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Technical note

Special Issue
Use of NORM-containing products in construction

Development of a European harmonised standard to determine the natural radioactivity concentrations in building materials

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HIGHLIGHTS

- A European harmonized standard for the determination of ^{226}Ra, ^{232}Th and ^{40}K in building materials.
- The standard considers existing regulations and standardized practices, such as ISO and NEN.
- The method is applicable to samples from products as well as its individual material constituents.

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ABSTRACT

In 2013 the European Commission has published its latest basic safety standards for protection against the dangers arising from exposure to ionising radiation (Council Directive 2013/59/Euratom). The council directive regulates radiation exposure from building materials through the presence of radioactivity (^{226}Ra, ^{232}Th and ^{40}K) in these materials. Pivotal to successful regulation is the availability of a harmonised test method for the determination of the radionuclide concentrations as these nuclides form the basis for dose assessment and compliance. In 2017 a Technical Specification (CEN, 2017) on the determination of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in construction products was published by the European Committee for Standardization (CEN). The purpose of this work is to give an outline of the proposed method, with the protocols for sampling, measurement and data processing as well as a summary of the robustness testing and the expert comments that have been received following the final consultation.

1. Introduction

Building materials of mineral origin are a known source of natural radioactivity [1]. This includes raw materials used as a constituent in end-products or as a building material in its own right. To limit the exposure to natural radioactivity in building materials a reference level of 1 mSv per year for the radiation dose of the inhabitant is imposed in the recently published Council Directive 2013/59/Euratom [2]. The radiation dose is derived from the ^{226}Ra (radium), ^{232}Th (thorium) and ^{40}K (potassium) nuclides, which are (a source of) gamma-emitting nuclides and will contribute to the external radiation dose [3]. The concept of reference level in the directive is – in a strict sense – not a limit value, instead it is a dose level above which it is inappropriate to allow exposures to occur. In the context of building materials this means that restrictions and regulatory control should be imposed for those materials potentially exceeding the reference level.

Pivotal in such regulatory system are robust and harmonised measurement protocols to determine the radiological characteristics needed to assess the radiation dose from building materials and ensure compliance. Various national standards and protocols on the measurement of the activity content from natural radioactivity already exist [4–7]. However, recently the European Committee for Standardization (CEN) has drafted a Technical Specification (TS) [8] on the measurement of ^{226}Ra, ^{232}Th and ^{40}K in building materials, and it is envisaged to be published as a harmonised European Norm (EN) by 2018. The purpose of this work is to give an outline of the proposed test method, with the protocols for

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sampling measurement and data processing as well as a summary of the robustness testing and the comments received by experts in the field.

2. Background and scope

In 2017 a TS [8] on the determination of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in construction products was published by CEN. The TS was prepared by the Technical Committee (TC) 351 on *Construction products: Assessment of release of dangerous substances*[1], and was developed under the Mandate M/366 EN issued by the European Commission [9]. This mandate was issued in the framework of the Construction Products Directive, now Construction Products Regulation [10], and the TS addresses the part of the mandate to provide for a horizontal test method for the determination of natural radioactivity in construction products. The TS is intended to become a harmonised European Norm (EN), and the information produced by applying this TS is used for purposes of CE marking and evaluation/attestation of conformity.

The Technical Specification describes a test method using gamma-ray spectrometry. It comprises of sampling procedures from a laboratory sample, sample preparation, and the sample measurement. Furthermore, it includes calibration procedures, analysis of the spectrum and calculation of the activity concentrations with the associated uncertainties, decision threshold and detection limit, and reporting of the results. Product specification, standardisation of representative sampling and procedures for any product-specific sample preparation are the responsibility of product Technical Committee's (TC's) and are not covered in this TS.

Existing regulations and standardised practices have been considered and are supported. As a result the TS is based on methods already described in standards, such as ISO 10703 [11], 18589-2 [12], ISO 18589-3 [13] and NEN 5697 [4]. The Technical Specification is intended to be non-product specific in scope; however, there are a limited number of product-specific elements such as the preparation of the laboratory sample and drying of the test portion. Finally, the method is applicable to samples from products as well as its individual material constituents.

3. Principles of the method

The test method is based on the use of high resolution semiconductor gamma-ray spectrometry. This type of detection technology typically consists of a detector made from a high purity Germanium crystal. Such detector collects the electric charges produced by the ionization of the semiconductor material, resulting from the interaction with the gamma-rays emitted by the test specimen. Detection of these gamma-rays results in an energy-spectrum, which gives basic information on the energy and intensity of the gamma-ray interactions. The activity of the gamma-emitting radionuclides present in the test specimen is determined using energies and peak areas obtained from the energy peaks that allow the identification and the quantification of these nuclides [14]. For this purpose the test method includes procedures for energy and efficiency calibrations. Such calibration is performed using a calibration standard with known activity of the nuclides to be measured, and is similar in chemical composition and density to the materials that are to be tested. The calibration is based on a preselected set of photopeaks used for the determination of the activity concentration. Selected photopeaks are either emitted by the radionuclide itself or by one of its progeny nuclides [15]. The activity concentration is measured using a homogeneous, mostly granular, test specimen held in a container with a predefined geometry.

For ^{226}Ra and ^{232}Th the activity concentration is determined using a progeny nuclide, while for ^{40}K the concentration is based on the photopeak from the nuclide itself. In those cases where the activity is determined using a progeny nuclide, a secular equilibrium between the progeny nuclide and its originating nuclide is necessary. To reach such equilibrium for ^{226}Ra the test specimen is stored in a radon-tight container [16] for a period of at least three weeks in order to ensure there is no degradation in the equilibrium due to a leakage of radon activity [17]. A normative annex is provided in the standard to assess the radon-tightness of the sealed container under worst-case conditions.

Despite the required waiting time of three weeks a disequilibrium in the ^{232}Th decay chain can be present. Such disequilibrium is caused by different dissolution ratios between thorium and radium, particular hydrogeological history and effects of industrial processes. In case of such disequilibrium the ^{232}Th activity is approximated [2].

4. Sampling, measurement and data processing

4.1. Sampling

The test method contains a dedicated section on sampling and sample treatment prior to the testing of the sample. The sampling procedure starts with a laboratory sample received by the laboratory, and results in one or more test specimens used for counting and a test portion for determining the dry fraction. For this purpose a random portion (test sample) of the laboratory sample is crushed – except for granular and fibrous materials where no crushing is needed – to a particle size smaller than 5 mm to obtain a test sample, and used for a test portion and one or more test specimens. Sampling and sample preparation needed to obtain a product sample, which is subsequently send to the laboratory lies outside the scope and is described in product standards. Nevertheless, reference is made to the CEN Technical Report 16220 [18] for guidance on product sampling.

Where preparation of a cement-based concrete product sample from fresh, wet concrete is required, reference is made to the sampling procedures in EN 12350-1 [19] and the production of hardened specimens in EN 12390-2 [20]. These procedures are additional to the sampling procedures described in the TR 16220 [18].

4.2. Calibration and measurement

The natural radioactivity concentrations of the test specimen are based on the density dependent photo peak efficiencies determined for the gamma-ray energies 352 keV (^{214}Pb, progeny of ^{226}Ra), 583 keV (^{208}Tl, progeny of ^{228}Th), 911 keV (^{228}Ac, progeny of ^{228}Ra) and 1461 keV (^{40}K). The gamma-ray energies are selected considering its counting efficiency, interference of other neighbouring gamma-ray's and its sensitivity to the chemical composition of the sample material [21]. Other relevant energies for the decay series ^{226}Ra, ^{232}Th are listed and can be used for verification purposes. The choice for the 352 keV photo peak from ^{214}Pb is because in the ^{226}Ra series this energy line provides for the highest detector efficiency, and gives around 25% more counts as compared with the 609 keV [15].

Determination of the mean concentration ^{232}Th is based on the findings from ^{228}Ra and ^{228}Th. Reason for this approach is that ^{232}Th with a half-life of $1.41 \cdot 10^{10}$ years has a line at 63.81 keV with a very low emission probability of 0,263% which overlaps a line of ^{234}Th at 63.28 keV with a higher emission probability of 4.1% so that ^{232}Th cannot be determined directly by gamma spectrometry. Determination through its decay radionuclides ^{228}Ac and ^{208}Tl can

[1] Further information can be found under www.centc351.org.