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Water quality — Radon-222 —

Part 4:
Test method using two-phase liquid scintillation counting

Qualité de l'eau — Radon 222 —

Partie 4: Méthode d'essai par comptage des scintillations en milieu liquide à deux phases

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Foreword

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies (ISO member bodies). The work of preparing International Standards is normally carried out through ISO technical committees. Each member body interested in a subject for which a technical committee has been established has the right to be represented on that committee. International organizations, governmental and non-governmental, in liaison with ISO, also take part in the work. ISO collaborates closely with the International Electrotechnical Commission (IEC) on all matters of electrotechnical standardization.

The procedures used to develop this document and those intended for its further maintenance are described in the ISO/IEC Directives, Part 1. In particular, the different approval criteria needed for the different types of ISO documents should be noted. This document was drafted in accordance with the editorial rules of the ISO/IEC Directives, Part 2 (see www.iso.org/directives).

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For an explanation of the voluntary nature of standards, the meaning of ISO specific terms and expressions related to conformity assessment, as well as information about ISO's adherence to the World Trade Organization (WTO) principles in the Technical Barriers to Trade (TBT), see www.iso.org/iso/foreword.html.

This document was prepared by Technical Committee ISO/TC 147, *Water quality*, Subcommittee SC 3, *Radioactivity measurements,* in collaboration with the European Committee for Standardization (CEN) Technical Committee CEN/TC 230, *Water analysis,* in accordance with the Agreement on technical cooperation between ISO and CEN (Vienna Agreement).

This second edition cancels and replaces the first edition (ISO 13164-4:2015), which has been technically revised.

The main changes are as follows:

— 3.2: index has been modified according to more recent standards;

— Clause 8: a note has been added;

— A.4.2: efficiency and repeatability data have been revised and updated;

— A.4.2: subclause on reproducibility has been added.

A list of all the parts in the ISO 13164 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

Radionuclides are present throughout the environment; thus, water bodies (e.g., surface waters, ground waters, sea waters) contain radionuclides, which can be of either natural or anthropogenic origin:

— naturally-occurring radionuclides, including ^{3}H, ^{14}C, ^{40}K and those originating from the thorium and uranium decay series, in particular ^{210}Pb, ^{210}Po, ^{222}Rn, ^{226}Ra, ^{228}Ra, ^{227}Ac, ^{231}Pa, ^{234}U, and ^{238}U, can be found in water bodies due to either natural processes (e.g. desorption from the soil, runoff by rain water) or released from technological processes involving naturally occurring radioactive materials (e.g. mining, mineral processing, oil, gas, and coal production, water treatment and the production and use of phosphate fertilisers);

— anthropogenic radionuclides such as ^{55}Fe, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{99}Tc, transuranic elements (e.g., Np, Pu, Am, and Cm), and some gamma emitting radionuclides such as ^{60}Co and ^{137}Cs can also be found in natural waters. Small quantities of anthropogenic radionuclides can be discharged from nuclear facilities to the environment as a result of authorized routine releases. The radionuclides present in liquid effluents are usually controlled before being discharged to the environment [1] and water bodies. Anthropogenic radionuclides used in medical and industrial applications can be released to the environment after use. Anthropogenic radionuclides are also found in waters due to contamination from fallout resulting from above-ground nuclear detonations and accidents such as those that have occurred at the Chornobyl and Fukushima nuclear facilities.

Radionuclide activity concentrations in water bodies can vary according to local geological characteristics and climatic conditions and can be locally and temporally enhanced by releases from nuclear facilities during planned, existing, and emergency exposure situations.[2],[3] Some drinking water sources can thus contain radionuclides at activity concentrations that could present a human health risk. The World Health Organization (WHO) recommends to routinely monitor radioactivity in drinking waters [4] and to take proper actions when needed to minimize the health risk.

National regulations usually specify the activity concentration limits that are authorized in drinking waters, water bodies, and liquid effluents to be discharged to the environment. These limits can vary for planned, existing, and emergency exposure situations. As an example, during either a planned or existing situation, the WHO guidance level for ^{222}Rn in drinking water is 1 Bq·l^{-1}, see NOTE. Compliance with these limits is assessed by measuring radioactivity in water samples and by comparing the results obtained, with their associated uncertainties, as specified by ISO/IEC Guide 98-3[5] and ISO 5667-20[6].

NOTE   The guidance level calculated in Reference [4] is the activity concentration that, with an intake of 2 l·d^{-1} of drinking water for one year, results in an effective dose of 0,1 mSv·a^{-1} to members of the public. This is an effective dose that represents a very low level of risk to human health and which is not expected to give rise to any detectable adverse health effects[4].

The ^{222}Rn activity concentration in surface water is very low, usually below 1 Bq·l^{-1}. In groundwater, the activity concentration varies from 1 Bq·l^{-1} up to 50 Bq·l^{-1} in sedimentary rock aquifers, from 10 Bq·l^{-1} up to 300 Bq·l^{-1} in wells, and from 100 Bq·l^{-1} up to 1 000 Bq·l^{-1} in crystalline rocks. The highest activity concentrations are normally measured in rocks with a high concentration of uranium[7].

High variations in the activity concentrations of radon in aquifers have been observed. Even in a region with relatively uniform rock types, some well water can exhibit radon activity concentration much higher than the average value for the same region. Significant seasonal variations have also been recorded (see ISO 13164-1:2013, Annex A[8]).

In circumstances where high radon concentrations might be expected in drinking-water, it is prudent to measure for radon and, if high concentrations are identified, consider whether measures to reduce the concentrations present are justified[2].

This document contains method(s) to determine ^{222}Rn in water samples. It has been developed to support laboratories that need either a certification or accreditation to determine ^{222}Rn in water samples. A certification or accreditation are sometimes required by local and national authorities as well as some customers. The certification and accreditation are provided by an independent body.

The method(s) described in this document can be used for various types of waters (see Clause 1). Minor modifications such as sample volume and counting time can be made if needed to ensure that the characteristic limit, decision threshold, detection limit, and uncertainties are below the required limits. This can be done for several reasons such as emergency situations, lower national guidance limits, and operational requirements.

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Water quality — Radon-222 —

Part 4:
Test method using two-phase liquid scintillation counting

WARNING — This document does not purport to address all of the safety issues, if any, associated with its use. It is the responsibility of the user to establish appropriate safety and health practices and to ensure compliance with any national regulatory conditions.

IMPORTANT — It is essential that tests conducted in accordance with this document be carried out by suitably qualified staff.

1 Scope

This document describes a test method for the determination of radon-222 (^{222}Rn) activity concentration in non-saline waters by extraction and liquid scintillation counting.

The ^{222}Rn activity concentrations, which can be measured by this test method utilizing currently available instruments, are above 0,5 Bq·l^{-1} which is the typical detection limit for a 10 ml test sample and a measuring time of 1 h.

It is the responsibility of the laboratory to ensure the validity of this test method for water samples of untested matrices.

Annex A gives indication on the necessary counting conditions to meet the required detection limits for drinking water monitoring.

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 5667-1, *Water quality — Sampling — Part 1: Guidance on the design of sampling programmes*

ISO 5667-3, *Water quality — Sampling — Part 3: Preservation and handling of water samples*

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

ISO/IEC 17025, *General requirements for the competence of testing and calibration laboratories*

3 Terms, definitions and symbols

3.1 Terms and definitions

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

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

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

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

3.2 Symbols

For the purposes of this document, the symbols given in ISO 80000-10 and the following apply.

a	Massic activity of sample	Bq·g^{-1}
a_S	Massic activity of standard solution at the measuring time	Bq·g^{-1}
$\tilde{a}$	Possible or assumed true quantity values of the measurand	Bq·g^{-1}
a^*	Decision threshold for the total massic activity	Bq·g^{-1}
$a^{\#}$	Detection limit for the total massic activity	Bq·g^{-1}
$a^{\triangleleft}, a^{\triangleright}$	Lower and upper limits of the probabilistically symmetric coverage interval of the measurand, respectively	Bq·g^{-1}
$a^<, a^>$	Lower and upper limits of the shortest coverage interval of the measurand, respectively	Bq·g^{-1}
c_A	Activity concentration	Bq·l^{-1}
m	Mass of the test sample	g
m_S	Mass of standard solution used for the preparation of the counting standard	g
N_0	Number of background counts measured from the LSC spectrum for a given time in the region of interest of the measurand.	
N_g	Number of counts measured from the LSC spectrum for a given time in the region of interest of the measurand.	
r_0	Blank sample count rate	s^{-1}
r_g	Sample gross count rate	s^{-1}
r_S	Count rate of the standard in the counting window (alpha + beta)	s^{-1}
t_0	Blank sample counting time	s
r_{net}	Net count rate	s^{-1}
t_g	Test sample counting time	s
$t_{1/2}$	Radioactive half-life of an isotope	
$u(a)$	Standard uncertainty associated with the measurement result	Bq·g^{-1}
U	Expanded uncertainty, calculated using $U = ku(a)$, with $k = 2$	Bq·g^{-1}
w	Coefficient equal to $1/(\varepsilon\, m)$	g^{-1}
ε	Total efficiency	
ρ	Density	g·l^{-1}
u_{rel}^2	Relative uncertainty	
$\tilde{u}(\tilde{a})$	Standard uncertainty of a as a function of its true value	Bq·g^{-1}
$\tilde{u}(a^{\#})$	Standard uncertainty of an estimate of the measurand when the true value is equivalent to the detection limit	
k_p	Quantile of the standardized normal distribution for the probability p (for instance $p = 1 - \alpha$, $1 - \beta$ or $1 - \gamma/2$)	
λ	Decay constant of the isotope	
ω	Auxiliary quantity	
y	Primary measurement result of measurand	
Φ	Distribution function of the standardized normal distribution	
α	Probability of the false positive decision	
β	Probability of the false negative decision	

4   Principle

Radon is extracted from an aqueous solution by means of a scintillation cocktail not miscible with water (without emulsifier) inside the scintillation vial and counted after the equilibrium is reached with its short-lived decay products [9][10][11][12].

The aqueous sample is drawn with a gas-tight syringe from inside the water volume (i.e., well below surface) to avoid radon losses during sampling and transferred into a scintillation vial containing the desired amount of scintillation cocktail. For the same reason, the water sample is injected below the cocktail surface. The vial is tightly capped, shaken and kept for 3 h, preferably in the dark and at controlled temperature. The sample is then counted by a liquid scintillation counter. Either total counts (alpha + beta) or alpha counts only can be considered. In these conditions ^{222}Rn and its short-lived progeny (^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po) are measured.

5   Sampling

5.1   General

Sampling, handling and storage of the water samples shall be done as specified in ISO 5667-1 and ISO 5667-3. It is important that the laboratory receives a sample that is truly representative and has not been damaged or modified during transportation or storage.

Since radon is easily desorbed from water sample, care should be taken to avoid analyte losses during the sampling.

5.2   Sampling with source preparation "on site"

Attach a plastic tube to a faucet with a proper fitting. Insert the other end of the tube in a wide-mouth flask (6.2.3). Allow a steady water stream to fill and the volume of water should be overflowed in terms of volume, such as twice or three times. Adjust the flow rate to avoid turbulence, bubbles, and empty volumes both in the tube and in the flask.

Draw the water sample aliquot with a gas-tight syringe (6.2.4) inserting the needle well below the surface. Sampling time shall be recorded to calculate decay correction.

Prepare the counting source following the method described in 7.5.

5.3   Sampling without "on site" source preparation

Attach a plastic tube to a faucet with a proper fitting. Insert the other end of the tube in a wide-mouth glass bottle (6.2.2). Allow a steady water stream to flow out and overflow the bottle for approximately 2 min. Adjust flow rate to avoid turbulence, bubbles, and empty volumes both in the tube and in the bottle. Gently extract the tube and screw tightly the cap avoiding any air head space. A 1 l bottle is generally suitable for the sampling. Sampling time shall be recorded to calculate decay correction.

The sample should be transported into the laboratory and analysed ideally within 48 h. The sample should not be frozen to prevent breaking of the container. If the bottle is completely filled without bubbles and tightly capped, then normal road transport can deliver sample without significant radon loss even if the samples are exposed to elevated ambient temperatures.

6   Reagents and apparatus

6.1   Reagents

All reagents shall be of recognized analytical grade and, except for 6.1.4, shall not contain any detectable alpha and beta activity.

6.1.1   Water, distilled or deionized.

Deionized water can contain detectable amounts of ^{222}Rn and short-lived progeny. It is, therefore, strongly recommended that water be boiled with vigorous stirring and allowed to stand for 1 day before use. Otherwise, purge it with nitrogen for about 1 h for 2 l water.

6.1.2   Scintillation cocktail, commercially available scintillation cocktails, not water miscible.

6.1.3   Ethanol, 95 %.

6.1.4   Radium standard solution.

^{226}Ra standard solutions shall be provided with calibration certificates containing at least the activity concentration, measurement uncertainty and/or statement of compliance with an identified metrological specification.

6.2   Apparatus

6.2.1   Balance.

6.2.2   Wide-mouth glass sample bottles, volume from 500 ml to 1 l.

6.2.3   Wide-mouth flask, volume from 500 ml to 1 l.

6.2.4   Gas-tight syringe.

6.2.5   Liquid scintillation counter, preferably with thermostated counting chamber and preferably ultra-low level counter to achieve better detection limits.

6.2.6   Polyethylene scintillation vials, PTFE coated, volume 20 ml.

6.2.7   Glass scintillation vials, low potassium glass, volume 20 ml.

NOTE   PTFE coated polyethylene vials are the better choice since they prevent both the diffusion of the cocktail into the wall of the vial, radon loss and the absorption of radon from the external environment. Glass vials have similar advantages but exhibit a considerably higher background due to ^{40}K content.

7   Procedure

7.1   Preparation of calibration sources

Transfer an accurately known amount, m_S, of the ^{226}Ra standard solution (6.1.4) into a 20 ml scintillation vial (6.2.6 or 6.2.7). Dilute with laboratory water (6.1.1) (see ISO 3696) to the previously chosen mass (e.g. 10 g). Add the scintillation cocktail (6.1.2). Store the sample for at least 25 days until secular equilibrium is reached. A standard solution of ^{222}Rn can also be used if available.

Vigorously shake the vial for a few seconds. A vortex mixer could also be used for shaking. After phase separation, a waiting time of at least 3 h shall be used before starting counting. Since ^{226}Ra is not extracted into the organic phase, its alpha emission would not affect the detection efficiency calibration for ^{222}Rn.

7.2   Optimization of counting conditions

Both alpha + beta counting or alpha counting using alpha-beta discrimination can be used (see manufacturer instructions).

When using alpha-beta discrimination, both alpha background and efficiency are usually lower; in practice it is found that a much lower detection limit can be achieved.

Set the counting window so that the channels affected by photo- and chemo-luminescence are excluded.

NOTE   Since no water is present in the scintillation cocktail phase, the quenching is low and constant, thus no quenching correction is needed.

7.3 Detection efficiency

Let the counting rate be r_S for the counts of the calibration source in the counting window (alpha + beta).

Determine the detection efficiency according to Formula (1):

$$\varepsilon = \frac{r_S - r_0}{a_S \cdot m_S} \tag{1}$$

Acceptance limits for efficiency should be defined.

NOTE ε includes both counting and extraction efficiency. Usual values are in the range of approximately 400 % to 500 % (^{222}Rn, ^{218}Po, ^{214}Po alpha emissions and ^{214}Pb, ^{214}Bi beta emissions). If using alpha-only counting, a lower ε value (≤300 %) is to be expected.

It is advisable to check the method linearity. The efficiency should be assessed using calibration sources whose activities should cover the whole working range.

A more accurate estimate of efficiency can be obtained by preparing and measuring a number of calibration sources.

Efficiencies should be verified with a periodicity established by the laboratory and whenever changes in materials (e.g., scintillation cocktail (6.1.2)) or when maintenance operations are performed on the scintillation counter (6.2.5). A verification or a recalibration is necessary when requirements of instrument quality control are not met.

7.4 Blank sample preparation and measurement

Transfer the chosen quantity (e.g., 10 g) of degassed laboratory water (6.1.1) into the scintillation vial (6.2.6 or 6.2.7). Add the scintillation cocktail (6.1.2) and shake vigorously the vial for a few seconds. A vortex mixer could also be used for shaking.

After phase separation, wait at least 3 h before starting counting. Count the blank sample using the chosen conditions. Let the measured counting rate in the counting window be r_0.

Acceptance limits for blank samples should be defined, also on the basis of the desired detection limit. For this purpose, the use of control charts is advisable.[13]

It is recommended to count blank samples for the same counting time as the test samples.

Blank measurements should be performed with a periodicity established by the laboratory (e.g., monthly) and whenever changes in materials (e.g., scintillation cocktail batch (6.1.2)) or when maintenance operations are performed on the scintillation counter (6.2.5). Verification or a recalibration is necessary when requirements of instrument quality control (see Clause 8) are not met.

7.5 Sample preparation and measurement

Transfer into the scintillation vial (6.2.6 or 6.2.7) the chosen amount of the scintillation cocktail (6.1.2) (e.g., 10 ml). Weigh the vial.

This operation should be done in the laboratory. The weighed, capped vial, containing the scintillation cocktail, can be transported to perform "on site" sampling.