data points are in between the considered extreme cases, for the higher moisture contents the data points are still somewhat outside the calculated range of variations. However, in view of the still limited amount of information available for the input parameters (especially the moisture distribution and the emanation factor) we conclude that the moisture dependence of radon release from

Figure 12: *Model calculations using eqn (??) for a homogeneous (solid line) m-distribution, and for m-distributions according to Fig. ?? (dashed line) and Fig. ?? (dashed-dotted line).*

concrete modelled on basis of the multi-phase radon-transport equation is consistent with the presented experimental results.

## Discussion

The radon-release rate of concrete was found to depend strongly on its moisture content. As differences of an order of magnitude may occur in the radon-release rate for a certain type of concrete it is recommended that in reporting the results of release (exhalation) rates of concrete also the moisture content during the measurement is provided.

The radon-exhalation rate of concrete in constructions is reported to sometimes decrease with age (Yu et al. 1995). Under the assumption that the radon release of other types of concrete have a similar moisture dependence as found in this study, this can be explained. Concrete in new constructions is very moist and will dry during its life time. If the first exhalation measurement is made on the left side of the maximum of the curve in Fig. ??, radon exhalation will appear to be decreasing as a function of age. Furthermore, long-term follow-up studies that most likely started with measurements on the wet side of the maximum often reveal a maximum in radon release after 0.5 to 1 year (Roelofs and Scholten 1994) as the concrete gradually dries and the radon-release rate takes values from left to right in Fig. ??.

The radon-diffusion coefficients of concrete found in this study are two orders of magnitude smaller than for soil-like materials. A factor of three of this large difference can be explained by the lower porosity (factor of three) of concrete. However, the main reason is probably the completely different pore-size distribution of concrete. Model calculations with DuCOM show that the pores in the hydrated cement (gel) that are approximately uniform in size ( $10^{-9}$  m) make up roughly 65% (Fig. ??) of the total

Figure 13: *Model calculations (solid line) using eqn (??) for a homogeneous m-distribution and with the alternative linear relation for  $\eta(m)$ .*

porosity<sup>7</sup>. Capillary pores in concrete constitute about 35% of the total porosity with a broad pore-size distribution ranging from  $10^{-6}$  to  $10^{-9}$  m. Such a pore-size distribution will most likely result in very tortuous transport path and moreover, transport in the very small pores will be dominated by Knudsen diffusion. Both effects result in lower diffusion coefficients. To make a first-order estimate of the effect of Knudsen diffusion, we assume a medium with straight cylindrical pores with radius  $r$  (m), then the Knudsen diffusion coefficient  $D_K$  ( $\text{m}^2\text{s}^{-1}$ ) is given by (Henry et al. 1967)

$$D_K = \frac{2}{3}r\bar{v} = 2D_a\frac{r}{\bar{\ell}} \quad (10)$$

where,  $\bar{v}$  ( $\text{ms}^{-1}$ ) is the mean molecular velocity and  $\bar{\ell}$  (m) is the mean free path of the atoms. In eqn (??) the relation for the unconstrained molecular-diffusion coefficient,  $D_a = \frac{1}{3}\bar{v}\bar{\ell}$  that follows from kinetic gas theory is used. The mean free path may be estimated as  $\bar{\ell}=1.2\times 10^{-7}$  m from (McQuarrie 1976)

$$\bar{\ell} = \frac{k_B T}{\sqrt{2}\pi\sigma_{Rn}^2 P} \quad (11)$$

with  $k_B = 1.38\times 10^{-23}$  J K<sup>-1</sup>,  $T = 293$  K;  $P = 10^5$  Pa and  $\sigma_{Rn} = 0.27$  nm, the atomic diameter of radon. Using this value for the mean free path and taking  $r = 10^{-9}$  m as a typical gel pore size in eqn (??) results in  $D_K \approx 2\times 10^{-7}$   $\text{m}^2\text{s}^{-1}$ . Converting this to a bulk diffusion coefficient (for a dry sample) by multiplying with the porosity ( $\epsilon=0.115$ ) of the concrete we arrive at  $D \approx 2\times 10^{-8}$   $\text{m}^2\text{s}^{-1}$ , which is of the same order of the measured

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<sup>7</sup>Sometimes a further distinction is made between three classes of pores namely, capillary, gel and interlayer pores. The latter are situated between the layers of calcium silicate hydrates making up the gel. In this paper the interlayer pores are lumped into the gel porosity.

Figure 14: Model calculations using eqn (??) for a homogeneous  $m$ -distribution with all parameters as in Fig. ?? (solid line) and with increase/decrease of  $1\sigma$  in the  $a$  and  $b$  parameters of  $\eta(m)$  (dashed lines) and  $D(m)$  (dotted lines).

diffusion coefficient at low moisture contents. Consequently, diffusive transport of radon through (intact) concrete is mainly dominated by Knudsen type diffusion in the gel pores.

The values for the bulk diffusion coefficient for radon in concrete found in this study are in the range  $5 \times 10^{-11}$  to  $2 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$ . This is at the lower end of ranges of values found<sup>8</sup> by Rogers et al. (1994) of  $0.4 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  to  $10 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  for residential concretes; by Rogers et al. (1995) of  $3 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  to  $13 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  for aged residential concretes; by Gadd and Borak (1995) of  $1.1 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  to  $4.4 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  for *in-situ* concrete and by Renken and Rosenberg (1995) of  $5 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  to  $14 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$ . The likely explanation for the lower values in this study is that the type of concrete used in this study is more ‘heavy’ and less porous than as used in the studies from the United States referenced above. In most of these studies porosities around 0.20 are quoted which is considerably larger than the value of 0.115 of the concrete in this study. Results from Europe of Klink et al. (1999) who found values in the range  $5 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$  to  $9 \times 10^{-8} \text{ m}^2 \text{ s}^{-1}$  for low porosity mortars and concretes and of Folkerts et al. (1984) who report a value of  $6.8 \times 10^{-10} \text{ m}^2 \text{ s}^{-1}$  for ‘heavy’ concrete are comparable to the results of this study.

Also the  $m$ -dependence of the emanation factor of concrete, that deviates from that of various soil types, is most likely a result of this pore-size distribution. It is hypothesized that in going from dry to wet conditions the pores are gradually filled, starting with the smallest pore sizes (and so enhancing the probability of recoiling radon to end up in the pore space instead of penetrating the mineral of an adjacent grain). At higher moisture contents more and more pores will be ‘activated’ leading to the more or less

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<sup>8</sup>Values of the pore-diffusion coefficients reported by Rogers et al. (1994) and Rogers et al. (1995) have been used to estimate the bulk radon-diffusion coefficient by multiplying by the porosities mentioned in these papers.

linear increase in emanation factor. Such a linear increase was also calculated by Sun and Furbish (1995) using a Monte Carlo model. However, in their numerical model a saturation plateau is reached between  $m=0.4$  and  $0.5$  for a porous material with a porosity of  $0.3$ . This is in contrast to the results presented here for concrete (Fig. ??) where no evidence for such a plateau is found.

The model calculations on basis of the multi-phase radon-transport equation using independently measured information on the input parameters are in good agreement with the measured moisture dependence of radon release rates from concrete. However, some not fully justified assumptions have been made in the process. One of these assumptions is that the radon surface-adsorption coefficient  $k_a$  has been set to zero. A non-zero value would increase the value of the partition-corrected porosity via an extra term in eqn (??). The bulk diffusion coefficient is calculated via eqn (??) from which a value for  $D/\beta$  is obtained and  $D$  is calculated by multiplying this value with  $\beta$ . Consequently,  $D$  (as input parameter obtained from independent measurements) would increase with the same factor as  $\beta$  in case of a non-zero value for  $k_a$ . A third input parameter that should be differently interpreted in case of non-zero surface adsorption is the radon emanation factor  $\eta$ . If during the radon release measurement on the crushed concrete parts radon adsorbs to the internal surfaces of the concrete the radon-release rate measured will be smaller than in case of zero surface adsorption. Thus, if  $k_a \neq 0$  than the actual value of  $\eta$  is higher than calculated under the assumption that all radon produced in the pore space is also released. The scaling factor in this case is most likely comparable to that for  $\beta$  and  $D$ . Due to this the total effect on the model calculations based on eqn (??) will be reduced because as  $D$ ,  $\beta$  and  $S$  (via  $\eta$ ) increase with the same factor, all three terms in the equation are affected in the same way and the effect can be divided out of the equation. So, even in case of non-zero adsorption of radon on surfaces, model calculations based on the input parameters as determined in this study are expected to represent radon release from and radon transport in concrete fairly well.

In the light of this conclusion the model will be used in the further part of this discussion to assess the effect of moisture content on the radon source and barrier aspects of concrete.

## Concrete as a radon source

A consequence of the radon-release rate of concrete being dependent on its moisture content is that the radon production of concrete that is actually present in a house (room) will depend on the indoor relative humidity (RH). To estimate the magnitude of this effect a relation is needed that links RH and the moisture content ( $m$ ) of concrete. Such a relation was developed on basis of the Kelvin equation (Scheidegger 1974) that gives the maximum pore radius (cylindrical pores are assumed) at which pores are completely saturated at a certain relative humidity. Water in partly saturated pores is taken into account by computing the thickness of the adsorbed water layer in these pores (as a function of RH) using a modification of the BET theory (Brunauer et al. 1938) proposed by Hillerborg (1985). The moisture content  $m$  as a function of relative humidity is then calculated using the distributions of gel and capillary pores (after 160 days of curing) and the ratio of gel to capillary porosity from DuCOM. Fig. ?? shows that the fraction of water saturation  $m$  of the total porosity increases relatively fast at low RH-values (RH<10%). Thereafter the decrease is more gradual upto RH=80%, the

final increase until saturation is again more steep ( $RH > 80\%$ ). The figure also shows that the gel porosity is already 80% ( $m=0.8$ ) saturated at  $RH=20\%$ , this is in sharp contrast with the behaviour of the capillary porosity that gradually increases to  $m=0.2$  upto  $RH=80\%$ , thereafter the increase to saturation is very steep.

Figure 15: *Relation between RH and m for concrete of similar composition as used in the radon-release rate experiments. The relation is constructed on basis of DuCOM calculations. Solid line: fraction of water saturation of total porosity; dotted line: idem, for gel porosity and dashed line: idem, for capillary porosity.*

The relation between RH and  $m$  can now be used to calculate the radon-release rate of concrete of which the water content is in equilibrium with a certain relative humidity. As in (Dutch) houses concrete is mostly present in large slabs (walls, floors) of approximately 20 cm thickness (Janssen et al. 1998) it was decided (to obtain results that can readily be converted to practical situations) to consider the radon release of a 20 cm thick slab (surface area  $1 \text{ m}^2$ ). Furthermore, as walls and floors usually have large surface areas, radon release will, to a good approximation, only take place from the surfaces that are directed towards the inside of the room and towards the cavity wall or the outside of the house. This implies that radon release is a 1-dimensional process. To simulate this, zero-flux boundary conditions were set at the four smaller area side surfaces of the concrete slab. Finally, only the indoor directed radon release was considered. Using the model parameters of the final modelling (as in Fig. ??) the surface exhalation rate  $E_s$  ( $\text{Bq m}^{-2} \text{ s}^{-1}$ ) was calculated for this 20 cm thick slab of concrete as a function of relative humidity (Fig. ??, dotted line).  $E_s$  is found to increase gradually in going from  $RH=0$  to 60-70% where a maximum is reached. Thereafter  $E_s$  decreases very rapidly, reflecting the fast filling of the porosity at the higher end of the  $m$ -RH curve (Fig. ??) which will result in a fast decrease of the diffusion coefficient.

If we consider the entire range of the relative humidity (0-100%) we conclude from Fig. ?? that the radon-exhalation rate can, in principle, range over an order of magnitude. However, for most houses the RH-value will most likely be within the range

30-70%. This implies that for real situations, variations of 10-15% are to be expected. In this range lower relative humidity will lead to a reduction of the radon concentration. Fig. ?? also shows that from a radon point of view it is beneficial to condition the indoor relative humidity to a value close to 100%. Obviously this has some serious drawbacks and is thus not a practical countermeasure.

Figure 16: *Surface radon-exhalation rate  $E_s$  of 20 cm thick concrete slab as a function of relative humidity  $RH$ , due to production in the concrete (dotted line); due to diffusive transport of radon from soil gas with a concentration of  $50 \text{ kBq m}^{-3}$  (dashed line) and the sum of both contributions (dashed-dotted line).*

### Concrete as a radon barrier

As a second application of the constructed model for radon transport and generation in concrete we considered the diffusive transport through a floor slab of 20 cm thick concrete as a function of the relative humidity of the slabs surroundings. It is realised that for such a floor slab the relative humidity of the environment is a much less well defined concept then for the walls of a dwelling and will strongly depend on the ground water management of the location where the dwelling is situated. However, the results of this assessment will give at least an impression of the effect of changing conditions. In this case we considered a slab with the same parameters as for the calculation of the radon-source aspects except that we set the radon production of the slab to zero ( $C_{Ra} = 0 \text{ Bq kg}^{-1}$ ). Fixed concentration boundary conditions were set at the top and bottom surface of the slab with zero radon concentration at the top and  $50 \text{ kBq m}^{-3}$  at the bottom. The first condition reflects the relatively low concentrations indoors and the second is taken as a typical soil-gas concentration (Hutter and Knutson 1998). The model calculations (Fig. ??, dashed line) show that the radon exhalation of the top surface due to transported radon originating from the soil gas decreases rapidly with increasing relative humidity. The figure shows that there might be an order of magnitude

difference in radon entry from the soil if the relative humidity of the environment changes from 10% (very arid soil) to 80% or higher (high ground-water table). For comparison, also the surface exhalation of radon originating from the concrete is plotted in the figure (dotted line) and for these choice of parameters it appears that (except for very dry soil), the entry of radon produced in the concrete dominates the entry from radon from soil gas diffusing through the slab. In this case the total entry (dashed-dotted line) is more or less constant upto 80% RH thereafter it steeply decreases. It should be noted that this behaviour is only valid for the situation considered, the relative contribution of radon from the concrete itself and radon originating from soil gas will be influenced by the actual radon concentration in the soil gas. However, the dependence on relative humidity of the latter contribution will not change its shape but the absolute value of the entry rate will simply scale linearly with the value of the soil-gas radon concentration.

Finally, it should be emphasized that the results in this study are valid for intact concrete. Cracks were not present in the samples and thus not taken into account. Although cracks will most likely not significantly change the radon production properties of concrete, the barrier action could change dramatically. Landman (1982) developed a model for diffusion through cracks in concrete and concluded that an intact concrete slab could reduce diffusive radon entry from the soil with a factor of 80; in case of cracks this reduction is much lower (typically a factor of 4). The reduction of radon entry achievable by a concrete barrier as calculated from results presented in this study should thus be considered as the maximum reduction possible.

## Conclusions

The radon-release rate of concrete was found to increase almost linearly upto moisture contents of 50 to 60%. At 70 to 80% a maximum occurs and for higher moisture contents the radon-release rate decreases very steeply.

This moisture dependence of radon release from concrete was succesfully modelled on basis of the multi-phase radon-transport equation using independently measured values for the input parameters. Interpretation of the results was aided and guided by model calculations with a model for concrete structure development (DuCOM). It is expected that combining cq. integrating the developed radon-transport model with a model for concrete structure (such as DuCOM) will lead to a very powerful modelling tool for the design of low-radon producing concrete.

The model was used to simulate the dependence of the radon source and barrier aspects as a function of relative humidity. It was concluded that although the radon exhalation rate from concrete may change over an order of magnitude over the range of 0 to 100% RH, for most houses where the relative humidity will be in the range 30-70%, variations will only be about 10-15%. As a radon barrier, it was concluded that concrete performs best at the higher end of the relative humidity range. Actually, only for very arid soils, diffusive entry of radon originating from the soil gas may be expected to dominate the production inside the concrete itself.

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## Appendix A

### Analytical solution for radon transport in a hollow cylindrical sample

For a hollow cylinder that has a top and bottom surface that is intranparent for radon only radial transport remains and the steady-state radon transport equation (in cylindrical coordinates) reduces to,

$$D \frac{\partial^2 C}{\partial r^2} + D \frac{1}{r} \frac{\partial C}{\partial r} - \lambda C + S = 0, \quad (12)$$

where  $S$  ( $\text{Bq m}^{-3}\text{s}^{-1}$ ) is the bulk radon-production rate of the material of the hollow cylinder (height  $h$ , inner radius  $a$  and outer radius  $b$ , see Fig. ??). In the situation with a radon source of strength  $R_s$  ( $\text{Bq s}^{-1}$ ) enclosed in the inner cylinder and the radon concentration at the outer surface set at zero, the boundary conditions for eqn (??) are:

$$C(r) = \frac{R_s + \beta D \frac{\partial C}{\partial r} 2\pi a h}{\lambda_{Rn} \pi a^2 h}; \quad r = a; \quad (13)$$

$$C(r) = 0; \quad r = b. \quad (14)$$

If we assume that the radon-production rate of the radon source is much larger than the production rate of the material of the cylinder we can neglect the production term  $S$  in eqn (??) and reformulate this equation as:

$$\frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r} - \alpha^2 C = 0; \quad (15)$$

$$\alpha^2 = \frac{\lambda \beta}{D}. \quad (16)$$

Eqn (??) is a modified Bessel equation of order zero. This type of equation is solved independently by the modified Bessel functions  $I_0(\alpha r)$  and  $K_0(\alpha r)$ , the general solution being a linear combination of both Bessel functions,

$$C(r) = A I_0(\alpha r) + B K_0(\alpha r). \quad (17)$$

The constants  $A$  en  $B$  are found by substitution of eqn (??) in the boundary conditions eqns (??) and (??). This results in:

$$C(r) = \frac{R_s}{\lambda \pi a^2 h} \left[ \frac{I_0(\alpha b) K_0(\alpha r) - K_0(\alpha b) I_0(\alpha r)}{(K_0(\alpha a) I_0(\alpha b) - K_0(\alpha b) I_0(\alpha a)) + \frac{2\beta}{\alpha} (I_0(\alpha b) K_1(\alpha a) + K_0(\alpha b) I_1(\alpha a))} \right] \quad (18)$$

The radon-release rate  $R_{cs}$  ( $\text{Bq s}^{-1}$ ) of the hollow cylinder with the source enclosed is found by multiplying the external lateral area with the bulk diffusive flux at  $r = b$ ,

$$R_{cs} = -2\pi b h D \frac{\partial C}{\partial r}; \quad r = b. \quad (19)$$

Substitution of eqn (??) in eqn (??) finally leads to:

$$\frac{R_{cs}}{R_s} = \frac{2b\beta}{a^2 \alpha} \left[ \frac{I_0(\alpha b) K_1(\alpha b) + K_0(\alpha b) I_1(\alpha b)}{(K_0(\alpha a) I_0(\alpha b) - K_0(\alpha b) I_0(\alpha a)) + \frac{2\beta}{\alpha} (I_0(\alpha b) K_1(\alpha a) + K_0(\alpha b) I_1(\alpha a))} \right] \quad (20)$$

## Appendix B

### Analytical profiles for a position-dependent moisture content

#### Zero-moisture content boundary condition

In this paragraph we construct a function  $m(x, y, z)$  that represents a moisture distribution in a cube with side  $2L$  of which the pores at all surfaces have  $m = 0$  and of which  $m$  reaches a maximum value  $m_{max}$  in the centre of the cube. From symmetry considerations we assume we can write the function as the following product,

$$m(x, y, z) = \prod_{i=x}^z m_i(i). \quad (21)$$

The functions  $m_i(i)$  are subject to the following boundary conditions,

$$m_i(0) = m_i(2L) = 0; i = x, y, z; \quad (22)$$

$$\frac{\partial m_i}{\partial i} \Big|_{i=l} = 0; i = x, y, z. \quad (23)$$

The first boundary condition reflects the zero moisture content at the surface, the second reflects the maximum in the centre. Due to the symmetry of eqns (??) and (??), the three functions  $m_i$  ( $i = x, y, z$ ) are actually identical. Thus using symmetry along the planes  $i = L$  ( $i = x, y, z$ ) we construct a two parameter function for the  $m_i$ 's that after insertion in eqn (??) force  $M(x, y, z)$  to satisfy eqns (??) and (??):

$$m_i(i) = f(i); 0 < i \leq L; i = x, y, z; \quad (24)$$

$$m_i(i) = f(2L - i); l < i \leq 2L; i = x, y, z; \quad (25)$$

$$f(\xi) = A(1 - e^{-\alpha\xi} - \alpha e^{-\alpha\xi}\xi) \quad (26)$$

where  $\alpha$  and  $A$  are parameters of the function.

The maximum moisture content is now given by,

$$m_{max} = A^3(1 - e^{-\alpha L} - \alpha e^{-\alpha L} L)^3 \quad (27)$$

and we will assume that it depends on the average moisture content  $\bar{m}$  according to:

$$m_{max} = \bar{m} + \bar{m}(1 - \bar{m}). \quad (28)$$

With the latter assumption we force the maximum moisture content in the centre of the cube to scale with the average moisture content and obey the logical constraints that  $m_{max} = 0$  if  $\bar{m} = 0$  and  $m_{max} = 1$  if  $\bar{m} = 1$ . The additional effect is that one of the two adjustable parameters of the function is now fixed. The average moisture content can now be calculated by inserting eqns (??-??) in eqn (??) and then from performing the integrals one obtains,

$$\bar{m} = 2 - L^3 \left( \frac{1 - e^{-\alpha L} - \alpha L e^{-\alpha L}}{L + \frac{1}{\alpha} e^{-\alpha L} - \frac{1}{2} \alpha L^2 e^{-\alpha L} - \frac{1}{\alpha}} \right)^3. \quad (29)$$

With eqn (??) values for  $\alpha$  can be generated for each value  $\bar{m}$  and using eqns (??) and (??) the corresponding value for  $A$  can be deduced. This procedure has been used to generate values of the fraction of water saturation of the pores at a certain position  $(x, y, z)$  in the concrete cubes.

## Non-zero moisture content boundary condition

A similar approach as in the preceding section will be used also for the case of a non-zero values of the moisture at the surfaces of a cube with side  $2L$ . However, in this case, the function  $m(x, y, z)$  is a sum of three terms in which the first term ( $m_0$ ) corresponds to the case of a homogeneous distribution:

$$m(x, y, z) = m_0 + \prod_{i=x}^z m_A(i) + \prod_{i=x}^z m_B(i); \quad i = x, y, z; \quad (30)$$

$$m_A(i) = f_A(i); \quad 0 < i \leq L; \quad (31)$$

$$m_A(i) = f(2L - i); \quad L < i \leq 2L; \quad (32)$$

$$m_B(i) = f_B(i); \quad 0 < i \leq L; \quad (33)$$

$$m_B(i) = f(2L - i); \quad L < i \leq 2L; \quad (34)$$

$$f_A(\xi) = A\xi \left( \sin \left( \frac{\xi\pi}{2L} \right) + \frac{2}{\pi} \cos \left( \frac{\xi\pi}{2L} \right) \right); \quad (35)$$

$$f_B(\xi) = B^3\xi(\xi - L)^2. \quad (36)$$

In this construction  $m_0$ ,  $A$  and  $B$  are adjustable parameters. It can be easily checked that  $m(x, y, z)$  satisfies the following set of boundary conditions, which are similar to those of the first case except for the non-zero value at the surfaces.

$$m(0, y, z) = m(x, 0, z) = m(x, y, 0) = m_0; \quad (37)$$

$$m(2L, y, z) = m(x, 2L, z) = m(x, y, 2L) = m_0; \quad (38)$$

$$\frac{\partial m}{\partial x} \Big|_{x=L} = \frac{\partial m}{\partial y} \Big|_{y=L} = \frac{\partial m}{\partial z} \Big|_{z=L} = 0 \quad (39)$$

The average moisture content  $\bar{m}$  for this function can be expressed as a function of the adjustable parameters,

$$\bar{m} = m_0 + \left( \frac{A}{L} \right)^3 \left( \frac{2L}{\pi} \right)^6 \left( 2 - \frac{2}{\pi} \right)^3 + \left( \frac{B}{L} \right)^3 \left( \frac{B^2 L^4}{12} \right)^3 \quad (40)$$

After some experimenting with the behaviour of  $m(x, y, z)$  as a function of the adjustable parameters it was decided to set  $B = 0.1 \text{ cm}^{-1}$ . Setting  $B$  at larger values sometimes resulted in a physically unwanted local maximum of the moisture distribution at positions between the surface and the centre of the cube.

For average moisture contents in the range from 0 to 1, values for  $m_0$  (gradually increasing, see Fig. ??) and  $A$  were selected so that eqn ?? was satisfied.

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