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Measurement of radioactivity in the environment — Air: radon-222 —

Part 10:
Determination of diffusion coefficient in waterproof materials using activity concentration measurement

Mesurage de la radioactivité dans l'environnement — Air: radon 222 —

Partie 10: Détermination du coefficient de diffusion du radon des matériaux imperméables par mesurage de l'activité volumique du radon

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Foreword

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International Standards are drafted in accordance with the rules given in the ISO/IEC Directives, Part 2.

The main task of technical committees is to prepare International Standards. Draft International Standards adopted by the technical committees are circulated to the member bodies for voting. Publication as an International Standard requires approval by at least 75 % of the member bodies casting a vote.

Attention is drawn to the possibility that some of the elements of this document may be the subject of patent rights. ISO shall not be held responsible for identifying any or all such patent rights.

ISO 11665-10 was prepared by Technical Committee ISO/TC 85, *Nuclear energy, nuclear technologies and radiological protection*, Subcommittee SC 2, *Radiological protection*.

ISO 11665 consists of the following parts, under the general title *Measurement of radioactivity in the environment — Air: radon 222*:

— *Part 1: Origins of radon and its short-lived decay products and associated measurement methods*

— *Part 2: Integrated measurement method for determining average potential alpha energy concentration of its short-lived decay products*

— *Part 3: Spot measurement method of the potential alpha energy concentration of its short-lived decay products*

— *Part 4: Integrated measurement method for determining average activity concentration using passive sampling and delayed analysis.*

— *Part 5: Continuous measurement method of the activity concentration.*

— *Part 6: Spot measurement method of the activity concentration*

— *Part 7: Accumulation method for estimating surface exhalation rate*

— *Part 8: Methodologies for initial and additional investigations in buildings*

— *Part 9:Test methods for determining exhalation rate of building materials*

— *Part 10: Determination of the diffusion coefficient in waterproof materials using activity concentration measurement*

— *Part 11: Method for soil gas with sampling at depth*

Introduction

Radon isotopes 222, 219 and 220 are radioactive gases produced by the disintegration of radium isotopes 226, and 224, which are decay products of uranium-238 and thorium-232 respectively, and are all found in the earth's crust. Solid elements, also radioactive, followed by stable lead are produced by radon disintegration [Nuclear Data Base issued from the Decay Data Evaluation Project. http://www.nucleide.org/DDEP_WG/DDEPdata.htm].

When disintegrating, radon emits alpha particles and generates solid decay products, which are also radioactive (polonium, bismuth, lead, etc.). The potential effects on human health of radon lie in its solid decay

products rather than the gas itself. Whether or not they are attached to atmospheric aerosols, radon decay products can be inhaled and deposited in the bronchopulmonary tree to varying depths according to their size.

Radon is today considered to be the main source of human exposure to natural radiation. The UNSCEAR (2006) report [UNSCEAR, E*ffects of ionizing radiation*, Vol. 1, 2006, Report to the General Assembly, with Scientific Annexes (New York : United Nations Publication 2008)] suggests that, at the worldwide level, radon accounts for around 52% of global average exposure to natural radiation. The radiological impact of isotope 222 (48%) is far more significant than isotope 220 (4%), while isotope 219 is considered negligible. For this reason, references to radon in this part of ISO 11665 refer only to radon-222.

Radon activity concentration can vary from one to more orders of magnitude over time and space. Exposure to radon and its decay products varies tremendously from one area to another, as it depends on the amount of radon emitted by the soil, weather conditions, and on the degree of containment in the areas where individuals are exposed [Kemski, J.; Klingel, R.; Siehl, A.; Neznal, M.; Neznal, M.; Matolín, M. (2012): Erarbeitung fachlicher Grundlagen zur Beurteilung der Vergleichbarkeit unterschiedlicher Messmethoden zur Bestimmung der Radonbodenluftkonzentration; Bd.2 Sachstandsbericht "Radonmessungen in der Bodenluft – Einflussfaktoren, Messverfahren, Bewertung", BfS Forchungsvorhaben 3609S10003, 2012, 122 pages].

As radon tends to concentrate in enclosed spaces like houses, the main part of the population exposure is due to indoor radon. Soil gas is recognized as the most important source of residential radon through infiltration pathways. Other sources are described in other parts of this standard (building materials) and ISO 13164 (water).

Radon enters into buildings mainly via convection mechanism, the so-called "stack effect" that is due to a difference in air temperature between the inside and the outside of the building, which generates a difference in pressure between the air in the building and the air contained in the underlying soil. Radon activity concentration depends on the building structure, the equipment (chimney, ventilation systems, among others), the environmental parameters of the building (temperature, pressure, etc.) and the occupants' lifestyle.

To limit the risk to individuals, a national reference level of 100 Bq.m^{-3} is recommended by the World Health Organization [WHO Handbook on indoor radon [WHO, 2009. A public Health Perspective]. Wherever this is not possible, the chosen level should not exceed 300 Bq.m^{-3}. COUNCIL DIRECTIVE 2013/59/EURATOM of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, (and repealing Directives 89/618/Euratom, 90/641/Euratom, 96/29/Euratom, 97/43/Euratom and 2003/122/Euratom) sets that Member States shall establish national reference levels for indoor radon concentrations. The reference levels for the annual average activity concentration in air shall not be higher than 300 Bq.m^{-3}.

To reduce the risk to the overall population, building codes should be implemented that require radon prevention measures in buildings under construction and radon mitigating measures in existing buildings. Radon measurements are needed because building codes alone cannot guarantee that radon concentrations are below the reference level.

When a building requires protection against radon from the soil, radon-proof insulation (based on membranes, coatings or paints) placed between the soil and the indoors may be used as a stand-alone radon prevention/remediation strategy or in combination with other techniques such as passive or active soil depressurization. Radon-proof insulation functions at the same time as the waterproof insulation [5], [6], [7].

Radon diffusion coefficient is a parameter that determines the barrier properties of waterproof materials against the diffusive transport of radon. Applicability of the radon diffusion coefficient for the design of radon-proof insulation is prescribed by national building standards and codes. Requirements for radon-proof insulation as regards the durability, mechanical and physical properties and the maximum value of the radon diffusion coefficient are also prescribed by national building standards and codes or independent certification bodies.

Radon diffusion coefficient is a material property. In homogeneous (single-layered) materials its value does not depend on the thickness of the tested material. Radon diffusion coefficient of layered materials produced in several thicknesses shall be determined separately for each thickness.

The radon diffusion coefficient of these membranes is currently assessed with two types of methods based on different assumptions:

— Rapid determination test method

— Long term test methods.

As no reference standards and reference materials are currently available for these types of materials and related values of radon diffusion coefficient, the metrological requirement regarding the determination of the performance of a method, as required by ISO 17025, cannot be directly met.

As it is the responsibility of the laboratory to carry out its testing in such a way as to meet the requirements of this International Standard and to satisfy the needs of the customer, this part of ISO 11665 gives, for each testing method described, assumptions and boundary conditions that shall be met.

NOTE   The origin of radon-222 and its short-lived decay products in the atmospheric environment and the measurement methods are described generally in ISO 11665-1.

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Measurement of radioactivity in the environment — Air: radon 222 — Part 10: Determination of the diffusion coefficient in waterproof materials using activity concentration measurement

1 Scope

This standard specifies the different methods intended for assessing the radon diffusion coefficient in waterproofing materials such as bitumen or polymeric membranes, coatings or paints, as well as assumptions and boundary conditions that shall be met during the test.

The test methods described in this standard allows estimating the radon diffusion coefficient in the range of 10^{-8} to 10^{-12} m2/s with an associated uncertainty from 10 to 40 %.

This standard is not applicable for porous materials, where radon diffusion depends on porosity and moisture content.

2 Normative references

The following referenced documents are indispensable for the application of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

ISO 921, *Nuclear energy — Vocabulary*

ISO 80000-10, *Quantities and units – Part 10: Atomic and nuclear physics*

ISO 11665-1, *Measurement of radioactivity in the environment ⎯ Air: Radon-222 ⎯ Part 1: Origins of radon and its short-lived decay products and associated measurement methods*

ISO 11665-5, *Measurement of radioactivity in the environment ⎯ Air: Radon-222 ⎯ Part 5: Continuous measuring method of the activity concentration*

ISO 11665-6, *Measurement of radioactivity in the environment ⎯ Air: Radon-222 ⎯ Part 6: Spot measurement method of the activity concentration*

ISO 11929, *Determination of the characteristic limits (decision threshold, detection limit and limits of the confidence interval) for measurements of ionizing radiation — Fundamentals and applications*

ISO/IEC Guide 98-3:2008, *Uncertainty of measurement — Part 3: Guide to the expression of uncertainty in measurement (GUM:1995)*

IEC 61577-2, *Radiation Protection Instrumentation — Radon and radon decay product measuring instruments — Part 2: Specific requirements for ^{222}Rn and ^{220}Rn measuring instruments*

3 Terms, definitions and symbols

3.1 Terms and definitions

For the purposes of this document, the terms and definitions given in ISO 11665-1, ISO 921 and ISO 80000-10 and the following apply.

3.1.1
material
material produced according to a certain technical specifications which is the object of the test

3.1.2
sample (of material)
certain amount of material chosen from the production batch for determination of the material properties

3.1.3
radon diffusion coefficient (bulk) D
radon activity permeating due to molecular diffusion through unit area of a layer of material of unit thickness per unit time at unit radon activity concentration gradient on the boundaries of this layer

3.1.4
radon diffusion length l
distance crossed by radon due to diffusion in which activity is reduced by "e" times because of decay (numeric "e" is the natural logarithm, equal to about 2.72)

Note 1 to entry: Radon diffusion length is expressed by the relationship given in Formula (1)

$$l = (D/\lambda)^{1/2} \tag{1}$$

where

l is the radon diffusion length, in metre

D is the radon diffusion coefficient of the sample, in square metre per second;

λ is the radon decay constant, in per second.

3.1.5
diffusive radon surface exhalation rate E
value of the activity of radon atoms that leave a material per unit surface per unit time

Note 1 to entry: For the purpose of this standard only the diffusion transport through the sample is taken into account. The diffusive radon exhalation rate in a homogeneous material is given by Formula (2) (Fick's law)

$$E(x) = -D\frac{\partial C(x)}{\partial x} \tag{2}$$

where

$E(x)$ is the distribution function along the axis "x" the radon exhalation rate in the sample, in becquerel per square metre per second

D is the radon diffusion coefficient of the sample, in square metre per second;

$C(x)$ is the distribution function along the axis "x" the radon activity concentration in the sample, in becquerel per cubic metre;

x is the coordinate on axis "x" (the axis is directed along radon transport and perpendicular to the sample surface), in metre.

3.1.6
non-stationary radon diffusion
the process of radon diffusion through the sample characterized by a change in time of radon activity concentration in the sample

one-dimensional non-stationary radon diffusion is described by the partial differential equation:

$$D \cdot \frac{\partial^2 C(x,t)}{\partial x^2} - \lambda \cdot C(x,t) = \frac{\partial C(x,t)}{\partial t} \tag{3}$$

where

  D is the radon diffusion coefficient of the sample, in square metre per second;

  $C(x,t)$ is the function changing in time along the axis "x" of radon activity concentration in the sample, in becquerel per cubic metre;

  x is the coordinate on axis "x" (the axis is directed along radon transport and perpendicular to the sample surface), in metre;

  λ is the radon decay constant, in per second.

Note 1 to entry: Non stationary radon diffusion occurs during the time when radon activity concentration in the source container is not steady and in the time interval that immediately follows the moment when the steady concentration in the source container is established (long term test methods).

3.1.7
stationary radon diffusion
time independent radon diffusion through the sample; stationary radon diffusion is characterized by a stable (time independent) radon distribution within the sample and consequently by a stable radon surface exhalation rate from the sample into the receiver container (long term test methods);

one-dimensional stationary radon diffusion is described by the differential equation:

$$D \cdot \frac{\partial^2 C(x)}{\partial x^2} - \lambda \cdot C(x) = 0 \tag{4}$$

where

  D is the radon diffusion coefficient of the sample, in square metre per second;

  $C(x)$ is the distribution function along the axis "x" the radon activity concentration in the sample, in becquerel per cubic metre;

  x is the coordinate on axis "x" (the axis is directed along radon transport and perpendicular to the sample surface), , in metre;

  λ is the radon decay constant, in per second.

3.1.8
decisive measurements
measurement results used to calculate the radon diffusion coefficient

3.1.9
decisive volume of the chamber (container)
volume of the chamber (container) used to calculate the radon diffusion coefficient

3.1.10
decisive sample area
material sample area used to calculate the radon diffusion coefficient

3.1.11

minimum duration of the decisive measurement for non-stationary radon diffusion

such period of time in the frame of the decisive measurement of radon activity concentrations in the source and receiver containers taken during the phase of non-stationary diffusion ensuring the uncertainty of the radon diffusion coefficient assessment lower than $\pm$ 20 % (long term test methods)

3.1.12

minimum duration of the decisive measurement for stationary radon diffusion

such period of time in the frame of the decisive measurement of radon activity concentrations in the source and receiver containers taken during the phase of stationary diffusion ensuring the uncertainty of the radon diffusion coefficient assessment lower than $\pm$ 20 % (long term test methods)

3.1.13

minimum radon activity concentration in the source container

such concentration of radon in the source container which for the particular sample characterised by the d/l ratio ensures values of radon activity concentration in the receiver container measurable with uncertainty lower than 10 % (long term test methods)

3.1.14

radon transfer coefficient

expresses radon transport in thin boundary layer of air near the surface of the sample; in this boundary layer radon activity concentration on the surface of the sample equalizes with radon activity concentration in the surrounding air; for waterproof materials the default value of the radon transfer coefficient is 0,1 m.s^{-1}

3.2 Symbols

For the purposes of this document, the symbols given in ISO 11665-1 and the following apply.

D	Radon diffusion coefficient of the sample, in square metre per second
$D^{\triangleleft}$	Lower limit of the confidence interval of radon diffusion coefficient of the sample, in square metre per second
$D^{\triangleright}$	Upper limit of the confidence interval of radon diffusion coefficient of the sample, in square metre per second
D_M	Radon diffusion coefficient of the material, in square metre per second
λ	Radon decay constant, in per second
λ_L	Radon leakage rate, in per second
$\hat{\lambda}_L$	Best estimate of the radon leakage rate, in per second
C	Radon activity concentration in the sample, in becquerel per cubic metre
C_{sd}	Radon activity concentration in a source-detect chamber, in becquerel per cubic metre
C_0	Radon activity concentration in a source-detect chamber at the initial time after injection of radon, in becquerel per cubic metre
C_{amb}	Radon activity concentration in the ambient air, in becquerel per cubic metre
C_m	Measured radon activity concentration in a source-detect chamber, in becquerel per cubic metre
d	Thickness of the sample, in metre
S_s	Decisive area of the sample, in square metre
V_{sd}	Decisive volume of the source-detect chamber, in cubic metre
h	Radon transfer coefficient, in metre per second

t	Time, in second
τ_i	Time of start of i measurement period, in second (or hour)
r_g	Gross count rate, in counts per second
r_g^{α}	Gross count rate from alpha-source, in counts per second
r_0	Background count rate, in counts per second
R_m	Measured rate of decrease of radon activity concentration in the chamber (of pulse count rate), rel
R_c	Calculated rate of decrease of radon activity concentration in the chamber, rel
R_L	Calculated rate of decrease of radon activity concentration in the chamber at verification of radon-tightness, rel
$R_{\min}$	Function of the minimum measured rate of decrease in the chamber, rel
$R_{\max}$	Function of the maximum measured rate of decrease in the chamber, rel
$u(y)$	Standard uncertainty of the value of y
$s(y)$	Standard deviation of the value of y
$u_{rel}(D)$	Relative uncertainty of radon diffusion coefficient in the sample, rel
k	Coverage factor
N	Number of samples of the test material

4 Principle

Two type of testing methods can be performed to assess the radon diffusion coefficient in waterproofing materials.

Rapid testing methods are based on the measurement of the decrease over time of the radon activity concentration in a source-detect container in contact with one side of the tested membrane. The test is performed in a non-stationary mode. These methods can be used for single-layer waterproof materials when testing results are needed rapidly such as for screening purposes and for quality control during the membrane production. They are not applicable for multi-layer waterproof materials that do not meet the requirements of 6.11, Fig. 7.

Long term testing methods are based on the measurement of the decrease over time of the radon activity concentration in a source container in contact with one side and the ingrowth of radon in a container on the opposite side of the tested membrane. The test can be performed either in a transitory mode or a non-stationary mode. This method can be used for single and multi-layered waterproof materials. Applicability range depends on the ability to deliver sufficiently high radon concentrations in the source container such that they are measurable with sufficient accuracy within the receiver container.

5 Preparation of samples – General consideration

At least 3 samples are required for testing accordingly to ISO11929. If the condition of Equation (35) is not satisfied after testing three samples, additional samples of the material should be prepared and tested until this condition is fulfilled.

The diameter of the circular sample or the side of a rectangular sample shall be at least 5 times greater than the thickness of the sample. The minimum decisive sample area shall be 0,004 m^2 at least.

The samples are cut out from the prefabricated membranes at a minimum distance of 100 mm from the edges of the membrane.

In the case of coatings, paints, sealants or other waterproof materials prepared on site, at least 4 samples are required for testing. Samples can be produced by applying a coating, paint or sealant on a non-absorbing flexible underlay material (for example wax-paper, cellophane foil etc.) that is removed from the sample after the drying process is completed. The underlay shall not react with the applied coatings, paints or sealants. Approximately uniform thickness of the samples can be achieved with the help of guide gibs (paint, coating or sealant is poured or pasted between the gibs of uniform height and the excessive material is removed by drawing the steel float over the gibs). The samples should not be tested until the drying and hardening processes are completed. The time between the sample preparation and the start of the measurement as well as the storing conditions shall correspond to the recommendation of the producer.

The thickness of each sample is measured with accuracy of $\pm$ 0,01 mm ($\pm$ 0,005 mm for thicknesses from 0,1 to 0,2 mm) at 4 points per 0,05 m^2 placed uniformly along the surface of the sample. The resulting thickness of each sample is the arithmetic mean of all measurements on the sample. If a radon-permeable surface coating is a part of the tested material, its thickness is not included in the thickness of the tested sample. This type of surface coating can be removed from the sample before performing the test. If a metallic foil is a part of the tested material, the thickness of the metallic foil only is considered as the thickness of the tested sample when calculating the radon diffusion coefficient.

If the aim of the test is to verify the radon-tightness of the joint between membranes, the effective dimension of the sample in the direction that is perpendicular to the longitudinal axis of the joint shall exceed the width of the overlap by at least 20 mm on each side of the overlap. The thickness of samples with a joint corresponds to the thickness of a membrane. If membranes with radon-permeable surface coatings are jointed and the application rules do not require removal of the surface coating at the place of the jointing, the radon-permeable surface coating shall not be removed from the sample before performing the test.
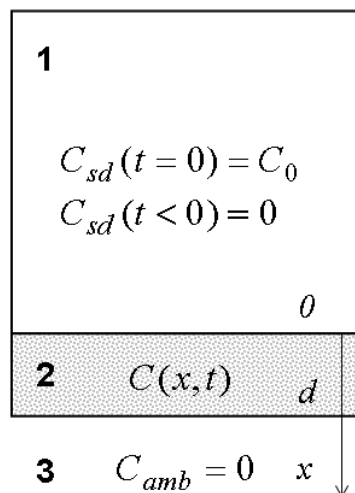
6 Rapid test method

6.1 Principle

A sample of the tested material is installed in the sealed end of the cylindrical chamber (See Figure 1). The detector of radon activity is located at the other end of the chamber.

Key

1   source-detect chamber

2   tested sample

3   ambient air

Figure 1 — Measurement scheme

In the beginning of the test, a highly active portion of radon is injected into the chamber through a special valve. Since then, radon activity concentration in the chamber begins to decline because of:

a)   diffusion of radon through the sample towards the ambient air;

b)   radon decay;

c)   leakage of radon from the chamber.

The chamber serves as a source radon and also allows to measure radon activity in the chamber.

The process of radon transfer from the chamber through the sample is described by the equations:

$$\frac{\partial C_{sd}(t)}{\partial t} = -\lambda \cdot C_{sd}(t) - \frac{S_s}{V_{sd}} \cdot h_1 \cdot \left[C_{sd}(t) - C(0,t) \right], \tag{5}$$

$$\frac{\partial C(x,t)}{\partial t} = D \frac{\partial^2 C(x,t)}{\partial x^2} - \lambda \cdot C(x,t), \ \ 0 \leq x \leq d, \ 0 \leq t < \infty, \tag{6}$$

with the following boundary conditions:

$$C_{sd}(t=0) = C_0, \ \ C(x,0) = 0, \tag{7}$$

$$-D \frac{\partial C(0,t)}{\partial x} = h_1 \cdot \left[C_{sd}(t) - C(0,t) \right], \tag{8}$$

$$D \frac{\partial C(d,t)}{\partial x} = h_2 \cdot \left[C(d,t) - C_{amb}(t) \right], \ (C_{amb}(t) = 0), \tag{9}$$

Equations (5) - (9) with respect to the values of radon activity concentration in the chamber are solved as the function:

$$C_{sd}(t) = C_0 \cdot f(t, D, \lambda, V_{sd}, S_s, d), \tag{10}$$

which is calculated as

$$C_{sd}(t) = C_0 \left[\int_0^t e^{\lambda(\tau - t)} F(\tau) d\tau + e^{-\lambda \cdot t} \right]. \tag{11}$$

The calculation by formula (11) is carried out by the algorithm described in Annex A.

During the test, the ratios between current radon activity concentrations in the chamber to radon activity concentration at the beginning of the decisive measurements are registered. These ratios determine the rate of radon activity concentration decrease in the chamber (6.5).

The radon diffusion coefficient in the sample is calculated according to 6.6, taking into account the effect of radon leakage from the chamber.

6.2   Equipment

The scheme of the test installation is shown in Figure 2.