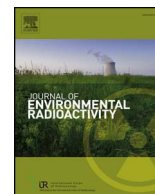




Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

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

Novel spectrometers for environmental dose rate monitoring

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ARTICLE INFO

Keywords:

Environmental dosimetry

Spectrometers

MetroERM

Early warning networks

ABSTRACT

A new generation of dosimeters, based on the scintillators LaBr₃, CeBr₃ and SrI₂, read out with conventional photomultipliers, to be used in the field of environmental gamma-radiation monitoring, was investigated. The main features of these new instruments and especially their outdoor performance, studied by long-term investigations under real weather conditions, are presented. The systems were tested at the reference sites for environmental radiation of the Physikalisch-Technische Bundesanstalt. The measurements are compared with that of well characterized classical dose rate reference instruments to demonstrate the suitability of new spectrometers for environmental dose rate monitoring even in adverse weather conditions. Their potential to replace the (mainly Geiger Müller based) dose rate meters operated in about 5000 European early warning network stations as well as in environmental radiation monitoring in general is shown.

1. Introduction

When dosimetric information is calculated from spectra on a routine basis, the applied instruments can be referred to as spectro-dosimeters. In the context of the European Metrology Research Programme (EMRP), the joint research project MetroERM, *Metrology for radiological early warning networks in Europe* (Neumaier et al., 2016), started in 2014 and finished in 2017. One major goal of the project was the advancement of spectro-dosimeters, based on scintillation detectors (LaBr₃, CeBr₃, SrI₂). This new generation of robust, commercially available, affordable and sensitive spectrometry systems with acceptable energy resolution (of about 3%–4% at 662 keV) (Quarati et al., 2013, 2007; Hawrami et al., 2008), compared to 10% of NaI based detectors, is capable of providing both spectral information (i.e. nuclide specific) and dose rate values. In addition the LaBr₃ and CeBr₃ based detectors have a faster decay time of the scintillation light and thus can measure higher dose rates. Due to the higher efficiency compared to NaI smaller scintillation crystals can be used. The possibility to identify radionuclides and to quantitatively analyse activity concentrations of ground contamination levels suggest to use these systems instead of classical dose rate meters.

The major application of the spectro-dosimeters is environmental radiation monitoring, especially by early warning networks (e.g. LaBr₃ (Toivonen et al., 2008)). Currently, about 450 nuclear power plant units (about 190 in Europe), more than 250 research reactors (100 in Europe), and many other nuclear installations are in operation. Therefore, legal regulations were implemented (e.g. Council Directives of the European Commission) laying down basic safety standards (BSS)

for the protection of the health of workers and the general public (2013/59/EURATOM) (European Commission, 2014). The EURATOM treaty and various IAEA (International Atomic Energy Agency) recommendations and UNSCEAR reports (United Nations Scientific Committee on the Effects of Atomic Radiation) underline the importance of appropriate environmental radiation monitoring. Radiological data provided by environmental radiation monitoring networks are a key input for counter measures in case of nuclear or radiological emergencies. Also during routine operation of nuclear installations, reliable and metrologically validated data on dose rate values and activity concentrations are indispensable.

Spectro-dosimeters are not limited to early warning networks and can also be used on unmanned aerial vehicles as described in (Kurvinen et al., 2005; Connor et al., 2016) for airborne radiation mapping. This is a major task for the follow up project called "Preparedness" where those systems are investigated under metrological conditions. Another application is the measurement in aquatic environments such as activity concentration in lakes or ocean waters (Povinec et al., 1996; Zhang et al., 2015). In the following paper the authors focus on the use of these systems as dosimeters in early warning networks.

The main disadvantage of a classic dose rate meter, like a Geiger Müller counter or an ionisation chamber, is the lack of information concerning the single components of the environmental radiation. This is illustrated by the following equation, describing the dose rate reading R of such an instrument:

$$R = R_0 + q_{\text{TR}} \cdot H_{\text{TR}} + q_{\text{SCR}} \cdot H_{\text{SCR}} + q_{\text{ART}} \cdot H_{\text{ART}} \quad (1)$$

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Received 31 May 2017; Received in revised form 8 December 2017; Accepted 23 January 2018

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where R_0 is the reading due to the inherent background of the instrument, $q_{TR} \cdot H_{TR}$ the reading due to natural terrestrial radiation, $q_{SCR} \cdot H_{SCR}$ the reading due to secondary cosmic radiation (SCR) and $q_{ART} \cdot H_{ART}$ the possible reading due to an artificial source of ionizing radiation, such as an enhanced dose rate caused by a nuclear installation (routine monitoring) and/or undesired contaminations (emergency monitoring).

As both, the terrestrial dose rate H_{TR} and the dose rate caused by secondary cosmic radiation H_{SCR} vary with time, it is impossible to derive the artificial dose rate H_{ART} directly from the instruments reading (if H_{ART} is in the same order of magnitude as H_{TR}). Moreover, the response factors q_{TR} , q_{SCR} and q_{ART} are often not well known and may deviate considerably from unity (note: most dosimeters show an over-response to secondary cosmic radiation, i.e. $q_{SCR} > 1$; the other two response factors depend on the energy spectra of the radiation). Finally, the inherent background can be in the same order of magnitude as the natural background radiation reading, which is, e.g., often true for Geiger Müller counter based instruments. These facts make it very difficult to identify small increases of the dose rate caused by artificial radiation. A comprehensive study of the problems of environmental radiation monitoring is given in (Zähringer et al., 2014).

This article describes outdoor benchmark experiments, carried out by the Physikalisch-Technische Bundesanstalt (PTB), the national metrology institute of Germany, at dedicated reference measuring sites, subsequently after the characterisation of the spectro-dosimeters. Basic features (energy resolution, angular dependence) of the scintillation based spectrometry systems LaBr₃, CeBr₃ and SrI₂, studied under metrological conditions, are already described in a former article (Kessler et al., 2016), primarily the energy dependent response function which converts counts to dose.

The technical stability of spectro-dosimeters installed outdoors has to be guaranteed to obtain reliable dose rate values, e.g. the temperature dependence of the gain caused by the photomultiplier (PM) needs to be compensated, otherwise an unwanted temperature dependence of the indicated dose rate is observed.

This publication presents the performance of three fully characterized scintillation detectors during a long-term outdoor campaign in a direct comparison with a conventional reference dose rate meter, which is traceable to PTB's primary standards.

2. Methods

2.1. Scintillator based spectro-dosimeters

The spectro-dosimeters investigated in this paper are based on commercially available parts on the basis of scintillation crystals (LaBr₃, CeBr₃ and SrI₂), read out by PMs in combination with digital spectrometer bases (including digital multi channel analysers (MCA), preamplifier and high voltage supply). Due to the relatively high efficiency of these detectors (caused by the high photon interaction cross section and the high density of the materials), already small crystals (e.g. cylinders of 2.54 cm in diameter and 2.54 cm in length) are as sensitive as conventional high pressure ionisation chambers or proportional counters. That means that under environmental conditions, a 600 s measurement has the capability to detect a change of the net dose rate of less than 10 nSv/h. The lower limit of the measuring range, which is in the order of some nSv/h (assuming 600 s measurements) is so low that the natural background radiation can be quantified with a sufficient precision. In order to be able to neglect the angular dependence of the response, the selected crystals have a cylindrical shape with an identical diameter and height. However, the small (in most cases negligible) energy dependent angular response of the detectors was both absolutely measured and relatively calculated by Monte Carlo simulations. Details about the materials and dimensions of the crystals as well as of the PM and readout electronics are listed in Table 1.

The detectors including the MCA were put inside a cylindrical aluminium casing with a height of 530 mm, a diameter of 90 mm and a

Table 1

Properties of the detector systems consisting of cylindrical shaped scintillation crystals, Hamamatsu Photomultipliers and read-out electronics.

Crystal type	Dimensions height × diameter in cm	Photomultiplier Hamamatsu type	Read-out electronics	Software
CeBr ₃	2.54 × 2.54	R6231-01	Ortec digiBase-E	MAESTRO
LaBr ₃	2.54 × 2.54	R6231-01	Ortec digiBase-E	MAESTRO
SrI ₂	2.54 × 2.54	R6231-01	GBS Base527	WinSPEC

wall thickness of 2 mm. Foam material was used to position the detectors in the tube casings and to insulate the detectors against moderately low temperatures (using the heat dissipation of the electronics). For the ongoing operation during winter time at potentially much lower temperatures, a heating system was developed. A temperature sensor was mounted inside the housing of the detector as close as possible to the scintillation crystal. That also allows the correction of the measured spectra on the basis of the measured temperature value.

2.2. Determination of dose rate values

According to the European basic safety standards, area doses have to be expressed in the quantity ambient dose equivalent, $H^*(10)$, because the quantity can be used to quantify photon and also particle radiation. Hence, doses and dose rates reported in this article are expressed in ambient dose equivalent and ambient dose equivalent rate, respectively.

The basic principle to derive dose rate values from pulse height spectra of spectrometers with isotropic response is the following: both, the dose and the pulse height spectrum are caused by the energy depended photon fluence of the external gamma radiation field. Therefore, there is a direct relation between the ambient dose equivalent $H^*(10)$ and the corresponding measured pulse height spectrum \vec{n} defining the response vector \vec{v} .

$$H^*(10) = \vec{v} \cdot \vec{n} \quad (2)$$

Based on this equation, there are several different methods to derive area doses from pulse height spectra described in the past. An overview of the main methods is presented in (Dombrowski, 2014). A common method is to analyse the photo-peaks of the energy spectrum, which is suited for spectrometers with a good energy resolution such as a high purity Ge-detectors. From the spectra the activity concentration of the isotope in question is calculated. (Beck et al., 1972). Another possibility is to determine the photon fluence from the spectra. Therefore incomplete energy absorption, mostly Compton-scattering, has to be taken into account and removed from the spectrum. This can either be done by stripping the spectrum from high energies to low energies or by the complete unfolding of the spectra. (Clouvas et al., 1998; Reginatto, 2010). After the application of one of these three methods a fluence spectrum is derived which can be converted to dose rate with the appropriate conversion factors (e.g. for $H^*(10)$).

The method used for this investigation is described in (Dombrowski, 2014) as: *Conversion of complete spectra without deconvolution* also described in (Toivonen et al., 2008). The counts of the spectra are directly converted into dose rate values. The recorded γ -spectrum is divided into several energy regions and a conversion factor from counts to dose for each region is experimentally determined by using mono-energetic sources. Additional conversion factors were calculated with the Monte Carlo code *EGSnrc* (Kawrakow, 2000). The spectra calculated with the MC-simulations for γ -energies of 30 keV, 2015 keV, 2315 keV, 2615 keV, 2915 keV and 3215 keV, were used to derive conversion factors that are practically impossible to access with mono energetic

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