Improvement of Incorporation Monitoring during Decommissioning of the Hot-Cell Facility at Karlstein

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Abstract

During the decommissioning of the hot-cell facility at Karlstein, which was contaminated with irradiated nuclear fuel, Co-60 and Am-241, it was necessary to closely monitor incorporation of the significant radionuclide Am-241.

Initially, only limited external capacity was available for the analysis of faecal samples, with results taking about 4 weeks to be returned. The setting up of an in-house excretion analysis section at Siemens' radiochemistry laboratory in Erlangen enabled sample analysis turnaround times to be reduced to 1 to 2 weeks. This made it possible to respond faster to new cases of incorporation and also to improve identification and elimination of incorporation sources, leading in turn to an overall reduction in doses received through incorporation.

A total of up to 125 persons per year were monitored, with over 6300 samples being analyzed to date, at the in-house laboratory. Only a small proportion of the monitored individuals exceeded 3% of the specified incorporation limit. Maximum intake over the last 3 years was well below 10% of the limit.

A new ICP-MS system now installed in the laboratory will cut sample analysis times down to just 2 to 5 days in the future.

Nuclides to be monitored and determination of the significant nuclide for incorporation monitoring

During operation of the hot-cell facility at Karlstein, Germany, in addition to investigations concerning fuel assemblies/fuel rods and individual pellets containing irradiated nuclear fuel, Co-60 sources for medical radiation therapy were also produced and americium nitrate was converted to americium oxide.

The nuclide vector comprised approximately 5 - 12% Am-241. Plutonium isotopes, excluding Pu-241, accounted for a significantly lower proportion (approximately 0.3 - 2 %). The Pu-241 fraction stood at approximately 16 - 40%. Cs-137 comprised approximately 10 - 30 % of the nuclide vector, and Co-60 approximately 0.3 - 3 %.

In accordance with the German Radiological Protection Ordinance, the following annual limit values apply for incorporation by inhalation in the case of Category A occupationally exposed workers (for oxides where applicable):

Co-60	4 E 05 Bq	Pu-238, Pu-239, Pu-240	4 E 02 Bq each
Cs-137	6 E 06 Bq	Pu-241	7 E 03 Bq
Am-241	1 E 02 Bq		

The limits for incorporation by ingestion are several orders of magnitude higher in the case of alpha-emitters in oxide form. The type of incorporation is therefore always conservatively assumed to be inhalation. As the particle size distribution of the inhaled aerosols is not known, an AMAD of 1µm was assumed in accordance with the specifications of the calculational basis.

Determination of the significant nuclide:

Nuclide	Fraction of nuclide vector	Annual Limit of Intake (ALI) for persons of Category A	Relative contribution to utilization of limits (fraction of nuclide vector/ ALI forCategory A)
Co-60	0.3 - 3 %	4 E 05	7.5 E-07 - 7.5 E-06
Sr-90	4 – 32 %	5 E 04	8.0 E-05 - 6.4 E-04
Cs-137	10 - 30 %	6 E 06	1.7 E-06 - 5.0 E-06
Am-241	5 – 12 %	1 E 02	5.0 E-02 - 1.2 E-01
Pu-238, 239, 240	0.3 - 2 %	4 E 02	7.5 E-04 - 5.0 E-03
Pu-241	16 - 40 %	7 E 03	2.3 E-03 - 5.7 E-03

Comparison of the nuclide fractions in the nuclide vector and the limit values for the nuclides shows that Am-241 makes the greatest contribution to the utilization of the incorporation limit, making it the significant or critical nuclide. The plutonium isotopes were identified as the nuclides which made the second-largest contribution to utilization of the limit.

Determination of a suitable process for incorporation monitoring:

The following processes are available for incorporation monitoring of Am-241 and plutonium isotopes:

- Body-counter analysis (lung measurements)
- Airborne activity analysis
- Excretion analysis (faeces or urine samples analysis)

The suitability of these processes is examined below:

· Monitoring via lung measurements

The minimum detectable Am-241 activity levels in the chest region (lung measurement) with the available measuring equipment are as follows, given acceptable measuring times of approximately 50 minutes:

- 60 kg bodyweight 5 Bq Am241
- 100 kg bodyweight 12 Bq Am241.

This method therefore only enables detection of single intakes of Am-241 at over 20 % - 50 % of the incorporation limit, if the measurement is carried out immediately after incorporation (i.e. within one day). Lung measurements are thus only suitable for detecting significantly increased intake after an incident, or for identifying increased long-term incorporation.

Lung measurements only enable detection of incorporation of plutonium at intake levels in the vicinity of the annual limit. Lung measurements are therefore not suitable for routine monitoring of plutonium incorporation.

· Monitoring via airborne activity analysis

Determination of intake via airborne activity analysis necessitates determination of the representative activity concentration in the breathing air in the workplace area. During the actual decommissioning work, workers wear protective clothing with respirators (full-face masks equipped with P3 filters, or air supply via self-contained breathing apparatus or air-line devices) to protect against incorporation and contamination. Intake therefore only occurs in the event of incidents in which respirators are no longer properly attached, or on removal of protective clothing in exit - airlocks. Steady-state air flow conditions cannot be assumed in such occurrences. Airborne activity analysis is therefore not suitable for incorporation monitoring.
During decommissioning work, continuous airborne activity analysis is always carried out in the rooms concerned, although this was not used for incorporation monitoring. Instead, it was used to detect activity releases to enable classification of the rooms into activity zones and assessment of the effectiveness of protective measures.

· Monitoring via excretion analysis

Two techniques (urine samples analysis and faeces samples analysis) are available with the excretion analysis method.

In the case of Am-241, the faeces excretion rate is still greater by a factor of four than the excretion rate in urine even twelve months after incorporation. This factor increases significantly with shorter intervals, rising to a factor of 50, for example, 30 days after intake. With even shorter intervals, the difference between faeces excretion rate and urine excretion rate is even greater. For this reason, faeces samples were considered the most suitable option for enabling detection of relatively low intake levels (down to 10 mBq) of Am-241 (incorporations of approximately 20 mBq can still be detected given a detection limit of 1 mBq and sample delivery within four days after incorporation).

Determining monitoring frequency

The type of incorporation monitoring and the frequency of analysis must be agreed with our competent licensing and supervisory authorities and the institute for radition protection of our workman`s compensation institute (Berufsgenossenschaft). The objective was to enable detection of individual intakes of 3% of the annual limit for activity incorporation. An additional objective was to verify that intakes do not exceed 10% of the incorporation limit within a single year, given normal activities (i.e. excluding incidents).

The detection limits for faeces samples were initially 1.5 mBq for Am-241, Pu-238 and Pu-239/240.

According to the specifications of the Guideline, intake at the mid-point of the monitoring interval should be assumed. Given the conservative assumption of a 1:1:1 ratio for the radionuclides Am-241, Pu-238 and Pu-239/240, 3% of the annual activity incorporation limit could be reliably detected for single individual intakes using faeces samples every 3 weeks (21 days).

The timings of sample delivery by the cohort of workers to be monitored were uniformly distributed over the three-week period. No samples were provided on Monday and Tuesday (following work-free weekends) and after extended work-free periods. This rolling process enabled rapid identification of incorporation sources and initiation of countermeasures without delay.

As a result of improved analysis of the excretion functions and the reduction of the detection limit to 1 mBq for the nuclides Am-241, Pu-238 and Pu-239/240, the monitoring intervals were extended to four weeks (28 days) after mid-1997. The distribution of sample delivery was maintained.

As a result of the large number of faeces samples analyzed, and the progress of decommissioning work, as well as the individual nuclide vector fractions determined, it was established in early 1998 that a ratio of only 1:1 should be applied to express the ratio of Am-241 to the plutonium isotopes Pu-238, 239 and 240. Optimization analyses (see Appendix 1) showed that, given a monitoring interval of 70 days, the sum of the possibly undetected intakes lies below 10% of the annual limit. With longer and shorter periods, the minimum detectable activity intake increases. Only relatively short monitoring intervals of seven days and less lead to lower detectable annual intake values. Such short monitoring intervals were not feasible for regular monitoring for reasons of acceptability to the workers concerned, and on cost grounds. The optimized monitoring intervals were adopted following approval by the supervisory authority and the institute for radition protection of our workman`s compensation institute (Berufsgenossenschaft).

As the number of faeces samples in which plutonium isotopes were detected, subsequently became relatively low, and because in recent years Pu has only ever been detected in the presence of Am-241, regular analysis of faeces samples for plutonium isotopes will cease in the future. They will only be carried out if Am-241 is detected in the sample. The corresponding parts of sample processing are preserved in our analysis station until such time as this occurs. This leads to a reduction in sample analysis effort.

The following applied to all monitoring intervals:

- If a sample analysis result is found to be above the detection limit, samples are taken at intervals of one week until the results fall below the detection limit.

- In the event of sample analysis results over 50 mBq Am-241, a lung analysis is carried out immediately at our body (lung) - counter - station at Hanau, Germany.

- Samples are taken prior to work interruptions which are longer than the monitoring interval.

- Initial samples are taken from external workers who have been exposed to alpha-emitters.

- Following identified incidents, a sample should be taken immediately on the following day. The health physics officer makes the decision on whether further samples are necessary.

Results of the monitoring program in recent years

The monitoring program has been implemented since 1994. Due to the comparability of the data, only the results for whole years are compiled below.

Year	1994	1995	1996	1997	1998
Number of persons monitored	163	115	125	101	68
Number of faeces samples	1780	1344	1314	1011	498
Number of faeces samples > detection limit	295	173	112	186	84
Number of persons > detection limit	90	56	52	56	39
Number of persons > 3 % of annual limit	16	15	5	3	9
Maximum annual intake [% of annual limit]	8.8	19.6	4.7	6.7	7.8
Maximum effective dose [mSv]	2.4	2.4	1.2	0.8	0.9

In the event of incorporations exceeding 3% of the annual limit, the monitoring data are notified to our regulatory authority, which checks our analyses, officially fixes the dose values and reports these to the German radiological protection record office (*Strahlenschutzregister*). Once we have received notification, we enter the data into the dose records (dose passport) of the persons concerned.

The figures show the influence of the extended monitoring interval (four weeks) on the number of faeces samples from mid-1997 onward.

The effectiveness of the radiation protection measures implemented is highlighted by comparing the figures for the level and number of incorporations over 3% in 1994 and 1995 with the following years. The protective measures have remained practically unchanged since then.

The increase in the number of persons over 3% of the annual limit and the corresponding maximum values for incorporation and dose in 1998 is due to the fact that, given longer monitoring intervals, higher incorporation and dose values result from equal excretion activity levels due to the longer time period which has to be taken into account.

Improvement of analysis technique for excretion samples

Over 6,300 faeces and urine samples have been analyzed to date in the accredited excretion analysis laboratory, part of Siemens KWU Group's radiochemical laboratory at Erlangen, Germany.

- Current analysis technique:

After incoming inspection, the samples are mixed with a suitable tracer (Pu-236 and Am-243) for alpha-spectrometry analysis. Faeces samples are then subjected to complete thermal mineralization at 500° in a furnace with the aid of nitric acid and hydrogen peroxide. Urine samples are first evaporated in the sand bath, and then subjected to further treatment in the furnace using nitric acid. The faeces and urine samples mineralized in this way are then further treated in the same fashion. The incineration residue is dissolved into nitric acid, and the relevant radionuclides (Pu and Am) are selectively separated by means of liquid-liquid extraction. The radionuclides isolated in this way are then electrochemically precipitated on a small stainless steel plate, on which the alpha-spectrometry analysis is subsequently carried out using a boundary layer detector. After a measuring period of 1-3 days, computer-aided analysis is carried out, which allows determining the activity of the relevant plutonium and americium isotopes (Pu-238, Pu-239/240 and Am-241) with the aid of the previously added tracer.

This kind of analysis of faeces or urine samples lasts approximately 5 - 6 days. Appendix 2 shows a diagrammatic representation of the analysis sequence.

- Future analysis technique (from end 1999):

After incoming inspection, the excretion samples are mixed, as before, with an appropriate tracer and completely mineralized, and the relevant radionuclides are selectively separated from the salt-rich solubilization solution by means of liquid-liquid extraction.

The Am-241 activity excreted is then determined by means of ICP-MS (Inductively Coupled Plasma - Mass Spectrometry). In this analytical technique, the solution containing americium is converted into an aerosol using atomizers, and completely ionized at 8000 K in the ICP part of the system. The ions generated in this way are then separated and detected in the MS part of the system on the basis of their mass-to-charge ratio. Am-241 analysis is also carried out via the previously added Am-243 tracer. ICP measurement and analysis takes approximately 20 minutes for each sample. If several samples are to be analyzed simultaneously, the degree of automation can be increased by the use of a sample changer. Plutonium is subjected to further alpha-spectrometry analysis if necessary, as the ICP-MS analysis system is unable to distinguish between the isobars of Pu-238 and the omnipresent U-238.

ICP-MS analysis of a single faeces or urine sample for Am-241 takes a total of approximately 2-3 days. If plutonium analysis is additionally carried out, the analysis time increases to 3-4 days. Appendix 3 shows a schematic representation of this analysis sequence.

References

1) Radiation Protection Ordinance, Bundesgesetzblatt Part I, No. 34, Bonn, Fed. Rep. Germany, 1321 - 1375 (1989)

2) Guidline for Physical Radiation Protection Monitoring, Gemeinsames Ministerialblatt (from 13. March 1997), Vol. 45, No. 7, 285 - 308, Bonn, Fed. Rep. Germany (1994)

3) Guidline for the Evaluation of Body Doses from Internal Radiation Exposure, Bundesanzeiger, Vol. 49, No. 122a, Bonn, Fed. Rep. Germany (1997)

APPENDIX 1. Determination of Optimum Monitoring Period

The minimum detectable annual activity intake Z (t), given regular monitoring, is calculated as a function of the length of the monitoring interval (t), the detection limit (DL) for the nuclide in the sample and the excretion function S (t) of the radionuclide concerned, **assuming that intake occurred at the start of the monitoring interval,** as follows:

$$Z(t) = \frac{DL}{S(t)} \times \frac{365}{t};$$

The detection limit in faeces samples for Am-241 is currently 1 mBq.

The excretion function S (t) is shown in the table below, which contains the data used in the German guideline for the evaluation of body doses from internal radiation exposure.

To determine the optimum length of the monitoring interval, the minimum values of Z (t) have to be determined as a function of the length of the monitoring interval (t).

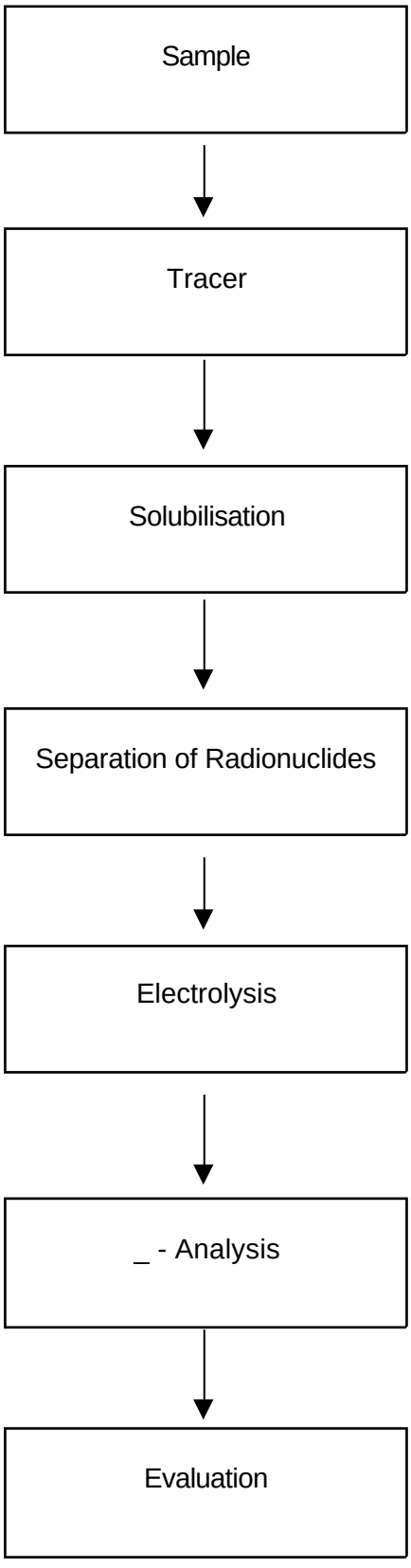
Some characteristic values for Z (t) are shown below.

Length of monitoring interval [d]	1	2	3	4	7	14	20	30	40	60	70	80	100
S (t) [mBq/day per Bq intake]	44	140	110	66	8.4	1.7	1.2	1.1	0.94	0.71	0.62	0.54	0.41
Z (t) [Bq]	8.3	1.3	**1.1**	1.4	6.2	15.3	15.2	11.1	9.7	8.6	**8.4**	8.5	8.9

It can be seen from the table that there are two minimum values, at 3 days and 70 days. The first minimum at 3 days cannot be utilized for long-term monitoring due to the unreasonably short monitoring period. The second minimum at 70 days enables verification of annual activity intake values below 10% of the annual limit of 10 Bq for Am-241. The value of 70 days was therefore selected as the optimum length of the monitoring period.

In the event of an actual intake, it is first assumed in accordance with the so called reference process of the guidline for the evaluation of body doses from internal radiation exposure, that the intake occurred at the mid-point of the monitoring period. Given an excretion of 1 mBq and a monitoring period of 70 days, an intake of 1 Bq Am-241 results, given an excretion function value for 35 days of S (35) = 1.0. An actual minimum determined intake of 5 Bq Am-241 per year is thus obtained with 5 monitoring periods per year.

APPENDIX 2. Schematic Representation of Alpha-Spectrometry Analysis of Americuim and Plutonium in Excretion Samples



Faeces, urine, blood, other materials

Pu 236, Am 243

Thermal at 500 ^{0} C and with concentrated HNO_3/H_2O_2

Liquid-liquid extraction using TIOA, TOPO as extraction agents

Electrolyte solution: 0,5 m $(NH_4)_2SO_4$
Duration : 1,5 hours

PIPS detectors, analysis time 20 - 50 hours
Detection limit: 1,0 mBq

Computer-aided using standard software
Routine monitoring, particularly in the event of „findings„

APPENDIX 3. Schematic Representation of ICP-MS Analysis of Americuim in Excretion Samples