

## 1 Radioactivity Measurements Method for Crude Drugs

(生薬の放射能測定法)

Crude drugs are natural products produced by harvesting cultivated plants/reared animals or collecting wild resources and processing them through washing and drying. This General Information describes the radioactivity measurement method of crude drugs that can be applied when there is a concern about the contamination of radioactive materials in more amounts exceeding that from natural origin.

The measurement methods described here are procedures to measure radioactivity by  $\gamma$ -ray spectrometry, and target nuclides are  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ .

### 1. Principle<sup>1)</sup>

In order to measure the radioactivity of a radionuclide in a sample, radioactive materials are identified based on the energy of radiation by measuring  $\alpha$ -rays being helium nuclei,  $\beta$ -rays being electrons and  $\gamma$ -rays being photons, emitted when radionuclides decayed, and radioactivity is determined from the number of radiations counted per unit time. Radiations have different penetrating powers depending on their kind and energy. Generally,  $\alpha$ -rays have weakest penetrating power and are shieldable by papers.  $\beta$ -Rays have stronger penetrating power than  $\alpha$ -rays, being shieldable by a light metal plate with a few millimeters thickness, and are classified to the weak penetrating power radiation. On the other hand,  $\gamma$ -rays have strong penetrating power, and shielding them needs a substance with a few to 10 centimeters which has high atomic number and high density such as lead.

Difference in penetration of radiation is an important factor in the measurement of radiation/radioactivity.  $\gamma$ -Rays are usually used to determine radionuclides.  $\alpha$ - and  $\beta$ -rays are easily self-shielded (absorbed) because of their weak penetrating radiation. They are suitable for the measurement of surface contamination, but the identification of radionuclides by their spectroscopies needs professional techniques for sample preparation, etc., and is not easy. On the other hand, most  $\gamma$ -rays do not lose their energy when penetrating a substance even in emission from inside the substance, and the information of the emitted  $\gamma$ -ray energy is obtained from the measured spectrum. Since  $\gamma$ -ray energy emitted from a radionuclide is different for each radionuclide, it is relatively easy to identify the radionuclide based on the obtained energy spectrum. For the measurement of radioactivity concentration in crude drugs, it is necessary to identify the radionuclide contained in the crude drug and to measure the concentration of the radionuclide, therefore measurement methods by  $\gamma$ -ray spectroscopy are recommended.

Semiconductor detectors and scintillators are known as detectors used for the radioactivity measurement methods by  $\gamma$ -ray spectroscopy. By injection of radiation, the former produce electron-hole pairs and the latter emit a light. Scintillator exhibits scintillation (flash and fluorescence), but the intensity of the light is very weak. Therefore, it is used with the combination of a photomultiplier tube, etc. which amplify an electric signal converted from photon. A germanium semiconductor detector (hereinafter referred to as "Ge detector"), one of semiconductors, has the highest performance as a detector that can measure the radionuclide in crude drugs. In addition, a thallium activated sodium iodide scintillation detector (hereinafter referred to as "NaI (Tl) detector") is easy to handle and can measure the radioactivity of crude drugs.

#### 1.1. Target radionuclide

The target nuclides are  $^{131}\text{I}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ .

##### 1.1.1. Ge detector

Radiation data necessary for measurement of radioactivity by  $\gamma$ -ray spectrometry using a Ge detector is shown in Table 1.

**Table 1** Radiation data of target nuclides for a Ge detector<sup>1)</sup>

Nuclide	Half-life	Energy	$\gamma$ -ray Emission Rate	$\gamma$ -ray that require correction of summing effect ( $\gamma$ -ray Emission Rate)
$^{131}\text{I}$	8.021 days	364.5keV	0.817	284.3keV(0.061), 637.0keV(0.072), etc.
$^{134}\text{Cs}$	2.065 years	604.7keV	0.976	569.3keV(0.154), 801.9keV(0.087) *, etc.
		795.9keV	0.855	
$^{137}\text{Cs}$	30.17 years	661.7keV	0.851	no (single $\gamma$ -ray)

\* When resolution is not high, the peaks of 795.9keV and 801.9keV can be treated as one peak (0.942).

##### 1.1.2. NaI (Tl) detector

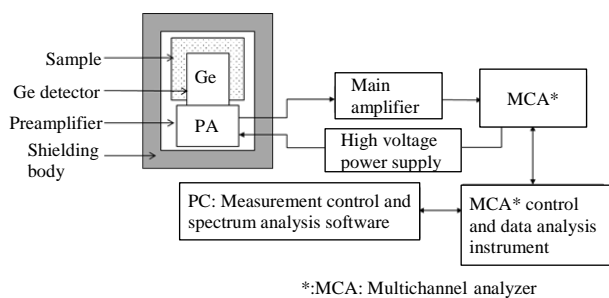
Radiation data necessary for the measurement of radioactivity by  $\gamma$ -ray spectrometry using a NaI detector is shown in Table 2. In the measurement using a NaI detector, radioactive Cs is treated as the sum of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  because it is difficult to quantify the radionuclides by precise distinction.

**Table 2** Radiation data of target nuclides for a NaI(Tl) detector<sup>1)</sup>

Nuclide	Half-life	Energy	$\gamma$ -ray Emission Rate	$\gamma$ value that require correction of summing effect ( $\gamma$ -ray Emission Rate)
$^{131}\text{I}$	8.021 days	364.5keV	0.817	284.3keV(0.061), 637.0keV(0.072), etc.
$^{134}\text{Cs}$	2.065 years	604.7keV	0.976	the peaks of 795.9keV and 801.9keV are treated as one peak (0.942).
		795.9keV	0.855	
		801.9keV	0.087	
$^{137}\text{Cs}$	30.17 years	661.7keV	0.851	no (single $\gamma$ -ray)

## 2. Apparatus

87 The system configuration of a  $\gamma$ -ray spectrometer is shown  
 88 in Figure 1. The apparatus generally consists of a detector, a  
 89 circuit part for measuring such as an amplifier, and an analy-  
 90 sis part (personal computer: PC) (Figure 1). In some com-  
 91 mercially available apparatuses, a circuit part for measuring  
 92 such as a high voltage power supply, amplifier and multi-  
 93 channel analyzer is integrated with a detector, and the result-  
 94 ant detection part which include a shielding body are combin-  
 95 ed with a PC for analysis. Details will be described later.  
 96



97  
 98  
 99 **Figure 1** The system configuration of a  $\gamma$ -ray spectrome-  
 100 ter<sup>2)</sup>

101 The Ge detector has a cooling system using liquid nitrogen.

### 102 3. Sampling, preparation, storage and transport

#### 103 3.1. Sampling

##### 104 3.1.1. Sampling container, tool and label

105 Fresh polyethylene bags are used for sampling containers.  
 106 Auxiliary tools for sampling are made of stainless, poly-  
 107 ethylene, or their equivalent materials. Parts which contact  
 108 with samples should be protected with polyethylene bags to  
 109 prevent contamination during transport. Because auxiliary  
 110 tools are used at sampling sites, pay attention to contamina-  
 111 tion from these tools when sampling are conducted at multi-  
 112 ple sites.

113 Fill out immediately the following items on sampling con-  
 114 tainers before or after sampling.

- 115 ① Sample number (lot)
- 116 (When collecting a same sample in multiple sample con-  
 117 tainers, each should be distinguished.)
- 118 ② Sample name
- 119 ③ Production area of sample
- 120 ④ Sampling date
- 121 ⑤ Sampler name
- 122 ⑥ Special notes
- 123 ⑦ Others necessary for evaluation
- 124

##### 125 3.1.2. Sampling and handling of samples

126 Random sampling is performed to collect samples repre-  
 127 senting a unit for measurement, and collected samples are ho-  
 128 mogenized by through mixing. As a general rule, one sample  
 129 is measured per one unit for measurement.

130 If direct sampling is difficult, collect samples using a  
 131 shovel, and transfer them to sampling containers, using a fun-  
 132 nel if necessary.

##### 133 3.1.3. Amount of sampling

134 About two times of amount required for testing is desirable.

#### 135 3.2. Preparation of sample

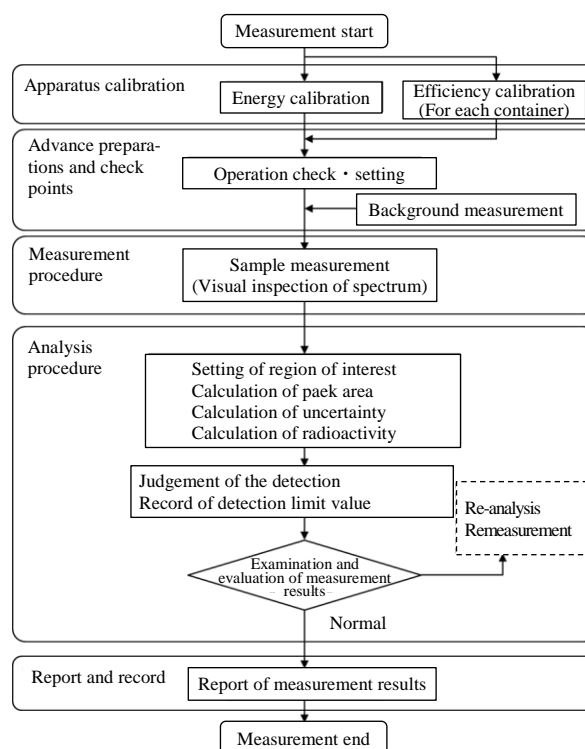
136 If necessary, adjust the size of samples to fit to each appa-  
 137 ratus. Crude drugs are produced using various parts of plants,  
 138 minerals, animals, and so on to cause various size, form and  
 139 hardness, so they are cut and crushed according to their char-  
 140 acteristics. Procedure such as washing that affect test results  
 141 must not be done after sampling.

#### 142 3.3. Storage and transport of sample

143 Test immediately after sampling. Make sure that sampling  
 144 containers are not broken and samples do not leak from sam-  
 145 pling containers. When testing is not performed immediately,  
 146 store samples avoiding moisture and insect damage.

### 147 4. Measurement of sample

148 An example of analysis by a  $\gamma$ -ray spectrometer is shown  
 149 in Figure 2.  
 150



151  
 152 **Figure 2.** An example of analysis flow  
 153

### 154 4.1. Measurement using a Ge spectrometer

#### 155 4.1.1. Characteristic of the measurement method

156 Because the measurement of radioactivity by a  $\gamma$ -ray spec-  
 157 trometer using a Ge detector has a very high energy resolu-  
 158 tion, it can determine energy accurately to identify a radionu-  
 159 chloride easily and certainly, and clearly analyze the energy by

160 separately from the other  $\gamma$ -rays with close energy. Moreover,  
161 because the spreading of  $\gamma$ -ray peaks is small and the ratio of  
162 the background to the peak is low, it is suitable for low level  
163 radioactivity.

#### 164 4.1.2. Apparatus, tool and so on

##### 165 4.1.2.1. Configuration of apparatus

###### 166 (1) Ge detector

167 The relative efficacy of a detector should be not less than  
168 20%.

169 The energy resolution is generally 1.8 to 2.0 keV as a half  
170 width.

###### 171 (2) Shielding body

172 Shielding a detector is very important in the measurement  
173 of low level radioactivity.  $\gamma$ -Rays derived from natural nu-  
174 clides ( $^{40}\text{K}$ , nuclides of the uranium and thorium series)  
175 should be sufficiently shielded.

176 A lead shielding body with 10 to 15 cm thick is generally  
177 used around a detector.

178 The size of the inner space of the shielding body must be  
179 enough to put a sample container.

180 There are vertical type (dip stick) and L-type (cryostat is  
181 attached to the side of a liquid nitrogen container) detectors,  
182 and the structure of the shielding body differs depending on  
183 both the form of a detector and the connection form between  
184 a detector and a liquid nitrogen container.

##### 185 4.1.2.2. Tools and so on

###### 186 (1) Sample container

187 Sample containers should have good sealing performance,  
188 high mechanical strength, resistance to acid and heat, and the  
189 internal sample should be visible. Sample containers include  
190 Marinelli containers with an internal volume of 1 to 2 L and  
191 cylindrical containers with an internal volume of 100 to 500  
192 mL. Sample containers should be selected based on sample  
193 volume.

###### 194 (2) Energy calibration source

195 Select some energy calibration sources to cover from 100  
196 keV to 2000 keV such as  $^{22}\text{Na}$  (511 keV, 1275 keV),  $^{54}\text{Mn}$   
197 (835 keV),  $^{60}\text{Co}$  (1173 keV, 1332 keV),  $^{88}\text{Y}$  (898 keV, 1836  
198 keV),  $^{137}\text{Cs}$  (662 keV) and  $^{139}\text{Ce}$  (166 keV). Each radioactiv-  
199 ity should be 1000 to 3000 becquerel (Bq).

###### 200 (3) Efficiency calibration source

201 Efficiency calibration sources are commercially available  
202 standard samples containing  $^{137}\text{Cs}$  and so on, and their vol-  
203 umes of the container and the medium are equal to those of a  
204 sample. Each radioactivity should be 1000 to 3000 Bq.

###### 205 (4) Software for spectral analysis

206 The software which can search peaks, identify nuclides,  
207 calculate peak areas and statistical uncertainties of count, is  
208 used. In addition, it is desirable to be able to correct self-ab-  
209 sorption and summing effect.

#### 210 4.1.3. Apparatus calibration

##### 211 4.1.3.1. Energy calibration

212 For energy calibration sources, correspondence relation-  
213 ship between  $\gamma$ -ray energy and a peak center channel is ob-  
214 tained as a linear equation according to the following proce-  
215 dure.

216 (1) Attach an energy calibration source to the regular po-  
217 sition of a detector, and measure the spectrum until the peak  
218 area of a main  $\gamma$ -ray reaches several thousand counts.

219 (2) Assuming that  $\gamma$ -ray energy ( $E$ ) and a peak center  
220 channel ( $p$ ) are in linear relationship, the following formula  
221 is obtained using spectral analysis software.

$$222 \quad E = a + b \times p$$

223 By setting the energy range of  $\gamma$ -rays to 0 to 2000 keV and  
224 the channel full scale of a multichannel analyzer to 4000 ch,  
225 the region of interest can be set easily even when the count  
226 value is low, and in this case “a” in the above formula is close  
227 to 0, and “b” is as close to 0.500 as possible.

228 (3) Record and save the above data.

##### 229 4.1.3.2. Efficiency calibration

230 In order to determine radioactivity from a measured  $\gamma$ -ray  
231 spectrum, counting efficiency to a peak (hereinafter referred  
232 to as “peak efficiency”) is necessary, and radioactivity anal-  
233 ysis postulates that the peak efficiency is correctly calibrated.

234 For efficiency calibration, use efficiency calibration  
235 sources of which concentration are known. Usually, standard  
236 sources containing various nuclides are measured to obtain a  
237 peak efficiency function with energy as a variable so that it  
238 can be applied to the energy range of approximately 50 to  
239 2000 keV. Since the peak efficiency varies depending on a  
240 sample container, it is necessary to perform efficiency cali-  
241 bration for each sample container when using multiple sam-  
242 ple containers.

##### 243 4.1.4. Procedure

###### 244 4.1.4.1. Preparations in advance and points to be 245 checked

###### 246 (1) Operation check of apparatus and settings

247 Before measuring a sample, use spectral analysis software  
248 to analyze the spectrum of the energy calibration source and  
249 confirm that the peak center channel, half width and peak  
250 count rate are normal for major  $\gamma$ -rays.

###### 251 (2) Background measurement

252 Measure a background under specified measurement con-  
253 ditions. In principle, the measurement is performed by plac-  
254 ing a sample for background measurement (enclose the same  
255 amount of water that does not contain the target radionuclide  
256 in the same sample container) that has the same conditions as  
257 the sample.

258 Since the analytical result of a background spectrum meas-  
259 ured recently is used for the radioactivity analysis of a sample,  
260 when a peak corresponding to the main  $\gamma$ -ray energy of a tar-  
261 get nuclide shown in Table 1 is observed, calculate the count  
262 rate and the statistical uncertainty of count and save the result

263 in preparation for measurement. Note that there is 609.3 keV  
 264 (0.426) emitted from  $^{214}\text{Bi}$  of the uranium series in a back-  
 265 ground spectrum and this spectrum is close to 604.7 keV of  
 266  $^{134}\text{Cs}$ .

#### 267 4.1.4.2. Measurement procedure

268 When filling a sample into a sample container, take care to  
 269 make the gap as small and uniform as possible. Therefore,  
 270 according to the characteristics of a crude drug to be meas-  
 271 ured, pretreat it by cutting or crushing as necessary.

272 Attach the same container filled with the same amount of  
 273 a sample as the standard sample used for the efficiency cali-  
 274 bration to the center of a detector. At this time, the deviation  
 275 from the center should be within about 1 cm for Marinelli  
 276 containers and within about 2 mm for cylindrical containers.

277 Start measurement after setting the measurement time of  
 278 the sample so that a target detection limit value can be ob-  
 279 tained based on the results of peak efficiency and background  
 280 measurement. Note that a peak shape may deteriorate when a  
 281 count rate is very high.

282 After the measurement, save the spectrum data.

#### 283 4.1.4.3. Analysis procedure

##### 284 (1) Setting of region of interest

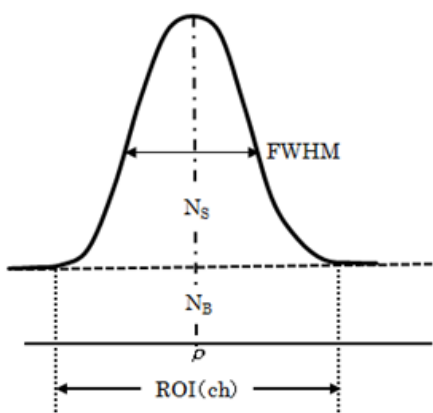
285 After the measurement is completed, set the region of in-  
 286 terest (ROI) for the  $\gamma$ -ray of the target nuclide using spectrum  
 287 analysis software. At this time, if the count is insufficient, the  
 288 variation of the count for each channel can be leveled by  
 289 smoothing process.

290 From the peak center channel ( $p$ ) and the full width at half  
 291 maximum (FWHM), it can be confirmed that the measured  
 292 spectrum is normal, but the value may vary for weak peaks.

##### 293 (2) Calculation of peak area

294 Peak areas ( $N_S$ ) are calculated based on a peak search in  
 295 ROI automatic setting by analysis software, but for especially  
 296 weak peaks, confirm whether the position and width of the  
 297 ROI are appropriate.

298



299

300

301 Figure 3 Setting of region of interest (ROI), and cal-  
 302 culation of peak area ( $N_S$ ) and background area ( $N_B$ )

##### 304 (3) Subtraction of background count rate and calculation 305 of statistical uncertainty of count

306 In usual  $\gamma$ -ray spectrometry, it is not always necessary to  
 307 subtract a background count rate  $n_{BG}$  (hereinafter referred to  
 308 as “BG count rate”), but if a detector and the inside of a shield  
 309 body are contaminated, it is necessary to subtract the BG  
 310 count rate. A net count rate  $n$  ( $s^{-1}$ ) is obtained by subtracting  
 311 the BG count rate in the same ROI from the sample count rate  
 312 ( $n_S = N_S / t_S$ ).

$$n = n_S - n_{BG}$$

314 The relation of a count error  $\sigma_n$  and a count rate ( $n$ ) is ex-  
 315 pressed as the following formula:

$$n \pm \sigma_n = n \pm (n/t)^{1/2}$$

317 The statistical uncertainty of count to a net count rate,  $\sigma_n$   
 318 ( $s^{-1}$ ), is expressed as the root sum squares of statistical uncer-  
 319 tainties ( $\sigma_S$  and  $\sigma_{BG}$ ) of each count rate, shown as follows:

$$\sigma_n = (\sigma_S^2 + \sigma_{BG}^2)^{1/2}$$

##### 321 (4) Calculation of radioactivity

322 The radioactivity  $A$  (Bq) and the radioactive concentration  
 323  $C$  (Bq/kg) of a sample are obtained by the following formulae:  
 324

$$A = \frac{n}{a\epsilon f_{SUM}} \quad C = \frac{A}{M}$$

326 In the above formulae, the abbreviations are as follows:

327  $n$ : Count rate

328  $a$ :  $\gamma$ -Ray emission rate ( $Bq^{-1}$ ) shown in Table 1

329  $\epsilon$ : Peak efficiency

330  $f_{SUM}$ : Correction factor for summing effect

331  $M$ : Mass (kg) of a sample in a sample container

332 However, when the measurement is compared with a  
 333 standard sample, “ $a$ ”, “ $\epsilon$ ”, and “ $f_{SUM}$ ” are the same, so there  
 334 is no need to consider. In other words, when the radioactivity  
 335 of the standard sample is  $A_{STD}$  and the count rate is  $n_{STD}$ , the  
 336 radioactivity  $A$  is obtained by  $A = (n / n_{STD}) A_{STD}$ .

##### 337 (5) Uncertainty of detection

338 If it is not necessary to consider the uncertainty of the mass  
 339 of a sample in a sample container, the statistical uncertainty  
 340 of count  $\delta_A$  (Bq) of the radioactivity of the sample and the  
 341 statistical uncertainty of count  $\delta_C$  (Bq / kg) of the radioactive  
 342 concentration are obtained by the following formulae:

$$\delta_A = \frac{\sigma_n}{n} A \quad \delta_C = \frac{\delta_A}{M}$$

343

344 If the measured radioactive concentration  $C$  exceeds  $3\delta_C$ ,  
345 it is considered statistically significant.

346 **(6) Record of detection limit value**

347 If no radioactivity is detected, record  $3\delta_C$  of the radioactiv-  
348 ity value, which would be measured in the analysis, as the  
349 detection limit value.

350 The detection limit value is affected by a BG count rate,  
351 sample measurement time, and sample mass. In radioactivity  
352 measurement, methods to calculate radioactivity from the  
353 count rate of the background part of the peak to be measured  
354 (usually calculated from the baseline region beside the peak),  
355 BG count rate, sample measurement time and background  
356 measurement time, sample mass, etc. are widely used.

357 **(7) Examination and evaluation of measurement results**

358 Summarize measurement results for each target nuclide,  
359 and confirm that it is normal based on the statistical uncer-  
360 tainty of count, peak center channel, FWHM, etc. If there is  
361 any doubt in the confirmation of results, remeasure as neces-  
362 sary.

363 **4.1.5. Points to note for measurement**

364 **4.1.5.1. Control of background**

365 When the same nuclide as a target nuclide is detected from  
366 a background, it is necessary to confirm the cause and sup-  
367 press the influence of the background as much as possible. In  
368 the case of indoor contamination, the influence can be sup-  
369 pressed by cleaning and checking shielding devices around  
370 an apparatus and performing appropriate shielding. If the in-  
371 side of a shield body or a detector itself is contaminated, de-  
372 contamination should be generally attempted. However, if  
373 decontamination is impossible, it is necessary to subtract the  
374 contribution from the background when calculating radioac-  
375 tivity.

376 **4.1.5.2. Contamination prevention of apparatus, tool  
377 and so on**

378 Cover a detector with polyethylene bag to prevent from  
379 contamination. If contamination should occur, cope by re-  
380 placing the polyethylene packaging. If the surface of a detec-  
381 tor is contaminated, wipe it off with a neutral detergent or  
382 gauze soaked in ethanol. Be careful not to let contaminants  
383 such as dust enter when opening a shield body.

384 Use a sample container after simple cleaning. When a sam-  
385 ple is placed into a sample container, it is also important to  
386 prevent the sample from adhering around the sample con-  
387 tainer.

388 Use a disposable container when measuring a high concen-  
389 tration sample or when decontamination is difficult. When a  
390 sample container is used repeatedly, it is recommended to ap-  
391 ply fluorine coating. It is also effective to use a plastic bag in  
392 a sample container.

393 **4.1.5.3. Routine maintenance of apparatus**

394 Regular performance tests of an entire measurement sys-  
395 tem is very important for the control of apparatuses. In the

396 performance tests, a  $\gamma$ -ray source for checking is placed at a  
397 fixed position on a detector, and a peak center channel,  
398 FWHM and peak count rate are obtained for low, medium  
399 and high energy  $\gamma$ -rays. Save these data as time-series data.  
400 The performance tests should be preferably performed daily,  
401 at least prior to a series of sample measurement, so that accu-  
402 rate energy calibration can always be used.

403 In addition, it is confirmed that there is no contamination  
404 around a detector and a sample container by regularly per-  
405 forming measurement with no sample or by placing an empty  
406 container.

407 One of detector troubles is vacuum loss in a cryostat. This  
408 can be judged from the consumption of liquid nitrogen, a de-  
409 crease in energy resolution, and visual inspection (the exist-  
410 ence of condensation at the neck of the cryostat).

411 **4.2. Measurement by NaI (TI) spectrometer**

412 **4.2.1. Characteristic of measurement method**

413 Scintillation detectors calculate the energy and number of  
414 radiation by converting a weak light generated at the time  
415 when radiation enters a solid crystal called a scintillator, into  
416 an electric signal using a photomultiplier. As the advantages,  
417 it is relatively inexpensive compared to Ge detectors and can  
418 be used at room temperature. Another characteristic is that  
419 the detection efficiency is determined by the size of crystals  
420 because the size of commercially available solid crystals is  
421 standardized. Some solid crystals include NaI and  $\text{LaBr}_3^{3)}$ .

422 **4.2.2. Apparatus, tool and so on**

423 **4.2.2.1. Configuration of apparatus**

424 A scintillation spectrometer is generally composed of a  
425 scintillation detector, a high-voltage power supply, an ampli-  
426 fier, a multichannel analyzer, and a PC for analysis. The scin-  
427 tillation spectrometer has the analytical function of a  $\gamma$ -ray  
428 spectrum and can perform processes from radioactive meas-  
429 urement to quantitative calculation.

430 **(1) Detector**

431 The energy resolution is not more than 8%.

432 **(2) Shielding body**

433 In order to reduce the influence of environmental radiation  
434 (background), it is desirable to have the structure of which  
435 detector surroundings are shielded with lead. It is more desir-  
436 able to surround the all surface with lead because the reduc-  
437 tion effect is low when shielding only the side surface of a  
438 detector.

439 **(3) Sample stage**

440 Set the sample stage that can fixes a detector and a sample  
441 in a fixed spatial relationship (geometry) and is stored in a  
442 shield body. In this case, it is desirable to place a beaker sam-  
443 ple container perpendicularly just above the detector in terms  
444 of detection efficiency and stability during measurement. In  
445 the case of a rectangular sample container, it is also possible  
446 to make the side of the container and the sample stage closely  
447 contact with the detector horizontally.

#### 448 4.2.2.2. Tool and so on

##### 449 (1) Sample container

450 Marinelli containers, plastic bottles, polyethylene tanks,  
451 etc. are used as sample containers. In the case of an emer-  
452 gency, quantification is possible by inserting a detector in a  
453 bucket containing a sample. However, it is necessary to cal-  
454 culate detection efficiency for each measurement container in  
455 advance.

##### 456 (2) Energy calibration source

457 Select some energy calibration sources to cover from 100  
458 keV to 2000 keV such as  $^{22}\text{Na}$  (511 keV, 1275 keV),  $^{54}\text{Mn}$   
459 (835 keV),  $^{60}\text{Co}$  (1173 keV, 1332 keV),  $^{88}\text{Y}$  (898 keV, 1836  
460 keV),  $^{137}\text{Cs}$  (662 keV), and  $^{139}\text{Ce}$  (166 keV). Obtain the rela-  
461 tionship between the  $\gamma$ -ray energy and the peak center chan-  
462 nel as a linear equation.

463 Because the energy resolution of NaI spectrometers is low  
464 unlike Ge detectors, the radiation source composed of some  
465 nuclides with close  $\gamma$ -ray energy is not used.

##### 466 (3) Efficiency calibration source

467 For efficiency calibration, use the efficiency calibration  
468 source whose radioactivity is known. Since peak efficiency  
469 varies depending on a sample container, it is necessary to cal-  
470 ibrate the efficiency for each sample container when using  
471 multiple sample containers. It is desirable to use the nuclide  
472 which emit one or two  $\gamma$ -rays, considering the energy resolu-  
473 tion of a spectrometer. It is desirable that the source includes  
474 target nuclides,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ .

##### 475 (4) Software for spectral analysis

476 Even if there is overlapping of peaks attributed to multiple  
477 nuclides, the software should be able to separate the peak of  
478 interest and calculate its area by peak function fitting, etc. It  
479 is desirable to be able to perform peak analysis conforming  
480 to "Radioactivity measurement series No.7,  $\gamma$ -Ray Spectrom-  
481 etry by Germanium Semiconductor Detector"<sup>4)</sup>. In addition,  
482 it is desirable to be able to calculate radioactive concentration  
483 from the radiation data of a nuclide to be quantified (half-life,  
484  $\gamma$ -ray emission rate) and detection efficiency.

#### 485 4.2.3. Apparatus calibration

##### 486 4.2.3.1. Energy calibration

487 An energy calibration equation is obtained by using sev-  
488 eral energy calibration sources after the channel width of a  
489 multichannel analyzer is set to about 1000 ch and adjusted so  
490 that  $\gamma$ -rays up to 2000 keV can be measured.

491 There is the following relationship between the  $\gamma$ -ray en-  
492 ergy ( $E$ ) and the peak center channel ( $p$ ):

$$493 \quad E = a + b \times p$$

494 In the above equation, "a" is desirable to be as close to 0  
495 as possible, and "b" is to be as close to 2.0 as possible, con-  
496 sidering the number of channels.

##### 497 4.2.3.2. Efficiency calibration

498 Because counting efficiency differs depending on the en-  
499 ergy of  $\gamma$ -rays, the efficiency calibration ( $\varepsilon$ ) is obtained as the  
500 function of  $\gamma$ -ray energy ( $E$ ) by using an efficiency calibration  
501 source composed of some nuclides with known amounts.  
502 There is the following relationship in the region of several  
503 hundreds to 2000 keV.

$$504 \quad \log(\varepsilon) = a + b \times \log(E)$$

505 If there is a calibration source containing  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$   
506 for quantification, the counting efficiency for a target  $\gamma$ -ray  
507 peak can be obtained directly.

#### 508 4.2.4. Procedure

##### 509 4.2.4.1. Preparations in advance and points to be checked

###### 511 (1) Operation check of apparatus and settings

512 Apply polarity and voltage specified by a manufacturer to  
513 a photomultiplier tube. When a source is brought close to a  
514 detector, it is desirable to check with an oscilloscope that out-  
515 put waveforms from a preamplifier meet specifications.  
516 However, it is also acceptable to refer to an instruction man-  
517 ual for a model to be used. Or connect a detector to a multi-  
518 channel analyzer, and confirm that there is no noise signals  
519 which are not normally observed and that a dead time meter  
520 does not scale out. The channel width of the multichannel an-  
521 alyzer is set to about 1000 ch. The range of measurement en-  
522 ergy is to be about 100 keV to 2000 keV.

523 For energy calibration, confirm that reference  $\gamma$ -rays (for  
524 example,  $^{137}\text{Cs}$  and  $^{40}\text{K}$ ) can be detected in a channel set in  
525 advance at the time of power-on and every day. If there is a  
526 significant deviation from the set channel, adjust the gain of  
527 the amplifier.

###### 528 (2) Background measurement

529 Perform measurement about once a week without sample  
530 or with an empty container, and confirm that there is no con-  
531 tamination around a detector and a sample container. If a  
532 peak is observed in the same channel as a  $\gamma$ -ray to be quanti-  
533 fied and decontamination is impossible, the counting rate  
534 should be obtained and recorded.

##### 535 4.2.4.2. Measurement procedure

536 Since the measurement procedure is basically the same as  
537 the method using a Ge detector, follow the operation of a  $\gamma$ -  
538 ray spectrometer using a Ge detector.

539 (1) Setting of measurement time: Determine the meas-  
540 urement time according to a target detection limit and the  
541 amount of a sample. To lower the detection limit, the reduc-  
542 tion of a background is most effective.

543 (2) Start and end procedures of measurement, and record  
544 of the times.

545 (3) Store of spectral data: File names should be such that  
546 samples and measured dates can be identified.

##### 547 4.2.4.3. Analysis procedure

548 Since the analysis procedure is basically the same as the  
549 method using a Ge detector, follow the operation of a  $\gamma$ -ray  
550 spectrometer using a Ge detector. Points to be noted in this  
551 analysis are as follows:

552 (1) Setting of ROI

553 Set the ROI where a significant count against a background  
554 is obtained as the  $\gamma$ -ray peak used for quantification. If the  
555 variation is large because of an insufficient count and the set-  
556 ting the region is difficult, set the ROI after leveling the count  
557 of each channel by smoothing process.

558 (2) Identification of nuclide

559 Prepare to convert data by an energy calibration curve in  
560 order to determine which channel corresponds to the  $\gamma$ -ray of  
561 a nuclide to be quantified. Prepare nuclear data books and  
562 environmental radiation spectra, and if an unknown peak is  
563 detected, investigate the  $\gamma$ -ray energy, identify the nuclide,  
564 and examine possibility of interference of the  $\gamma$ -ray used for  
565 quantification.

566 (3) Calculation of peak area

567 Subtract the count of a background under a peak from the  
568 total count of the peak region. If the peak is too multiple to  
569 quantify by the method, the peak area is calculated after peak  
570 separation using a peak function fitting method.

571 (4) Calculation of radioactivity

572 In the calculation of radioactivity it is necessary to estimate  
573 the calculation result of radioactivity considering natural ra-  
574 dioactive isotopes contained in a sample and a background.  
575 In this case, a net count ( $n$ ) is obtained by subtracting a count  
576 in the region corresponding to the  $\gamma$ -ray peak of a nuclide  
577 used for quantification.

578 The radioactivity  $A$  (Bq) and the radioactive concentration  
579  $C$  (Bq/kg) of a sample are obtained from the count rate ( $n$ ),  
580 which is obtained by dividing the net count by the measure-  
581 ment time, by the following formulae.

$$582 \quad A = \frac{n}{\epsilon \epsilon f_{\text{SUM}}} \quad C = \frac{A}{M}$$

583 In the above formula, the abbreviations are as follows.

584  $n$ : Count rate

585  $a$ :  $\gamma$ -Ray emission rate (Bq<sup>-1</sup>) shown in Table 2

586  $\epsilon$ : Peak efficiency

587  $f_{\text{SUM}}$ : Correction factor for summing effect. The summing  
588 effect must be corrected for <sup>134</sup>Cs, but if correction is  
589 not performed, state that.

590  $M$ : Mass (kg) of a sample in a sample container

591 (5) Calculation of detection limit value

592 The detection limit value is calculated on the assumption  
593 that the  $\gamma$ -ray of the target nuclide exists in the channel of a  
594 background spectrum. The 3 folds value of the count error in  
595 background count of the peak region is expressed as the de-  
596 tection limit. The detection limit of commercially available

597 spectrometers with a shield body is about 30 Bq/kg for <sup>131</sup>I  
598 and <sup>137</sup>Cs, but differs largely depending on the detector size,  
599 shield thickness and sample volume.

600 The peak detection limit value in an actual sample depends  
601 largely on the spectrum of the sample. When other nuclides  
602 coexist in the sample, their Compton background may also  
603 affect the detection limit value to result in being large in some  
604 cases.

605 (6) Examination and evaluation of measurement results

606 Summarize measurement results for each target nuclide,  
607 and confirm that it is normal based on statistical uncertainty  
608 of count, peak center channel, FWHM, etc. If there is any  
609 doubt in the confirmation of results, remeasure as necessary.

610 **4.2.5. Points to note for measurement**

611 **4.2.5.1. Effect of temperature**

612 In the case of a NaI (TI) spectrometer, the peak center  
613 channel may vary depending on a temperature around a de-  
614 tector. In particular, keep room temperature constant because  
615 room temperature tends to vary at night and in winter. If a  
616 sample is stored at low temperature, return it to room temper-  
617 ature before measurement.

618 **4.2.5.2. Control of background**

619 Apply 4.1.5.1.

620 **4.2.5.3. Contamination prevention of apparatus**

621 Cover a detector with polyethylene bag to prevent from  
622 contamination. If contamination should occur, cope by re-  
623 placing the polyethylene packaging. If the surface of a detec-  
624 tor is contaminated, wipe it off with a neutral detergent or  
625 gauze soaked in ethanol. Be careful not to let contaminants  
626 such as dust enter when opening a shield body.

627 Use a sample container after simple cleaning, if necessary.  
628 A polyethylene bag can be used in a container. When a sam-  
629 ple solution is placed into a sample container, do not contam-  
630 inate the surroundings of the sample container.

631 **5. Report and record**

632 Examples of items to be described are as follows.

- 633 ① Information concerning apparatus used: Apparatus  
634 name (detector size, resolution), number of measure-  
635 ment channels, analysis software type, processing  
636 method
- 637 ② Sample information: Sample name (number), collection  
638 site, collection date and time, collection volume, type of  
639 collection container, name of person in charge of collec-  
640 tion
- 641 ③ Measurement conditions: Type of sample container,  
642 sample amount, geometry
- 643 ④ Measurement records: Start date and time of measure-  
644 ment, measurement time (Live Time, Real Time)
- 645 ⑤ Analysis records: Peak center channel, FWHM, peak  
646 area and its statistical uncertainty of count, sample count  
647 rate and its statistical uncertainty of count, BG count rate  
648 and its statistical uncertainty of count, peak efficiency,

649 attenuation correction coefficient, radioactivity and ra-  
650 dioactive concentration and their statistical uncertainties  
651 of each count, radioactivity of detection limit or radio-  
652 active concentration of detection limit, name of person  
653 in charge of measurement/analysis.

654 For analysis records, the report of analysis software  
655 used can be used as it is to avoid transcription mistakes.  
656 For numerical values, the number of significant digits of  
657 radioactivity or radioactive concentration is “reduced”  
658 based on the number of significant digits of the statisti-  
659 cal uncertainty of count.

660 ⑥ Measurement result: Name of nuclide, radioactive con-  
661 centration (Bq/kg), detection limit value

662 When measurement work is entrusted, in principle,  
663 write and report the measurement results in a format  
664 specified by the measurement work consignor, and store  
665 it together with the original data.

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