Applied Radiation and Isotopes 95 (2015) 13–22

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

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

Production of reference sources of radioactive aerosols in filters for proficiency testing

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HIGHLIGHTS

- Proficiency testing campaigns organized by IRSN.
- Special preparation and characterization of filter sources.
- Production bench description for aerosols calibrated for size and activity.
- Measuring filters and assigned values.

ARTICLE INFO

Article history:
Received 27 January 2014
Received in revised form
18 September 2014
Accepted 26 September 2014
Available online 7 October 2014

Keywords:
Measurement
Environment
Radioactivity
Air
Aerosol
Filter
ICARE bench

ABSTRACT

In the framework of the organization of proficiency testing, filters with deposits of ^{137}Cs and $^{90}Sr+^{90}Y$ radioactive aerosols have been submitted to laboratories for radionuclide measurement. Procedures for the special preparation and characterization of filters have been developed. The different steps of filter preparation, determination of the deposited radionuclide activity and characterization of the homogeneity of these deposits are presented. This method of filter preparation can also be used in the production of secondary standards, whose properties are more adapted to the needs of laboratories measuring radioactivity in filters than are the solid sources that they typically use.

1. Introduction

Determining the activity deposited in aerosol sampling filters in the context of monitoring the environment and facilities and their releases remains a major metrological challenge because of the diversity of filters and their conditions for use.

The lack of truly suitable standard filters precludes satisfactorily linking the radioactivity measurement of these filters to a primary reference. The majority of laboratories involved in this type of measurement use solid sources to calibrate their instruments. These sources are not representative of aerosol sampling on a filter. Indeed, the detection efficiency of the instrument is highly influenced by numerous parameters related to aerosol deposits in the filtering medium. Several current studies (Geryes, 2009; Geryes, 2009) show that, even in the favorable case of measuring filters with a small surface deposit, inappropriate calibration can result in under-estimating deposited activity by 40% in some cases. Such results underline the importance of developing global modeling that correlates the differences noted in nuclear measurements with the airborne particle deposition mechanisms or the parameters linked to the filter exposure conditions. Such studies are complicated by the wide variability of media used (type of fiber used in the filters, compacity, thickness, etc.) (Sioutas, 2001); sampling conditions, such as filtering velocities; aerosol characteristics (Lippmann, 2001); etc.

Under proficiency testing campaigns organized by the Institut de Radioprotection et de Sûreté Nucléaire (IRSN), especially for the needs of the national network for radioactivity measurements of the environment, preparing and characterizing reference filters for

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http://dx.doi.org/10.1016/j.apradiso.2014.09.014
0969-8043/

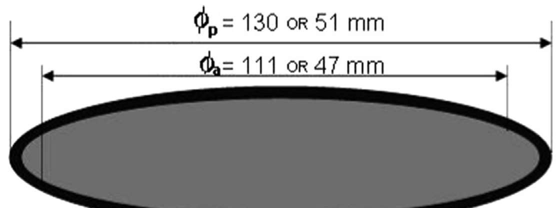

Fig. 1. Geometry of the proposed reference sources in the C569 filter and in the B132 filter.

laboratory measurement conditions is also an issue of prime importance. Therefore, for the first time, filters with a deposit of a radioactive aerosol have been submitted for testing.

The work required to prepare these references is detailed in the remainder of this document. After this study, IRSN has become an accredited PT provider following ISO 17043 requirements (ISO/IEC 17043, 2010).

2. Characteristics of the test item

The medium required for the deposition of the aerosols was the first decision made to build representative test items. Two types of filters were chosen as representative test items for aerosol deposition: B132 and C569 (B. Dumas company[1]). This choice was driven by the fact that these filters are used by a majority of laboratories carrying out environmental measurements in filters and by the monitoring networks of most plant operators in the nuclear industry. Two different materials could also be explored with this choice:

- A so-called "blue" filter (B132) that is made up of pure cellulose
- and a so-called "yellow" filter (C569) composed of a blend of glass fibers and cellulose.

The materials' "physical" diameter is different from the B132 ($\varnothing_p$=51 mm) and C569 ($\varnothing_p$=130 mm) filters (see Fig. 1). By considering the filter holder, the active diameter ($\varnothing_a$) upon which the radioactive aerosols are deposited is also defined as 47 mm for the B132 filter and as 111 mm for the C569 filter.

The technical characteristics of the two chosen filters, such as the collection efficiency for standard fluorescein aerosols (uranine) with a mass median aerodynamic diameter (MMAD) of 0.18 μm (NF M60-760, 2001), are shown in Table 1. For example, the collection efficiency measured using the NFX44-011 standard (NFX 44-011, 1972) for a sampling velocity of 1 cm/s is 96% for the C569 filter but only 65% for the B132 filter.

The pressure drop and the collection efficiency for uranine aerosols versus the sampling velocity were measured using the same type of filter, and these measurements are shown in Figs. 2 and 3 (NF M60-760, 2001).

The chosen radionuclides were ^{137}Cs (β/γ emitter) and ^{90}Sr (pure β emitter). An aerosol deposit was made from two successive layers using the ICARE test bench (described in the next paragraph), starting with the aerosols labeled with ^{137}Cs.

The radioactive aerosols were then protected to ensure the survival over time of the test items and to make them easier to handle.

3. Production bench for aerosols calibrated for size and activity (ICARE)

The ICARE test bench (Monsanglant-Louvet et al., 2012; Ruzer et al., 2005), which is schematized in Fig. 4, is a test bench used to generate radioactive aerosols calibrated for particle size and activity concentration.

The principle of aerosol generation is based on the ultrasonic spraying of a solution made up of carrier salts and radioactive products. Choosing the salt is tricky because it must be readily dissolvable in water and allow for labeling by the desired radionuclides. For the aerosols labeled with ^{137}Cs, cesium chloride (CsCl) is used. The production of aerosols labeled with ^{90}Sr+^{90}Y is more difficult because secular equilibrium must be maintained between the two radionuclides to obtain a stable source over time. It was decided to prepare a mix of strontium chloride ($SrCl_2$) and yttrium chloride (YCl_3). The concentrations of the carrier salts in the sprayed solutions were on the order of 1 g/L.

The expected size distribution is centered on an activity median aerodynamic diameter (AMAD) of 4 μm. The size distribution of the produced aerosol was measured using an ELPI (Electrical Low-pressure Impactor) to experimentally determine its MMAD and the geometric standard deviation (σ_g). Fig. 5 shows that for the two types of solutions produced, the MMAD is approximately 4.2 μm with σ_g=1.5 for the $SrCl_2+YCl_3$ aerosols and approximately 3.0 μm with σ_g=1.7 for the CsCl aerosols.

A reference filter (FSLW from the company Millipore[2]) is used as the control of the activity concentration by sampling the aerosol for 20 min in the reference channel (see Fig. 4) of the ICARE test bench and subsequently by global beta counting connected to the primary standards using the activity. Figs. 6 and 7 show the data obtained for the ^{137}Cs and ^{90}Sr+^{90}Y volumic activities for the B132 filter (A) and for the C569 filter (B); each point corresponds to the mean volumic activities upon being sampled by the FSLW filter for 20 min. Table 2 shows the average values of the activity concentration based on different sequences of the generation of aerosols produced and how these values are dispersed. The dispersion is characterized by two parameters, the standard deviation s_i and the repeatability S_r (NF ISO 5725-2, 1994), which are defined as

$$s_i = \sqrt{\frac{\sum_{i=1}^{n_i}(x_i-\bar{x})^2}{n_i-1}}, \tag{1}$$

and

$$S_r = \sqrt{\frac{\sum_{i=1}^{p}(n_i-1).s_i^2}{\sum_{i=1}^{p}(n_i-1)}}, \tag{2}$$

with n_i being the number of repetitions of the experiment, x_i being the activity concentration for each n_i repetition, $\bar{x}$ being the average activity concentration for each experiment, and p being the number of experiments of the same type.

This monitoring shows that the repeatability of the activity concentration for aerosol generation has a maximum 0.59 Bq/m^3 for activity concentrations on the order of one Becquerel per cubic meter to generate aerosols labeled with ^{137}Cs. This repeatability has, in this case, a maximum of 1.01 Bq/m^3 for generating aerosols labeled with ^{90}Sr+^{90}Y. This dispersion of activity concentration and, therefore, the dispersion of related samplings prompted us to allocate an assigned activity value for each radioactive fiber filter produced.

4. Preparing test items

The filter-sampling device is made up of two filter holders in series monitored by a flowmeter and a sampling pump. The first filter allows for the collection of the aerosol sample, while the second filter, which is an ULPA filter in Teflon (FSLW), allows for the measurement of the first filter efficiency. The flowmeter is used to monitor the flow rate through the filters in real time. The

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[2] Millipore Corporate Headquarters: 290 Concord Road, Billerica, MA 01821.