Monitoring of the radioactivity of gaseous gamma-emitting iodine isotopes in the air

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1 Purpose

The aim of the procedure described is the nuclide-specific determination of gaseous radionuclides of iodine via their gamma radiation by direct measurement of the sample after enriching the iodine on an adsorbent. In the simplest of cases, no distinction is made between gaseous, elemental iodine (I_2) and organic iodine compounds (e. g. CH_3I).

The procedure complies with the requirements of the Directive on the Monitoring of Emissions and Immissions of Nuclear Plants (REI) [1] in normal operation and with the requirements of the General Administrative Regulation on IMIS (AVV-IMIS) [2].

2 Measurand, unit of measurement and limit of detection to be required

2.1 Measurand

The measurand is the activity concentration of gamma-emitting gaseous iodine, averaged over the sampling period and related to the middle of the collection interval.

2.2 Unit of measurement

The unit of measurement is the radioactivity concentration in $Bq \cdot m^{-3}$.

2.3 Required limit of detection

According to REI, in normal operation of nuclear plants, a limit of detection of $5.0 \cdot 10^{3}$ $Bq \cdot m^{-3}$, related to a collection interval of 14 days, is required for I-131 [1]. Within the scope of the routine measuring program in the Integrated Measuring and Information System (IMIS) for the Monitoring of Environmental Radioactivity, a limit of detection of $2.0 \cdot 10^{-2}$ $Bq \cdot m^{-3}$ applies, related to a collection period of seven days with one hour measuring time [2].

3 Method of measurement

3.1 Sampling

By means of a pump, the air is first pre-cleaned from airborne particle by a filter for suspended particles and then, for enrichment of the gaseous radionuclides of the iodine, passes through a KI-impregnated activated carbon granulate [3, 4]. The airflow rate should be set to 1 to 3 $m^3 \cdot h^{-1}$ in order to obtain a sufficiently long contact time of more than 0.2 seconds between the flowing air and the adsorber material. This way, an almost 100 % adsorption of the iodine and the iodine compounds is achieved. In order to prevent condensation and to improve the collection efficiency, a heater is to be provided for increasing the temperature of the supply air. The collection efficiency for I_2 and CH_3I amounts to more than 99.9% at a temperature of 45°C and a relative humidity of 77 %.

Constant pre-heating of the air must be guaranteed to ensure equally good adsorption properties of the adsorber. In order to reduce adsorption losses, a short supply duct of appropriate material (e. g. stainless steel or polytetrafluorethylen, e. g. Teflon®) with a smooth surface is to be used.

The adsorber material is filled into a cylindrical stainless steel cartridge (see figure).

Figure 1: Iodine adsorber cartridge, filled with KI-impregnated carbon granulate

If collection times are long, at least two cartridges should be connected one after another so that the filter efficiency can be determined. The cartridges arranged behind are also analyzed to determine the filter efficiency when iodine is measured in the first cartridge.
By connecting two cartridges with adsorbers of different selectivity one after another, both the activity concentration of the gaseous elemental iodine and the activity concentration of the gaseous, organically bound iodine can be determined.

3.2 Sample preparation

The adsorbent is filled into an appropriate vessel (calibrated geometry for gamma spectrometry) and evenly mixed. For this purpose, e. g. 250 ml plastic bottles or Marinelli cups can be used. If the adsorbent is filled into the vessel in situ right after the end of the mixing, the vessel must be dimensionally stable up to 60°C as the special steel cartridge and the adsorbent are still hot.

3.3 Measurement and analyzes

3.3.1 Gamma-ray spectrometry

The adsorbent substance is measured by gamma spectrometry. In order to obtain limits of detection in the order of magnitude of approx. 1 $mBq \cdot m^{-3}$, high-purity germanium detectors with a relative efficiency of at least 40 %, related to a 3" x 3" NaI(TI) crystal, and a half width smaller than 2.0 keV, related to the 1332 keV gamma line of the radionuclide ^{60}Co, are to be used. For the measurement of very low activities ("low level area"), it is recommended using a shield for the detector which should be made of low-activity lead (^{210}Pb content < 50 Bq/kg) with a wall thickness of at least 10 cm. A multi-purpose gamma spectrometry measuring set-up is usually provided with a copper plate installed between the lead shield and the detector in order to suppress lead rays at 75 keV and 85 keV, and additionally with acrylic glass for the adsorption of the beta particles scattered on the copper plate. This additional shield for the measurement of higher activities is, however, disturbing when very low activities are measured because the Compton background is unnecessarily increased. For measuring tasks with lower sensitivity, a less complex apparatus is sufficient.

3.3.2 Calibration

Calibration for the gamma spectrometry is carried out with a special gel-type mixed standard of equal density and volume containing several radionuclides of known activity. Starting from the assumption of a homogeneously distributed activity in the sample, the activity standard for calibration is measured directly on the detector. Here, also the necessary coincidence summation corrections are to be taken into account.

It is assumed that comparability is given between the homogeneously distributed activity in a solution and the activity on the surfaces of a homogeneously distributed solid adsorber.

3.3.3 Computation of the analysis results

The computation of the activity concentration c_r, related to the middle of the collection period, is carried out according to equation (1):

$$c_r = \frac{N_n}{\varepsilon_r \cdot p_\gamma \cdot t_m \cdot V} \cdot f_0 \cdot f_1 \cdot f_2 \cdot f_3 \quad (1)$$

or

$$c_r = \frac{R_n}{\varepsilon_r \cdot p_\gamma \cdot V} \cdot f_0 \cdot f_1 \cdot f_2 \cdot f_3 \quad (1a)$$

where

$$R_n = \frac{N_n}{t_m} \quad (1b)$$

with

c_r : activity concentration of the radionuclide r in $Bq \cdot m^{-3}$

R_n : net count rate

N_n : net pulse number

ε_r : detection probability for radionuclide r in $Bq^{-1} \cdot s^{-1}$

p_γ : emission probability per nuclear transformation

t_m measuring time of sample in s

V : air volume having passed through filter in m^3

λ_r : decay constant of the radionuclide r

t_A : period of time between sampling (related to a mean collection time) and the start of measurement in s

f_0 : correction factor for differences in the self-attenuation of the iodine adsorber from the gel used for calibration

f_1 : coincidence summation correction factor

f_2 : correction factor for the decay of the radionuclides within the period starting in the middle of the collection period until the beginning of the measurement

f_3 : correction factor for the decay of the radionuclides during the measurement

For point and area sources positioned directly on the top of the endcap, the coincidence summation correction factor f_1 in the case of pulse losses usually ranges between 1.1 and 1.5.

The activity at a reference time earlier than the time of measurement by a time period t_A is obtained by multiplication by f_2 according to equation (2):

$$f_2 = e^{\lambda_r \cdot t_A} \quad (2)$$

For the correction factor f_3 the following applies according to equation (3):

$$f_3 = \frac{\lambda_r \cdot t_m}{1 - e^{-\lambda_r \cdot t_m}} \quad (3)$$

If the period of time between the collection time and the measuring time and/or the measuring time is much smaller than the half-life of the radionuclide to be measured, $f_2 = 1$ and/or $f_3 = 1$ applies.

For the standard measurement uncertainty $s(R_n)$ of the net counting rate, the following is valid:

$$s(R_n) = \sqrt{\frac{R_n}{t_m} + \frac{b \cdot \overline{R_0}(E_\gamma)}{t_m} \cdot \left(1 + \frac{b}{2L}\right)} \tag{4}$$

where

$s(R_n)$	:	standard measurement uncertainty assigned to the net counting rate in s^{-1}
Rn	:	net counting rate in s^{-1}
b	:	base width of the gamma line in number of channels
$R_0(E_\gamma)$:		mean counting rate of background per channel in s^{-1}
E_γ	:	energy of gamma line
L	:	number of channels for the determination of the background

For the standard measurement uncertainty assigned to the activity concentration $s(c_r)$, the following equation applies:

$$s(c_r) = s(R_n) \cdot \frac{c_r}{R_n} = s(R_n) \cdot \frac{1}{\varepsilon_r \cdot p_\gamma \cdot V} \cdot f_0 \cdot f_1 \cdot f_2 \cdot f_3 \tag{5}$$

3.3.4 Arithmetic example

For the computation of the concentration of the overall gaseous I-131 activity, the following numerical values for a sample submitted to suction for 14 days are entered:

N_n	=	1200
R_n	=	$2.08 \cdot 10^{-2}$ s^{-1}
$\varepsilon_{I\text{-}131}$	=	0.02987 $Bq^{-1} \cdot s^{-1}$
p_γ(364 keV)	=	0.817
b	=	10
L	=	5
E_γ	=	364.5 keV
$\overline{R}_0(E_\gamma)$	=	0.022 s-1
t_m	=	57600 s
V	=	840 m^3 (2.5 $m^3 \cdot h^{-1}$)
t_A	=	604808 s
$\lambda_{I\text{-}131}$	=	$1.00 \cdot 10^{-6}$ s^{-1}
f_0 (364 keV)	=	0.87
f_1	=	1

with

$$f_2 = e^{1.00 \cdot 10^{-6} \cdot 604808} = 1.83$$

and

$$f_3 = \frac{1.00 \cdot 10^{-6} \cdot 57600}{1 - e^{-1.00 \cdot 10^{-6} \cdot 57600}} = 1.03$$

The following is obtained for the activity concentration of I-131:

$$c_{I-131} = \frac{1200}{0.02987 \cdot 0.816 \cdot 57600 \cdot 840} \cdot 0.87 \cdot 1 \cdot 1.83 \cdot 1.03 \ Bq \cdot m^{-3} = 1.67 \cdot 10^{-3} Bq \cdot m^{-3}$$

The standard measurement uncertainty assigned to the net counting rate is:

$$s(R_n) = \sqrt{\frac{2.08 \cdot 10^{-2}}{57600} + \frac{10 \cdot 0.022}{57600} \cdot 2} \ s^{-1} = 2.83 \cdot 10^{-3} \ s^{-1}$$

This way, for the standard measurement uncertainty assigned to the activity concentration of I-131, the following is obtained:

$$s(c_{I-131}) = 2.83 \cdot 10^{-3} \cdot \frac{1.67 \cdot 10^{-3}}{2.08 \cdot 10^{-2}} Bq \cdot m^{-3} = 2.27 \cdot 10^{-4} \ Bq \cdot m^{-3}$$

The following results for the mean activity concentration of I-131:

$c_{I-131} = (1{,}67 \pm 0{,}23)\ 10^{-3}$ Bq·m^{-3}

3.4 Standard measurement uncertainty and limit of detection

3.4.1 Combined standard measurement uncertainty of the procedure

The combined standard measurement uncertainty mainly consists of the measurement uncertainties arising from sampling (filter efficiency, adsorption losses due to different air humidities, determination of the airflow rate), the preparation of samples (intermixing of the activated carbon), the detector calibration and the statistical contribution to the combined uncertainty. Without taking into account the statistical contribution, it amounts to approx. 10 %. The magnitude of the statistical measurement uncertainty depends on the magnitude of the activity and the background. Due to the confidence level required, the statistical measurement uncertainty exceeds all other uncertainties in the vicinity of the limit of detection.

3.4.2 Limit of detection

For the limit of detection G_N of the activity concentration, the following applies to activity measurements by gamma spectrometry (a precondition is that the background counting rates are not too small and $t_0 = t_m$):

$$G_N = \frac{c_r}{R_n} \cdot \left(k_{1-\alpha} + k_{1-\beta}\right) \cdot \sqrt{\frac{2 \cdot b \cdot \overline{R_0}\left(E\gamma\right)}{t_m}} \qquad (6)$$

where

G_N : limit of detection in Bq·m^{-3}
$k_{1-\alpha}$: quantile of normal distribution
$k_{1-\beta}$: quantile of normal distribution

With $k_{1-\alpha} = 3$ and $k_{1-\beta} = 1.645$ as well as with the numerical values given in section 3.3.4, the limit of detection of I-131 is obtained:

$$G_N = \frac{1.67 \cdot 10^{-3}}{2.08 \cdot 10^{-2}} \cdot \left(3 + 1.645\right) \cdot \sqrt{\frac{2 \cdot 10 \cdot 0.022}{57600}} \ Bq \cdot m^{-3} = 1.03 \cdot 10^{-3} \ Bq \cdot m^{-3}$$

This way, a value of $1.03 \cdot 10^{-3}$ Bq·m^{-3} results for the limit of detection after 16 hours measuring time. The required limit of detection of $5 \cdot 10^{-3}$ Bq·m^{-3} is thus obtained according to the specifications of REI.

4 Evaluation of the method

Apart from determining gaseous I-131, the procedure allows at the same time the detection of all gaseous gamma-emitting radioactive nuclides of iodine. The reliability of the procedure is to be ensured by quality assurance measures which have to be carried out at regular intervals. This also applies to the regular check of background and calibration.

4.1 Time needed

The time needed for a measurement of I-131 according to the Measuring Program for the Environmental Monitoring of a Nuclear Power Plant in normal operation can be broken down as follows:

Preparation of samples: 15 min
Collection period: 14 d
Measuring time: > 1 h

In the routine measuring program according to AVV-IMIS, the following results by analogy:
Preparation of samples: 15 min
Collection period: 7 d
Measuring time: > 4 h

4.2 Equipment (taking DWD as an example)

- Iodine collection system:
  Pump with pre-heating of the sucked-in air with a volume-flow rate of 1 to 3 m3·h-1, iodine collector consisting of vacuum pump, flowmeter, pre-heating of the air flow, cylindrical stainless steel cartridge with V = 170 ml for taking up the iodine adsorber, with support grid for a pre-filter for suspended particles.

- Consumables:
  Activated carbon
  KI- impregnated activated carbon, granulate with a diameter of d = 3.5 mm and a length of l = 4 to 10 mm
  Filling volume: 167 ml, bulk density in absorber vessel: 0.51 g/cm³
  Separation efficiency for I_2 and CH_3I:
  Ø 99.9 % at a temperature of 45 degrees and a relative humidity of 77 %
  Ø 90 % at a temperature of 50 degrees and a relative humidity up to 100 %

  filter for suspended particles

  Glassfibre filter free of bonding agent, 47 mm, separation efficiency for I_2 < 1 %, CH_3I < 1 % at room temperature

  Liquid nitrogen to cool the germanium detector

- Calibration source

- Gamma-spectrometer set-up

5 Documentation

5.1 Reporting

As specified in REI or AVV-IMIS:
Beginning of sampling, end of sampling, place of sampling, measured values for the radionuclide, measuring uncertainty, limit of detection achieved.

5.2 Other data to be recorded

Data concerning the measuring time and the volume-flow rate

5.3 Storage times

The measured adsorbent substances are stored about 2 months in closed vials.
According to REI, the recorded measurement results must be stored for 30 years.

6 Literature

[1] Federal Ministry for the Environment, Nature Conservation and Reactor Safety: Directive on the monitoring of emissions and immissions of nuclear plants (REI), GMBl Nr. 29, 19.08.1993, page 502 ff and GMBl Nr. 9/10, 20.03.1996, page 195 ff and revision from 07.12.2005, GMBl Nr. 14-17, 23.03.2006, page 254, (in German)

[2] Federal Ministry for the Environment, Nature Conservation and Reactor Safety: General administrative regulation on the Integrated Measuring and Information System according to the Law on Preventive Radiation Protection, Bundesanzeiger 2006, Volume 47, Number 244a, Dec 13, 2006 (in German)

[3] Federal Ministry for the Environment, Nature Conservation and Reactor Safety: Measuring instructions for the monitoring of radioactivity in the environment and for the determination of radioactive emissions from nuclear plants, ISSN 1865-8725, online available (in German), see URL: http://www.bmu.de/strahlenschutz/ueberwachung_der_umweltradioaktivitaet/messanleitungen/doc/42042.php

[4] Sampling for the monitoring of radioactivity in the air – Volume 3: Sampling procedures, DIN 25423-3 (in German)

Please refer to the note in the loose leaf 1.3 "Notes to the loose-leaf collection".