L7
Analysis Techniques

Follow-up Training Course on Environmental Radioactivity Monitoring 2025

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Integrated Support Center for Nuclear Nonproliferation Security
and Human Resource Development
JAPAN ATOMIC ENERGY AGENCY

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Job history:

2000-	Start of work in the Japan Nuclear Cycle Development Institute (JNC)
2005-	Inauguration of the Japan Atomic Energy Agency (JAEA)
2008-2009	Study in Technical University of Denmark (DTU) Risø Campus
2009-	Restart of work in the JAEA
2018-2022	Work in International Commission on Radiological Protection (ICRP) as an Assistant Scientific Secretary
2022-	Restart of work in the JAEA

Education:

2006 Ph. D (Doctor of Science), Niigata University

Majors:

- Environmental monitoring around the reprocessing plant
- Effluent monitoring of the reprocessing plant, etc.
- Bioassay of radiation worker
- Radiation control of plutonium and radioisotopes treating facility
- Dosimetry using natural and artificial materials
- Radiochemistry

Contents:

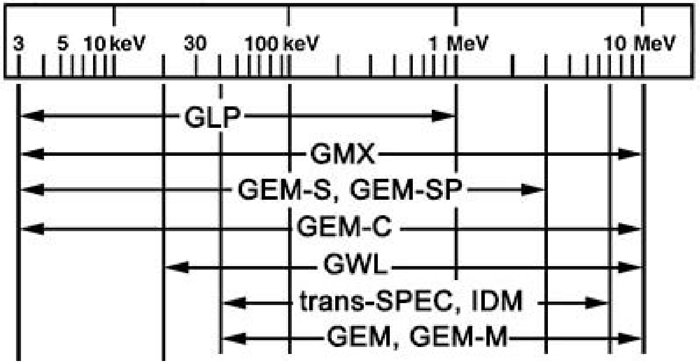
- \- Gamma spectrometry, Alpha/beta counting, Mass Spectrometry and Advanced Methods
- \- Radiochemical separation basics
- \- Quality assurance

Gamma spectrometry

Gamma rays exhibit minimal energy loss during transmission through matter, enabling accurate measurement of the energy spectrum originating from radioactive isotopes. Each radioactive isotope emits gamma rays with characteristic energies. Consequently, the obtained energy spectrum allows for relatively straightforward identification of the radioisotopes.

Gamma-ray spectrometers utilize two primary detector types: semiconductor detectors that generate electron-hole pairs upon radiation interaction, and scintillators. Scintillators emit faint light, which is then amplified using devices such as photomultiplier tubes (PMTs). Among detectors suitable for measuring radioactive isotopes in environmental samples, germanium semiconductor detectors offer the highest performance.

Gamma spectrometry



High Purity Germanium (HPGe) Radiation Detectors

https://www.ortec-online.com/products/radiation-detectors/high-purity-germanium-hpge-radiation-detectors

Detector: 5”x5” NaI(Tl) scintillator
Detection limit (Cs-134, Cs-137): 10 Bq/kg
(Sample 1 kg, Measurement time: 10 min.)

https://www.adfutec.com/pdf/Catalog_160108-1_AFTNDA2.pdf

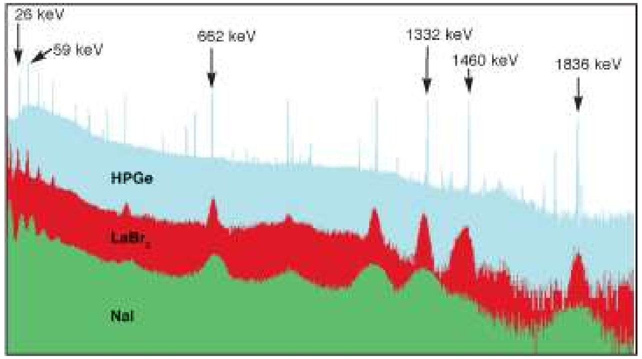
Gamma spectrometry

$CeBr_3$

Resolution: 4% (@662 keV)
Density: 5.2 g/cc
Decay time: 17 ns
Absorbency: Yes

https://www.sii.co.jp/hubfs/CeBr_CatalogV1.3-1.pdf

$LaBr_3$(Ce)



Resolution: 2.8-4.0% (@662 keV)
Density: 5.29 g/cc
Decay time: 26 ns

https://www.ortec-online.com/products/radiation-detectors/scintillation-radiation-detectors/scintillation-detector-types/lanthanum-bromide-detectors

Photoelectric Effect

When a γ-ray enters the sensitive region of a germanium semiconductor detector, it transfers its total energy ($E\gamma$) to the orbital electrons (electrons in K- or L-shell with binding energy I) of a germanium atom. Consequently, fast electrons (photoelectrons) with a kinetic energy of ($E\gamma - I$) are generated. This phenomenon is called the photoelectric effect.

When $E\gamma$ is larger than the binding energy (IK) of orbital electrons in K-shell, approximately 80 % of the total interaction corresponds to the interaction with orbital electrons in K-shell, whereas approximately 20 % corresponds to the interaction with other orbital electrons.

Because the probability of the photoelectric effect (including all orbital electrons) is proportional to the −3.2th power of the energy of a γ-ray, the photoelectric effect is an interaction predominant in low-energy γ-rays

Compton Scattering

There is a case wherein a part of the energy of an incident γ-ray is transferred to an electron (orbital or free electron), and the γ-ray is not extinguished but scattered with the remaining energy.
At that time, the γ-ray gives a part of its energy to an electron in the sensitive region of the detector, generating a fast electron (sometimes called the Compton electron), and the γ-ray changes to the scattered γ-ray with a lower energy ($E\gamma'$).
The energy ($Ee = E\gamma - E\gamma'$) of the generated fast electron is not constant because the energy differs at each interaction depending on the scattering angle of the γ-ray. Consequently, the energy exhibits a continuous distribution. For a scattering angle of 180°, the Compton electron has the maximum energy, corresponding to the high-energy edge of the continuous distribution called the Compton edge.
The probability of the Compton scattering in germanium is almost constant in the range of the γ-ray energy from dozens of keV to approximately 100 keV. At energies higher than this region, the probability becomes a gradual decreasing function.

Electron pair creation

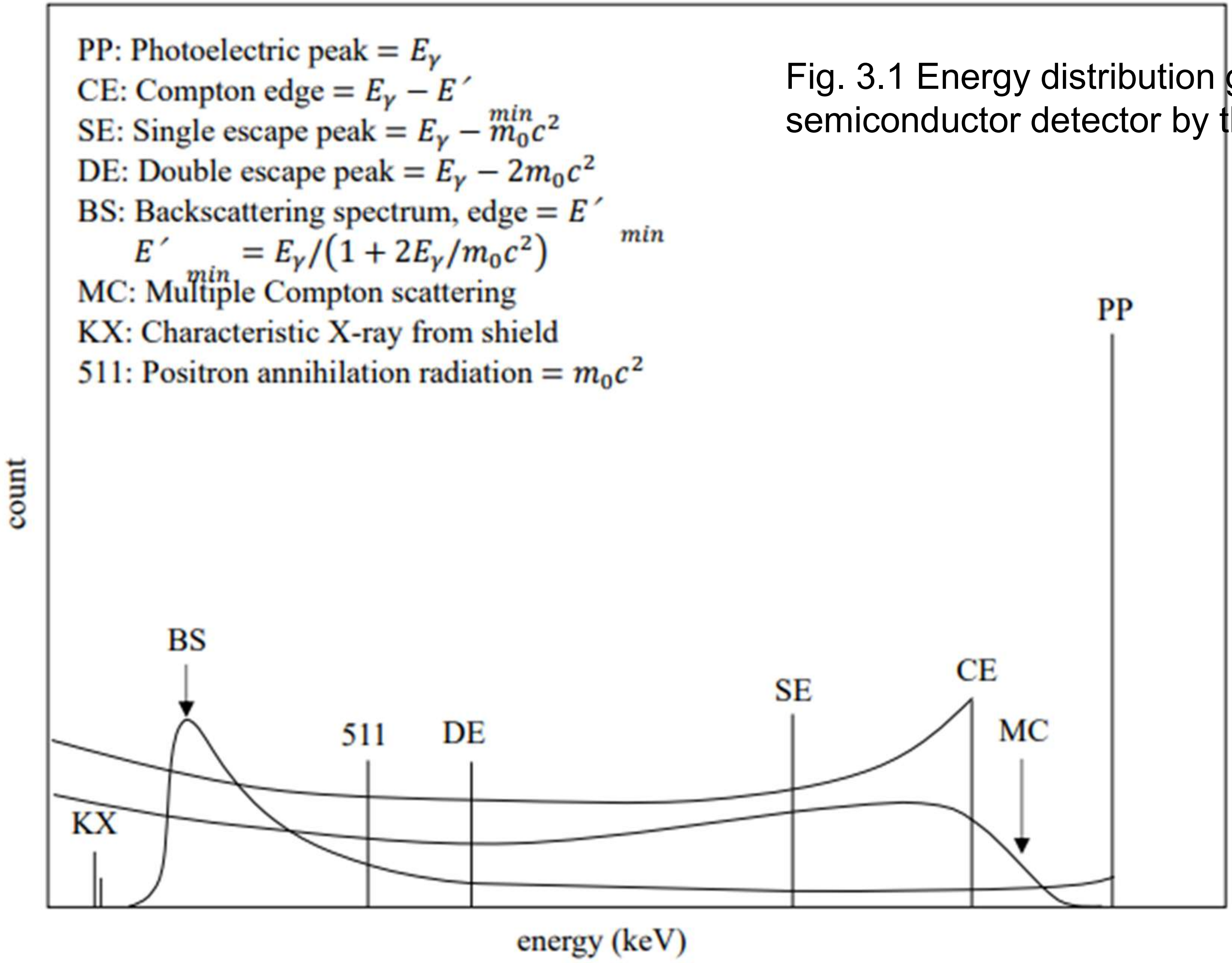
When a γ-ray with an energy of $2m_0c^2$ (corresponding to the static mass of the two electrons = 1.002 MeV) or higher, goes through the Coulomb field near a nucleus, the γ-ray is sometimes extinguished, producing an electron–positron pair. In this case, the nucleus and orbital electron energies do not change.

The sum of the kinetic energy of the generated electron and positron is equal to the value $(E\gamma - 2m_0c^2)$ that the energy corresponding to the static mass of the two electrons is subtracted from the γ-ray energy.

Because an electron pair production is an interaction that occurs for γ-rays with an energy of $2m_0c^2$ or higher, its probability increases with increasing γ-ray energy. However, its probability is only one-hundredth of the total interaction even at 1.5 MeV, implying that an electron pair production is an interaction predominant at further high energy.

Gamma spectrometry

Fig. 3.1 Energy distribution generated in a germanium semiconductor detector by the interaction with γ-rays



PP: Photoelectric peak = E_γ
CE: Compton edge = $E_\gamma - E'_{min}$
SE: Single escape peak = $E_\gamma - m_0c^2$
DE: Double escape peak = $E_\gamma - 2m_0c^2$
BS: Backscattering spectrum, edge = E'_{min}
$E'_{min} = E_\gamma / (1 + 2E_\gamma / m_0c^2)$
MC: Multiple Compton scattering
KX: Characteristic X-ray from shield
511: Positron annihilation radiation = m_0c^2

https://www.envraddb.go.jp/wp-content/uploads/2020/12/No7.pdf

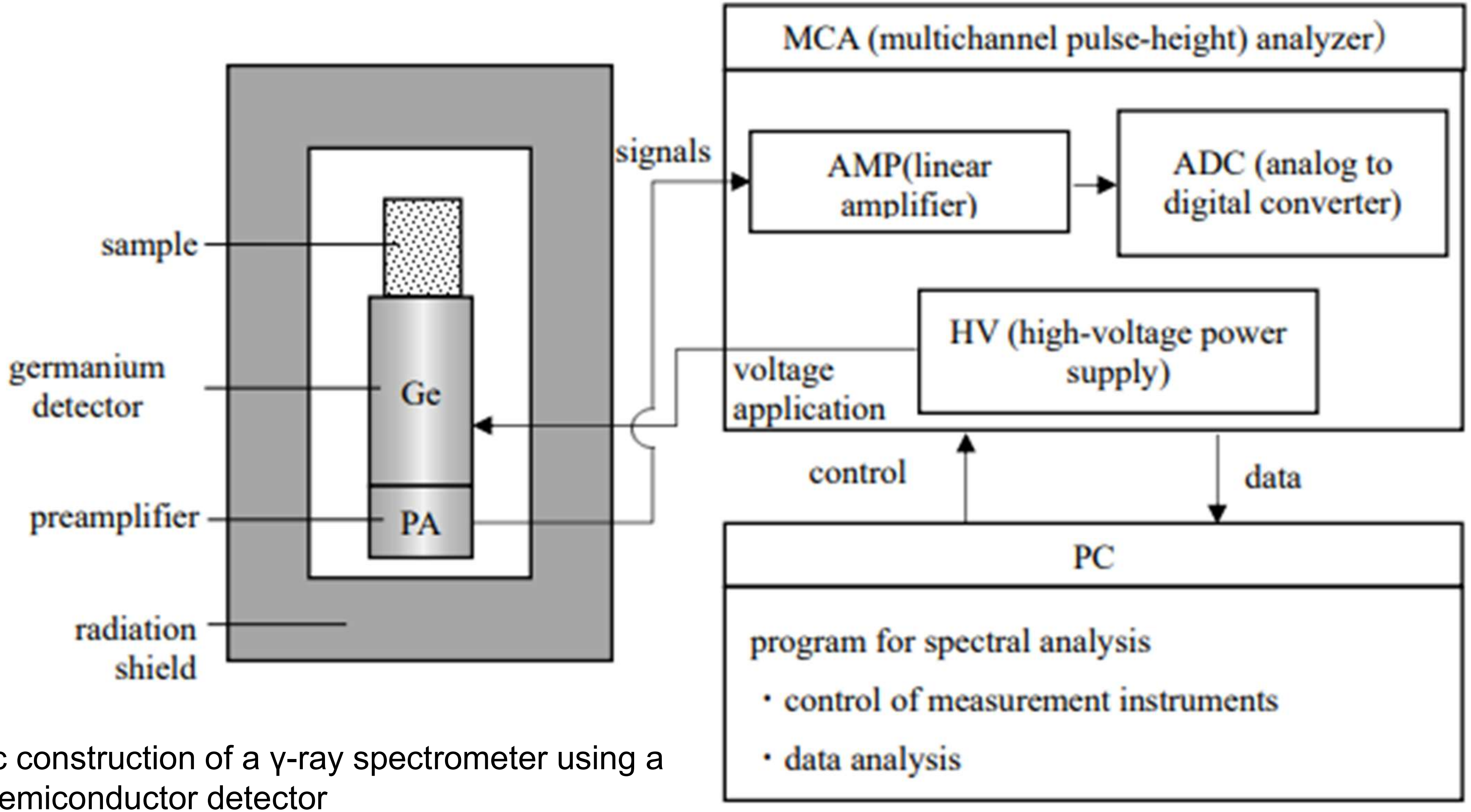


Fig. 4.1 Basic construction of a γ-ray spectrometer using a germanium semiconductor detector

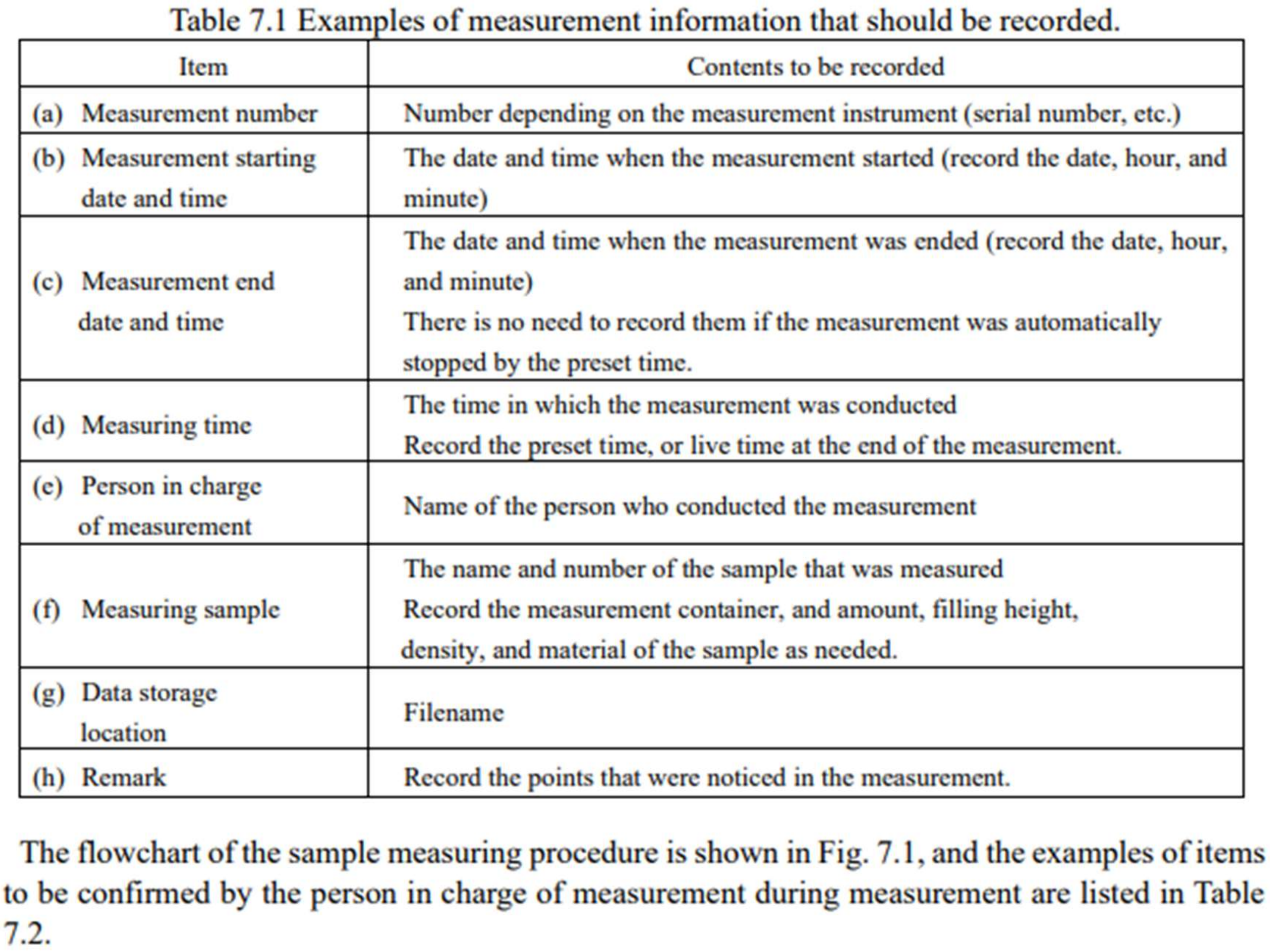


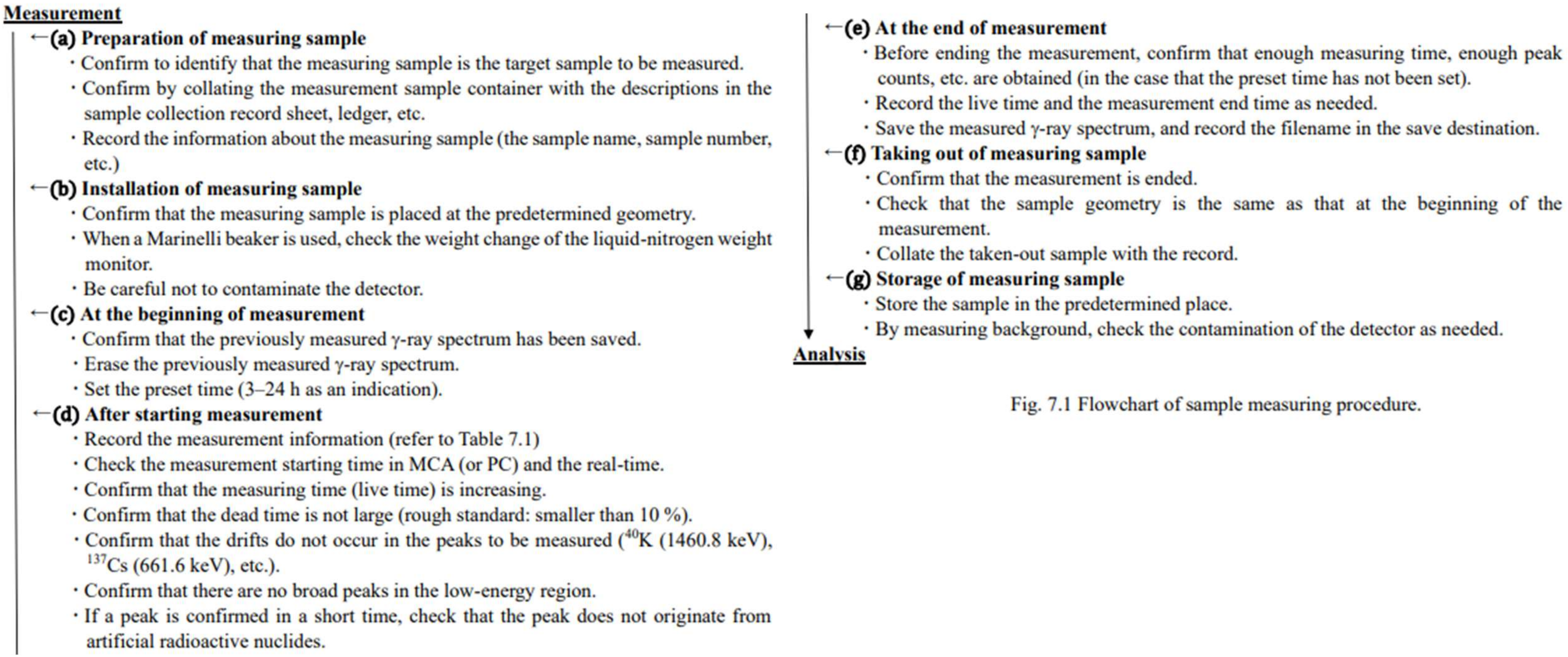
Table 7.1 Examples of measurement information that should be recorded.

Item	Contents to be recorded
(a) Measurement number	Number depending on the measurement instrument (serial number, etc.)
(b) Measurement starting date and time	The date and time when the measurement started (record the date, hour, and minute)
(c) Measurement end date and time	The date and time when the measurement was ended (record the date, hour, and minute) There is no need to record them if the measurement was automatically stopped by the preset time.
(d) Measuring time	The time in which the measurement was conducted Record the preset time, or live time at the end of the measurement.
(e) Person in charge of measurement	Name of the person who conducted the measurement
(f) Measuring sample	The name and number of the sample that was measured Record the measurement container, and amount, filling height, density, and material of the sample as needed.
(g) Data storage location	Filename
(h) Remark	Record the points that were noticed in the measurement.

The flowchart of the sample measuring procedure is shown in Fig. 7.1, and the examples of items to be confirmed by the person in charge of measurement during measurement are listed in Table 7.2.

https://www.envraddb.go.jp/wp-content/uploads/2020/12/No7.pdf

Gamma spectrometry



Measurement

←**(a) Preparation of measuring sample**

- Confirm to identify that the measuring sample is the target sample to be measured.
- Confirm by collating the measurement sample container with the descriptions in the sample collection record sheet, ledger, etc.
- Record the information about the measuring sample (the sample name, sample number, etc.)

←**(b) Installation of measuring sample**

- Confirm that the measuring sample is placed at the predetermined geometry.
- When a Marinelli beaker is used, check the weight change of the liquid-nitrogen weight monitor.
- Be careful not to contaminate the detector.

←**(c) At the beginning of measurement**

- Confirm that the previously measured γ-ray spectrum has been saved.
- Erase the previously measured γ-ray spectrum.
- Set the preset time (3–24 h as an indication).

←**(d) After starting measurement**

- Record the measurement information (refer to Table 7.1)
- Check the measurement starting time in MCA (or PC) and the real-time.
- Confirm that the measuring time (live time) is increasing.
- Confirm that the dead time is not large (rough standard: smaller than 10 %).
- Confirm that the drifts do not occur in the peaks to be measured (^{40}K (1460.8 keV), ^{137}Cs (661.6 keV), etc.).
- Confirm that there are no broad peaks in the low-energy region.
- If a peak is confirmed in a short time, check that the peak does not originate from artificial radioactive nuclides.

←**(e) At the end of measurement**

- Before ending the measurement, confirm that enough measuring time, enough peak counts, etc. are obtained (in the case that the preset time has not been set).
- Record the live time and the measurement end time as needed.
- Save the measured γ-ray spectrum, and record the filename in the save destination.

←**(f) Taking out of measuring sample**

- Confirm that the measurement is ended.
- Check that the sample geometry is the same as that at the beginning of the measurement.
- Collate the taken-out sample with the record.

←**(g) Storage of measuring sample**

- Store the sample in the predetermined place.
- By measuring background, check the contamination of the detector as needed.

Analysis

Fig. 7.1 Flowchart of sample measuring procedure.

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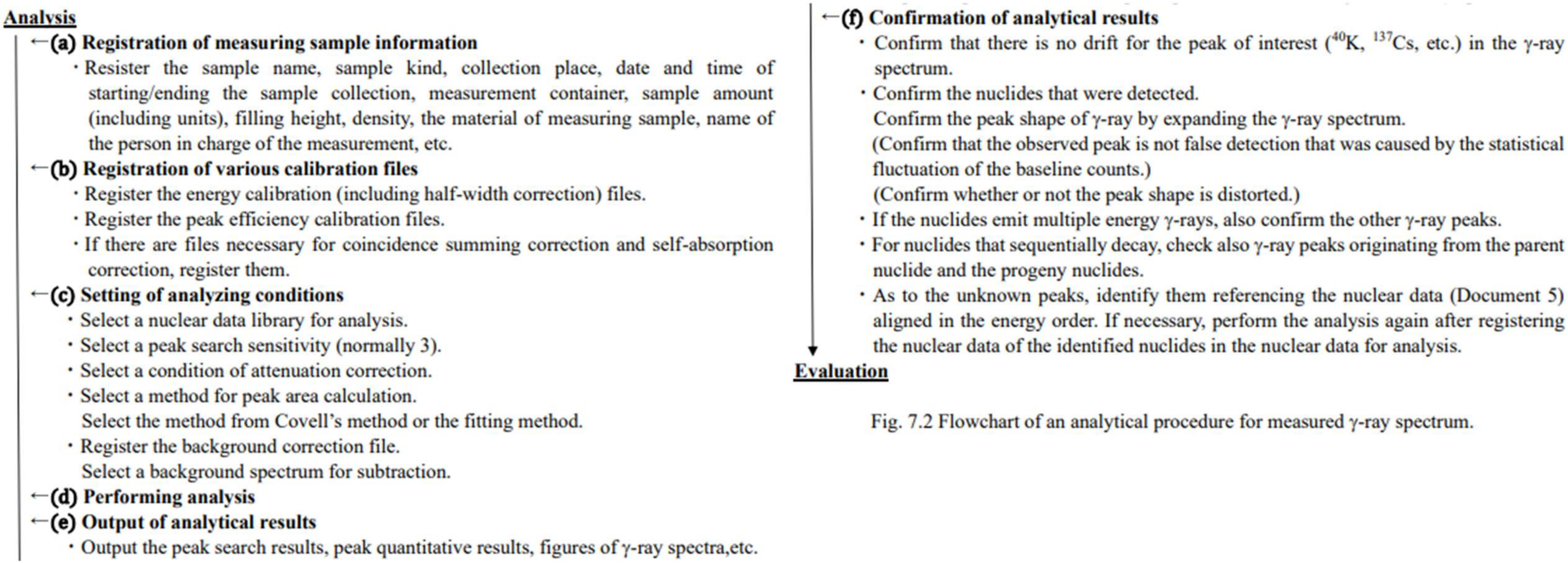
Gamma spectrometry



Analysis

←**(a) Registration of measuring sample information**

- Resister the sample name, sample kind, collection place, date and time of starting/ending the sample collection, measurement container, sample amount (including units), filling height, density, the material of measuring sample, name of the person in charge of the measurement, etc.

←**(b) Registration of various calibration files**

- Register the energy calibration (including half-width correction) files.
- Register the peak efficiency calibration files.
- If there are files necessary for coincidence summing correction and self-absorption correction, register them.

←**(c) Setting of analyzing conditions**

- Select a nuclear data library for analysis.
- Select a peak search sensitivity (normally 3).
- Select a condition of attenuation correction.
- Select a method for peak area calculation.
  Select the method from Covell's method or the fitting method.
- Register the background correction file.
  Select a background spectrum for subtraction.

←**(d) Performing analysis**

←**(e) Output of analytical results**

- Output the peak search results, peak quantitative results, figures of γ-ray spectra,etc.

←**(f) Confirmation of analytical results**

- Confirm that there is no drift for the peak of interest (^{40}K, ^{137}Cs, etc.) in the γ-ray spectrum.
- Confirm the nuclides that were detected.
  Confirm the peak shape of γ-ray by expanding the γ-ray spectrum.
  (Confirm that the observed peak is not false detection that was caused by the statistical fluctuation of the baseline counts.)
  (Confirm whether or not the peak shape is distorted.)
- If the nuclides emit multiple energy γ-rays, also confirm the other γ-ray peaks.
- For nuclides that sequentially decay, check also γ-ray peaks originating from the parent nuclide and the progeny nuclides.
- As to the unknown peaks, identify them referencing the nuclear data (Document 5) aligned in the energy order. If necessary, perform the analysis again after registering the nuclear data of the identified nuclides in the nuclear data for analysis.

Evaluation

Fig. 7.2 Flowchart of an analytical procedure for measured γ-ray spectrum.

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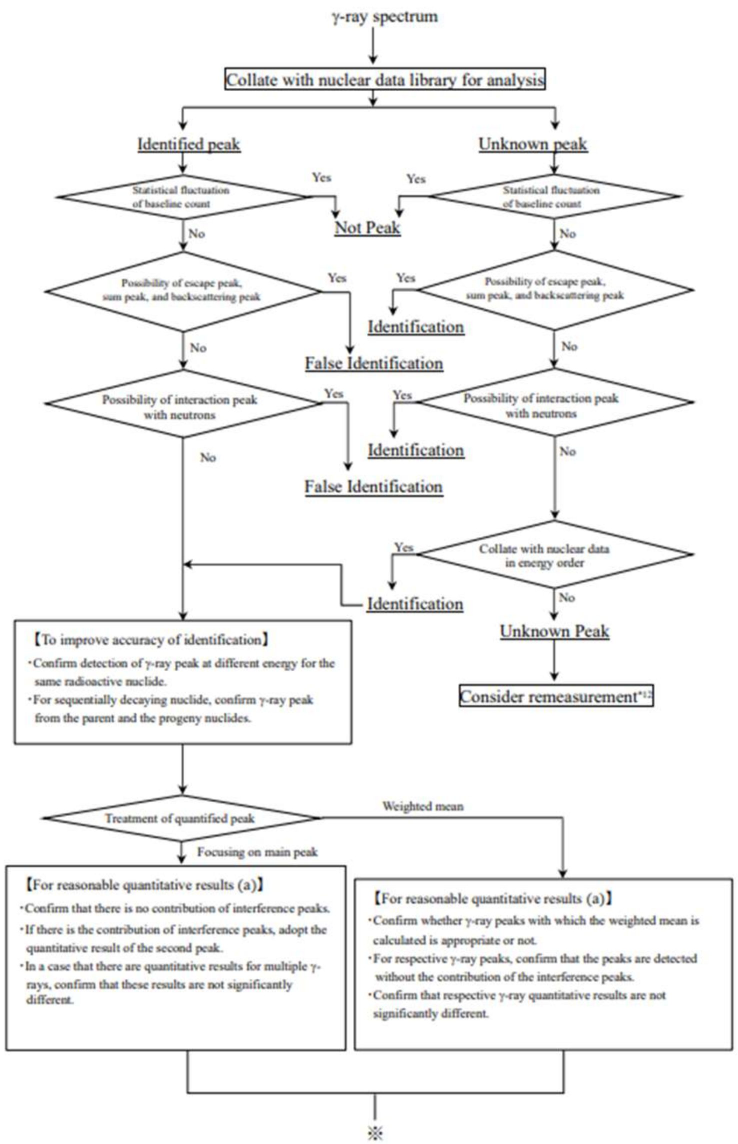
Gamma spectrometry



【To improve accuracy of identification】

・Confirm detection of γ-ray peak at different energy for the same radioactive nuclide.

・For sequentially decaying nuclide, confirm γ-ray peak from the parent and the progeny nuclides.

【For reasonable quantitative results (a)】

・Confirm that there is no contribution of interference peaks.

・If there is the contribution of interference peaks, adopt the quantitative result of the second peak.

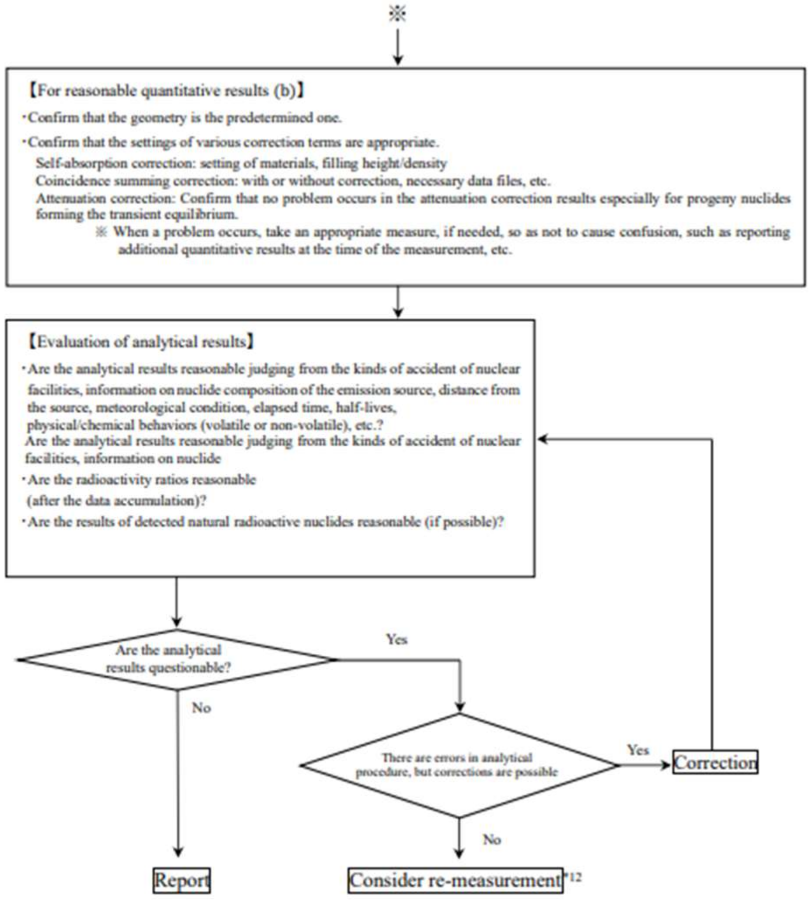
・In a case that there are quantitative results for multiple γ-rays, confirm that these results are not significantly different.

【For reasonable quantitative results (a)】

・Confirm whether γ-ray peaks with which the weighted mean is calculated is appropriate or not.

・For respective γ-ray peaks, confirm that the peaks are detected without the contribution of the interference peaks.

・Confirm that respective γ-ray quantitative results are not significantly different.



【For reasonable quantitative results (b)】

・Confirm that the geometry is the predetermined one.

・Confirm that the settings of various correction terms are appropriate.

Self-absorption correction: setting of materials, filling height/density
Coincidence summing correction: with or without correction, necessary data files, etc.
Attenuation correction: Confirm that no problem occurs in the attenuation correction results especially for progeny nuclides forming the transient equilibrium.

※ When a problem occurs, take an appropriate measure, if needed, so as not to cause confusion, such as reporting additional quantitative results at the time of the measurement, etc.

【Evaluation of analytical results】

・Are the analytical results reasonable judging from the kinds of accident of nuclear facilities, information on nuclide composition of the emission source, distance from the source, meteorological condition, elapsed time, half-lives, physical/chemical behaviors (volatile or non-volatile), etc.?
Are the analytical results reasonable judging from the kinds of accident of nuclear facilities, information on nuclide

・Are the radioactivity ratios reasonable (after the data accumulation)?

・Are the results of detected natural radioactive nuclides reasonable (if possible)?

Fig.7.3.1 Example of flowchart until reporting analysis results (continued).

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Gamma spectrometry

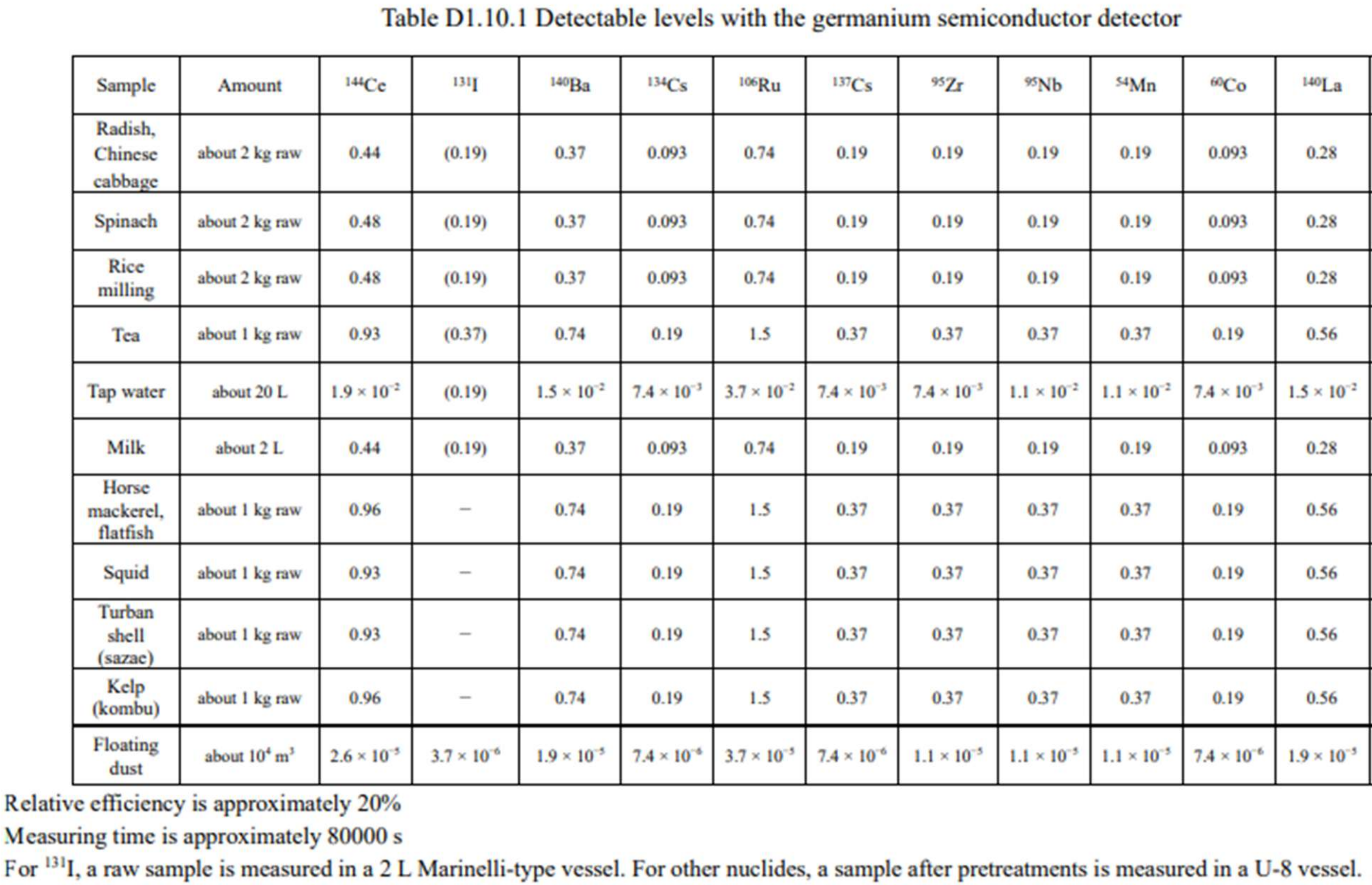

Table D1.10.1 Detectable levels with the germanium semiconductor detector

Sample	Amount	^{144}Ce	^{131}I	^{140}Ba	^{134}Cs	^{106}Ru	^{137}Cs	^{95}Zr	^{95}Nb	^{54}Mn	^{60}Co	^{140}La	Unit
Radish, Chinese cabbage	about 2 kg raw	0.44	(0.19)	0.37	0.093	0.74	0.19	0.19	0.19	0.19	0.093	0.28	Bq/kg raw
Spinach	about 2 kg raw	0.48	(0.19)	0.37	0.093	0.74	0.19	0.19	0.19	0.19	0.093	0.28	Bq/kg raw
Rice milling	about 2 kg raw	0.48	(0.19)	0.37	0.093	0.74	0.19	0.19	0.19	0.19	0.093	0.28	Bq/kg raw
Tea	about 1 kg raw	0.93	(0.37)	0.74	0.19	1.5	0.37	0.37	0.37	0.37	0.19	0.56	Bq/kg raw
Tap water	about 20 L	1.9×10^{-2}	(0.19)	1.5×10^{-2}	7.4×10^{-3}	3.7×10^{-2}	7.4×10^{-3}	7.4×10^{-3}	1.1×10^{-2}	1.1×10^{-2}	7.4×10^{-3}	1.5×10^{-2}	Bq/L
Milk	about 2 L	0.44	(0.19)	0.37	0.093	0.74	0.19	0.19	0.19	0.19	0.093	0.28	Bq/L
Horse mackerel, flatfish	about 1 kg raw	0.96	–	0.74	0.19	1.5	0.37	0.37	0.37	0.37	0.19	0.56	Bq/kg raw
Squid	about 1 kg raw	0.93	–	0.74	0.19	1.5	0.37	0.37	0.37	0.37	0.19	0.56	Bq/kg raw
Turban shell (sazae)	about 1 kg raw	0.93	–	0.74	0.19	1.5	0.37	0.37	0.37	0.37	0.19	0.56	Bq/kg raw
Kelp (kombu)	about 1 kg raw	0.96	–	0.74	0.19	1.5	0.37	0.37	0.37	0.37	0.19	0.56	Bq/kg raw
Floating dust	about 10^4 m^3	2.6×10^{-5}	3.7×10^{-6}	1.9×10^{-5}	7.4×10^{-6}	3.7×10^{-5}	7.4×10^{-6}	1.1×10^{-5}	1.1×10^{-5}	1.1×10^{-5}	7.4×10^{-6}	1.9×10^{-5}	Bq/m^3

Relative efficiency is approximately 20%

Measuring time is approximately 80000 s

For ^{131}I, a raw sample is measured in a 2 L Marinelli-type vessel. For other nuclides, a sample after pretreatments is measured in a U-8 vessel.

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Gamma spectrometry

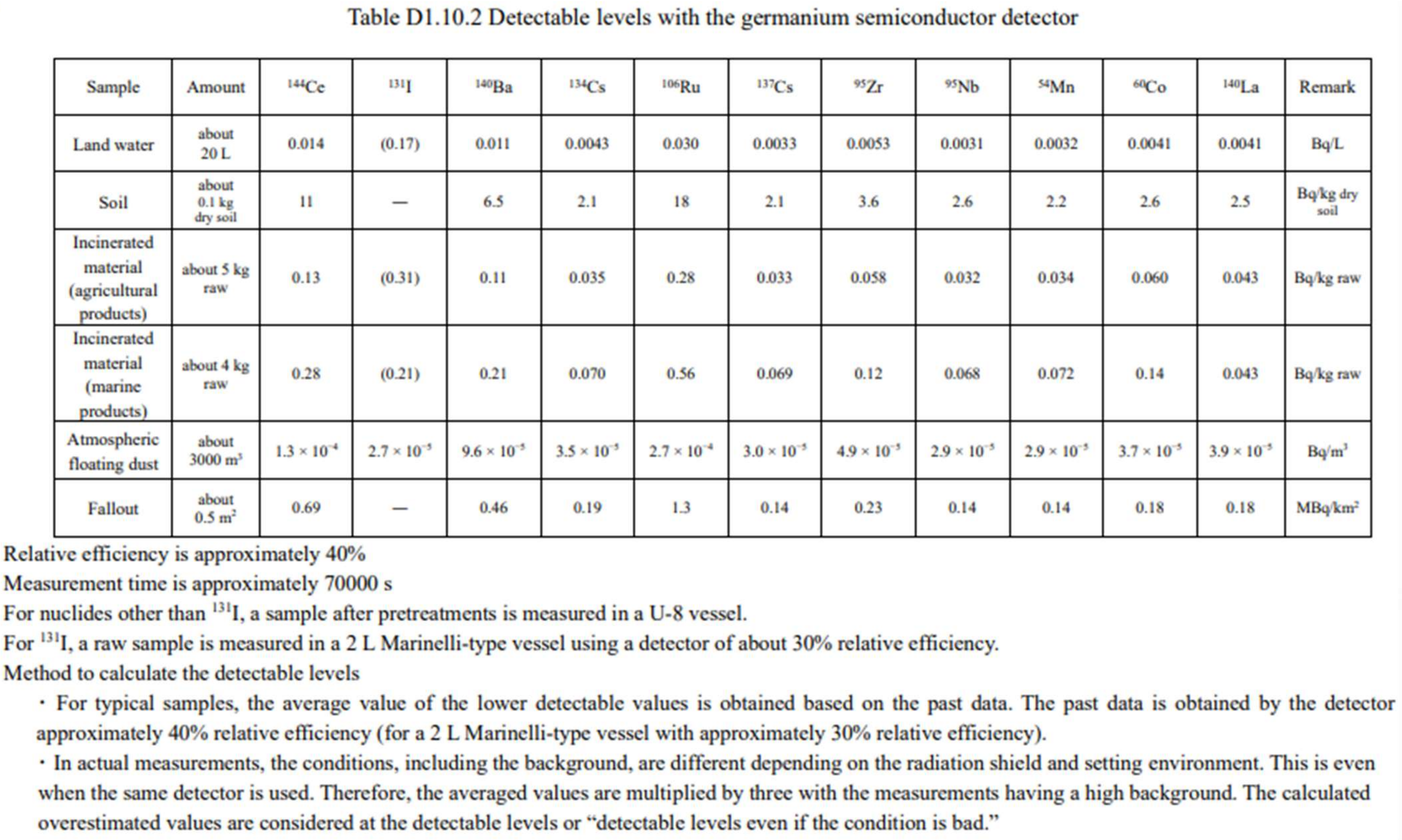

Table D1.10.2 Detectable levels with the germanium semiconductor detector

Sample	Amount	^{144}Ce	^{131}I	^{140}Ba	^{134}Cs	^{106}Ru	^{137}Cs	^{95}Zr	^{95}Nb	^{54}Mn	^{60}Co	^{140}La	Remark
Land water	about 20 L	0.014	(0.17)	0.011	0.0043	0.030	0.0033	0.0053	0.0031	0.0032	0.0041	0.0041	Bq/L
Soil	about 0.1 kg dry soil	11	—	6.5	2.1	18	2.1	3.6	2.6	2.2	2.6	2.5	Bq/kg dry soil
Incinerated material (agricultural products)	about 5 kg raw	0.13	(0.31)	0.11	0.035	0.28	0.033	0.058	0.032	0.034	0.060	0.043	Bq/kg raw
Incinerated material (marine products)	about 4 kg raw	0.28	(0.21)	0.21	0.070	0.56	0.069	0.12	0.068	0.072	0.14	0.043	Bq/kg raw
Atmospheric floating dust	about 3000 m^3	1.3×10^{-4}	2.7×10^{-5}	9.6×10^{-5}	3.5×10^{-5}	2.7×10^{-4}	3.0×10^{-5}	4.9×10^{-5}	2.9×10^{-5}	2.9×10^{-5}	3.7×10^{-5}	3.9×10^{-5}	Bq/m^3
Fallout	about 0.5 m^2	0.69	—	0.46	0.19	1.3	0.14	0.23	0.14	0.14	0.18	0.18	MBq/km^2

Relative efficiency is approximately 40%

Measurement time is approximately 70000 s

For nuclides other than ^{131}I, a sample after pretreatments is measured in a U-8 vessel.

For ^{131}I, a raw sample is measured in a 2 L Marinelli-type vessel using a detector of about 30% relative efficiency.

Method to calculate the detectable levels

- For typical samples, the average value of the lower detectable values is obtained based on the past data. The past data is obtained by the detector approximately 40% relative efficiency (for a 2 L Marinelli-type vessel with approximately 30% relative efficiency).
- In actual measurements, the conditions, including the background, are different depending on the radiation shield and setting environment. This is even when the same detector is used. Therefore, the averaged values are multiplied by three with the measurements having a high background. The calculated overestimated values are considered at the detectable levels or "detectable levels even if the condition is bad."

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Various corrections (factors affecting spectral analysis)

Summing･coincidence effect

When nuclides that emit multiple γ-rays simultaneously at the disintegration time are measured, there is a case that multiple photons simultaneously entering the detector are detected. Moreover, the output of the sum of the two-photon signals is generated. This phenomenon is called the summing･coincidence effect or summing effect.

The generation of such a summing effect decreases the counting rate of the total energy absorption peak to be measured. Thus, correcting this effect in the quantitative analysis of the spectrum is necessary.

Self-absorption

Self-absorption is a complex phenomenon that depends on the γ-ray energy, sample materials, sample shape, shape and size of the detector, and geometry between the detector and sample. Because self-absorption in a bulk sample is sometimes dozens of percentages or more, the correction is indispensable. To correct self-absorption, a method depending on the sample shape and materials is obtained by measuring the standard sources of various media.

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Alpha counting

Plutonium, americium, and curium are separated from a slurry using the Neodymium co-precipitation method and collected on a membrane filter. The resulting dried samples are then subjected to alpha particle measurement using a ZnS(Ag) scintillator-based counting system.

Alpha Particle Detection Systems:

- ZnS(Ag) Scintillation Counter
- 2π Gas-Filled Proportional Counter
- Silicon Semiconductor Alpha Spectrometer

Alpha counting: ZnS(Ag) Scintillation Counter

Detector: ZDS-451B
ZnS(Ag) scintillator (Detecting area: Φ50.6 mm)

Measurable sample:
Φ25.4 mm, Φ50.6 mm sample dish
Φ60 mm filter

https://www.aloka.co.jp/radiation/detector/jdc1813/

Alpha counting:
ZnS(Ag) Scintillation Counter

Measurement Procedure:

The following procedures (1)~4)) are routine maintenance tasks to be performed periodically.

1) Make a plateau curve using an alpha source to determine the applied voltage.
2) Set the voltage to the value determined in step 1 and allow it to stabilize for at least one hour.
3) Measure the background radiation for four hours.
4) Determine the counting efficiency by counting a standard source (e.g., U_3O_8, Am-241).
5) Count the alpha-emitting sample for approximately four hours at the same distance from the detector as the standard source (±0.1mm).
6) Record the sample name/number, count rate, start time, and counting duration.

https://www.envraddb.go.jp/wp-content/uploads/2020/12/No31.pdf

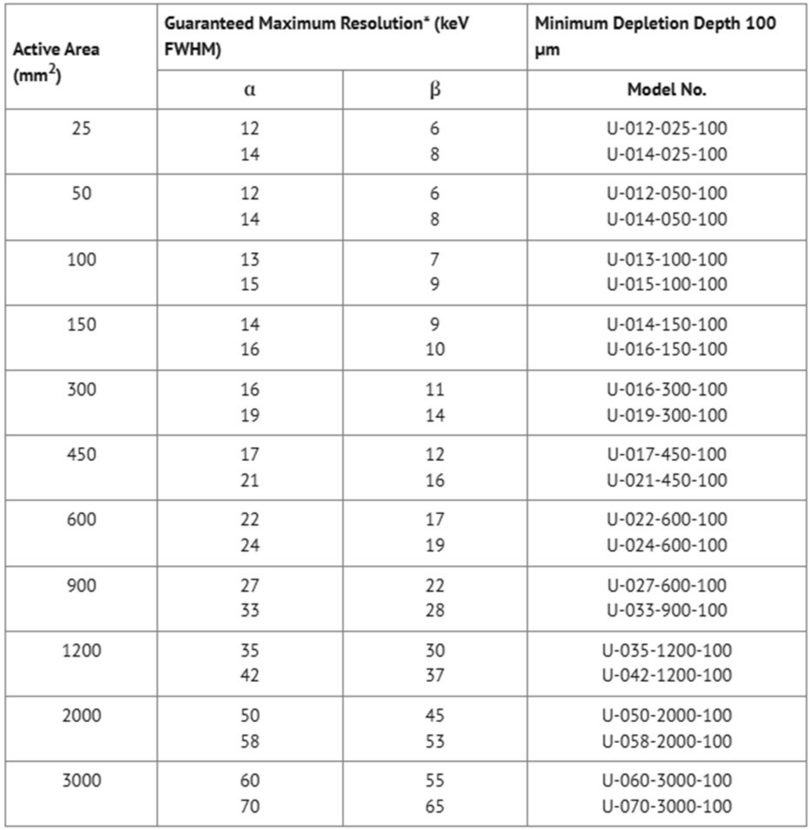
Alpha counting:
2π Gas-Filled Proportional Counter

Detector: LBC-5200 series
2π Gas-Filled Proportional Counter with a shield

Measurable sample:
Φ25.4 mm x 3.2, 6.2 mm sample dish
Φ50.6 mm x 3.3, 6.3 mm sample dish
Φ50, 60 mm filter

https://www.aloka.co.jp/radiation/detector/lbc51005200series/

Alpha counting:
Silicon Semiconductor Alpha Spectrometer



Active Area (mm^2)	Guaranteed Maximum Resolution* (keV FWHM)		Minimum Depletion Depth 100 μm
	α	β	Model No.
25	12 14	6 8	U-012-025-100 U-014-025-100
50	12 14	6 8	U-012-050-100 U-014-050-100
100	13 15	7 9	U-013-100-100 U-015-100-100
150	14 16	9 10	U-014-150-100 U-016-150-100
300	16 19	11 14	U-016-300-100 U-019-300-100
450	17 21	12 16	U-017-450-100 U-021-450-100
600	22 24	17 19	U-022-600-100 U-024-600-100
900	27 33	22 28	U-027-600-100 U-033-900-100
1200	35 42	30 37	U-035-1200-100 U-042-1200-100
2000	50 58	45 53	U-050-2000-100 U-058-2000-100
3000	60 70	55 65	U-060-3000-100 U-070-3000-100

https://www.sii.co.jp/jp/segg/products/archives/application/alpha-spectroscopy/

Alpha counting:
Calculation of radioactivity concentration

The total alpha activity concentration (A) is calculated using the following formula:

$$A = \frac{n \cdot 100}{E \cdot W} \qquad \sigma_A = \frac{\sigma_S}{n} A$$

Where:

A: Total alpha activity concentration (Bq/kg, Bq/L, etc.)
W: Sample weight (kg, L, etc.)
σA: Counting error in total alpha activity concentration (Bq/kg, Bq/L, etc.)
E: Counting efficiency (%)
n: Net count rate (counts per second, cps)
σS: Counting error in the net count rate (cps)

Beta counting

The objectives of total beta activity measurement include: (1) providing data to inform decisions regarding the necessity of precise isotope analysis; (2) providing data to determine whether regulatory standards have been exceeded; (3) monitoring temporal and spatial trends in radioactivity levels of specific targets, such as assessing the evolution of contamination levels due to radioactive waste; and (4) providing a general overview of radioactivity trends in the environment for public information dissemination.

Beta-ray Detection Systems:

- GM counter
- Plastic Scintillation Counter
- 2π Gas-Filled Proportional Counter

https://www.envraddb.go.jp/wp-content/uploads/2020/11/No1.pdf

Beta counting: GM Counter

Detector: JDC-1937-DET
GM tube (Detecting area: Φ50 mm)

https://www.aloka.co.jp/radiation/detector/jdc1813/

Beta counting:
GM Counter

Measurement Procedure:

The following procedure should be followed for measurement:

(1) Record the following parameters: Instrument model and name, counting chamber type, sample plate material and dimensions (inner diameter and depth), mica window thickness of the GM detector, sample plate position within the counting chamber, sample geometry and weight.

(2) Perform a measurement using a reference sample to verify the proper operation of the counting apparatus.

(3) Acquire background radiation counts for 15-30 minutes.

(4) Measure the sample for 15-30 minutes.

(5) After the complete measurement sequence, acquire a final background radiation count and calculate the average of the initial and final background values.

https://www.envraddb.go.jp/wp-content/uploads/2020/11/No1.pdf

Beta counting:
GM Counter

Measurement Procedure (continued):

(6) Calculate the true count rate (CPM) and standard deviation by subtracting the background count rate from the sample count rate using the following equations.

$$\left(\frac{N_s}{t_s}-\frac{N_b}{t_b}\right)\pm\sqrt{\left(\frac{N_s}{t_s^2}+\frac{N_b}{t_b^2}\right)}$$

Where:
Ns: Total counts
ts: Counting time of sample (min.)
Nb: Background counts
Tb: Counting time of background (min.)

(7) Calculate the radioactivity based on the obtained CPM.

Beta counting: Plastic Scintillation Counter

Detector: JDC-1918-DET
Plastic scintillator (Detecting area: Φ50 mm)

https://www.aloka.co.jp/radiation/detector/jdc1900/index_jdc1918.html

Beta counting:
2π Gas-Filled Proportional Counter

Detector: LBC-5100 or 5200 series
2π Gas-Filled Proportional Counter with a shield

Measurable sample:
Φ25.4 mm x 3.2, 6.2 mm sample dish
Φ50.6 mm x 3.3, 6.3 mm sample dish
Φ50, 60 mm filter

https://www.aloka.co.jp/radiation/detector/lbc51005200series/

Mass Spectrometry

Mass spectrometry is an analytical technique that elucidates the atomic mass, molecular mass, molecular structure, quantity (concentration), and physical state of chemical substances by ionizing the substance and measuring the mass-to-charge ratio of the resulting ions.
Measurements are performed using a mass spectrometer, which comprises an inlet system for sample introduction, an ionization source, a mass analyzer, a detector, a vacuum system, and a data system for instrument control and data processing.
During measurement, the introduced sample is ionized in the ion source, converting it into gaseous ions. These ions exhibit varying trajectories based on their mass-to-charge ratio (m/z). Therefore, an analyzer employing various principles (primarily electromagnetic techniques) separates the ions according to their m/z, and a detector is used to detect them.

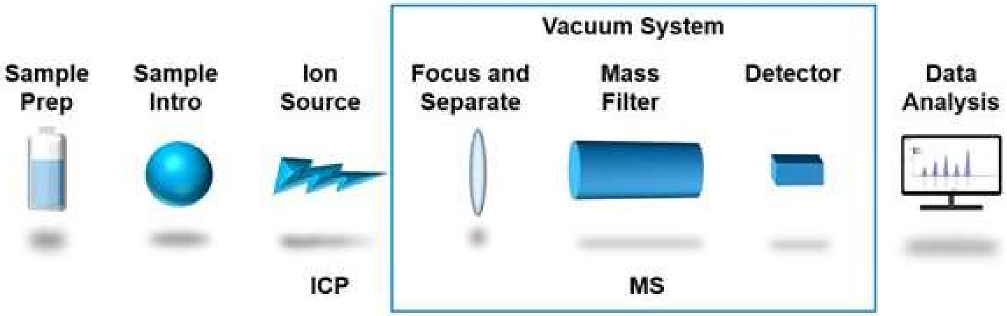
ICP-MS

ICP-MS uses an argon (Ar) plasma – the ICP – to convert the sample into ions that are then measured using a mass spectrometer – the MS. ICP-MS is similar to inductively coupled plasma optical emission spectroscopy (ICP-OES), but ICP-OES uses an optical spectrometer to measure the light emitted from elements as they pass through the plasma, whereas ICP-MS measures the elements (ions) directly. Both techniques provide fast analysis of multiple elements in a sample, but ICP-MS provides much lower detection limits than ICP-OES, so it’s a better choice for trace element analysis.

https://www.agilent.com/en/product/atomic-spectroscopy/inductively-coupled-plasma-mass-spectrometry-icp-ms/what-is-icp-ms-icp-ms-faqs

ICP-MS

An ICP-MS instrument consists of the ion source (the ICP), a mass spectrometer (MS) – usually a scanning quadrupole mass filter, and a detector. The ICP is at atmospheric pressure, while the MS and detector operate in a vacuum chamber, so an ICP-MS also requires a vacuum pump, a vacuum interface, and some electrostatic ion “lenses” to focus the ions through the system. Modern ICP-MS systems also typically contain some device or mechanism to resolve spectral interferences



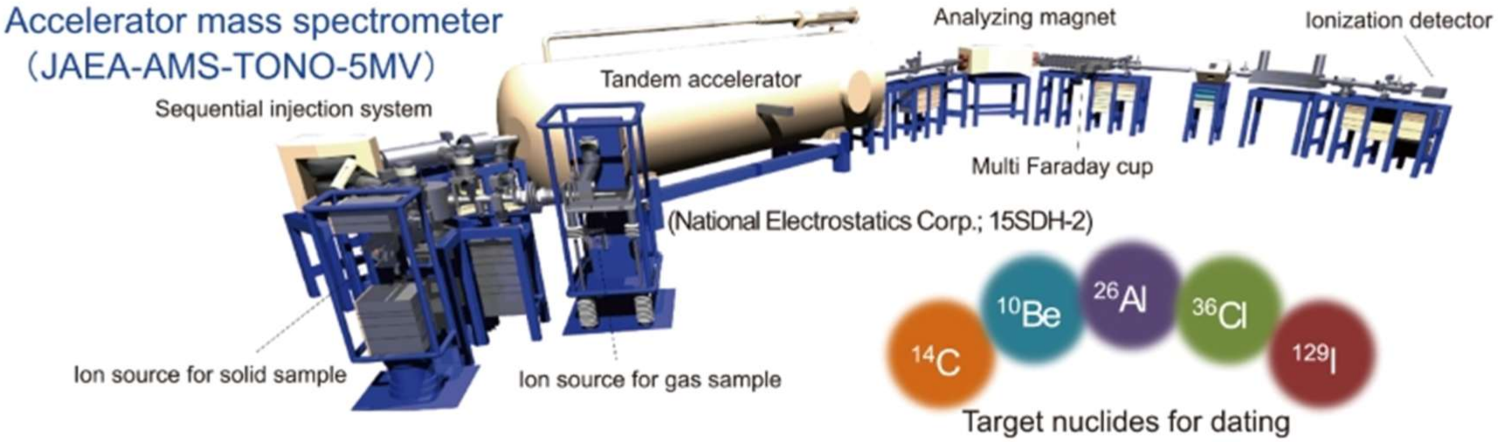
https://www.agilent.com/en/product/atomic-spectroscopy/inductively-coupled-plasma-mass-spectrometry-icp-ms/what-is-icp-ms-icp-ms-faqs

Advanced Methods

Accelerator Mass Spectrometry (AMS) is a technique where the ions of interest are accelerated to high energies (in the MeV range per nucleon) using an accelerator before undergoing mass analysis. Unlike conventional mass spectrometers (typically in the keV range), AMS can effectively eliminate interfering isotopes and isobaric molecules by leveraging the high kinetic energy imparted by the accelerator. This results in an extremely high-sensitivity method for analyzing specific nuclides.

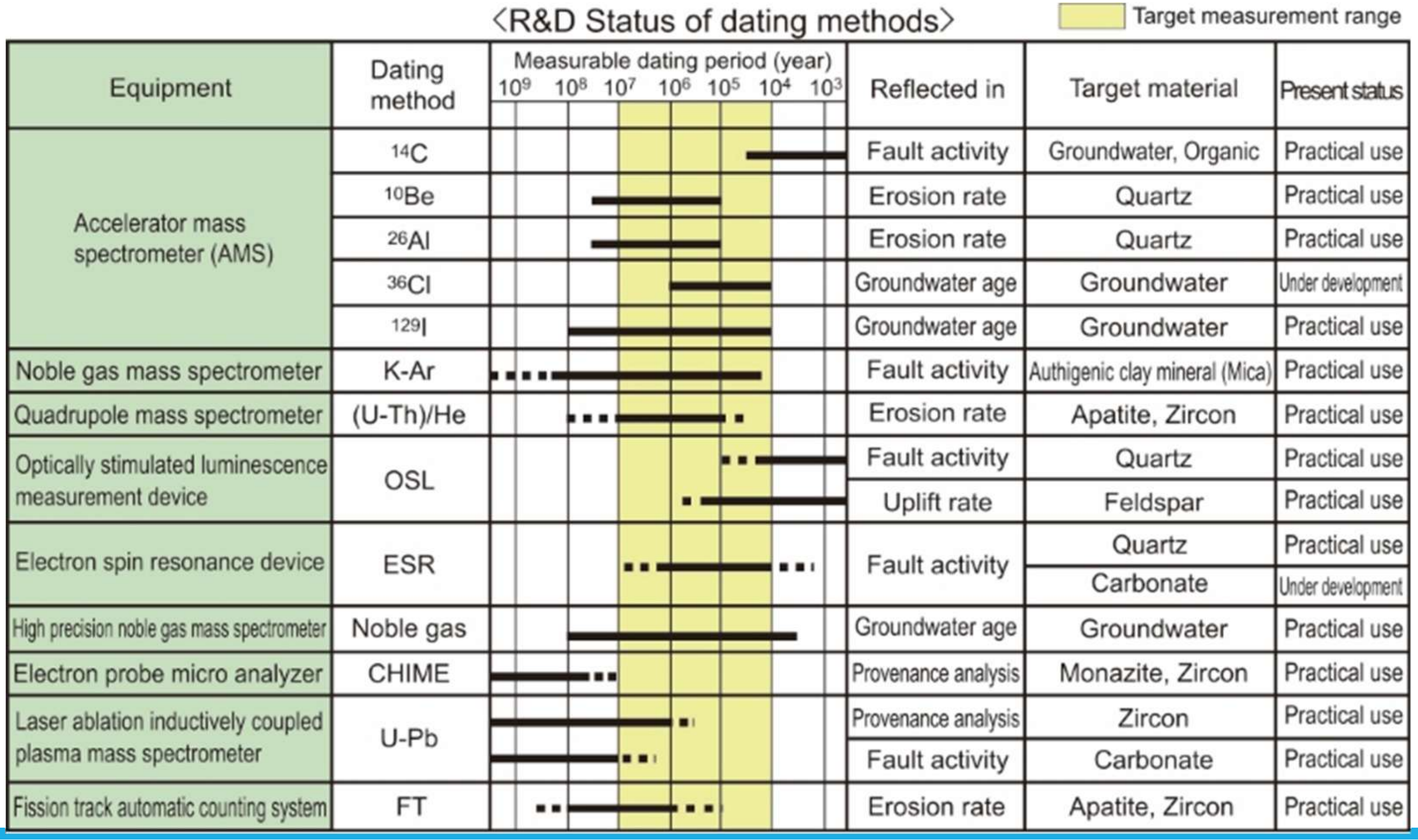
AMS

The basic components of an AMS system are: ① an ion source, ② a low-energy analyzer, ③ an accelerator, ④ a high-energy analyzer (including a system for stable isotope measurements), and ⑤ a detector (for detection of the target nuclide). Currently, most operational AMS installations worldwide utilize a tandem electrostatic accelerator as the accelerator component.



https://www.jaea.go.jp/04/tono/en/research/stability/stability3.html

AMS



〈R&D Status of dating methods〉

🟨 Target measurement range

Equipment	Dating method	Measurable dating period (year) 10^9 10^8 10^7 10^6 10^5 10^4 10^3	Reflected in	Target material	Present status
Accelerator mass spectrometer (AMS)	^{14}C		Fault activity	Groundwater, Organic	Practical use
	^{10}Be		Erosion rate	Quartz	Practical use
	^{26}Al		Erosion rate	Quartz	Practical use
	^{36}Cl		Groundwater age	Groundwater	Under development
	^{129}I		Groundwater age	Groundwater	Practical use
Noble gas mass spectrometer	K-Ar		Fault activity	Authigenic clay mineral (Mica)	Practical use
Quadrupole mass spectrometer	(U-Th)/He		Erosion rate	Apatite, Zircon	Practical use
Optically stimulated luminescence measurement device	OSL		Fault activity	Quartz	Practical use
			Uplift rate	Feldspar	Practical use
Electron spin resonance device	ESR		Fault activity	Quartz	Practical use
				Carbonate	Under development
High precision noble gas mass spectrometer	Noble gas		Groundwater age	Groundwater	Practical use
Electron probe micro analyzer	CHIME		Provenance analysis	Monazite, Zircon	Practical use
Laser ablation inductively coupled plasma mass spectrometer	U-Pb		Provenance analysis	Zircon	Practical use
			Fault activity	Carbonate	Practical use
Fission track automatic counting system	FT		Erosion rate	Apatite, Zircon	Practical use

https://www.jaea.go.jp/04/tono/en/research/stability/stability3.html

Advanced Methods

A Triple Quadrupole Inductively Coupled Plasma Mass Spectrometer (ICP-MS) features a structural configuration with two quadrupoles flanking a collision/reaction cell. This design enhances sensitivity and allows for complex isotopic analysis.

Figure 1. Cutaway diagram of the Agilent 8900 ICP-QQQ.

https://www.agilent.com/cs/library/applications/appcompendium_icp-qqq-5991-2802en-us-agilent.pdf

ICP-QQQ

In MS/MS operation, where both quadrupoles are operated as unit mass filters, ions at the target analyte mass are selected by the first quadrupole (Q1) and passed to the ORS cell, where the analyte ions are separated from overlapping interfering ions. The resulting product ions that emerge from the cell are then filtered by the second quadrupole (Q2) before being passed to the detector.
This configuration releases the full potential of reaction cell gas methods to resolve spectroscopic interferences including isobaric and doubly-charged interferences, as well as polyatomic ion overlaps.
As a result, ICP-QQQ can determine a wider range of analytes at much lower concentrations with greater reliability and higher confidence.

https://www.agilent.com/cs/library/applications/appcompendium_icp-qqq-5991-2802en-us-agilent.pdf

Radiochemical separation basics

Separation methods based on phase equilibria

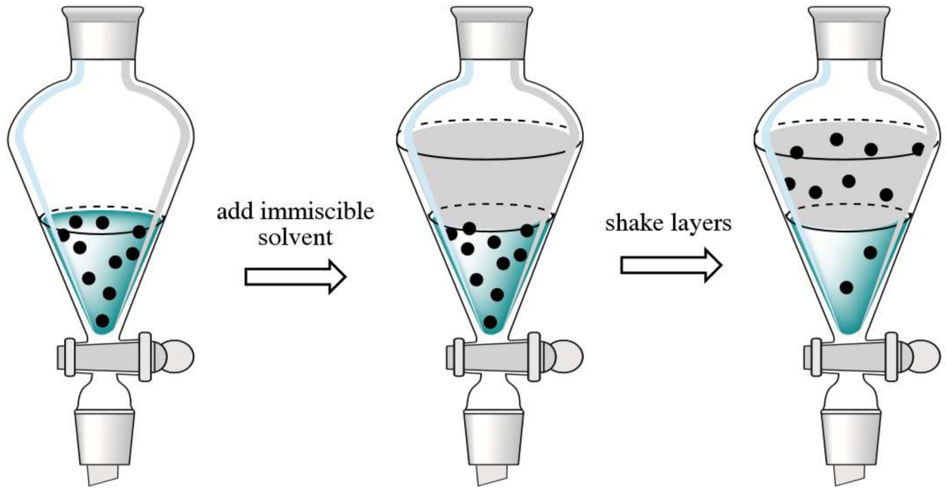
Gas - Liquid	Gas - Solid	Liquid - Liquid	Liquid - Solid
Disk	Adsorption	Solvent Extraction	Precipitation
Gas Chromatograph	Sublimation	Exclusion	Fractional Crystallization
	Molecular Sieves		Ion Exchange
	Gas Chromatograph		Extraction Chromatography
			Adsorption
			Ion exclusion

Particle Separation Methods

Filtration	Particle Electrophoresis
Sedimentation	Electrostatic Precipitation
Elutriation	Flotation
Centrifugation	Screening

Solvent extraction

This technique involves the equilibration of a solute between an aqueous phase and an immiscible organic solvent at a constant temperature, resulting in a partitioning of the solute in a defined ratio between the two phases.



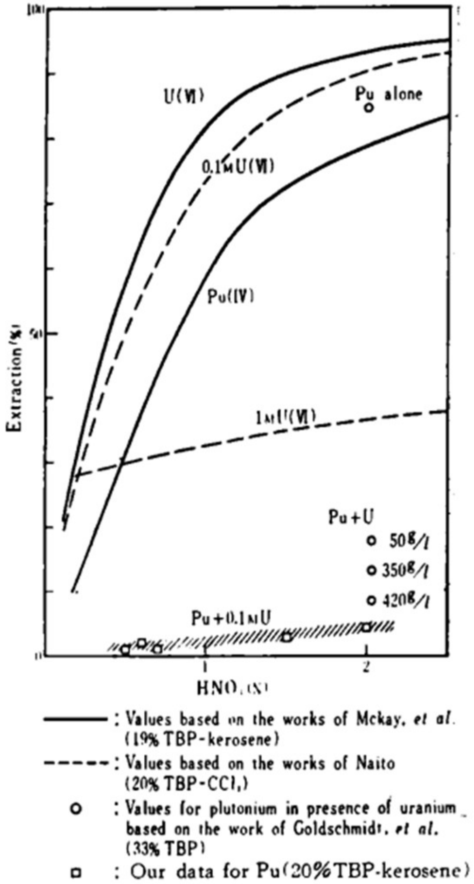
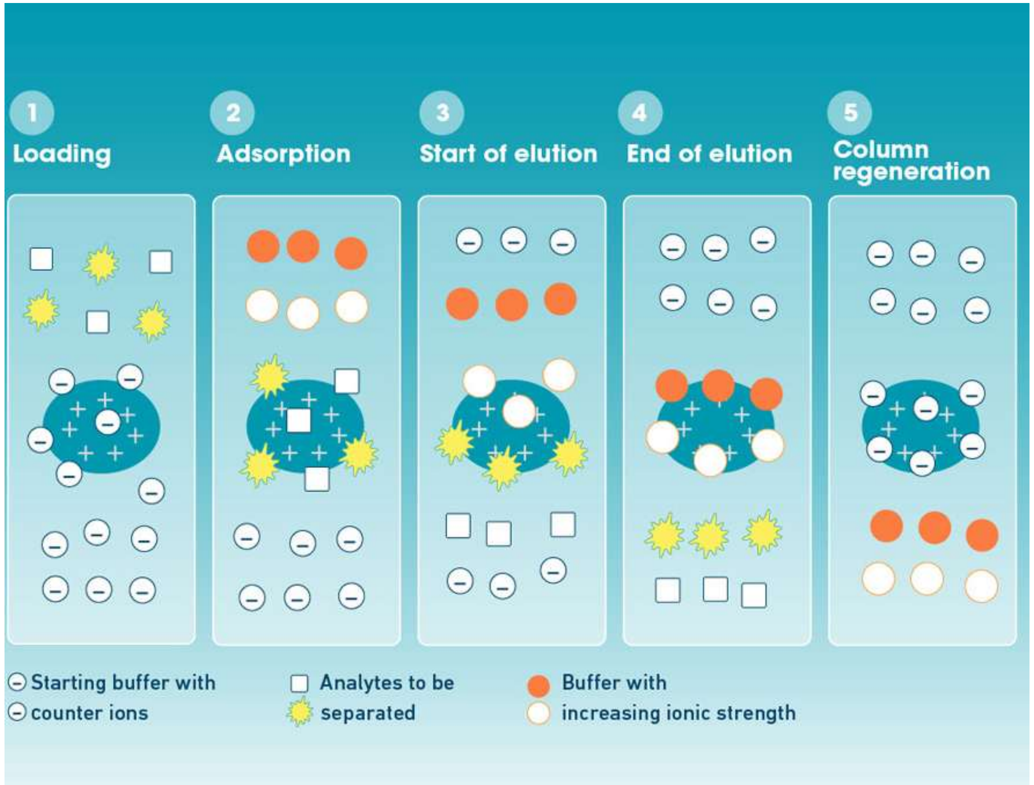


Fig.2 Extractions of U and Pu with TBP solutions from nitric acid solution

https://www.jstage.jst.go.jp/article/jaesj1959/3/7/3_7_507/_pdf/-char/ja

Ion Exchange



The chromatographic separation begins with equilibration of the stationary phase in the initial buffer to ensure analyte interaction. Samples dissolved in the same buffer are then loaded onto the column, with buffer pH and ionic strength optimized for analyte binding while minimizing impurity retention. A wash step removes unbound and similarly charged molecules, stabilizing the baseline. Elution is achieved using either a salt or pH gradient. Salt gradients elute analytes based on their binding strength, while pH gradients release analytes at their pI values. For ion elution, pH is adjusted accordingly. Finally, the column is regenerated with a high ionic strength buffer to restore capacity for subsequent runs, followed by conditioning with the initial buffer.

https://www.technologynetworks.com/analysis/articles/exploring-the-principle-of-ion-exchange-chromatography-and-its-applications-357491

Ion Exchange

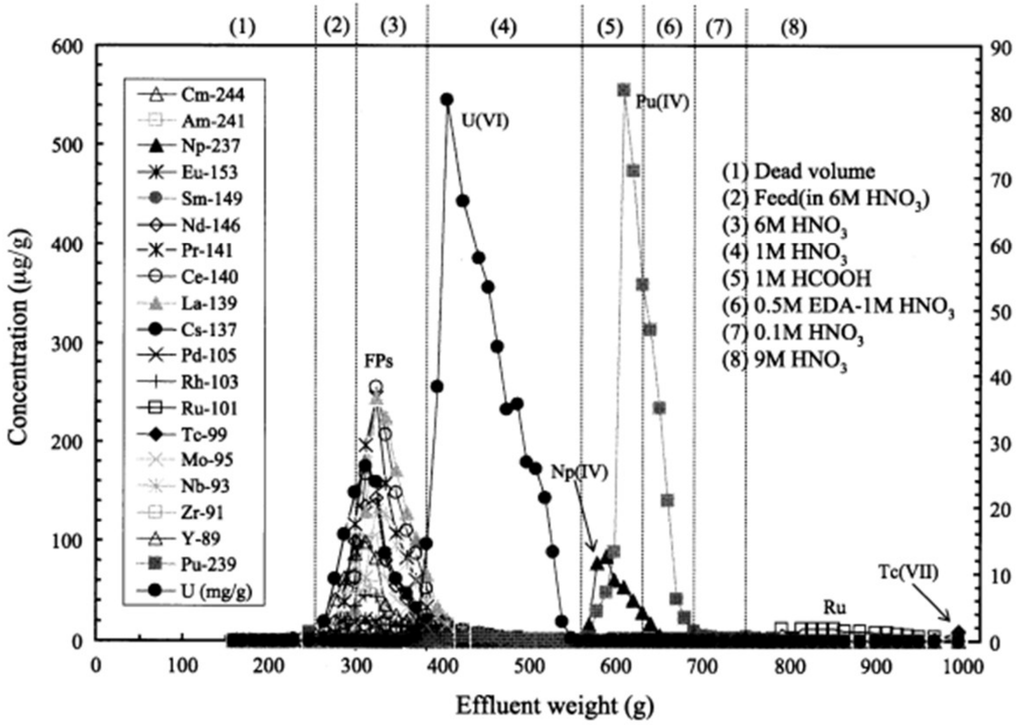


Fig. 12 Results of separation experiment for the nitric acid solution of a spent BWR-fuel by the 1st AR–01 column (Column: ϕ20 mm ×h1000 mm, Flow rate: 3.8 m/h, Temp.: 296 K).

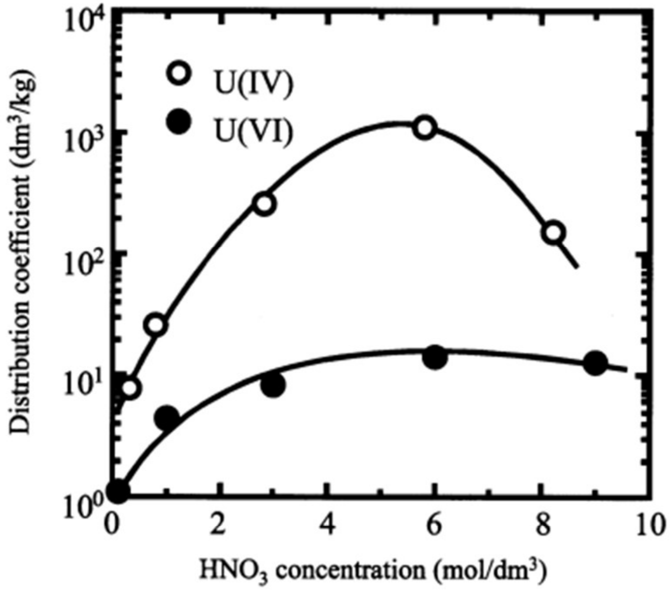


Fig. 7 Adsorption behavior of U from nitric acid solution onto AR–01 anion exchanger (2 h, 333 K).

https://www.jstage.jst.go.jp/article/jaie1990/16/2/16_2_102/_pdf/-char/ja

Extraction Chromatography

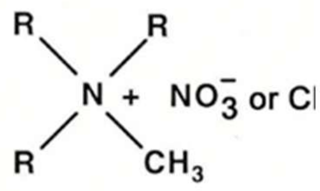
Extraction chromatographic resins allow the separation and determination of radionuclides (e.g. actinides and fission products) from environmental matrices e.g. soil, sediment, vegetation and seawater samples.

Their use allows a rapid and precise determination of radionuclides in emergency and routine situations, as well as, combined with suitable sample preparation methods, to obtain very low detection limits through analysis of large volume samples.

TEVA Resin

Figure 1

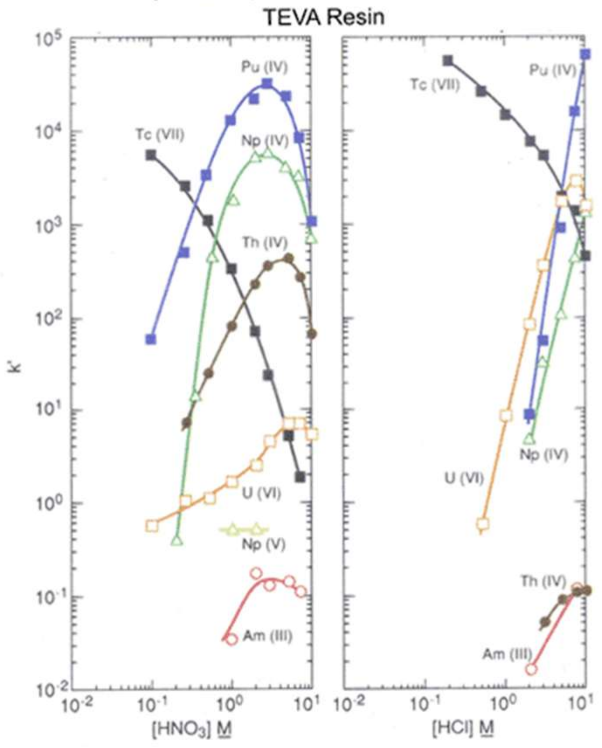
Trialkyl, methylammonium nitrate (or chloride)



R = C_8H_{17} and $C_{10}H_2$

Figures 2 & 3

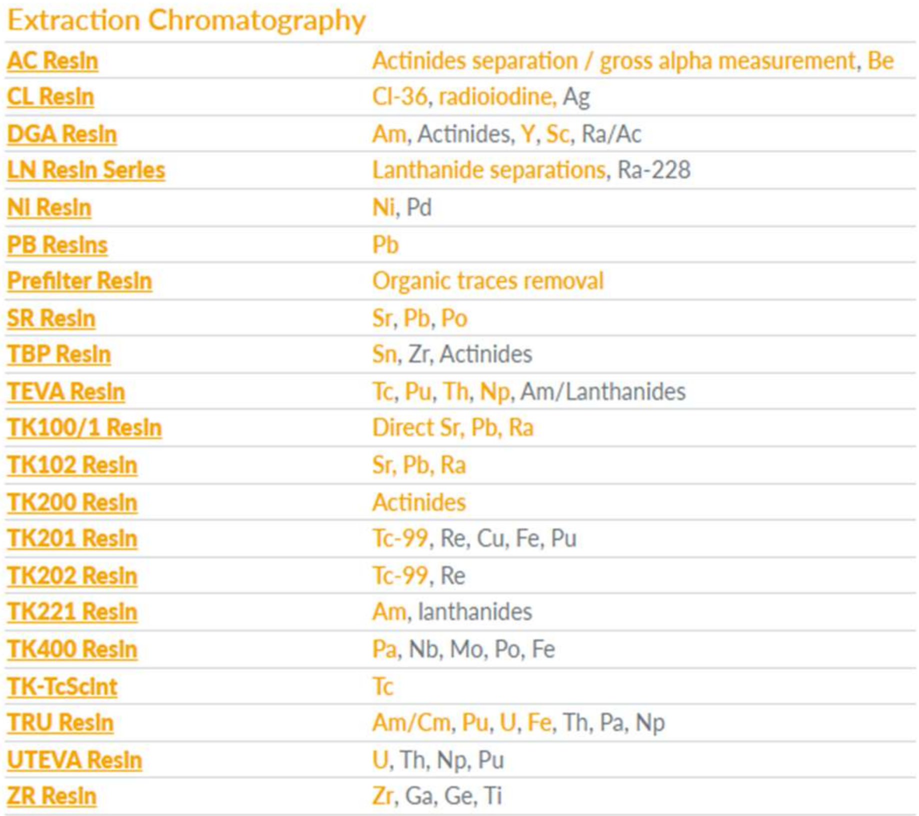
Acid dependency of k' for various ions at 23°C.



Horwitz, et al. (HP195)

https://www.eichrom.com/products/teva-resin/

Extraction Chromatography



Extraction Chromatography	
AC Resin	Actinides separation / gross alpha measurement, Be
CL Resin	Cl-36, radioiodine, Ag
DGA Resin	Am, Actinides, Y, Sc, Ra/Ac
LN Resin Series	Lanthanide separations, Ra-228
NI Resin	Ni, Pd
PB Resins	Pb
Prefilter Resin	Organic traces removal
SR Resin	Sr, Pb, Po
TBP Resin	Sn, Zr, Actinides
TEVA Resin	Tc, Pu, Th, Np, Am/Lanthanides
TK100/1 Resin	Direct Sr, Pb, Ra
TK102 Resin	Sr, Pb, Ra
TK200 Resin	Actinides
TK201 Resin	Tc-99, Re, Cu, Fe, Pu
TK202 Resin	Tc-99, Re
TK221 Resin	Am, lanthanides
TK400 Resin	Pa, Nb, Mo, Po, Fe
TK-TcScint	Tc
TRU Resin	Am/Cm, Pu, U, Fe, Th, Pa, Np
UTEVA Resin	U, Th, Np, Pu
ZR Resin	Zr, Ga, Ge, Ti

Acid dependency of k' for various ions at 23-25°C.
TRU Resin

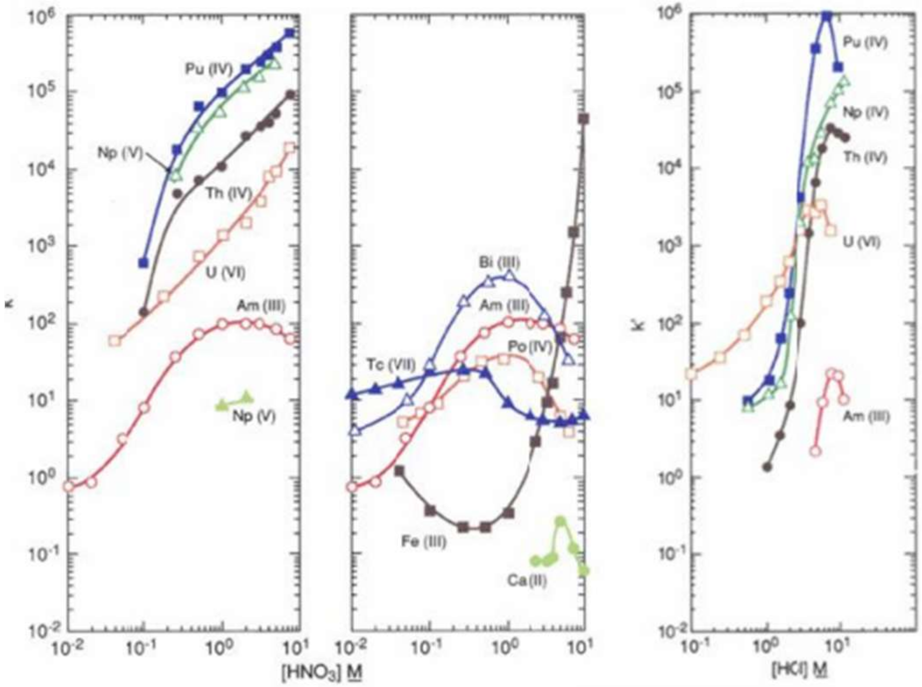
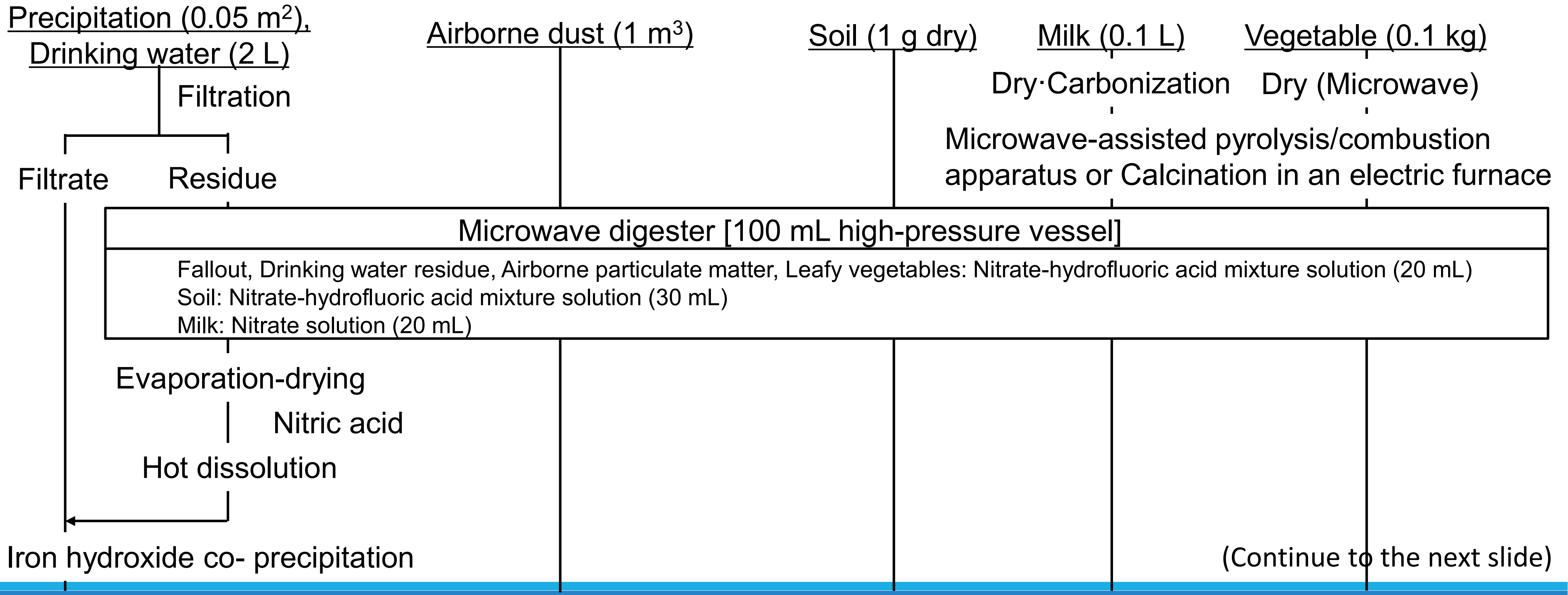


Figure 2 : k' values of different elements in HNO$_3$ and HCl media on TRU Resin (1)

https://www.triskem-international.com/scripts/files/6698d219daf470.41267302/PS_TRU-Resin_EN_161007.pdf

Rapid Analysis of Gross Alpha Radioactivity in Environmental Samples



Precipitation (0.05 m²), Drinking water (2 L)

Filtration

Filtrate

Residue

Airborne dust (1 m³)

Soil (1 g dry)

Milk (0.1 L)

Dry·Carbonization

Vegetable (0.1 kg)

Dry (Microwave)

Microwave-assisted pyrolysis/combustion apparatus or Calcination in an electric furnace

Microwave digester [100 mL high-pressure vessel]

Fallout, Drinking water residue, Airborne particulate matter, Leafy vegetables: Nitrate-hydrofluoric acid mixture solution (20 mL)
Soil: Nitrate-hydrofluoric acid mixture solution (30 mL)
Milk: Nitrate solution (20 mL)

Evaporation-drying

Nitric acid

Hot dissolution

Iron hydroxide co- precipitation

(Continue to the next slide)

https://www.envraddb.go.jp/wp-content/uploads/2020/12/No31.pdf

Rapid Analysis of Gross Alpha Radioactivity in Environmental Samples

Adjustment of Pu^{4+} (Using sodium nitrite)

↓

Iron(III) hydroxide co-precipitation

↓

Separation and purification of Pu, Am, and Cm using TRU resin

↓

Organic decomposition·Evaporation-drying

↓

Neodymium fluoride precipitation

↓

Alpha-ray measurement (ZnS(Ag) scintillation detector)

https://www.envraddb.go.jp/wp-content/uploads/2020/12/No31.pdf

Rapid Analysis of Gross Alpha Radioactivity in Environmental Samples

Sample	Sample amount	ZnS(Ag) scintillation counter[1)]	2π gas-flow counter[1)]	Si semiconductor detector[1)]	Unit	Reference level for radiological protection (Bq/kg)[2)]
Airborne dust	1 m^3	5	10	5	mBq/m^3	-
Soil	1g dry soil	5	10	5	Bq/kg dry soil	-
Precipitation	0.05 m^2, 1 d	0.1	0.2	0.1	$Bq/m^2/d$	-
Drinking water	2 L	0.002	0.005	0.002	Bq/L	1
Milk	0.1 L	0.05	0.1	0.05	Bq/L	1
Vegetable	0.1 kg fresh	0.05	0.1	0.05	Bq/kg fresh	10

1) 4-hour measurement; 2) Plutonium and transuranic alpha-emitting radionuclides

https://www.envraddb.go.jp/wp-content/uploads/2020/12/No31.pdf

Quality assurance

Adherence to the established analytical procedures is paramount. Equally critical is the implementation of routine maintenance to ensure traceability to national standards and the integrity of analytical instruments. Consistent execution of these practices provides demonstrable assurance of analytical result quality. Traceability typically involves the use of standard reference materials distributed by international organizations. To further guarantee result quality, adherence to the principles of ISO/IEC 17025 is recommended, encompassing both internal quality control (e.g., analyst proficiency testing) and external quality assurance (e.g., participation in proficiency testing schemes).

Quality assurance

Internal Quality Control:

- Traceability Assurance
- Routine Maintenance/Checks

External Quality Assessment/Control:

- Interlaboratory Comparison
- Proficiency Testing/Skill Assessment

Thank you for your attention!