भारतीय मानक Indian Standard

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पर्यावरण मे रेड़ियोधर्मिता का मापन — वायु : रेडॉन - 222 भाग 13 जलरोधी सामग्री मे प्रसार गुणांक का निर्धारण झिल्ली की दोनों तरफ गतिविधि सांद्रता की मापन विधि Measurement of Radioactivity in the

Environment — Air Radon - 222

Part 13 Determination of the Diffusion Coefficient in Waterproof Materials Membrane Two-Side Activity Concentration Test Method

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NATIONAL FOREWORD

This Indian Standard which is identical with ISO 11665-13 : 2017 'Measurement of radioactivity in the environment — Air : radon-222 — Part 13: Determination of the diffusion coefficient in waterproof materials: membrane two-side activity concentration test method' issued by the International Organization for Standardization (ISO) was adopted by the Bureau of Indian Standards on the recommendation of the Nuclear Energy for Peaceful Applications Sectional Committee and approval of the Chemical Division Council.

Under the general title 'Measurement of radioactivity in the environment — Air : radon-222' this Indian Standard is being published in several other parts. This part is an adoption ISO 11665-13 : 2017 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.

This Indian Standard is published in several parts. The other parts in this series are:

- a) Part 1 Origins of radon and its short-lived decay products and associated measurement methods
- b) Part 2 Integrated measurement method for determining average potential alpha energy concentration of its short-lived decay products
- c) Part 3 Spot measurement method of the potential alpha energy concentration of its short-lived decay product
- d) Part 4 Integrated measurement method for determining average activity concentration using passive sampling and delayed analysis
- e) Part 5 Continuous measurement methods of the activity concentration
- f) Part 6 Spot measurement methods of the activity concentration
- g) Part 7 Accumulation method for estimating surface exhalation rate
- h) Part 8 Methodologies for initial and additional investigations in buildings
- j) Part 9 Test methods for exhalation rate of building materials
- k) Part 11 Test method for soil gas with sampling at depth
- m) Part 12 Determination of the diffusion coefficient in waterproof materials: membrane one-side activity concentration measurement method

The text of ISO Standard has been approved as suitable for publication as an Indian Standard without deviations. Certain conventions and terminologies are, however, not identical to those used in Indian Standards. Attention is particularly drawn to the following:

- a) Wherever the words 'International Standard' appear referring to this standard, they should be read as 'Indian Standard'
- b) Comma (,) has been used as a decimal marker in the International Standard, while in Indian Standards, the current practice is to use a point (.) as the decimal marker.

In this adopted standard, reference appears to certain International Standards for which Indian Standards also exist. The corresponding Indian Standards, which are to be substituted in their respective places, are listed below along with their degree of equivalence for the editions indicated:

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Introduction

Radon isotopes 222, 219 and 220 are radioactive gases produced by the disintegration of radium isotopes 226, 223 and 224, which are decay products of uranium-238, uranium-235 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^[5].

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. UNSCEAR^[Z] 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 document 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.

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 ISO 11665 and ISO 13164 series for water^[2].

Radon enters into buildings via diffusion mechanism caused by the all-time existing difference between radon activity concentrations in the underlying soil and inside the building, and via convection mechanism inconstantly generated by a difference in pressure between the air in the building and the air contained in the underlying soil. Indoor radon activity concentration depends on radon activity concentration in the underlying soil, 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 $Bq\cdotm^{-3}$ is recommended by the World Health Organization^[8]. Wherever this is not possible, this reference level should not exceed 300 $Bq\cdotm^{-3}$. This recommendation was endorsed by the European Community Member States that shall establish national reference levels for indoor radon activity concentrations. The reference levels for the annual average activity concentration in air shall not be higher than 300 $Bq\cdotm^{-3}$ [9].

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.

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 radon-proof insulation can be prescribed by national building standards and codes. Requirements for radon-proof insulation as regards the durability, mechanical and physical properties and the maximum

design value of the radon diffusion coefficient can also be prescribed by national building standards and codes.

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 the different methods described in ISO/TS 11665-12 and this document, as required by ISO/IEC 17025^[3], cannot be directly met.

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

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Indian Standard

MEASUREMENT OF RADIOACTIVITY IN THE ENVIRONMENT — AIR RADON - 222 PART 13 DETERMINATION OF THE DIFFUSION COEFFICIENT IN WATERPROOF MATERIALS MEMBRANE TWO-SIDE ACTIVITY CONCENTRATION TEST METHOD

1 Scope

This document 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.

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

2 Normative references

The following documents are referred to in the text in such a way that some or all of their content constitutes requirements 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 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 measurement 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 application

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

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

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 and ISO 80000-10 and the following apply.

ISO and IEC maintain terminological databases for use in standardization at the following addresses:

- IEC Electropedia: available at <u>http://www.electropedia.org/</u>
- ISO Online browsing platform: available at <u>https://www.iso.org/obp</u>

3.1.1

material

product 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 radon diffusion coefficient

3.1.3

radon diffusion coefficient

D

radon activity permeating due to molecular diffusion through unit area of a monolayer material of unit thickness per unit time at unit radon activity concentration gradient on the boundaries of this material

3.1.4

equivalent radon diffusion coefficient

 D_{eqv}

radon diffusion coefficient of the multilayer material that numerically equals to the radon diffusion coefficient of a homogeneous material of the same thickness as the layered material through which radon penetrates in the same amount as through the layered material

3.1.5

radon diffusion length

l

distance crossed by radon due to diffusion in which activity is reduced by "e" times because of decay

Note 1 to entry: Numeric "e" is the natural logarithm, equal to about 2,72.

Note 2 to entry: Radon diffusion length is expressed by the relationship given in the following formula:

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

(1)

where

- *l* is the radon diffusion length, in metres;
- *D* is the radon diffusion coefficient of the sample, in square metres per second;
- λ is the radon decay constant, in per second.

3.1.6

diffusive radon surface exhalation rate

Ε

value of the activity concentration of radon atoms that leave a material per unit surface per unit time

Note 1 to entry: For the purpose of this document, only the diffusion transport through the sample is taken into account. The diffusive radon exhalation rate is given by the following formula (Fick's law):

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

where

- *E*(*x*) is the distribution function along the axis "x" of the radon exhalation rate in the sample, in Becquerel per square metre per second;
- *C*(*x*) is the distribution function along the axis "x" of the radon activity concentration in the sample, in Becquerel per cubic metre;
- *D* is the radon diffusion coefficient of the sample, in square metre per second;
- *x* is the coordinate on axis "x" (the axis is directed along radon transport and perpendicular to the sample surface), in metre.

3.1.7

non-stationary radon diffusion

time-dependent radon diffusion through the sample when the radon activity concentration within the sample is changing (in dependence on time, distance from the surface exposed to radon and the radon activity concentration in the source container) and the radon surface exhalation rate from the sample into the receiver container is also changing

Note 1 to entry: One-dimensional non-stationary radon diffusion is described by the partial differential equation:

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

3.1.8

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)

Note 1 to entry: One-dimensional stationary radon diffusion is described by the differential equation:

$$D \cdot \frac{\partial^2 C(x)}{\partial x^2} - \lambda \cdot C(x) = 0$$
⁽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" of 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.

3.1.9

decisive measurement of radon activity concentrations

measurement of the time courses of radon activity concentrations in the source and receiver containers used for calculating the radon diffusion coefficient

Note 1 to entry: The duration of the decisive measurement can be shorter or the same as the duration of the test.

3.1.10

decisive volume of the container

V

volume of the container used to calculate the radon diffusion coefficient

3.1.11

decisive sample area

 $S_{\rm S}$

material sample area used to calculate the radon diffusion coefficient

3.1.12

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

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 $\pm 20~\%$

3.1.13

minimum duration of the decisive measurement for stationary radon diffusion

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 ± 20 %

3.1.14

minimum radon activity concentration in the source container

concentration of radon in the source container which for the particular sample characterized by the d/l ratio ensures values of radon activity concentration in the receiver container measurable with uncertainty lower than 10 %

3.1.15

radon transfer coefficient

radon transport in thin boundary layer of air near the surface of the sample

Note 1 to entry: In this boundary, layer radon activity concentration on the surface of the sample equalizes with radon activity concentration in the surrounding air.

Note 2 to entry: For waterproof materials, the default value of the radon transfer coefficient is 0,1 m $\cdot s^{-1}$

3.1.16

standard uncertainty of a variable

Χ

standard deviation of a variable *X*

3.1.17 relative uncertainty of a variable X $u(X) = k \cdot s(X)/EX$

where

- *EX* is the expected value of a variable *X*;
- k is the shrinkage factor (k = 1,96 by default for 95 % confidence interval).

3.2 Symbols

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

- λ radon decay constant, in per second
- λ_V radon leakage rate characterizing the ventilation of the receiver container, in per second
- *C* radon activity concentration in the sample, in Becquerel per cubic metre
- *C*_a radon activity concentration in a particular container of the measuring device, in Becquerel per cubic metre
- *C*_s radon activity concentration on the surface of the sample, in Becquerel per cubic metre
- *C*_{rc} radon activity concentration in the receiver container, in Becquerel per cubic metre
- *C*_{sc} radon activity concentration in the source container, in Becquerel per cubic metre
- *D* radon diffusion coefficient of the monolayer sample, in square metre per second
- D_{eqv} equivalent radon diffusion coefficient of the multilayer sample, in square metre per second
- *d* thickness of the sample, in metre
- *E* diffusive radon surface exhalation rate, in Becquerel per square metre per second
- $E_{\rm rc}$ diffusive radon surface exhalation rate from the sample to the receiver container, in Becquerel per square metre per second
- *h* radon transfer coefficient, in metre per second
- *l* radon diffusion length, in metre
- $S_{\rm s}$ decisive area of the sample, in square metre
- *t* time, in second
- Δt duration of the considered time step between time t_{i-1} and t_i , in second
- *V* decisive volume of the receiver container, in cubic metre
- *x* distance within the tested sample measured from the surface of the sample exposed to radon, in metre
- *u*(*X*) relative uncertainty of a variable *X*, in relative units
- *s*(*X*) standard uncertainty of a variable *X*, in same units as variable X

4 Principle of the test method

The sample of the tested material is placed between the air-tight source and the receiver containers, and the joint is carefully sealed.

Radon activity concentration in both containers shall be measured using continuous or spot measurement methods as specified in ISO 11665-5 and ISO 11665-6.

By means of the radon source with stable radon production rate, the radon activity concentration in the source container is kept on a high level (usually within the range $1 \text{ MBq} \cdot \text{m}^{-3}$ to $100 \text{ MBq} \cdot \text{m}^{-3}$). The radon that diffuses through the sample is monitored using calibrated radon monitor in the receiver container.

Using an appropriate mathematical process (either analytical or numerical), the radon diffusion coefficient is afterwards calculated from the time-dependent courses of the radon activity concentrations measured in the source and receiver containers, and the area and thickness of the tested sample. In case of multilayer samples, the above described principle results in determination of the equivalent radon diffusion coefficient D_{eqv} .

5 Measuring system

5.1 Components of the measuring system

The measuring system for determining the radon diffusion coefficient in the waterproof materials shall comprise the following components:

- a) at least two air-tight containers (source and receiver), each with a minimum air volume of $0,5 \times 10^{-3}$ m³ or when the spot measurement method for radon activity concentration is going to be used, the minimum air volume should be at least 10 times larger than the total volume of spot samples taken from each of the containers during the test performance, and made from metal materials (for example, aluminium, stainless steel, etc.) of a thickness at least 5×10^{-4} m that effectively eliminates radon transport between the air inside and outside the containers; each container shall be equipped with a test area of at least 5×10^{-3} m² surrounded by flanges for fixing the tested material; the minimum width of the flanges shall be 0,01 m and their arrangement shall eliminate the transport of radon from the source container to the receiver container; each container shall be further equipped with an appropriate number of valves intended for ventilating the containers, for measuring the pressure differences between the containers, for extracting air samples for control measurements of radon activity concentration and for connecting to the radon source;
- b) a measuring instrument capable of determining the thickness of the tested sample with accuracy $\pm 0,01$ mm (maximum standard relative uncertainty of measurement 5 %);
- c) a source of radon with stable radon production rate capable of creating a radon activity concentration in the source container within the range $1 \text{ MBq} \cdot \text{m}^{-3}$ to $100 \text{ MBq} \cdot \text{m}^{-3}$;
- d) an air-tight flow pump with the range of air flow rates $6 \times 10^{-3} \text{ m}^3/\text{h}$ to $30 \times 10^{-3} \text{ m}^3/\text{h}$ that is used in some measurement methods in a closed circuit with a radon source and a source container;
- e) a calibrated measuring device for monitoring the radon activity concentration in the receiver container with standard relative uncertainty 10 % and a dynamic measuring range from $500 \text{ Bq} \cdot \text{m}^{-3}$ to 1,0 MBq $\cdot \text{m}^{-3}$;
- f) a calibrated measuring device for monitoring the radon activity concentration in the source container with standard relative uncertainty 10 % and a dynamic measuring range from $10 \text{ kBq}\cdot\text{m}^{-3}$ to 100,0 MBq·m⁻³;
- g) a measuring instrument for determining the relative pressure difference between the air volume in the source container and the air volume in the receiver container with standard relative uncertainty of 10 % and a dynamic measuring range from 1 Pa to 150 Pa;

h) suitable sensors and a data storage system capable of continuously monitoring the temperature and relative humidity of air, atmospheric pressure and radon activity concentration in the place where the measuring device is positioned.

5.2 Configuration of the measuring system

In the simplest case, the measuring system can comprise one source container, one receiver container and a radon source connected to the source container (see Figure 1). If more than one sample is to be measured under equal conditions, it is convenient to use a measuring system comprising more than one receiver container assembled on one source container (see Figure 2), or a set of pair containers (source + receiver) connected to the radon source in a parallel circuit (see Figure 3) or connected to each other and to the radon source through the source containers in a serial circuit.



Кеу

- 1 receiver container
- 2 radon detector
- 3 tested sample
- 4 source container
- 5 pump
- 6 radon source

Figure 1 — Measuring system comprising one receiver container and one source container



Key

- 1 receiver container
- 2 radon detector
- 3 tested sample
- 4 source container
- 5 pump
- 6 radon source

Figure 2 — Measuring system comprising two receiver containers assembled on one source container



Key

- 1 receiver container
- 2 radon detector
- 3 tested sample
- 4 source container
- 5 pump
- 6 radon source

Figure 3 — Measuring system comprising two pair containers (source + receiver) connected to the radon source in a parallel circuit

The radon source shall be connected to the source container and the source containers shall be interconnected by flexible pipes that are as radon-tight as possible.

Radon can be transported from the radon source to the source container only by diffusion or with the help of a flow pump. If a flow pump is used, the radon source, the source container and the flow pump shall be in a single, closed circuit. A flow pump shall not be applied if the radon diffusion through the tested sample is influenced by the pressure difference between the source and receiver containers (this can be seen as rapid drop or rise of radon activity concentration in the receiver container after applying the pump).

If the measuring system is made up of a set of pair containers, the maximum number of pairs that can be connected to one radon source in a single closed circuit is four. A flow pump shall be an indispensable part of the circuit. Only samples of one material shall be tested in a single circuit at the same time.

If the measuring system is made up of more than one receiver container assembled on a single source container, only samples of one material shall be tested in this system at the same time.

Radon source can also be placed inside the source container. This arrangement is convenient for measuring systems comprising several receiver containers assembled on one source container.

If the source and/or receiver containers serve as radon detectors as well, then the sensitivity of these detectors to the gamma radiation should be taken into account in the arrangement of the source of radon inside the source container.

6 Test methods

6.1 General information

A suitable test method is selected from the following options in dependence on the measuring system, sampling method and properties of the tested material (especially its thickness and the assumed radon diffusion coefficient value). Methods A and B are convenient for continuous monitoring of radon activity concentrations and method C for spot measurements. Method A shall be used when the stationary radon diffusion is not established due to the characteristics of the tested material and/or the measuring device, lack of time, etc. Method B is usually preceded by method A. Method C gives the highest radon exhalation rates and therefore it should be applied every time when very low radon diffusion coefficient is expected. Radon diffusion coefficient determined by methods A, B and C requires numerical solution as described in 7.4. The linear part of build-up curve obtained at the beginning of the decisive measurement as described in methods B and C permits the use of analytical solution if conditions described in 7.5, fourth paragraph and 7.6, third paragraph are satisfied.

6.2 Method A — Determining the radon diffusion coefficient during the phase of nonstationary radon diffusion

After placing the sample between the source and receiver containers, both containers are closed and radon is admitted into the source container. The decisive measurement of radon activity concentrations in both containers begins at this moment (see Figure 4).



Кеу

- X time (without scale)
- Y radon concentration (without scale)
- 1 source container
- 2 receiver container
- 3 decisive measurement under non-steady-state conditions

Figure 4 — Test procedure according to method A

6.3 Method B — Determining the radon diffusion coefficient during the phase of stationary radon diffusion

After placing the sample between the source and receiver containers, both containers are closed and radon is admitted into the source container. The time-dependent increase in radon activity concentrations in both containers is monitored. After establishing stationary radon diffusion through the sample (time needed for establishing stationary radon diffusion can be taken from Figure 7), the receiver container is flushed with radon-poor ambient air. Flushing is stopped when the radon activity concentration in the receiver container decreases below the operational threshold (at least below 1 kBq·m⁻³). The decisive measurement of radon activity concentrations in both containers begins at this moment (see Figure 5).



Key

- X time (without scale)
- Y radon concentration (without scale)
- 1 source container
- 2 receiver container
- 3 decisive measurement under steady-state conditions
- 4 flushing

Figure 5 — Test procedure according to method B

6.4 Method C — Determining the radon diffusion coefficient during the phase of stationary radon diffusion established during ventilation of the receiver container

After placing the sample between the source and receiver containers, radon is admitted into the source container and the time-dependent increase in the radon activity concentrations in both containers is monitored. The radon activity concentration in the receiver container is held at values below the operational threshold (at least below 1 kBq·m⁻³) by means of continuous ventilation of the receiver container. After establishing the stationary radon diffusion through the sample (time needed for establishing stationary radon diffusion can be taken from Figure 7), the ventilation of the receiver container is stopped. The decisive measurement of radon activity concentrations in both containers begins at this moment (see Figure 6).



Кеу

- X time (without scale)
- Y radon concentration (without scale)
- 1 source container
- 2 receiver container
- 3 decisive measurement under steady-state conditions
- 4 flushing

Figure 6 — Test procedure according to method C

7 General application procedures

7.1 Preparation of samples

The diameter or the side of a rectangular sample shall be at least five times greater than the thickness of the sample. The minimum effective area of the sample (the area exposed to radon) shall be 0,005 m² at least.

The samples are cut out from the prefabricated membranes at a minimum distance of 100 mm from the edges of the membrane. At least three samples are required for testing.

In the case of coatings, paints, sealants or other waterproof materials prepared on site, at least four 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 shall 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 at four points per 0,05 m² 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.

Radon diffusion coefficient of monolayer materials produced in several thicknesses shall be determined for one selected thickness only and this value applies for all thicknesses. Radon diffusion coefficient of multilayer materials produced in several thicknesses shall be determined separately for each thickness.

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.

7.2 Fixing the samples in the measuring device

Samples of multilayer materials shall be placed in the measuring device in such a position that radon diffuses through the sample in the direction corresponding to real conditions. If in this position a radonpermeable surface coating is exposed to the receiver container, the surface coating shall be removed from the sample to avoid radon loss through this coating.

The sample is placed between the flanges of both containers on which a permanently elastic sealant (for example, on the acrylic, silicone, bee wax basis, etc.) has been applied. Over the whole duration of the test, the sealant shall not be subjected to physical or chemical changes and shall not cause physical or chemical damage to the sample. In horizontal positions, samples that are not self-supporting (for example, thin layers of paints, thick and heavy layers of unreinforced bitumen coatings that tend to deflect due to their high weight) shall be placed on the self-supporting wire screen fixed by the sealant to the flanges of the source container. The wire screen supports the sample over the whole duration of the test. The wire screen should be of such a construction that its influence on the decisive area of the sample can be neglected.

7.3 Test of radon-tightness, assessment of the radon leakage rate of the receiver container

The radon leakage rate of the receiver container shall be determined after installing the sample between the source and the receiver container to check the radon-tightness of the joint between the sample and the container. A high radon activity concentration is injected through the valve into the receiver container, and then the valve is closed. The decrease in radon activity concentration in the receiver container is monitored. The intensity of the ventilation (the radon leakage rate) shall be determined from deviations from the decay law (the radon transport through the sample shall be incorporated into the total radon losses). If the fixing of the sample in the measuring system is radon-tight, the radon leakage rate shall be lower than 0,003 78 h⁻¹ (half of the radon decay constant).

The sealing ability of different sealants and the radon-tightness of the receiver container can be verified in a similar manner. In this case, the sample of insulating material is replaced by a metal sheet.

7.4 Determining the radon diffusion coefficient according to method A

The decisive measurement of radon activity concentrations performed simultaneously in both containers begins after placing the sample in the measuring device and not later than 1 h after admitting radon into the source container. Time interval between successive records of concentrations shall not exceed 6 h during the first 5 d of the decisive measurement and 12 h during the following days. The duration of the decisive measurement shall be longer than the minimum duration of the decisive measurement for non-stationary diffusion, according to Figure 7 (see 7.7).

For at least two thirds of the duration of the decisive measurement, the radon activity concentration in the source container shall be higher than the minimum radon activity concentration, according to Figure 8 (see 7.7).

The radon diffusion coefficient is determined by an iterative procedure based on repeated numerical solutions of <u>Formula (3)</u>. During this calculation, the value of radon diffusion coefficient as the variable

in the numerical solution gradually grows from the assumed lower limit to the assumed upper limit. The final radon diffusion coefficient is the value that results in the numerical solution of Formula (3) for which the differences between the calculated and measured concentrations in the receiver container are minimal.

In the numerical solution of <u>Formula (3)</u>, the time-dependent boundary conditions on both surfaces of the tested sample shall be respected according to <u>Formula (5)</u>:

$$-D\frac{\partial C}{\partial x} = h \cdot (C_{\rm s} - C_{\rm a}) \tag{5}$$

The radon activity concentration, C_a , means either the radon activity concentration in the source container, C_{sc} , or the radon activity concentration in the receiver container, C_{rc} . In the calculation process, the radon activity concentration in the source container, C_{sc} , is always taken in accordance with the measured values. The initial radon activity concentration in the receiver container, $C_{rc,0}$, at the beginning of the calculation at time t = 0 is considered as the concentration measured at that time. In the following time steps of the numerical solution, the radon activity concentration in the receiver container is calculated according to Formula (6):

$$C_{\mathrm{rc},i} = C_{\mathrm{rc},i-1} \cdot e^{-(\lambda + \lambda_{\mathrm{v}}) \cdot \Delta t} + \frac{E_{\mathrm{rc},i-1} \cdot S_{\mathrm{s}}}{V \cdot (\lambda + \lambda_{\mathrm{v}})} \cdot (1 - e^{-(\lambda + \lambda_{\mathrm{v}}) \cdot \Delta t})$$
(6)

No solution of <u>Formula (4)</u> describing stationary radon diffusion can be used for determining the radon diffusion coefficient according to method A.

7.5 Determining the radon diffusion coefficient according to method B

After placing the sample in the measuring device and admitting radon into the source container, the radon activity concentrations in the source and receiver containers are simultaneously recorded at time intervals not longer than 12 h. Flushing of the receiver container with radon-poor ambient air starts after establishing stationary radon diffusion through the sample. Flushing is brought to an end when the radon activity concentration in the receiver container decreases below the operational threshold (at least below 1 kBq·m⁻³) that takes into consideration variable measurement conditions, different measurement methods and detection principles and corresponding minimal detection limits. Ventilation of the receiver container shall not influence the steady concentration in the source container. After bringing the ventilation to an end and closing the receiver container, decisive measurement begins of the radon activity concentrations, performed simultaneously in both containers. The radon activity concentrations performed simultaneously in both containers. The radon activity concentration between successive records of concentrations and the duration of the determination of the receiver depend on the mathematical procedure applied for the determination of the radon diffusion coefficient.

If the procedure stated for method A is used, the time interval between successive records of concentrations shall not exceed 6 h and the duration of the decisive measurement shall be longer than 72 h. The numerical solution shall take into account that the process of saturation of the sample with radon was finished before flushing the receiver container, and therefore, it cannot be restricted to the time interval in which the decisive measurement was performed. Before starting the iterative process for the time interval of the decisive measurement, the theoretical distribution of the radon activity concentrations in the source and receiver containers measured before flushing. Only then can the surface of the sample exposed to the receiver container be subjected to the boundary condition with $C_{\rm rc,0}$ corresponding to the radon activity concentration measured in the receiver container after flushing.

Decisive measurement is in case of method B performed during stationary radon diffusion, which enables to shorten its duration to at least 72 h in comparison with method A.

If after flushing, the increase in radon activity concentration in the receiver container is linear, the radon diffusion coefficient can also be determined by an analytical solution of Formula (4) under

boundary conditions taking into account the real situation. This approach can be used only under the following conditions, which shall be fulfilled at the same time:

- a) the radon activity concentration in the receiver container is recorded at time intervals not longer than 1 h and at least four records are required;
- b) the value of the correlation coefficient, *R*, of the measured radon activity concentration in the receiver container is at least 0,9 and the measured slope is significantly >0.

Finding an analytical solution is not possible without assuming constant boundary conditions. To minimize the errors and uncertainties caused by this simplifying assumption, the duration of the decisive measurement shall be as short as possible. Thus the decisive measurement for analytical solution shall not be longer than approximately 6 h. This limits the time interval between successive records of radon activity concentration measurements to 1 h in maximum. On the other hand, accuracy of a numerical solution with time-dependent boundary conditions [see Formula (5)] increases with increasing duration of the decisive measurement. Since the minimum required duration of the decisive measurement is 72 h, intervals between successive records can be prolonged up to 6 h.

Throughout the duration of the decisive measurement, the radon activity concentration in the source container shall be in the steady state (the coefficient of variation for data recorded during the last 24 h shall be lower than 10 %), and at the same time it shall be higher than the minimum radon activity concentration, according to Figure 8 (see 7.7).

7.6 Determining the radon diffusion coefficient according to method C

After placing the sample in the measuring device and admitting radon into the source container, the radon activity concentrations in the source and receiver containers are simultaneously recorded at time intervals not longer than 12 h. The radon activity concentration in the receiver container is held at values as low as achievable for particular measurement conditions, different measurement methods and detection principles and corresponding detection limits (at least below 1 kBq·m⁻³) by continuously ventilating the receiver container. After establishing stationary radon diffusion through the sample, ventilation of the receiver container is brought to an end and the container is closed. At this moment, the decisive measurement of radon activity concentrations begins, and is performed simultaneously in both containers. The radon activity concentrations shall be read for the first time not later than 10 min after closure of the receiver container. The time interval between successive records of concentrations and the duration of the decisive measurement depend on the mathematical procedure applied for the determination of the radon diffusion coefficient.

If the procedure stated for Method A is used, the time interval between successive records of concentrations shall not exceed 6 h and the duration of the decisive measurement shall be longer than 72 h. The numerical solution shall take into account that the process of saturation of the sample with radon was finished during ventilation of the receiver container, and it therefore cannot be restricted to the time interval in which the decisive measurement was performed. Before starting the iterative process for the time interval of the decisive measurement, the theoretical distribution of the radon activity concentration within the sample shall correspond to the radon activity concentrations in the source and receiver containers measured before closure of the receiver container.

If, after closing the receiver container, the increase in radon activity concentration in the container is linear, the radon diffusion coefficient can also be determined by an analytical solution of Formula (4), in accordance with the conditions stated for method B.

Throughout the duration of the decisive measurement, the radon activity concentration in the source container shall be in a steady state (the coefficient of variation for data recorded during the last 24 h shall be lower than 10 %), and at the same time it shall be higher than the minimum radon activity concentration, according to Figure 8 (see 7.7).

NOTE For a specific device and initial and boundary conditions, the solution of Formula (4) is described in Annex A.

7.7 General requirements for performing the tests

Measurements according to any method should be performed under stable laboratory conditions characterized by the temperature of the surrounding air in the place where the measuring system is installed 24 °C \pm 5 °C and the relative humidity of the surrounding air in range between 20 % and 70 %.

The relative pressure difference between the source and receiver containers should not differ from 0 of more than 5 Pa.

The minimum duration of the decisive measurement for non-stationary radon diffusion (method A) shall be assessed in dependence on the d/l ratio (thickness of the sample divided by the assumed radon diffusion length value) and the rate of the radon activity concentration increase in the source container, according to Figure 7. The diffusion length is assumed with respect to the results of previous measurements of samples of similar chemical composition^[10]. If the chemical composition is not known, the minimum duration of the decisive measurement shall be at least 400 h. Duration of the decisive measurement defined according to Figure 7 ensures that the radon diffusion coefficient determined for non-stationary diffusion does not differ more than of 10 % from the value determined for stationary radon diffusion.

For the purpose of methods B and C, it is assumed that time needed for establishing stationary radon diffusion can be also taken from Figure 7.

The curve S in Figure 7 shall be applied for slow increase of radon activity concentration in the source container (the steady state in the source container is reached at the time t = 700 h after radon has been admitted into the source container), the curve F for fast increase of radon activity concentration in the source container (the steady state in the source container is reached at the time t = 200 h after radon has been admitted into the source container) and the curve K shall be used if the sample is exposed to the steady state radon activity concentration in the source container.

General conditions applied for the construction of <u>Figure 7</u> are the following:

- the radon activity concentration in the receiver container should exceed 1 000 Bq·m⁻³ during the measurement interval of 800 h;
- the radon leakage rate of the receiver container is lower than the decay constant of radon;
- S_s/V ratio is around 0,07 cm⁻¹.

The minimum duration of the decisive measurement for stationary radon diffusion (methods B and C) shall be assessed in dependence on the mathematical procedure applied for the determination of the radon diffusion coefficient. Details are presented in <u>7.5</u> and <u>7.6</u>.



Figure 7 — Minimum duration of the decisive measurement for non-stationary radon diffusion

The minimum radon activity concentration in the source container shall be determined in dependence on the thickness of the sample and the assumed radon diffusion coefficient value, according to Figure 8. The radon diffusion coefficient is assumed with respect to the results of previous measurements of samples of similar chemical composition^[8]. General conditions applied for the construction of Figure 8 are identical to those for Figure 7.



Figure 8 — Minimum radon activity concentration in the source container

If the assessed value of the radon diffusion coefficient of the sample is lower than expected when the minimum duration of the decisive measurement is assessed according to Figure 7 and the minimum radon activity concentration in the source container assessed according to Figure 8, and consequently values of these parameters are underestimated of more than 30 %, the measurement should be repeated, adjusting the duration and the radon activity concentration in the source container to the ascertained radon diffusion coefficient.

8 Influence quantities

The main factor influencing the test results is radon leakage from the receiver container. Therefore, the radon leakage rate, λ_{v} , of the receiver container shall be derived for any tested material according to 7.3.

Extra attention should be also paid to the following quantities:

- the relative pressure difference between the source and receiver container;
- the temperature of the surrounding air where the measuring system is installed;
- the relative humidity of the surrounding air where the measuring system is installed;
- the atmospheric pressure of the surrounding air where the measuring system is installed;
- the occurrence of α , β and/or γ sources in the place where the measuring system is installed, which are not a subject of measurement during the whole period of decisive measurement (α , β and γ radiation may influence the response of some detectors and therefore their rate should be measured and the response corrected);
- all other quantities which may influence the accuracy of devices used for measurement.

It is recommended to provide measurement conditions such that none of the above-mentioned influencing factors induces variability in the result higher than ± 25 %.

9 Expression of results

9.1 Relative uncertainty

The uncertainties of all input parameters shall be derived in accordance with ISO/IEC Guide 98-3.

For such a complex system as the diffusion of radon through material, the best option is Monte Carlo simulation. There exists no explicit solution to the second Fick's law of diffusion and the functional relationship between all the input quantities, input errors, etc. and the measurement is very complex. According to JCGM 100:2008^[5], a feasible way to evaluate the uncertainty (Type A evaluation) is from data obtained experimentally (experimentally obtained values of diffusion coefficient of radon) under repeatability conditions or from data obtained via the simulation methods. To obtain sufficient amount of experimental data is not feasible within reasonable time period, hence, the simulation methods are recommended. All of the sources of uncertainty should be considered in such simulation. The major sources of uncertainty of measurement of the radon diffusion coefficient are measurement of radon concentration in the source and receiver container in each of the time intervals, the uncertainty of measurement of the tested sample, and the uncertainty of radon leakage rate.

Detailed description should be prepared by particular laboratories and should reflect uncertainties of the measurement technique and arrangements applied in the particular laboratory. Based on several tests and measurements, the laboratory should obtain a range of input values for the above mentioned uncertainties. These data then serve as input for Monte Carlo simulation to obtain the standard uncertainty of radon diffusion coefficient.

Based on experimental data, the following basic formula for calculation of relative uncertainty of calculated radon diffusion coefficient may be used as first approach for laboratories before establishing their own model:

$$u(D) = \frac{k}{5} \sqrt{37 [u(C)/k]^2 + 34 [u(C)/k]^4 + 31 [u(d)/k]^2}$$
(7)

where

k is the shrinkage factor (*k* = 1,96 by default for 95 % confidence interval);

u(*D*) is the relative uncertainty of *D*, relative units;

u(*d*) is the relative uncertainty of measured thickness, *d*, relative units;

u(C) is the relative uncertainty of radon activity concentration measurement, C, relative units.

Standard uncertainty of calculated radon diffusion coefficient, *D*, can be calculated as Formula (8):

$$s(D) = u(D) \cdot D \tag{8}$$

where

u(D) is the relative uncertainty of calculated radon diffusion coefficient calculated via Monte Carlo simulation or from Formula (7) for k = 1.

9.2 Decision threshold and detection limit

The characteristic limits of radon activity concentration measurement shall be assessed for each of the detection methods in accordance with ISO 11929.

9.3 Limits of the confidence interval

The lower and upper limits, D^l and D^u , of $1 - \alpha$ confidence interval for radon diffusion coefficient is calculated using Formulae (9) and (10):

$$D^{l} = D - k \cdot s(D) \tag{9}$$

$$D^u = D + k \cdot s(D) \tag{10}$$

where

- *D* is the calculated radon diffusion coefficient;
- *s*(*D*) is the standard uncertainty of calculated radon diffusion coefficient;
- *k* is the shrinkage factor equal to $1 \alpha/2$ quantile of standard normal distribution. Usually k = 1,96 to obtain a 95 % confidence interval.

10 Quality management and calibration of the test device

The metrological traceability of the measuring system shall be ensured by the calibration assessment of the radon activity concentration in the reference atmosphere with the known level of radon activity concentration, and shall be carried out at an authorized metrological centre or laboratory. The test device may be subject of comparison measurement on the national and/or international level.

11 Test report

The test report shall contain the following entries:

- a) a reference to this document, i.e. ISO/TS 11665-13;
- b) identification of the testing laboratory;
- c) identification of tested material (name, producer, composition);
- d) identification of the test method (A, B or C);
- e) details about samples: thickness, decisive area, whether the sample contains joints, information whether the sample was prepared by the testing laboratory or by the client;
- f) duration of the test (decisive measurement);
- g) average and the standard deviation of radon activity concentration in the source container after the steady state in the source container is reached;
- h) information about the experimental conditions during the test: air temperature and relative humidity, pressure difference between the source and receiver container;
- i) result of determining the radon diffusion coefficient, $D = D \pm k \cdot s(D)$ with the associated k value, and radon diffusion length of the material;
- j) date of test performance.

The following information shall be archived by the laboratory to be submitted when needed:

- a) details about sample preparation (surface, thickness), description of joints (type of joint, joint width), identification of the time between the sample preparation and the start of the test (not applicable for prefabricated membranes), storing conditions; radon diffusion coefficient of each sample;
- b) test arrangement;
- c) deviations from standard practice which may influence the test result;
- d) information about the measuring device;
- e) maximum, minimum, average and standard deviation radon activity concentration in the source container after the steady state in the source container is reached.

Annex A (informative)

Determining the radon diffusion coefficient during the phase of stationary radon diffusion according to method C

A.1 General

This annex deals only with the determination of the radon diffusion coefficient during the phase of stationary radon diffusion using a grab sampling method for the recording of the radon activity concentration in the useful volumes of the receiver and source containers.

Determining the radon diffusion coefficient of a tested sample involves measuring changes in the radon activity concentration in a given time, in the source and receiver containers with known geometric characteristics and with one open face applied to the surface of the sample under investigation.

It is assumed that back diffusion, radon leakage rate of the receiver container and radon decay are negligible in the time interval of the decisive measurement.

Radon leakage rate can be neglected only, if the test of radon-tightness performed according to 7.3 confirms that it is below 0,003 78 h⁻¹ and the duration of the decisive measurement is no longer than 6 h. Radon decay can be neglected, if the duration of the decisive measurement is also not longer than 6 h. The above-stated conditions ensure that the influence of both parameters on the slope of the linear regression straight line expressing the increase of radon concentration in the receiver container is negligible.

A.2 Additional terms and definitions

For the purpose of this annex, the following symbols and the symbols given in <u>Clause 3</u> apply.

- *N* number of counts per counting time Tc
- *N*⁰ scintillation cell background counts per the counting time Tc
- n_{α} number of alpha emitters present in the lucas cell per Becquerel of radon after time t elapsed between filling and counting the cell ($n_{\alpha} \approx 3$ at t = 3 h for 1 Bq of radon)
- *p* slope of the linear regression straight line, in Becquerel per cubic metre and per second
- *R*t calibration factor under the conditions of secular equilibrium between the radon and its short lived decay products, in counts per Becquerel
- *T*_c counting time, in per second
- *V*_f sensitive volume of the scintillation cell, in cubic metre

A.3 Equipment

The measuring system comprises two air-tight containers (see Figure A.1), each of them equipped with three valves intended for ventilating the containers, for extracting air samples for measurement of radon activity concentrations and for connecting to a certified radon source.



Key

- 1 receiver container
- 2 valve
- 3 tested sample
- 4 source container
- 5 pump
- 6 radon source

Figure A.1 — Measuring system for determining the radon diffusion coefficient using grab sampling method

A.4 Measurement procedure

The measurement is performed as follows:

- preparing the sample of the waterproofing material;
- measuring the sample thickness;
- placing the sample between the source and receiver containers;
- ensuring air-tightness between the containers and the sample;
- connecting the certified radon source to the source container and admitting radon into the source container;
- ventilating the receiver container continuously with radon-free air;
- recording radon activity concentrations simultaneously in the source and receiver containers at the time intervals not longer than 12 h by extracting air samples from the containers and introducing them into the scintillation cells placed under vacuum in advance;
- measuring the radon activity concentration in the scintillation cells;
- recording the date and time of sampling;
- waiting for establishing stationary radon diffusion through the sample;
- stopping the ventilation of the receiver container;
- performing the accumulation of radon in the receiver container and starting the decisive measurement of the radon activity concentrations in both containers by extracting air samples from the containers and introducing them into the scintillation cells placed under vacuum in advance. Radon activity concentrations shall be read for the first time not later than 10 min after closure of

the receiver container. Successive records of radon activity concentration in the receiver container shall be made at time intervals not longer than 1 h and at least four records are required;

- measuring the radon activity concentration in the scintillation cells;
- recording the date and time of sampling;
- calculating the radon diffusion coefficient.

A.5 Expression of results

A.5.1 Radon activity concentration measurement using scintillation cells

The spot radon activity concentration, *C*, at the time of counting is given by Formula (A.1):

$$C = \frac{N - N_0}{T_{\rm C} \cdot R_{\rm t} \cdot n_{\alpha} \cdot V_{\rm f}} = \frac{N - N_0}{\omega} \tag{A.1}$$

with $\omega = T_{\rm C} \cdot R_{\rm t} \cdot n_{\alpha} \cdot V_{\rm f}$

Reference [10] gives indications on the potential corrections needed to assess the counting uncertainty.

A.5.2 Radon diffusion coefficient

The radon activity concentration in the receiver container increases over time depending on the radon exhalation rate from the sample to the receiver container, area of the sample and volume of the receiver container. Under steady state conditions and considering the reduction in the radon activity concentration in the receiver container due to its radioactive decay, back diffusion and ventilation as negligible, the linear increase of radon activity concentration in the receiver container is described by Formula (A.2):

$$C_{\mathrm{rc},i} = C_{\mathrm{rc},i-1} + \frac{E \cdot S_{\mathrm{s}}}{V} \cdot \Delta t \tag{A.2}$$

and the diffusive radon exhalation rate by Formula (A.3):

$$E = \frac{V(C_{rc,i} - C_{rc,i-1})}{S_s \Delta t} = p \cdot \frac{V}{S_s}$$
(A.3)

where

p is the slope of the linear regression straight line expressed in Becquerel per cubic metre and per second.

Assuming constant radon activity concentration in the source container and negligible radon activity concentration in the receiver container at the beginning of the decisive measurement, the diffusive radon exhalation rate from the sample is expressed by the relationship:

$$E = \frac{2 \cdot C_{\rm sc} \cdot l \cdot \lambda}{e^{d/l} - e^{-d/l}} \tag{A.4}$$

The radon diffusion length is subsequently calculated from Formulae (A.3) and (A.4) by means of an iterative process.

The radon diffusion coefficient is derived from:

$$D = \lambda l^2 \tag{A.5}$$

A.6 Example: Determination of the radon diffusion coefficient of the HDPE membrane with the thickness 0,47 mm

The variations in the radon activity concentrations in the source and receiver containers are presented in <u>Table A.1</u> and <u>Figure A.2</u>.

Time (s)	Radon activity concentrations in the			
	Source container (Bq·m ⁻³)	Relative uncertainty	Receiver container (Bq·m ⁻³)	Relative uncertainty
		<i>u</i> (Bq·m⁻3)		<i>u</i> (Bq·m⁻3)
300	22 141 050	1 107 053	990	50
3 600	22 029 050	1 101 453	3 820	191
7 200	21 881 690	1 094 085	6 900	345
10 800	21 805 060	1 090 253	9 980	499
14 400	21 796 220	1 089 811	13 070	654
18 000	21 887 580	1 094 379	17 370	869



Кеу

X time (s)

Y radon concentration (Bq/m³)

Figure A.2 — Linear extrapolation of concentrations measured in the receiver container

The slope of the linear regression calculated from the measured activity concentrations is:

 $p = 0.943 \text{ Bq} \cdot \text{m}^{-3} \cdot \text{s}^{-1}$,

which is significantly greater than 0.

The correlation coefficient, *R*, of the measured radon activity concentrations in the receiver container is:

R = 0,993 7,

which is greater than 0,9.

The average radon activity concentration in the source container throughout the duration of the measurement was:

 $C_{\rm sc}$ = 21 923 442 ± 123 649 Bq·m⁻³,

which means that the coefficient of variation is lower than 10 %, and that C_{sc} is higher than the minimum radon activity concentration, according to Figure 8.

Based on the geometric characteristics of the receiver container:

useful volume: V = 2 319,0 cm³ and

area of the tested sample: $S_s = 152,0 \text{ cm}^2$,

the diffusive radon exhalation rate, *E*, from the sample into the receiver container obtained from Formula (A.3) is:

 $E = 0,1439 \text{ Bq} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$

The radon diffusion length calculated from <u>Formulae (A.3)</u> and <u>(A.4)</u> by means of an iterative process is:

 $l = 1,25 \times 10^{-3} \text{ m}$

The radon diffusion coefficient derived from Formula (A.5) is:

 $D = 3.1 \times 10^{-12} \text{ m}^{2} \cdot \text{s}^{-1}$

The laboratory has to provide the uncertainty of the values obtained for the purpose of the diffusion coefficient assessment, considering all the sources of uncertainty related to the experimental arrangement and the mathematical methods chosen.

In general, the standard errors propagation model cannot be applied due to the fact that the radon diffusion length is calculated from Formula (A.3) and Formula (A.4) by means of an iterative process. The standard uncertainty estimate of *D* has been calculated using the numerical model application on the set of data obtained during the internal quality control testing under the repeatability conditions (the same measurement apparatus and the same operator). The relative uncertainty of measured thickness u(d) was assessed to be u(d) = 0,01 and the relative uncertainty of radon activity concentration measurement u(C) was obtained by repeated measurement and is equal to u(C) = 0,05.

NOTE The systematic error assessment cannot be done due to the fact that the traceable standards for this specific measurement do not exist as a part of common traceability measurement.

Expression of the result of radon diffusion coefficient assessment with the standard uncertainty of measurement:

 $D = D \pm 1,96s(D)$

Using <u>Formula (7)</u>:

 $D = 3.1 \pm 0.186 \times 10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$

Annex B (informative)

Determining the radon diffusion coefficient during the phase of non-stationary radon diffusion

B.1 General

This annex deals only with the determination of the radon diffusion coefficient according to method A when the decisive measurement is performed during the phase of non-stationary radon diffusion using a continuous method for the recording of the radon activity concentration in the useful volumes of the receiver and source containers. Application of the mathematical procedure for methods B and C when the decisive measurement is performed during the phase of stationary radon diffusion is also possible.

Determining the radon diffusion coefficient of a tested sample involves measuring variations in the radon activity concentration as a function of time, in the source and receiver containers with known geometric characteristics and with one open face applied to the surface of the sample under investigation.

Back diffusion, ventilation of the receiver container and radon decay are not excluded from the mathematical processing of the measured data.

B.2 Additional terms and definitions

For the purpose of this annex, the following symbols and the symbols given in <u>Clause 3</u> apply.

Cn	vectorofactivityconcentrationsinnodesofthefiniteelementmesh, inBecquerelspercubicmetre
C _{ssc}	activity concentration on the surface of the sample exposed to the source container, in Bec- querels per cubic metre
C _{src}	activity concentration on the surface of the sample exposed to the receiver container, in Becquerels per cubic metre
de	length of one-dimensional finite element, in metres
∂∕∂n	derivative in the direction of external normal to the boundary
E _m	mean energy released by the radiation in the air of the chamber for a temperature $T(K)$ and pressure p (hPa), in joules
F _C	calibration factor, in Becquerels per cubic metre per ampere
F_{T}	correction factor for temperature, dimensionless
Fp	correction factor for atmospheric pressure, dimensionless
Ι	ionization current, in Amperes
I ₀	background ionization current, in Amperes
Kc	capacity matrix

K _d	conductance matrix
K _h	boundary conditions matrix
<i>K</i> _{h,s}	boundary conditions matrix of the finite element exposed to the source container
<i>K</i> _{h,r}	boundary conditions matrix of the finite element exposed to the receiver container
K_{λ}	radioactive decay matrix
n	radon leakage rate of the receiver container, in per second
Ν	vector of interpolation functions
ω _{air}	mean energy expended for the production of an ion pair in the air of the chamber, in joules
e _c	unit electron charge in coulomb ($e_c = 1,602 \times 10^{-19}$ Coulomb)
P _m	atmospheric pressure when operating the chamber, in hectoPascal
Pe	atmospheric pressure when calibrating the chamber, in hectoPascal
$q_{ m h}$	boundary conditions vector
$q_{\mathrm{h,s}}$	boundary conditions vector of the finite element exposed to the source container
$q_{ m h,r}$	boundary conditions vector of the finite element exposed to the receiver container
<i>T</i> _m	temperature of the ambient air when operating the chamber, in Kelvin
Te	temperature of the ambient air when calibrating the chamber, in Kelvin
V _{ch}	volume of air present in the chamber, in cubic metres

B.3 Equipment

The measuring system comprises two air-tight containers (see Figure B.1) that serve at the same time as ionization chambers operating in current mode. Both containers are equipped with three valves intended for ventilating the containers, for extracting air samples for control measurement of radon activity concentrations and for connecting to a certified radon source. A fully automatic measuring unit ensuring continuous monitoring of radon activity concentrations in both containers at very short time intervals is a part of the measuring system.



Key

- 1 ionization chamber in the form of the receiver container
- 2 valve
- 3 tested sample
- 4 ionization chamber in the form of the source container
- 5 pump
- 6 radon source
- 7 measuring unit
- 8 collecting electrode
- 9 steel case

Figure B.1 — Measuring system for determining the radon diffusion coefficient using continuous method for monitoring radon activity concentrations

B.4 Measurement procedure

In case of method A, the measurement is performed as follows:

- preparing the sample of the waterproofing material;
- measuring the sample thickness;
- placing the sample between the source and receiver containers;
- ensuring air-tightness between the containers and the sample;
- connecting the certified radon source to the source container and admitting radon into the source container;
- performing the accumulation of radon in the receiver container and monitoring the radon activity concentrations in both containers by continuous measuring device for a period longer than the minimum duration of the decisive measurement for non-stationary radon diffusion, according to Figure 7. The first reading of the radon activity concentrations shall be carried out not later than 1 h after radon is admitted into the source container. Time interval between successive records of concentrations shall not exceed 6 h during the first 5 d of measurement and 12 h during the following days;
- reading the data (radon activity concentrations and time) recorded during the accumulation process;
- calculating the radon diffusion coefficient.

B.5 Expression of results

B.5.1 Radon activity concentration measurement using the current ionization chamber

The radon activity concentration is obtained from Formula (B.1):

$$C = (I - I_0) \cdot \omega \tag{B.1}$$

with $\omega = F_{\rm C} \cdot F_{\rm p} \cdot F_{\rm T}$

$$F_{\rm C} = \frac{1}{V_{\rm ch}} \cdot \frac{\omega_{\rm air}}{E_{\rm m} \cdot e_c} \tag{B.2}$$

$$F_{\rm p} = \frac{p_{\rm e}}{p_{\rm m}} \tag{B.3}$$

$$F_{\rm T} = \frac{T_{\rm m}}{T_{\rm e}} \tag{B.4}$$

Radon diffusion coefficient determination using finite element method (FEM)

The governing formula, which describes radon distribution in the tested sample, is a standard onedimensional partial differential equation:

$$\frac{\partial}{\partial x} \left(D \frac{\partial C}{\partial x} \right) - \lambda \cdot C = \frac{\partial C}{\partial t}$$
(B.5)

where *x* ranges from 0 (the lower surface of the sample) to *d* (the upper surface of the sample). The origin of time *t* in Formula (B.5) is set to the beginning of the decisive measurement. At that time, $t_0 = 0$ s, the initial condition within the whole x-range is taken as:

$$C = C_0 = 0 \tag{B.6}$$

The initial radon activity concentration in the sample is taken as 0 Bq·m⁻³ because it is assumed that the sample is only fixed to the measuring device at that moment. The boundary condition for Formula (B.5) can be expressed on both sides of the sample (i.e. for x = 0 as well as for x = d) as:

$$-D\frac{\partial C}{\partial x} = h \cdot (C_{\rm s} - C_{\rm a}) \tag{B.7}$$

The value of *C*_a can be either the known radon activity concentration in the source container:

$$C_{\rm a} = C_{\rm sc} \tag{B.8}$$

(applicable for x = 0) or the initially unknown radon activity concentration in the receiver container (applicable for x = d). This value shall be calculated for the time step (*i*) from the previous step (*i* - 1) using Formula (B.9):

$$C_a = C_{\mathrm{rc},i} = C_{\mathrm{rc},i-1} \cdot e^{-(\lambda+n)\cdot\Delta t} + \frac{E_{i-1}\cdot S_s}{V\cdot(\lambda+n)} \cdot \left[1 - e^{-(\lambda+n)\cdot\Delta t}\right]$$
(B.9)

The diffusive radon exhalation rate from the sample to the receiver container in the time step (i - 1) can be calculated from:

$$E_{i-1} = h \cdot \left(C_{\operatorname{src},i-1} - C_{\operatorname{rc},i-1} \right)$$
(B.10)

with $C_{rc,0}$ taken as a known initial value (usually close to 0 Bq·m⁻³) and the radon transfer coefficient *h* derived using the procedure described in Reference [12].

The numerical solution of <u>Formula (B.5)</u> with conditions <u>Formulae (B.6)</u> and <u>(B.7)</u> can be obtained by means of the finite element method (FEM) using the standard Galerkin approach. This approach belongs among weighted residuals methods and thus the derivation of the finite element formulation starts with <u>Formula (B.11)</u>:

$$\int_{0}^{d_{e}} \left[\frac{\partial}{\partial x} \left(D \frac{\partial C}{\partial x} \right) - \lambda \cdot C - \frac{\partial C}{\partial t} \right] N \cdot dx = 0$$
(B.11)

which is a mathematical expression of the requirement that the residual of the numerical solution of Formula (B.5) shall be orthogonal to the interpolation functions, *N*. The unknown function, *C*, in Formula (B.11) is taken as an approximation:

$$C = N^{\mathrm{T}} \cdot C_{\mathrm{n}} \tag{B.12}$$

where C_n is a vector of unknown radon activity concentrations in the nodes of the created FEM mesh and *N* is a vector of known interpolation functions, which are dependent on the types of chosen finite elements. In this case, it is possible to use the simplest one-dimensional finite elements with two nodes and linear approximation (they correspond to the partial fractions of the thickness of the tested sample). Two interpolation functions are used for each finite element:

$$N_1 = 1 - \frac{x}{d_e} \text{ and } N_2 = \frac{x}{d_e}$$
 (B.13)

with *x* ranging from 0 (node 1) to d_e (node 2).

The general finite element formulation can be finally derived using three steps:

- a) substitution of <u>Formula (B.12)</u> into <u>Formula (B.11)</u>;
- b) integration by parts applied to the first term in <u>Formula (B.7)</u>;
- c) introduction of the boundary condition, <u>Formula (B.7)</u> into modified <u>Formula (B.11)</u>. As a result, the FEM formulation can be written in the form:

$$\left(K_{d} + K_{\lambda} + K_{h}\right) \cdot C_{n} + K_{c} \frac{\mathrm{d}C_{n}}{\mathrm{d}t} = q_{h}$$
(B.14)

where the conductance matrix, K_d , is defined as:

$$K_{\rm d} = \int_{0}^{d_{\rm e}} D \cdot \left(\frac{\mathrm{d}N}{\mathrm{d}x} \cdot \frac{\mathrm{d}N^{T}}{\mathrm{d}x} \right) \mathrm{d}x = \frac{D}{d_{\rm e}} \begin{bmatrix} 1 & -1\\ -1 & 1 \end{bmatrix}$$
(B.15)

the radioactive decay matrix, K_{λ} , as:

$$K_{\lambda} = \int_{0}^{d_{e}} \lambda \cdot N \cdot N^{T} dx = \frac{\lambda \cdot d_{e}}{6} \begin{bmatrix} 2 & 1 \\ 1 & 2 \end{bmatrix}$$
(B.16)

and the capacity matrix, *K*_c, as:

$$K_{\rm c} = \int_{0}^{d_{\rm e}} N \cdot N^{T} dx = \frac{d_{\rm e}}{6} \begin{bmatrix} 2 & 1 \\ 1 & 2 \end{bmatrix}$$
(B.17)

The final formulations of the matrices presented in <u>Formula (B.15)</u>, <u>Formula (B.16)</u> and <u>Formula (B.17)</u> were derived using the interpolation functions defined in <u>Formula (B.13)</u>.

The boundary related matrix, K_h , and vector, q_h , in Formula (B.14) are non-zero for two finite elements only. The first one is located on the lower side of the tested sample, which means that it is exposed to the source container and its matrix, K_h , and vector, q_h , are therefore:

$$K_{\mathrm{h},\mathrm{s}} = \begin{bmatrix} h & 0\\ 0 & 0 \end{bmatrix} \text{ and } q_{\mathrm{h},\mathrm{s}} = \begin{cases} h \cdot C_{\mathrm{sc}}\\ 0 \end{cases}$$
(B.18)

with radon activity concentration, C_{sc} , defined in Formula (B.8). The second finite element, with nonzero boundary matrix and vector, is located on the opposite side of the tested sample. Consequently, its matrix K_h and vector q_h have different definitions:

$$K_{\rm h,r} = \begin{bmatrix} 0 & 0 \\ 0 & h \end{bmatrix} \text{ and } q_{\rm h,r} = \begin{cases} 0 \\ h \cdot C_{\rm rc} \end{cases}$$
(B.19)

where radon activity concentration, C_{rc}, is calculated for each time step from Formula (B.9).

The general FEM formulation Formula (B.14) is valid for each finite element as well as for the entire analysed region (i.e. in this case for the tested sample). Relevant matrices and vectors for the entire region are created by localization of the individual element matrices and vectors.

The last problem of the numerical solution of Formula (B.14) is the time-dependency of the boundary vector, q_h , and the vector of unknown nodal values, C_n . To handle this difficulty, it is assumed that both vectors can be expressed in the time interval from (*i* – 1) to (*i*) as

$$C_{n} = \varepsilon \cdot C_{n,i} + (1 - \varepsilon) \cdot C_{n,i-1}$$
(B.20)

$$q_{\rm h} = \varepsilon \cdot q_{\rm h,i} + (1 - \varepsilon) \cdot q_{\rm h,i-1} \tag{B.21}$$

where ε is chosen as $0.5 \le \varepsilon \le 1$ in order to ensure the numerical stability of the calculation. The usual recommended value is $\varepsilon = 0.5$, which was also used in the presented numerical solution of Formula (B.5).

Substitution of Formula (B.20) and Formula (B.21) into Formula (B.14) leads finally to a system of linear algebraic formulae for unknown nodal values $C_{n,i}$:

$$\begin{bmatrix} \left(K_{d} + K_{\lambda} + K_{h}\right) \cdot \varepsilon + \frac{K_{c}}{\Delta t} \end{bmatrix} \cdot C_{n,i} = q_{h,i} \cdot \varepsilon + q_{h,i-1} \cdot (1-\varepsilon) + \\ + \begin{bmatrix} \frac{K_{c}}{\Delta t} - \left(K_{d} + K_{\lambda} + K_{h}\right) \cdot (1-\varepsilon) \end{bmatrix} \cdot C_{n,i-1}$$
(B.22)

where initial radon activity concentrations, $C_{n,0}$, are taken as radon activity concentrations, C_0 , from Formula (B.6). The complete calculation process for time step (*i*) starts with the determination of boundary conditions, particularly with the calculation of the radon activity concentration in the receiver container, $C_{rc,i}$, using Formula (B.9). The boundary vectors, $q_{h,i}$, for both boundary finite elements are subsequently calculated from Formula (B.18) and Formula (B.19). The right-hand side of Formula (B.22) is then entirely known and the solution of Formula (B.22) can be found using standard Gauss elimination. Known values from the previous time step (*i* - 1) are used throughout this process.

The radon diffusion coefficient is determined by an iterative procedure based on repeated numerical solutions of Formula (B.5). During this calculation, the radon diffusion coefficient gradually grows from the assumed lower limit to the assumed upper limit. In this first step, the radon diffusion coefficient is taken as the chosen lower limit. The calculated time-dependent results of radon activity concentration in the receiver container are afterwards compared with the relevant measured data and the differences between both data sets are recorded. In the following steps, the whole procedure is repeated with continuously growing radon diffusion coefficient until its upper limit is reached. The final radon diffusion coefficient is the value that results in the numerical solution of Formula (B.5) for which the differences between the calculated and measured concentrations in the receiver container are minimal.

B.6 Example: Determination of the radon diffusion coefficient of the HDPE membrane (of different composition than assumed in the example A.7) with the thickness 1,63 mm

The variations in the radon activity concentrations in the source and receiver containers are presented in Figure B.2. The duration of the measurement 380 h is longer than the minimum duration of the decisive measurement for non-stationary radon diffusion estimated from Figure 7 for the ratio d/l = 2,5and fast accumulation of radon in the source container (HDPE material was considered). The minimum radon activity concentration required in the source container according to Figure 8 is 1 MBq·m⁻³. This value was exceeded 3 h after the start of the measurement.



Key

- Y1 Rn concentration in the source container (kg·Bq/m³)
- Y2 Rn concentration in the receiver container (kg·Bq/m³)
- A source container
- B receiver container

Figure B.2 — Variations in radon activity concentrations for the source and receiver containers

Radon activity concentrations from Figure B.2 are used for the determination of the boundary conditions according to Formulae (B.8) and (B.9) and the diffusive radon exhalation rate from the sample to the receiver container according to Formula (B.10).

With the use of the geometric characteristics of the receiver container, useful volume: V = 2 319,0 cm³ and area of the tested sample: $S_s = 152,0$ cm², the iterative procedure based on repeated numerical solutions of Formula (B.5) yields the final radon diffusion coefficient:

 $D = 1.3 \times 10^{-12} \text{ m}^{2} \text{ s}^{-1}$

Differences between the calculated and measured radon activity concentrations in the receiver container are plotted in Figure B.3. The average difference between these two data sets is 189,6 Bq·m⁻³.



Кеу

- X time (h)
- Y radon concentration (Bq/m³)
- A measured values
- B calculated values

Figure B.3 — Differences between the radon activity concentrations calculated in the receiver container for the final radon diffusion coefficient $D = 1,3 \times 10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$ and the measured concentrations

The laboratory has to provide the uncertainty of the values obtained for the purpose of the diffusion coefficient assessment, considering all the sources of uncertainty related to the experimental arrangement and the mathematical methods chosen.

In general, the standard errors propagation model cannot be applied due to the fact that a non-liner time-dependent differential equation needs to be solved by means of numerical modelling tools. The standard uncertainty estimate of *D* has been calculated using the numerical model application on the set of data obtained during the internal quality control testing under the repeatability conditions (the same measurement apparatus and the same operator). The relative uncertainty of measured thickness u(d) was assessed to be u(d) = 0,01 and the relative uncertainty of radon activity concentration measurement u(C) was obtained by repeated measurement and is equal to u(C) = 0,05.

NOTE The systematic error assessment cannot be done due to the fact that the traceable standards for this specific measurement do not exist as a part of common traceability measurement.

The standard uncertainty of *D* has been calculated using the numerical model application on the set of data obtained during the internal quality control testing under the repeatability and reproducibility conditions.

Expression of the result of radon diffusion coefficient assessment with the standard uncertainty of measurement gives Formula (B.23):

$$D = D \pm 1,96s(D) \tag{B.23}$$

Using Formula (7): $D = 1,3 \pm 0,078 \times 10^{-12} \text{ m}^2 \cdot \text{s}^{-1}$

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International Standards/Documents

Corresponding Indian Standard

IS/ISO 80000-10 : 2019 Quantities and units:

Degree of Equivalence

Identical with

ISO 11665-1 : 2019 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 80000-10 Quantities and units-

Part 10: Atomic and nuclear physics

IS 18066 (Part 1) : 2022 Measurement of ISO 11665-1:2019 radioactivity in the environment - Air : radon-222: Part 1 Origins of radon and its short-lived decay products and associated measurement methods

> Identical with ISO 80000-10:2019

The technical committee has reviewed the provisions of the following International Standards/ documents referred in this adopted standard and has decided that they are acceptable for use in conjunction with this Standard:

Part 10 Atomic and nuclear physics

International Standards/ Documents	Title
ISO/IEC Guide 98-3	Uncertainty of measurements — Part 3: Guide to the expression of uncertainty in measurement (GUM : 1995)
ISO 11665-5	Measurement of radioactivity in the environment — Air : radon-222 — Part 5: Continuous measurement 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" in this table.
ISO 11929	Determination of the characteristic limits (decision threshold, detection limit, and limits of the confidence interval) for measurements of ionizing radiation-Fundamentals and application

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