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A comparison of methods for the determination of the natural radioactivity content and radon exhalation

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HIGHLIGHTS

- Activity concentrations and radon exhalation of concrete is measured using methods applicable in the Netherlands and Israel.
- The methods are equivalent and appropriate for testing building materials that fall under radiation protection regulation.
- The methods are on these grounds also suitable for use in harmonised standards that are presently under development.

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ABSTRACT

In this work a comparison of the activity content and radon exhalation rate of five concrete mixtures is carried out by NRG (NL) and SOREQ (IL) using the measurement standards applicable in the Netherlands and Israel, respectively. Comparison of the activity concentrations obtained by NRG and SOREQ for all concrete mixtures comply with proficiency requirements in literature and demonstrate that the results agree within a 99% confidence level. Variations in the weighted sum of the activity concentrations – computed according to the European and Israeli gamma indices – between the two laboratories are even smaller and well within the reported one standard deviation uncertainty.

The measured radon exhalation rates agree within a confidence level of 90%, despite considerable differences in the applied methods and uncertainties in the material's radon exhalation that are beyond the measurement protocol. This includes the effects from humidity and aging, and it is mentioned that future guidance on the sample representativeness would be welcomed. Based on the findings of this work it is concluded that the methods for determining the activity concentration and the radon exhalation are equivalent methods and appropriate for testing of building materials that typically fall under radiation protection regulation. Furthermore, the methods can also be recommended for use in harmonised standards that are presently under development.

1. Introduction

Fly ash – a by-product of burning pulverized coal in an electrical generating station – is widely used in the production of cement and concrete. Its potential as a supplementary cementitious material has been known almost since the start of the last century (Anon, 1914) and offers significant environmental and technological benefits in terms of reduction of carbon dioxide emission accompanying the manufacture of portland cement clinker, increased strength and reduced permeability of concrete, which improves durability of concrete structures. In this case the fly ash content of the final concrete product is usually 2–3% (by mass), assuming a 15–25% cement replacement rate (Kovler, 2012). Use of fly ash as a partial replacement of cement is favourable in structures made of mass concrete – because it reduces cement hydration heat.

As a partial replacement of sand (more accurately – of the fine fraction of sand), fly ash can be introduced in normal-weight concrete mixes by much larger amount, and this feature is especially important in the regions suffering from the lack of good quartz sand as an important concrete constituent (Kovler, 2017). In addition, fly ash as a replacement of fine sand improves workability and

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pumpability of fresh concrete mixes. At the same time, the level of 120 kg m^{-3} is usually not exceeded in concrete mixes applied in construction of habitable structures. Although high-volume fly ash (HVFA) concrete mixes containing about 200 kg m^{-3} of fly ash are known, such mixes are usually cast in pavements, foundations and road construction applications, where they have technological and economic success. However, these applications are not of our concern from a radiological point of view (Kovler, 2012).

As for all materials of mineral origin, fly ash is a source for natural radioactivity. To limit radiation exposure from building materials restrictions are imposed on the use of radioactive material from natural origin through national legislation (IM, 2017; SI, 2010). Such regulation is commonly based on international guidelines as there are the International Basic Safety Standard (BSS) (IAEA, 2014) and the EU BSS (EC, 2013). For example, the EU Basic Safety Standard sets specific requirements to limit the external dose from building materials due to its presence of ^{226}Ra (radium), ^{232}Th (thorium) and ^{4} K (potassium). These radionuclides are a source of gamma-emitting (decay) products, which will contribute to the external radiation dose. Other regulations also take account of the internal radiation dose from radon (ÖNORM, 1998; SI, 2010) due to the presence of ^{226}Ra – the parent nuclide of radon – which contributes to the radon exposure in dwellings.

Pivotal in good regulatory control are robust measurement protocols that provide for material testing with good repeatability and limited uncertainty as its results are needed to assess the radiation dose from building materials and ensure compliance. This includes measurement of the radioactivity concentration from ^{226}Ra, ^{232}Th and ^{4} K as well as the radon exhalation rate. Various national standards and protocols on the measurement of the activity content from natural radioactivity exist (SSM, 1998; UNI, 1999; NEN, 2001a; SI, 2010). Recently the EU has drafted a Technical Specification (CEN, 2017) on the measurement of ^{226}Ra, ^{232}Th and ^{4} K from building materials, and it is envisaged to be published as a European Norm (EN) by 2017. Measurement of radon exhalation rate is also subject of numerous national standards and protocols (NEN, 2001b; SI, 2010) and recently the International Standardization Organisation (ISO) has completed an ISO norm on this theme (ISO, 2016).

The purpose of this work is to perform a comparison on the measurement of the natural activity concentrations and radon exhalation rates from various concrete mixtures with and without fly ash, using the measurement standards applicable in the Netherlands and Israel, respectively. The standards in the Netherlands and Israel are well tested (Blaauw et al., 2001; Haquin et al., 2010; Kovler, 2011) and published by their national standardization organisations. As a result, the standards have a good international standing, e.g. the Dutch standards on natural radioactivity formed the basis for the international EN (CEN, 2017) and ISO (ISO, 2016) standard. For this reason, the proposed comparison will give insight in the consistency of the measured radiation properties that can be expected when assessing regular building materials for the purpose of regulatory control.

In this study a total of five types of concrete are prepared and analysed by the two different laboratories. The laboratories performed measurements according to their own national methods and protocols. Subsequently, the results are compared and a proficiency test is performed to investigate consistency in the measurement data reported by the laboratories.

2. Materials and methods

A series of measurements are performed to determine the activity concentrations of ^{226}Ra, ^{232}Th (^{228}Ra, ^{228}Th) and ^{4} K, and radon exhalation rate from five different concrete mixtures. The measurements are performed by the two nuclear research institutes, NRG and SOREQ. The measurements by NRG are performed in accordance with the Dutch NEN standards NEN-5697 (NEN, 2001a) and NEN-5699 (NEN, 2001b) for activity content and radon exhalation. SOREQ has performed its measurements according to the Israeli standard SI-5098 (SI, 2010).

2.1. Samples

The set of concrete mixtures consists of a reference mixture without fly ash followed by four mixtures containing fly ashes of different origin. For this purpose two sets of samples are prepared. Each laboratory received a set and measured the activity content in three identical samples (0.1 × 0.1 × 0.1 m^3) per concrete mixture that were crushed by the laboratory. The radon exhalation rate is measured in a single test using a separate set of three identical samples (0.1 × 0.1 × 0.2 m^3). These samples were circulated between the laboratories to perform the radon measurements on the same sample material. The fly ash content in each mixture is broadly similar for all cases and is approximately 120 kg m^{-3}, which corresponds with around 40% of the cement content. An overview of the concrete mixtures together with the origin of the fly ash is shown in Table 1. In addition a total of two concrete samples (A.1 and A.2) have been tested on their radon exhalation rate twice to demonstrate the effects from aging. The samples have also dimensions of 0.1 × 0.1 × 0.2 m^3 and are tested after respectively 6 and 23 months.

2.2. Determination of the activity concentrations

2.2.1. NEN 5697

The natural radioactivity concentrations of the specimens are determined according to a standard method published under NEN 5697 (NEN, 2001a). According to this method the density dependent photo peak efficiencies are determined for the gamma-ray energies 352 keV (^{214}Pb, parent ^{226}Ra), 583 keV (^{208}Tl, parent ^{228}Th), 911 keV (^{228}Ac, parent ^{228}Ra) and 1461 keV (^{4} K). Four calibration standards are assembled with increasing densities. The materials used are stearic acid, starch, gypsum and quartz sand, homogeneously mixed with certified amounts of ^{238}U and ^{232}Th, in equilibrium with their daughter nuclides, and ^{4} K. The standards are placed into Marinelli beakers with a volume of about 1 L, weighted and closed radon-tight. To obtain secular equilibrium, a waiting time of at least three weeks is taken into account before counting the samples. All samples are counted using an HPGe detector in a low-background facility. The samples of the material are analysed in an identical way as the calibration standards with respect to geometry, waiting time and radon-tightness of the beaker. The photo-peak efficiencies of the samples are deduced from the efficiency curves of the standard samples by interpolation. The results are expressed per unit of dry weight. Prior to the measurements,

Table 1
Overview of the tested samples.

Sample	Density (kg·m^{-3})	Fly ash (kg·m^{-3})	Origin (–)
1	2420	–	–
2	2440	120	Indonesia
3	2420	120	Australia
4	2430	120	Russia
5	2430	120	Colombia
A.1	2370	–	–
A.2	2260	140	South Africa