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Characterization and calibration of a novel detection system for real time monitoring of radioactive contamination in water processed at water treatment facilities

P. Carconi[a], P. De Felice[a,*], A. Fazio[a], A. Petrucci[a], M. Lunardon[b], S. Moretto[b], L. Stevanato[b], D. Cester[b], P. Pastore[c]

[a] ENEA, National Institute of Ionizing Radiation Metrology, Casaccia Research Centre, Via Anguillarese, 301 - S.M. Galeria, Galeria, I-00123 Roma, Italy
[b] University of Padova, Department of Physics and Astronomy, Via Marzolo 8, I-35131 Padova, Italy
[c] University of Padova, Department of Chemical Sciences, Via Marzolo 1, I-35131 Padova, Italy

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ABSTRACT

Characterization and calibration measurements were carried out at the National Institute of Ionizing Radiation Metrology of ENEA on the TAp WAter RAdioactivity (TAWARA) Real Time Monitor system recently developed for real time monitoring of radioactive contamination in water processed at water treatment facilities. Reference radiations and radionuclides were chosen in order to reflect energy ranges and radiation types of the major water radioactive contaminants possibly arising from environmental, industrial or terroristic origin. The following instrument parameters were tested: sensitivity, selectivity, background, short/long term stability, linearity with respect to activity.

1. Introduction

A novel detection system was designed and constructed in the frame of the TAp WAter RAdioactivity Real Time Monitor (TAWARA_RTM) project, financed by the EU under the FP7 research programme (FP7-SEC-2012-1). The TAWARA_RTM system is intended to be used for real time monitoring of radioactive contamination in water processed at water treatment facilities providing fast radiological alarm and radionuclide identification. The system is based on three different units. An Early Alarm Detector (EAD), placed at the water intake and based on a gamma-ray monitor, for gamma radioactivity early detection. A Real Time Monitor (RTM), placed just after the water treatment and based on a number of plane windowless ZnS scintillation detectors, used for continuous monitoring of gross alpha and beta radioactivity in water. In case of any positive detection (gamma, alpha or beta) water samples are further analysed by a Spectroscopy Detector (SPEC) equipped with ion-exchange resin concentrator and aiming at radionuclide identification and quantification. Signals from all TAWARA detectors are processed by modern digitizing techniques. Dedicated software as well as Information and Communication Technology (ICT) infrastructure was developed for system operation and alarm management. The TAWARA_RTM platform was conceived to detect a gross alpha/beta activity of the order of 1 Bq/l in several tens of minutes. Detailed description of the design and construction are reported elsewhere (TAWARA, 2016).

A number of characterization tests and calibration measurements were carried out during the last part of the project at the Italian Primary Metrology Institute for ionizing radiation, National Institute of Ionizing Radiation Metrology of ENEA (ENEA-INMRI). Results of TAWARA characterization and calibration tests have been reported in ENEA-INMRI official calibration certificates.

The testing and calibration work started with a careful selection of reference radiations and radionuclides, then single-nuclide aqueous solutions of the selected radionuclides were standardised and a set of reference materials and standard sources were prepared. Special radioactive-water generators were constructed to introduce radioactive reference material samples into the instruments, simulating water sampling from a pipeline. Calibrations were carried out in field-close conditions simulating, as far as possible, the application of the instrument for the intended use.

The following instrument parameters were tested: sensitivity, selectivity, background, short/long term stability, linearity with respect to activity. The testing and calibration work for the TAWARA system is described in the present paper.

As far as possible, tests were carried out with reference to available national and international Standards such as those issued by European

* Corresponding author.
E-mail address: pierino.defelice@enea.it (P. De Felice).

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Table 1
Reference nuclides used for EAD, RTM and SPEC testing and calibration.

System	Alpha (main α energy, keV)	Beta (max β energy, keV)	X/gamma (x, γ energy, keV)
EAD	–	–	Am-241 (59) Co-60 (1173) Co-60 (1333) K-40 (1461)
RTM	Am-241 (5486)	Co-60 (317) K-40 (1311) Sr-90 (546) Y-90 (2280) F-18 (633)	Am-241 (59) F-18 (511) Co-60 (1173) Co-60 (1333) K-40 (1461)
SPEC	–	–	Am-241 (59) Cd-109 (88) Co-57 (122) Ce-139 (166) Sn-113 (392) Cs-137 (662) Y-88 (898) Co-60 (1173) Co-60 (1333) Y-88 (1836)

Committee for Standardization (CEN), International Organization for Standardization (ISO), International Electrotechnical Commission (IEC) and Bureau International des Poids et Mesures (BIPM). In particular, terminology and definitions are in agreement with ISO/IEC (2012). All nuclear data used in the present work were taken from DDEP/BIPM (BIPM, 2013). Measurement uncertainties were evaluated and reported according to ISO/IEC (2008). Combined standard uncertainties are reported with a coverage factor k = 1, approximately corresponding to the 68% confidence level.

2. Aqueous reference materials and standard sources

Reference radiations and radionuclides for testing and calibration were chosen in order to reflect energy ranges and radiation types of the major water radioactive contaminants possibly arising from environmental, industrial or terroristic origin. A further consideration in radionuclide selection was the need to avoid internal contamination of the RTM detectors. A list of reference nuclides for EAD, RTM and SPEC testing and calibration is reported in Table 1.

A set of 3.6 cm^3 standard aqueous liquid solutions with single high-purity nuclides were prepared in flame-sealed glass ampoules. The mass of each solution was gravimetrically determined with high accuracy (combined standard uncertainty lower than 0.1%). Master single-nuclide standard solutions were then calibrated at ENEA-INMRI, with relative combined standard uncertainties lower than 1%, by the secondary standard well-type Centronic IG11 ionization chamber, traceable to the Italian primary standards of radionuclide activity (Capogni, 2009). Accurate radionuclide purity checks were performed by high-resolution gamma-ray spectrometry on all master solutions used. No radioactive impurity was found with activity higher than 0.1% of the reference nuclide activity.

While detectors used in the EAD and SPEC systems are hermetically sealed and direct contact of water solution with the sensitive detector elements is not possible, detectors used in the RTM system are designed to be in direct contact with the water solution under measurement. For this reason a special study and experiments were needed to identify the chemical composition of the aqueous reference material to be used for the RTM system to avoid detector surface damage and radionuclides adsorption on the detector surfaces with possible radioactive internal contamination of the system. To this regard, acid components and ionic form radionuclides were avoided in defining and realising the chemical composition of the RTM diluents. Special complexing agents (DTPA and EDTA) were added to avoid radioactive ion adsorption on surfaces. Stable carriers, with concentration of 25 µg per gram of solution, were also added in some cases. Although unlikely, internal contamination during system operation in the final installation cannot be excluded a priori. In this case the sensitive detector elements should be replaced.

Table 2
Aqueous reference materials prepared for EAD and RTM testing and calibration. Each reference material was prepared with the total volume of 10 L.

Source code	Radionuclide	Chemical composition	Activity concentration (Bq/kg)
BLANK	None	Distilled water, 1 MΩ	–
EAD1	Am-241	Distilled water + 0.1N DTPA carrier free	139
EAD2	Co-60	Distilled water + 0.1N EDTA carrier free	42.1
EAD3	K-40	Distilled water carrier free	105
RTM1	Am-241	Distilled water + 0.1N DTPA carrier free	54.4
RTM2			64.8
RTM3			110
RTM4			132
RTM5			166
RTM6	Co-60	Distilled water + 0.1N EDTA carrier free	23.1
RTM7			39.8
RTM8			962
RTM9	F-18	Distilled water carrier free	1581
RTM10	K-40	Distilled water carrier free	38.8
RTM11			105
RTM12	Sr-90	Distilled water + 0.1N DTPA carrier free	26.6
RTM13			67.8

The single-nuclide aqueous solutions were gravimetrically diluted or deposited to prepare a total number of 17 different spiked water reference materials traceable to ENEA-INMRI, as summarized in Table 2. Activity concentration levels for each radionuclide ranged from 23 Bq/L up to 1581 Bq/L with combined standard uncertainties of 2%. The activity concentration and stability of all these sources were also checked by gamma spectrometry and liquid scintillation counting. All the measured activity values were found in agreement with corresponding values obtained from gravimetric determination with maximum deviations of the order of ± 1%.

Aqueous reference materials were complemented by standard sources prepared on plastic tube annular supports and hermetically sealed. These sources were used to check system stability, energy/efficiency calibration of the SPEC system and to perform the initial and periodic system tuning. They were intended to simulate the spiral shaped tube, filled with ion exchange resin that will be installed in the final SPEC assembly. To this purpose, a ring shaped tube (same thickness and diameter as the spiral tube) was filled with standard radioactive solution and sealed.

A total number of 3 different annular standard sources were prepared as reported in Table 3. Activity levels for each radionuclide ranged from 245 Bq up to 330 Bq with combined standard uncertainties of 2%. A picture of the annular sources is reported in Fig. 1.

3. Laboratory set up

Early Alarm Detector makes use of a classic NaI(Tl) detector as a gamma-ray early monitor. The detector was surrounded by a dedicated INOX cylindrical tank, with lower inlet and upper outlet, which may contain up to 10 L of aqueous solution. The tank itself and the data acquisition module were surrounded by 2.5 cm thick lead shield. Read-out was provided by the new embedded data acquisition system “Gammastream”, developed by CAEN, hosting a digitizer with 62.5 Ms/s, 12 bit (CAEN, 2016).