Construction and Building Materials xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Gamma exposure from building materials – A dose model with expanded gamma lines from naturally occurring radionuclides applicable in non-standard rooms

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HIGHLIGHTS

- The number of considered gamma emission lines influences the absorbed dose rate in air (D_A).
- A novel model including 1845 gamma emission lines (EGDA) was developed.
- The sensitivity of the EGDA model for different parameters was evaluated.
- Density and thickness are the most important parameters influencing the D_A.
- The EGDA allows performing more accurate dose assessments as alternative for the ACI.

ARTICLE INFO

Article history:
Received 4 June 2017
Received in revised form 9 October 2017
Accepted 10 October 2017
Available online xxxx

Keywords:
Euratom basic safety standards
Dose models
Naturally occurring radionuclides
Naturally occurring radioactive materials
Radioactivity
Building materials
Concrete
Inorganic polymers
Alkali-activated materials
External gamma exposure

ABSTRACT

Building materials are a significant source of gamma rays exposure due to the presence of naturally occurring radionuclides. In order to protect the public from harmful radiation, the European Basic Safety Standards (Council directive 2013/59/Euratom) (European Council, 2014) introduced a one-size-fits-all building(s) (materials) activity concentration index (ACI) based on a limited set of gamma lines. The ACI is considered "*as a conservative screening tool for identifying materials that may cause the reference level (i.e. 1 mSv/y) laid down in article 75(1) to be exceeded*". Regarding calculation of dose, many factors such as density and thickness of the building material, as well as factors relating to the type of building, and the gamma emission data need to be taking into account to ensure accurate radiation protection. In this study the implementation of an expanded set of 1845 gamma lines, related to the decay series of ^{238}U, ^{235}U and ^{232}Th as well as to ^{40}K, into the calculation method of Markkanen (1995), is discussed. The expanded calculation method is called the Expanded Gamma Dose Assessment (EGDA) model. The total gamma emission intensity increased from 2.12 to 2.41 and from 2.41 to 3.04 for respectively the ^{238}U and ^{232}Th decay series. In case of ^{40}K a decrease from 0.107 to 0.106 is observed. The ^{235}U decay series is added, having a gamma emission intensity of 3.1. In a standard concrete room, the absorbed dose rates in air (D_A) per unit of activity concentration of 0.849, 0.256, 1.08, 0.0767 nGy/h per Bq/kg are observed. The use of weighted average gamma lines increased the D_A with 6.5% and 1% for respectively the ^{238}U and ^{232}Th decay series. A decrease of 4.5% is observed in the D_A of ^{235}U decay series when using the weighted average gamma lines in comparison to its non-averaged variant. The sensitivity of the EGDA model for density, wall thickness, presence of windows and doors and room size is investigated. Finally, a comparison of the index and dose calculations relevant for the dose assessment within the European legislative framework applicable towards building materials is performed. In cases where the ACI and density and thickness corrected dose calculation of Nuccetelli et al. (2015) cannot provide guidance, the EGDA allows performing more accurate dose assessment calculations leading to effective doses which can be several 100 μSv/y lower.

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https://doi.org/10.1016/j.conbuildmat.2017.10.051
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Please cite this article in press as: T. Croymans et al., Gamma exposure from building materials – A dose model with expanded gamma lines from naturally occurring radionuclides applicable in non-standard rooms, Constr. Build. Mater. (2017), https://doi.org/10.1016/j.conbuildmat.2017.10.051

1. Introduction

Building materials are a significant source of indoor gamma dose [4]. The importance to address the exposure originating from building materials is underlined in article 75 of the Euratom basic safety standards (EU-BSS) (Council directive 2013/59/Euratom), which must be transposed to national law by EU Member States before February 2018 [1]. This article states that "*The reference level applying to indoor external exposure to gamma radiation emitted by building materials, in addition to outdoor external exposure, shall be 1 mSv per year*". This European legislation was developed to establish basic standards, applicable in EU member states, for the protection against exposure of ionising radiation for workers and the general public. In a broader context, this legislation supports several launched initiatives of the European Commission for turning waste into a resource and promoting re-use and recycling with focus on the building industry in the framework of the Europe 2020 strategy [5–7]. In this context, the EU-BSS aims towards a safe use of by-products, originating from NORM (Naturally Occurring Radioactive Material)-processing industries, like metallurgical slags, fly and bottom ash, phosphogypsum and red mud. These residues are used or investigated to use in cement-based matrixes as supplementary cementitious materials (SCM) on a large scale [8–12]. In addition, more and more research is conducted to use these residues in cement alternatives which generate less CO_2, like inorganic polymers (IPs) [8–10]. This fits with the aim to reduce the usage of primary resources. It is expected that future building materials used for dwellings will shift more and more towards these secondary raw materials that can potentially be rich in naturally occurring radionuclides (NORs): therefore the impact on the external gamma exposure of the use of these secondary raw materials needs to be assessed [10,13,14].

In order to assess the impact on external gamma exposure of building materials, different calculation methods, based on Monte Carlo simulations, integration and simple index and dose formulas, have been developed in the past [2,15–26]. Different dose assessment calculations have been developed based on gamma-ray attenuation and build-up factors [2,16,17,22,27]. These calculations allow specifying the physical parameters of the room and the material it is constructed out, in a straightforward way. The density and wall thickness are identified as the most critical parameters. Modifying these parameters, for the evaluation of non-standard rooms, can generate dose rate differences up to 40% compared to a standard concrete room [27]. Seeking for a standardized approach, the EU-BSS [2] proposes a screening index, named activity concentration index (ACI). This index was originally developed by Markkanen [2] and is described in the technical guide Radiation Protection (RP)-112 [28]. The ACI is based on a number of assumptions that are not all necessarily valid. The ACI assumes a concrete room (400 cm × 500 cm × 280 cm) with a density of 2350 kg/m^3 and thickness of 20 cm for all surfaces (walls, floor and ceiling). In the last years, in order to get a reliable screening tool, that will allow for a realistic discrimination of building materials, a new density and thickness corrected index I(ρd) was developed by Nuccetelli et al. [3]. The available dose assessment models focus on the standard composition of concrete, however the increased usage of residues, which have an *a priori* chemical compositions differing from conventional raw materials (like OPC and gravel), can result in structures with very different compositions. Some models consequently apply a correction factor to compensate for the different composition [29]. In addition, disequilibrium in the ^{238}U and ^{232}Th decay series chain can be present for residues from NORM-processing industries. Information regarding disequilibrium can be valuable for gaining insight into environmental or industrial processes. However, when dealing with the dose assessments of building materials one should assess how meaningful the consideration of disequilibrium is. Up to now, to the authors' knowledge, in none of the existing dose calculations, disequilibrium situations are taken into account. In contrast RP-122 [30] suggests using the highest activity concentration of a radionuclide present in a certain decay series to specify the activity concentration of that whole decay series. In none of the existing tools the presence of ^{235}U and its decay products is considered.

The above mentioned calculation methods have in common that they only use a fraction of the gamma emission lines known today. In practice, this means that often dose models use a specific set of major gamma lines or that the set of several major gamma lines is reduced to one or several averaged gamma lines with the gamma intensity as weighing factor. Whereas, the gamma emission intensity of this averaged gamma line is the sum of the individual gamma emission intensities. This technique is performed to provide simplicity. However, progress has been made in the characterization of the gamma emissions of radionuclides. The Laboratoire National Henri Becquerel has built an online database providing continuously updated information on the gamma emission lines of a wide range of radionuclides that allows going beyond this simplified approach [31]. Implementation of this database into a dose calculation method allows a more accurate safety assessment to evaluate if construction products can be used from a radiation protection point of view [3]. Both sample parameters, like density and composition, as well as room parameters like thickness of the walls, ceiling and floor, number of walls present, the sample composition of each wall etc. impact the final received dose [15,27]. An adaptable dose assessment calculation allows taking these parameters into account.

Using an flexible dose or index calculation, in contrast to a screening index, for the evaluation of building materials fits better with the 1 mSv dose requirement of article 75 of the EU-BSS [2], in particular when dealing with non-standard room and building material parameters. In addition, the implementation of non-standard room and building material parameters deals with the requirement of annex VII of the EU-BSS [2], that states "*The calculation of dose needs to take into account other factors such as density, thickness of the material as well as factors relating to the type of building and the intended use of the material (bulk or superficial)*". The current study implements improvements, based on scientific data available in literature, into the existing and validated Markkanen room model [2]. A sensitivity analysis of the different parameters impacting the calculated absorbed dose rate in air is performed. For the different improvements implemented in the dosimetric evaluation, the impact and practicality for industrial implementation is discussed.

2. Materials & methods

2.1. Materials

For the evaluation of the dose model the composition of concrete, defined by NIST [32], is used, except when mentioned differently.

2.2. Model

2.2.1. Model description

To assess the absorbed dose rate in air (D_A), the room model of Markkanen [2] (see Eq. (1)) is used.

$$D_A = 5.77\text{x}10^{-7}\frac{AC_\rho}{4\pi}\sum_i \gamma_i\left(\frac{\mu_{en}}{\rho}\right)_i E_i \int B_i \frac{e^{-\mu_i s}}{l^2}\text{d}V \quad (1)$$

with D_A the absorbed dose rate in air in Gy/h, AC the activity concentration of a radionuclide incorporated in the material of concern

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