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

Part 12:
Determination of the diffusion coefficient in waterproof materials: membrane one-side activity concentration measurement method

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

Partie 12: Détermination du coefficient de diffusion des matériaux imperméables: méthode de mesure de l'activité volumique d'un côté de la membrane

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Foreword

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

A list of all parts in the ISO 11665 series can be found on the ISO website.

Any feedback or questions on this document should be directed to the user's national standards body. A complete listing of these bodies can be found at www.iso.org/members.html.

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[4].

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[5] 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 a diffusion mechanism caused by the all-time existing difference between radon activity concentrations in the underlying soil and inside the building, and via a 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·m^{-3} is recommended by the World Health Organization[6]. Wherever this is not possible, this reference level should not exceed 300 Bq·m^{-3}. This recommendation that was endorsed by the European community member states establishes national reference levels for indoor radon activity concentrations. The reference levels for the annual average activity concentration in air cannot be higher than 300 Bq·m^{-3}[8].

To reduce the risk to the overall population, building codes which require radon prevention measures in buildings under construction and radon mitigating measures in existing buildings should be implemented. 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.

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

As no reference standards and no reference materials are currently available for these types of materials, and related values of the radon diffusion coefficient, the metrological requirement regarding the determination of the performance of the different methods described in ISO/TS 11665-13 and in this document, as required by ISO 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.

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

Part 12:
Determination of the diffusion coefficient in waterproof materials: membrane one-side activity concentration measurement method

1 Scope

This document specifies the method 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 which will be met during the test.

The test method described in this document allows to estimate the radon diffusion coefficient in the range of 10^{-5} m^2/s to 10^{-12} m^2/s[8][9] with an associated uncertainty from 10 % to 40 %.

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 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, ISO 80000-10 and the following apply.

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

— ISO Online browsing platform: available at https://www.iso.org/obp

— IEC Electropedia: available at http://www.electropedia.org/

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

3.1.2
sample
certain amount of *material* (3.1.1) chosen from the production batch for determination of the *radon diffusion coefficient* (3.1.3)

3.1.3
radon diffusion coefficient
D
<bulk> 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* (3.1.1)

[SOURCE: ISO/TS 11665-13:2017, 3.1.3, modified — "<bulk>" has been added as the domain of the definition.]

3.1.4
decisive measurements
measurement results used to calculate the *radon diffusion coefficient* (3.1.3)

3.1.5
decisive volume of the chamber
volume of the chamber used to calculate the *radon diffusion coefficient* (3.1.3)

3.1.6
decisive area of the sample
material (3.1.1) *sample* (3.1.2) area used to calculate the *radon diffusion coefficient* (3.1.3)

3.1.7
radon transfer coefficient
radon transport in thin boundary layer of air near the surface of the *sample* (3.1.2)

Note 1 to entry: The default value of the radon transfer coefficient is 0,001 m/s to 0,1 m/s.

[SOURCE: ISO/TS 11665-13:2017, 3.1.15, modified — Note 1 to entry has been removed. Note 2 to entry has become Note 1 to entry and has been slightly rephrased.]

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
$D^{<}$	Lower limit of the confidence interval of the radon diffusion coefficient of the sample, in square metre per second
$D^{>}$	Upper limit of the confidence interval of the 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)
R_{c}	Calculated rate of decrease of radon activity concentration in the chamber
R_{L}	Calculated rate of decrease of radon activity concentration in the chamber at verification of radon-tightness
R_{min}	Function of the minimum measured rate of decrease in the chamber
R_{max}	Function of the maximum measured rate of decrease in the chamber
$u(y)$	Standard uncertainty of the value of y
$s(y)$	Standard deviation of the value of y
$u_{\mathrm{rel}}(D)$	Relative uncertainty of the radon diffusion coefficient in the sample
k	Coverage factor
N	Number of samples of the test material

4 Principle

The one-side method is based on the measurement of the decrease over time of the radon activity concentration in a source-detect chamber in contact with one side of the tested membrane. The test is performed in a non-stationary mode. This test method can be used for single-layer waterproof materials when testing results are needed rapidly. They are not applicable for multi-layer waterproof materials that do not meet the requirements of Figure 7.

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

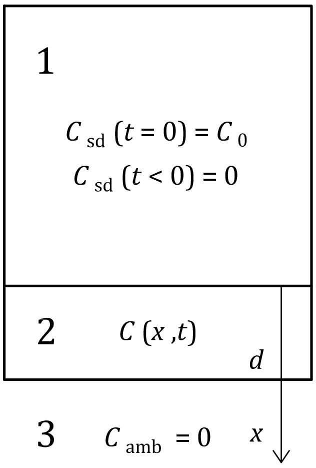
In the beginning of the test, a highly active portion of radon is injected into the chamber through a special valve. Since then, radon activity concentration in the chamber begins to decline because of

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

b) radon decay, and

c) leakage of radon from the chamber.

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



Key

1 source-detect chamber
2 tested sample
3 ambient air

Figure 1 — Measurement scheme

The process of radon transfer from the chamber through the sample is described by Formulae (1) and (2):

$$\frac{\partial C_{sd}(t)}{\partial t} = -\lambda \cdot C_{sd}(t) - \frac{S_s}{V_{sd}} \cdot h_1 \cdot \left[C_{sd}(t) - C(0,t)\right] \tag{1}$$

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

with the following boundary conditions in Formulae (3) to (5):

$$C_{sd}(t=0) = C_0,\ C(x,0) = 0 \tag{3}$$

$$-D\frac{\partial C(0,t)}{\partial x} = h_1 \cdot \left[C_{sd}(t) - C(0,t)\right] \tag{4}$$

$$-D\frac{\partial C(d,t)}{\partial x} = h_2 \cdot \left[C(d,t) - C_{amb}\right],\ \left[C_{amb}(t) = 0\right] \tag{5}$$

Formulae (1) to (5) with respect to the values of radon activity concentration in the chamber are solved as the function expressed by Formula (6):

$$C_{sd}(t) = C_0 \cdot f(t, D, \lambda, V_{sd}, S_s, d) \tag{6}$$

which is calculated as in Formula (7):

$$C_{sd}(t) = C_0\left[\int_0^t e^{\lambda\,(\tau - t)} F(\tau)\mathrm{d}\tau + e^{-\lambda \cdot t}\right] \tag{7}$$

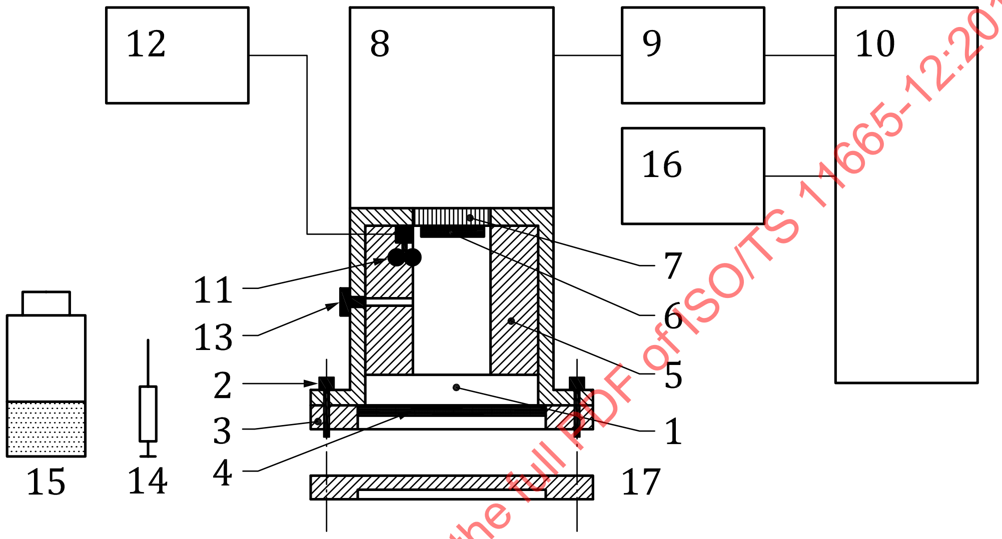
The calculation by Formula (7) is carried out by the algorithm described in Annex A.

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 (Clause 8).

The radon diffusion coefficient in the sample is calculated according to 9.1, taking into account the effect of radon leakage from the chamber.

5 Equipment

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



Key

1	chamber
2	bolt (with washer)
3	holder
4	sample
5	insert
6	scintillation plate
7	light-transmissive window
8	photomultiplier unit
9	signal converter
10	computer
11	micro-fan
12	power supply of the micro-fan
13	valve
14	syringe
15	radon source
16	thermometer
17	cap

Figure 2 — The scheme of the test installation

The installation includes an aluminium cylindrical chamber (1). The lower end of the chamber is a flange with a sealing gasket which is hermetically connected to the aluminium holders (3) with a test sample (4) by bolts (2).

Aluminium cylindrical inserts (5) are used to reduce the decisive volume of the chamber.

Scintillation plate (6) with sensitive ZnS(Ag) layer and light-transmissive window (7) made of polymethyl methacrylate are hermetically embedded in the upper end of the chamber.

The alpha radiation of radon and its progeny, interacting with the sensitive layer of the scintillator plate, causes light flashes (scintillations) which are converted into electrical pulses by the photomultiplier unit (8). These pulses are converted by the device (9) and transmitted to the computer (10). The computer software registers the average count rate at predetermined time intervals.

The micro-fan (11) provides stability in registering radon activity inside the chamber[8]. The micro-fan is equipped with a brushless motor and a consumption current of less than 20 mA at a voltage not exceeding 5 V. The power of the micro-fan is provided by a power supply of stable voltage (12).

The sealed valve (13) for injecting radon inside the chamber with the syringe (14) is located on the sidewall of the chamber. The radon portion in the syringe is introduced from the radon source (15). The activity and the volume of the radon source should be 10 kBq to 100 kBq and 0,2 l to 0,5 l, respectively.

The thermometer (16) is connected to the computer.

The test installation is equipped with at least 3 holders and one aluminium cap (17).

To register the activity concentration, a semiconductor or another type of detector, located inside the chamber and meeting the requirements of Clauses 7 and 10, can be also used.

The installation kit also includes

a) a torque wrench with predetermined torque between 1 Nm and 10 Nm,

b) a measuring instrument capable of determining the thickness of the tested sample with accuracy of ±0,01 mm (maximum standard relative measurement uncertainty of 5 %),

c) an epoxy adhesive,

d) alcohol, acetone, or another degreasing agent.

The chamber is set upright and the flange downwards (Figure 2) to avoid problems for radon exhalation from the sample.

Light shall not enter the chamber when the photomultiplier is on.

6 Sample preparation

6.1 General consideration

If the condition of Formula (29) is not satisfied after testing 3 samples, additional samples of the material should be prepared and tested until this condition is fulfilled.

The samples are cut out from the prefabricated membranes at a minimum distance of 20 mm from the edges of the membrane.

In the case of coatings, paints, sealants or other waterproof materials prepared on site, at least 3 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 at 4 points per 0,01 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 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.

6.2 Fixing the sample in the holder

The sample is cut into a disk and fixed in the holder (Figure 3) so that the direction of the radon flow through the sample during the test corresponds to the real flow direction through the material in operational conditions.

The sample is fixed in the holder without slack via an epoxy adhesive that is first applied to the shelf of the holder, and then poured into the annular gap (1 mm to 2 mm) between the sample and the sidewall of the holder.

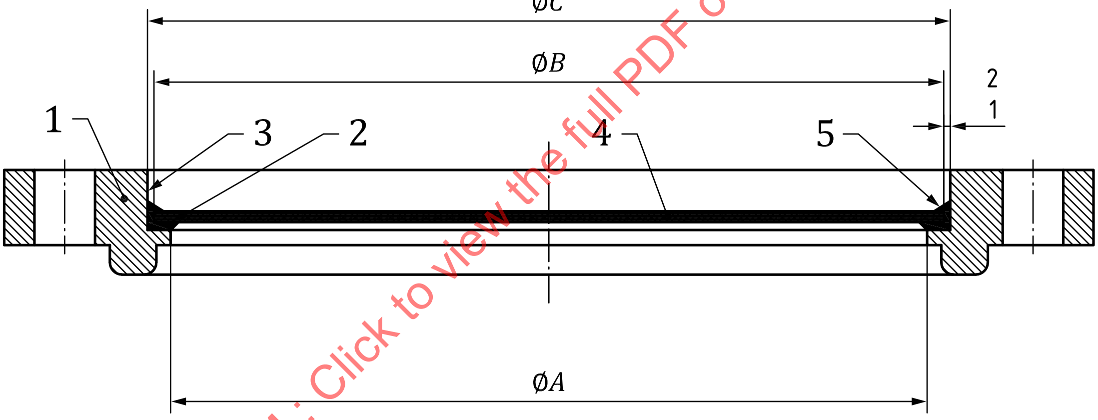
The surfaces of the sample and the holder to which the adhesive is applied, should be pre-treated by the degreasing agent.

During the preparation of the epoxy adhesive, the hardener is added to the epoxy resin in an amount which provides elasticity to the adhesive composition in order to avoid excessive hardness and brittleness after solidification. The prepared glue fixes the sample holder reliably for at least one week.

Before reuse, the holder is purified from the epoxy adhesive by using a solvent of epoxy resin, or by immersion in boiling water, or by a lathe.

The holder is attached to the chamber not earlier than 24 h after fixing the sample.

Dimensions in millimetres



Key

1	holder
2	shelf of the holder of 4 mm to 5 mm width
3	sidewall of the holder
4	sample
5	epoxy adhesive
ØA	decisive diameter of the sample, equal to the inner diameter of the chamber
ØB	diameter of the sample
ØC	diameter of the mating part of the holder

Figure 3 — Scheme of the sample fixing in the holder

6.3 Connection of the holder (cap) with the chamber

The holder (or cap) is connected to the chamber by the bolts uniformly tightened with a torque wrench to a torque of about 4 Nm. First the bolts are tightened by hand, and then using a torque wrench following the sequence, starting at any bolt, as shown in Figure 4. The bolt should be tightened by turning the wrench not more than 45° at a time.

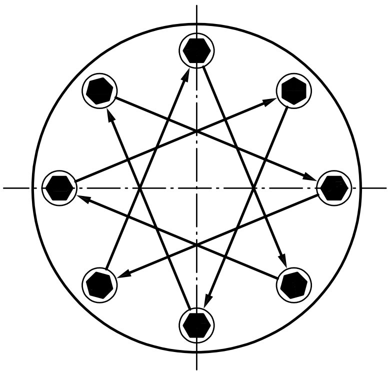


Figure 4 — The sequence of tightening the bolts

7 Control measurements

7.1 Verification of radon-tightness

The tightness of the chamber shall be checked before starting the sample testing in the following order:

a) attach the cap to the chamber according to 6.3;

b) inject radon;

c) measure the rate of decrease of radon activity concentration R_m in accordance with Clause 8.

The radon-tightness of the chamber is considered satisfactory when the measured rate of decrease of radon activity concentration in the chamber is approximated by the rate calculated with Formula (8) below:

$$R_\mathrm{L}(\tau_i) = \exp[-(\lambda + \lambda_\mathrm{L})\cdot(\tau_i - \tau_{i=41})],\ i = 41, 42, 43,\dots,100 \tag{8}$$

at values $\lambda_\mathrm{L} < 0{,}2\cdot\lambda$.

If this condition is satisfied, then the obtained value λ_L is stored and used later to assess the best estimate value $\hat{\lambda}_\mathrm{L}$ and uncertainty $s(\hat{\lambda}_\mathrm{L})$, which are calculated according to Formulae (9) and (10):

$$\hat{\lambda}_\mathrm{L} = \frac{1}{Q}\cdot\sum_{q=1}^{Q}\lambda_{\mathrm{L}q},\ q = 1, 2, \dots, Q,\ \text{at}\ Q \geq M_\mathrm{L} \tag{9}$$

$$s(\hat{\lambda}_\mathrm{L}) = \sqrt{\frac{\sum_{q=1}^{Q}(\hat{\lambda}_L - \lambda_{Lq})^2}{Q\cdot(Q-1)}},\ q = 1,2,\dots,Q,\ \text{at}\ Q \geq M_\mathrm{L} \tag{10}$$

where

λ_L is the calculated value of radon leakage rate at the satisfactory control measurement q;

Q is the number of satisfactory control measurements of radon leakage over the entire period of installation operation;

M_L is the minimum number of measurements in which both of the following conditions are simultaneously met:

$$2 \cdot s(\hat{\lambda}_L) < \hat{\lambda}_L \tag{11}$$

$$2 \cdot s(\hat{\lambda}_L) + \hat{\lambda}_L < 0{,}2 \cdot \lambda \tag{12}$$

7.2 Calibration

Calibration of the detector means determining the coefficient of proportionality (calibration factor) between the net count rate and radon activity concentration in the chamber.

The one-side method of the determination of the radon diffusion coefficient, does not require the detector calibration.

7.3 Detector background

Check the detector background when needed, but at least once a year.

Checking the detector background is carried out first without the insert, and then with each insert separately.

To check the background of the detector:

a) ventilate the chamber by air with radon activity concentration not exceeding 20 Bq·m^{-3};

b) attach the cap to the chamber in accordance with 6.3;

c) measure gross count rate r_g in accordance with Clause 8, but without radon injection into the chamber.

Detector background is considered satisfactory if the average count rate in the range of decisive measurements does not exceed 4 per second.

The average value of the count rate is then considered as r_0.

7.4 Instrument statistical fluctuation

Check the instrument statistical fluctuation when needed but at least once a year.

To check the instrument statistical fluctuation:

a) set the chamber without the insert, flange up;

b) place the alpha-source approximately in the middle of the scintillation plate;

c) attach the cap to the chamber in accordance with 6.3, while manually tightening the bolts;

d) measure the gross count rate r_g in accordance with Clause 8, but without radon injection into the chamber.

Instrument statistical fluctuation is considered satisfactory if the condition described in Formula (13) in the range of decisive measurements is fulfilled:

$$\frac{s(r_g)}{r_g^{\alpha}} < 0{,}01 \tag{13}$$

where, as shown in Formulae (14) and (15):

$$r_{\mathrm{g}}^{\alpha} = \frac{1}{60}\cdot\sum_{i=41}^{i=100} r_{\mathrm{g}}(\tau_i),\ i = 41, 42, 43, \dots, 100 \tag{14}$$

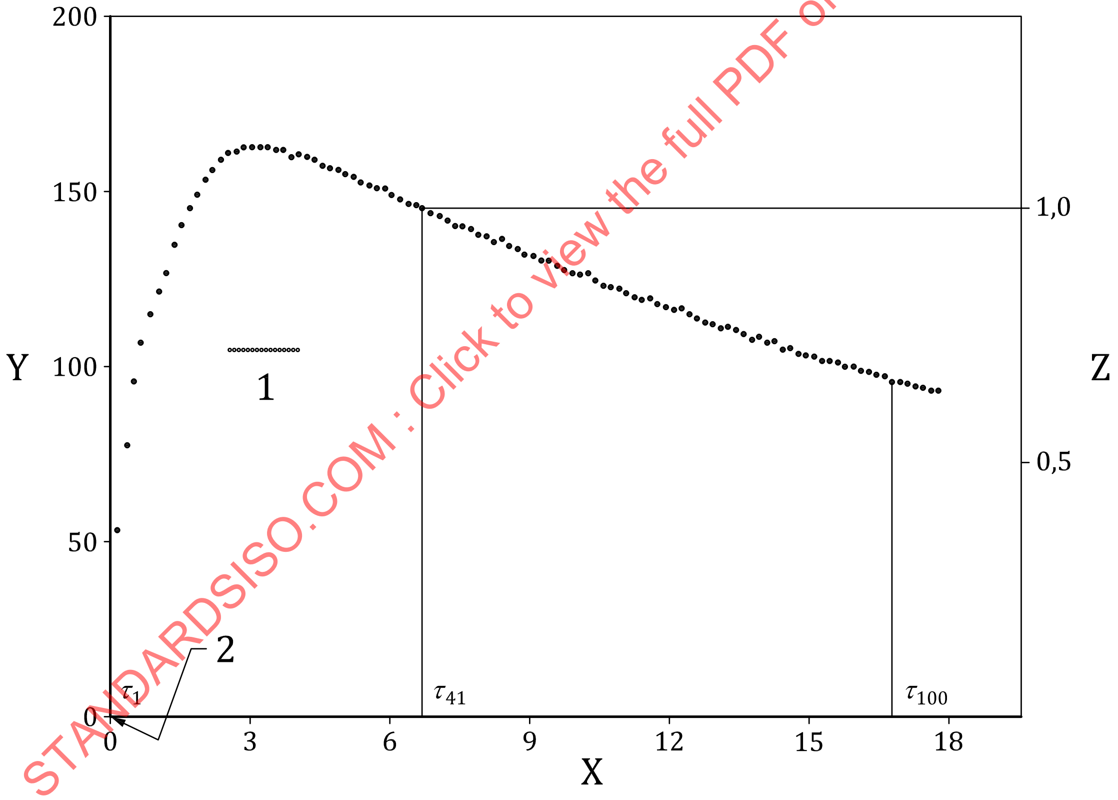
$$s(r_{\mathrm{g}}) = \sqrt{\frac{\sum\limits_{i=41}^{i=100}\left(r_{\mathrm{g}}^{\alpha} - r_{\mathrm{g}}(\tau_i)\right)^2}{60-1}},\ i = 41, 42, 43, \dots, 100 \tag{15}$$

8 Measurement of radon activity concentration

After injection of radon in the chamber immediately close the valve, switch on the micro-fan, the photomultiplier unit and run on a computer the program for registering time and pulse count rate.

The recommended time interval from the beginning to the end of the test is 100 periods, about 600 s each.

The beginning of the first period τ_1= 0 corresponds to the time of radon injection into the chamber (Figure 5). The forty-first period τ_{41} corresponds to the time of achieving satisfactory radioactive equilibrium between radon and its progeny in the chamber. The interval [τ_1, τ_{41}] is 6 h to 7 h.



Key

X time, h

Y counting rate, s^{-1}

Z rate of radon activity concentration decrease, rel

1 reference level of the minimum counting rate

2 portion radon inject

Figure 5 — Example of the data recorded during a test

Decisive measurements are performed in the time interval [τ_{41}, τ_{100}], the duration of which is from 10 h to 11 h. Thus, the total duration of the test of one sample does not exceed 18 h.

The measured rate of decrease of pulse count rate $R_m(\tau_i)$ is equal to the rate of radon activity concentration decrease in the chamber and is determined by Formula (16):

$$R_{\mathrm{m}}(\tau_i)=\frac{r_{\mathrm{g}}(\tau_i)-r_0}{r_{\mathrm{g}}(\tau_{i=41})-r_0}=\frac{C_{\mathrm{m}}(\tau_i)}{C_{\mathrm{m}}(\tau_{i=41})} \qquad (16)$$

where

i = 41,42,43,...,100	is the serial number of the measurement period;
$r_g(\tau_i)$	is the gross count rate in the i time period;
r_0	is the background count rate, determined in accordance with 7.3;
$C_m(\tau_i)$	is the measured average value of radon activity concentration in the i time period.

During the measurements, the following parameters shall be recorded:

- the number of the measurement period;
- the start time (up to one second);
- the count rate in the current period;
- the rate of decrease of radon activity concentration;
- the temperature of ambient air.

9 Processing and expression of the results for the sample

9.1 Determination of the radon diffusion coefficient in the sample

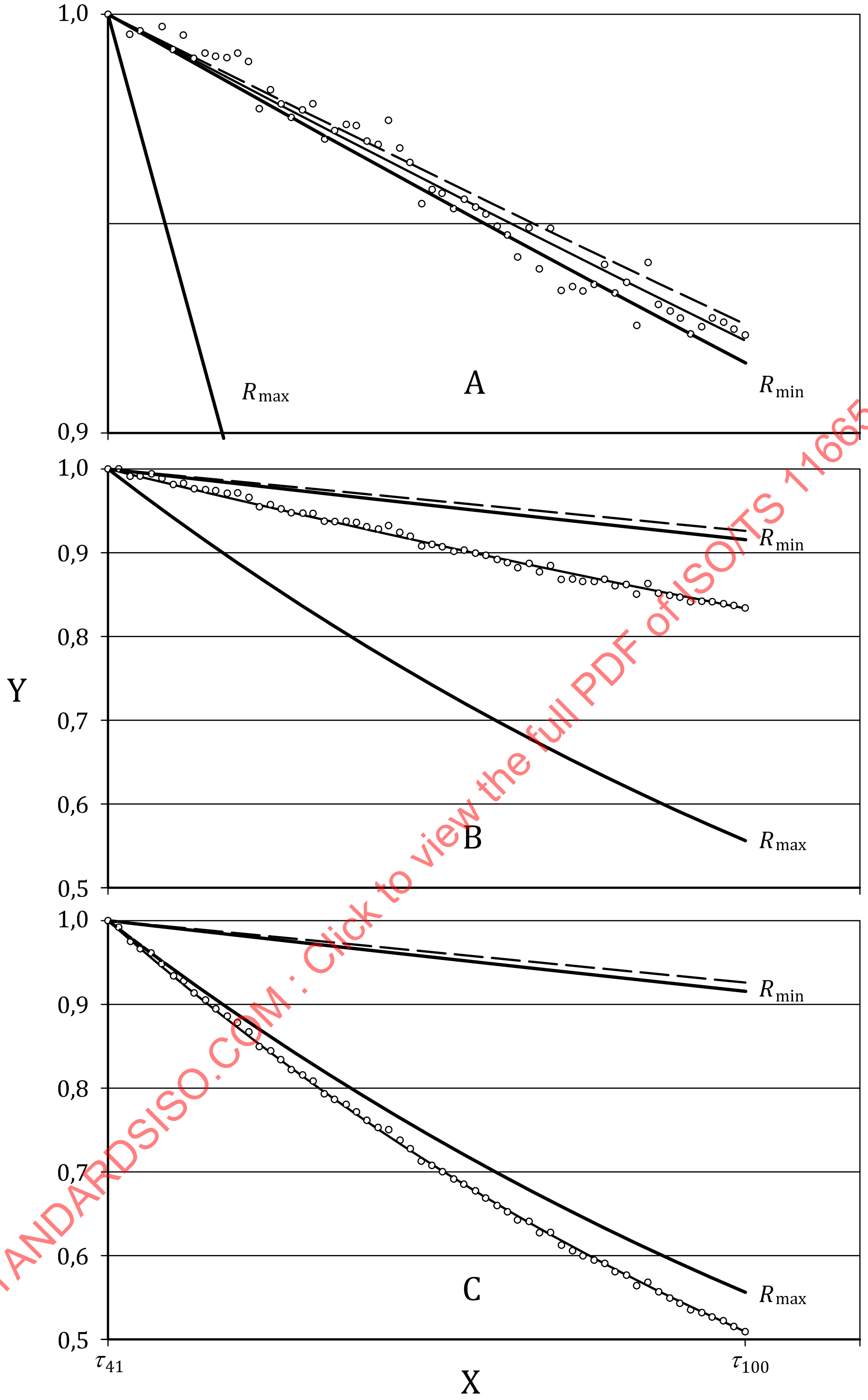
The value of the radon diffusion coefficient of the sample is determined by selecting such a value D at which the calculated rate of radon activity concentration decreases in the chamber by Formula (17):

$$R_{\mathrm{c}}(\tau_i)=\frac{C_{\mathrm{sd}}(t=\tau_i)}{C_{\mathrm{sd}}(t=\tau_{i=41})}\cdot\exp\left[-\lambda_{\mathrm{L}}\cdot(\tau_i-\tau_{i=41})\right],\ i=41,42,43,...,100 \qquad (17)$$

under condition $\lambda_{\mathrm{L}}=\hat{\lambda}_{\mathrm{L}}$ approximates the experimental function $R_m(\tau_i)$ (see Clause 8) in the range of decisive measurements [τ_{41}, τ_{100}] at the given volume of the chamber, thickness and area of the sample.

9.2 Characteristics of measurement limits

The behaviour of the function $R_m(\tau_i)$ depends on the radon diffusion coefficient of the sample, the sample size and the decisive volume of the chamber, and may correspond to one of the three options (A, B, C), shown in Figure 6.



Key

X	time
Y	rate of radon activity concentration decrease, rel
○	function R_m
———	approximation (fitting) curve R_c
———	functions R_{min} and R_{max}

— — — rate of decrease of radon activity concentration in the chamber due to radon decay only

Figure 6 — Options A, B and C for the behaviour of the function $R_{\mathrm{m}}(\tau_i)$

Option A - high uncertainty of the test results [$u_{\mathrm{rel}}(D) > 1$] for (see Formula 18):

$$R_{\mathrm{m}}(\tau_i) \geq R_{\mathrm{min}}(\tau_i), \tag{18}$$

Where, as shown in Formula (19):

$$R_{\mathrm{min}}(\tau_i) = \exp\left[-(\lambda + \hat{\lambda}_{\mathrm{L}} + 2 \cdot s(\hat{\lambda}_{\mathrm{L}})) \cdot (\tau_i - \tau_{i=41})\right], \; i = 41{,}42{,}43{,}\ldots 100 \tag{19}$$

Option B - satisfactory uncertainty of the test results according to Formula (20):

$$R_{\mathrm{min}}(\tau_i) > R_{\mathrm{m}}(\tau_i) \geq R_{\mathrm{max}}(\tau_i) \tag{20}$$

where, as shown in Formula (21):

$$R_{\mathrm{max}}(\tau_i) = \exp[-1{,}6 \cdot 10^{-5} \cdot (\tau_i - \tau_{i=41})], \; i = 41{,}42{,}43{,}\ldots 100 \tag{21}$$

Option C - unsatisfactory test results, see 10 g), according to Formula (22):

$$R_{\mathrm{m}}(\tau_i) < R_{\mathrm{max}}(\tau_i), \; i = 41{,}42{,}43{,}\ldots 100 \tag{22}$$

9.3 Estimation of confidence interval and uncertainty

The lower limit of the confidence interval of the radon diffusion coefficient of the sample $D^{<}$ is determined ($P = 0{,}95$) according to 9.1, but under condition $\lambda_{\mathrm{L}} = \hat{\lambda}_{\mathrm{L}} + 2 \cdot s(\hat{\lambda}_{\mathrm{L}})$.

The upper limit of the confidence interval of the radon diffusion coefficient of the sample $D^{>}$ is determined ($P = 0{,}95$) according to 9.1, but under condition $\lambda_{\mathrm{L}} = \hat{\lambda}_{\mathrm{L}} - 2 \cdot s(\hat{\lambda}_{\mathrm{L}})$.

The relative uncertainty of the radon diffusion coefficient of the sample is calculated ($P = 0{,}95$) by Formula (23):

$$u_{\mathrm{rel}}(D) = \frac{D^{>}}{D} - 1 \tag{23}$$

The confidence interval of the radon diffusion coefficient should be assessed, when the uncertainty of the sample thickness in the materials such as coatings or paints, exceeds 5 %. In this case, the combined uncertainty is equal to the positive square root of the sum of the squares of the uncertainties associated with radon leakage and the thickness of the sample.

9.4 Expression of the results

The results of the determination of the radon diffusion coefficient of the sample is expressed in accordance with Table 1.

Table 1 — Expression of results

Option (see 9.2)	Confidence interval (P = 0,95) of the radon diffusion coefficient of the sample in m^2/s		
	Lower limit	Best estimate	Upper limit
Option A	$D^<$ = 0 (the measurement results cannot be approximated)	D is calculated in accordance with 9.1 (D = 0, if the measurement results cannot be approximated)	$D^>$ is calculated in accordance with 9.3
Option B	$D^<$ is calculated in accordance with 9.3	D is calculated in accordance with 9.1	$D^>$ is calculated in accordance with 9.3
NOTE The relative uncertainty $u_{\mathrm{rel}}(D)$ is calculated according to 9.3 at $D \neq 0$.			

10 Requirements for the test

a) The test installation shall not be exposed to sunlight and bright light, before and during the test. During the test, the ambient temperature shall be between 18 °C and 28 °C and shall not vary by more than 5 °C, at a relative humidity not exceeding 75 %.

b) In order to avoid pressure fluctuations in the chamber due to external thermal effects, it is required to place the insert into the chamber and to keep the sample holder (or cap) close to the installation at least one day prior to measurement. When installing the holder or cap, it is necessary to minimize heat transfer from the hands of the operator.

c) The testing of each new sample shall begin after at least one day after the end of the testing of the previous sample (or controlling the radon leakage). After the end of the test, the sample holder (or cap) shall be immediately disconnected from the chamber. The insert, if there is no need to replace it, may be left in the chamber. The repetition of the tests of the same sample shall be performed at least one week after the end of the previous test.

d) The holder with the fixed sample (or cap) shall be connected with the chamber immediately prior to the measurement.

e) The time interval between the injection of radon into the chamber and the beginning of the measurement shall not exceed 20 s. The activity of injected radon (A_{inj}, Bq) shall ensure that, 3 h to 4 h after the start of the measurement, the measured count rate is higher than the "reference level of the minimum counting rate" (more than 100 s^{-1}) (Figure 5). The required count rate is achieved, if $A_{\mathrm{inj}} > 1\,000$ Bq. The volume injected into the chamber portion radon (V_{inj}, ml) is determined by Formula (24):

$$V_{\mathrm{inj}} = A_{\mathrm{inj}} \cdot \frac{V_{\mathrm{rs}}}{A_{\mathrm{rs}}} \tag{24}$$

where

V_{rs} is the internal air volume of the radon source in l;

A_{rs} is the activity of the radon source in kBq.

f) The thickness and diameter of the sample, as well as the decisive volume of the chamber shall be determined with a maximum standard relative measurement uncertainty of 5 %.

g) Start testing samples at a minimum volume of the chamber, if there is no information that the material is highly permeable. If the measured rate of decrease in the counting rate corresponds to option C (9.2), the test shall be repeated with the same sample at an increased decisive volume of the chamber or at a reduced working area of the sample.

h) Prior to the test of the material, radon leakage shall be checked in accordance with 7.1 at the minimum volume of the chamber. If the inspection results are satisfactory, the obtained value of radon leakage shall be stored and used later on to calculate the average radon leakage and its uncertainty. If the inspection results are not satisfactory, it is necessary to find and eliminate the causes of elevated radon leakage, then to repeat the leak test of the chamber. In order to find the reasons of radon leakage, it is to take into account the guidance [see 6.3 and Clause 10 a) to e)], and check the background of the detector in accordance with 7.3 and the instrument statistical fluctuation in accordance with 7.4.

i) Control the background of the detector in accordance with 7.3 as necessary, but at least once a year. If the results are satisfactory, the resulting value of the background shall be stored and is used later in the calculations. If the results are not satisfactory, it is necessary to find and eliminate the causes of high background (such as ^{226}Ra pollution of the scintillation plate or insert from the radon source, or accumulated ^{210}Po), and then, repeat the background check.

j) Control the instrument statistical fluctuation in accordance with 7.4 as necessary, but at least once a year. A point source in the form of a disk (diameter of not more than 5 cm) containing radionuclide ^{239}Pu or ^{241}Am, which provides the count rate of at least 40 s^{-1} (source activity at least 100 Bq), shall be recommended as an alpha-source. If the inspection results are not satisfactory, it is necessary to clarify and eliminate the causes of the bad instrument statistical fluctuation, and then repeat the test.

11 Influencing factors

The main factor influencing the test results is the radon leakage, λ_L. Other factors can influence the results, such as

a) statistical uncertainty of the rate of decrease of radon activity concentration in the chamber;

b) instability of the registration of radon activity in the chamber due to uneven deposition of radon decay products on the scintillation plate;

c) instability of the instrument statistical fluctuation;

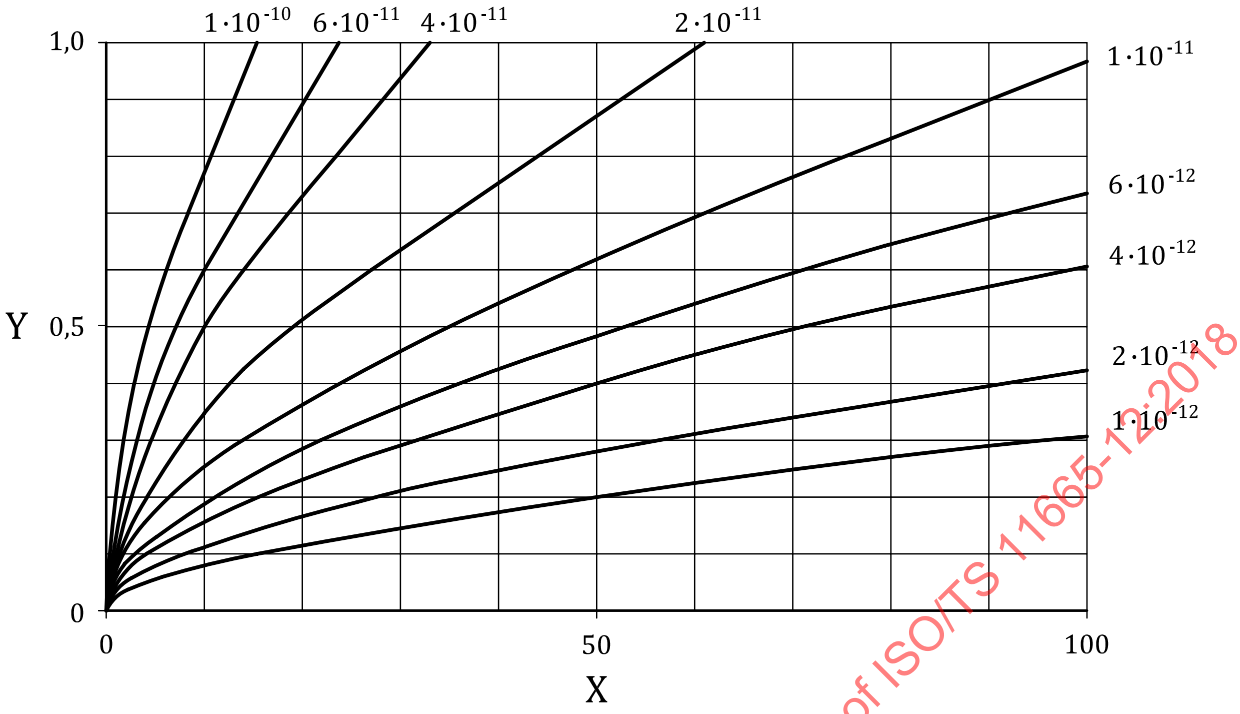
d) changes in atmospheric pressure and ambient temperature.

While the cumulative effect of all these factors is reflected in the value of λ_L, their influence is incomparably smaller.

The uncertainty of the radon diffusion coefficient of the sample can be reduced by

- increasing the number of tests of the same sample with an estimate of the results in accordance with Clause 12,
- increasing the number of control measurements of radon leakage,
- reducing the magnitude and variations of radon leakage by a tight connection of the sample with the chamber,
- reducing the decisive volume of the chamber,
- increasing the decisive area of the sample,
- increasing the duration of measurement periods, for example, from 600 s to 3 600 s. The time interval between the moment of radon injection into the chamber and the start of the first period of decisive measurements, regardless of its sequence number, shall be no less than 6 h, and the number of periods for decisive measurements is set to 60, thus in this case, the duration of the test of a single sample increases significantly from 18 h to 66 h.

When testing multilayer samples, the accuracy of the results, according to 9.3, is ensured with a duration of the test following the nomogram in Figure 7, depending on the thickness of the sample and the value indicative of the test material diffusion coefficient.



Key

X total test duration of a single sample, h

Y thickness of the multilayer sample, mm

NOTE The magnitude of the radon exhalation rate from the sample into the ambient air is not less than 10 % of its maximum value.

Figure 7 — Test duration vs. thickness of the multi-layer sample and the radon diffusion coefficient (m^2/s)

12 Expression of the results and assessment of the standard uncertainty for the material

The radon diffusion coefficient in the tested material is expressed, as specified in ISO/IEC Guide 98-3, as in Formula (25):

$$D_M \pm k \cdot u(D_M) \text{ with } k = 2 \tag{25}$$

with the average value of the radon diffusion coefficient in the material, in square metre per second, as in Formula (26):

$$D_M = \frac{1}{N} \cdot \sum_{j=1}^{N} D_j \tag{26}$$

where

D_j is the radon diffusion coefficient in the j^{th} sample, in square metre per second;

N is the number of samples of the test material.

The combined standard uncertainty of the radon diffusion coefficient in the material, in square metre per second is computed following Formula (27):

$$u(D_\mathrm{M}) = \sqrt{s^2(D_\mathrm{M}) + (0{,}05 \cdot D_\mathrm{M})^2} \tag{27}$$

with the standard deviation of the radon diffusion coefficient in the material, in square metre per second, as in Formula (28):

$$s(D_\mathrm{M}) = \sqrt{\frac{1}{N(N-1)} \cdot \sum_{j=1}^{N} (D_\mathrm{j} - D_\mathrm{M})^2} \tag{28}$$

When the first three tests conduct to Formula (29):

$$\frac{s(D_M)}{D_M} < 0{,}2 \tag{29}$$

additional samples should be tested until this condition is fulfilled. The test results are determined by Formulae (25) to (29), taking into account the data for all samples tested.

13 Quality management and calibration of the test device

In accordance with 7.2, the detector calibration is not required for the one-side method.

To check the correct application of this document, periodical control tests are recommended by using one of the following references:

a) the reference single-layer material with a thickness of 0,5 mm to 2,0 mm with a known radon diffusion coefficient, or

b) the calibrated hole in the cap or valve (Figure 2), according to A.5, provides the same low rate of radon activity concentration decrease in the chamber (R_m) and the same uncertainty of results [$u_\mathrm{rel}(D)$] as in the test of the sample of insulating material with a radon diffusion coefficient of about 10^{-11} m^2/s.

If these references are absent, repeatability of the test results is recommended using a material for which the radon diffusion coefficient is about 10^{-11} m^2/s.

14 Test report

The test report shall contain the information below.

14.1 The test report for material

a) a reference to this document, i.e. ISO/TS 11665-12:2018;

b) identification of the testing laboratory;

c) identification of tested material (name, producer, composition, technical specification);

d) identification of the test method (one-side method);

e) date of test performance;

f) results of determining the radon diffusion coefficient for each sample;

g) results of determining the radon diffusion coefficient in material.

14.2 The test report for each sample

a) identification of tested material (name, producer, composition, technical specification);

b) details about sample: number (code), whether the sample contain joints, information on whether the sample was prepared by the testing laboratory or by the client;

c) geometric parameters of the test;

d) duration of the test (decisive measurement);

e) information about the experimental conditions during the test: air, temperature and relative humidity;

f) the results of measurements and calculations of the main parameters (in tabular and graphical form), that were used to determine the radon diffusion coefficient of the sample;

g) results of determining the radon diffusion coefficient of the sample.

14.3 Archived material

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), information whether the sample was prepared by the testing laboratory or by the client, identification of the time between the sample preparation and the start of the test (not applicable for prefabricated membranes), storing conditions;

b) test arrangement;

c) the results of the control measurements (including the results of the reference material or calibrated hole);

d) deviations from standard practice which may influence the test results;

e) all experimental information associated with the testing of material: the results of measurements of values of count rate in tabular and graphical form, the values of influence quantities, the results of radon leakage control, and the results of calculations and analysis of data on each sample.

Annex A
(informative)

Determination of the radon diffusion coefficient of the sample

A.1 Scope

Annex A describes the method of calculation of radon activity concentration in the source-detect chamber and provides an example of determining the radon diffusion coefficient of the sample of radon-proof material (polyolefin) with a thickness of 0,60 mm.

Symbols are described in 3.2.

The equipment is described in Clause 5.

The procedure for the preparation of the sample for testing is described in Clause 6.

The procedure for the preparation of the equipment for the test is described in Clause 7.

The procedure for testing is described in Clause 8.

The determination of the radon diffusion coefficient of the sample is described in Clause 9.

A.2 Method of calculation of radon activity concentration in the source-detect chamber

The system of Formulae (1) to (5) has a solution expressed by Formulae (A.1) and (A.2):

$$C_{sd}(t) = C_0 \left[\int_0^t e^{\lambda(\tau - t)} F(\tau) d\tau + e^{-\lambda \cdot t} \right] \tag{A.1}$$

$$C(x,t) = C_0 \cdot \sum_{i=0}^{\infty} \left[A_i(x) \cdot e^{-q_i \cdot t} \cdot \left(1 + \int_0^t e^{q_i \cdot \tau} F(\tau) d\tau\right) \right] \tag{A.2}$$

The parameters and functions appearing in Formulae (A.1) and (A.2) are defined in the following sequence:

a) The input data are defined as C_0($C_0 = 1$ or any other value), t, D, V_{sd}, d, S_s, λ, h_1, h_2($h_1 = h_2 = 0{,}01$).

b) The following parameters are calculated:

$$H_1 = \frac{h_1}{D},\ H_2 = \frac{h_2}{D}$$

$$\psi_1 = \frac{H_1 H_2}{d \cdot H_1 H_2 + H_1 + H_2},\ \psi_2 = -\frac{H_1 \cdot (d \cdot H_2 + 1)}{d \cdot H_1 H_2 + H_1 + H_2}$$

c) The following solutions of the following Formula are found:

$$tg(\mu_i d) = \frac{(H_1 + H_2) \cdot \mu_i}{\mu_i^2 - H_1 H_2},\ i = 1,2,\ldots 8;$$

d) The sequences of the following numbers are determined:

$$q_0 = \lambda\,,\ q_i = D\,\mu_i^2 + \lambda\,,\ i = 1,2,\ldots,8,$$

$$g_i = -\frac{(\mu_i\psi_1 dH_1 + \mu_i\psi_2 H_1 - \mu_i\psi_1)\cos(\mu_i d) - (\mu_i^2\psi_2 + \mu_i^2\psi_1 d + \psi_1 H_1)\sin(\mu_i d) + \mu_i(\psi_1 - \psi_2 H_1)}{\mu_i^3}\ ;$$

$$L_i = \frac{\mu_i^4 d + \mu_i^2(dH_1^2 + dH_2^2 + H_1 + H_2) + H_1H_2(H_1 + H_2 + dH_1H_2)}{2\cdot(H_2^2 + \mu_i^2)\cdot\mu_i^2}\,,\ i = 1,2,\ldots,8,$$

$$\phi_0 = -(1 + \psi_2)\,,\ \phi_i = \frac{g_i}{L_i}\,,\ i = 1,2,\ldots,8;$$

$$A_0(x) = -(\psi_1\cdot x + \psi_2)\,,\ x \in [0;d]\,;\ A_i(x) = \phi_i\cdot\left(\cos(\mu_i x) + \frac{H_1}{\mu_i}\cos(\mu_i x)\right);\ i = 1,2,\ldots,8;$$

e) The following integral formula is solved:

$$F(t) = \int_0^t K(t,\ \tau)F(\tau)\,d\tau + P(t)$$

$$K(t,\tau) = \frac{S_s}{V_{sd}}\,h_1\sum_{i=0}^{i=8}\varphi_i\cdot e^{-q_i(\tau\text{-}t)}\,,\ P(t) = \frac{S_s}{V_{sd}}\,h_1\sum_{i=0}^{i=8}\varphi_i\cdot e^{-q_i t}\,,$$

for which we construct the following sequence of functions:

$$F_0(t) = P(t)\,,\ F_\mathrm{k}(t) = \int_0^t K(t,\tau)F_{\mathrm{k}\text{-}1}(\tau)\,d\tau\,,\ k = 1,2,\ldots,8;$$

f) The solution of the integral formula can be written in the following form:

$$F(t) = \sum_{k=0}^{k=8} F_\mathrm{k}(t)$$

which determines the functions of interest (A.1) and (A.2).

A.3 Example: Determination of the radon diffusion coefficient of the sample from the waterproof material (polyolefin) with a thickness of 0,60 mm

The test report for sample includes:

a) identification of the tested material:

- name: membrane "…";
- producer: "…";
- composition: polyolefin;

b) details about the sample:

- number (code): 1;
- contains joints: no;
- prepared by the testing laboratory "…";

c) geometrical parameters are given in Table A.1.

Table A.1 — Geometrical parameters

Sample thickness m	Sample diameter m	Chamber volume m^3
0,000 60	0,117	0,000 27

d) duration of the test: 17,0 h (decisive measurement: 10,4 h);

- — start date and time of the sample preparation: 28.09.2012 at 15:00
- — start date and time of the testing: 01.10.2012 at 18:58

e) information about the experimental conditions during the test

- — air temperature: (18 to 23) °C;

f) the results of measurements and calculations of the basic quantities used to determine the radon diffusion coefficient of the sample are shown in Table A.2 and Figure A.1.

Table A.2 — Measurement results of the basic quantities

No. of time interval (i)	Time, (τ_i) h	Rate of decrease of the radon activity concentration in the chamber		Deviation of the measured values from the calculated ones %
		Measured $R_m(\tau_i)$	Calculated $R_c(\tau_i)$	
41	6,604	1,000 0	1,000 0	0,00
42	6,779	1,000 1	0,998 3	0,18
43	6,954	0,993 7	0,996 6	−0,29
…	…	…	…	…
98	16,599	0,908 5	0,907 3	0,12
99	16,776	0,910 1	0,905 8	0,48
100	16,952	0,904 1	0,904 2	−0,01