방사선방어학회지
제14권 제1호 1989

■ 特別寄稿 ■

Recent State of Monitoring Techniques for Environmental Radiation and Radioactivity in Japan

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1. Introduction

For the development and the use of the nuclear power, the first priority is safety. Among various aspect of sustaining the safety of the nuclear systems, the monitoring of the environmental radiation and radioactivity is one of the essential requirements. This plays a role of an antenna to catch the abnormal operation of the nuclear systems, and also always acts a sheriff to protect the environmental conditions and human health from unexpected radioactivity contamination and/or radiation exposure. The confident systems and their pertinent operation for environmental radiation/radioactivity monitoring are indeed most important to keep the public acceptance for the development of nuclear power. The monitoring of the radioactivity contamination due to the accidents in the overseas or the nuclear explosion testings are also important. Especially after the Chernovyl, the importance of such monitoring was much more realized and emphasized.

At the present stage, in Japan 36 huge nuclear power stations are in operation, and nearly 30 percent of energy was supplied from this geneous superstar. Big facilities for fuel reprocessing are also under construction. In such circumstances, the Japanese government has paid great attention to establish such monitoring systems for past three decades since the environmental radioactivity pollution due to the nuclear explosion test at Bikini atoll on lst March, 1954.

Hereafter are introduced briefly some aspect of Japanese systems of environmental radiation and/or radioactivity monitoring and surveilance.

2. Administrative system for monitoring environmental radiation and radioactivity monitoring in Japan

In Japan the monitor of radiation and/or radioactivity in environment is all conducted by the Science and Technology Agency(STA), and the routine monitorings are carried out mostly by

1) Japan Chemical Analysis Center(JCAC)
2) National Institute of Radiological Sciences(NIRS) and
3) 13 prefectural institutes of regional bodies, surrounding of nuclear power facilities.

Among them, JCAC was established on 1974, and specially designated by the government to perform environmental radioactivity analysis for monitoring the possible contaminations originated from the nuclear facilities and radioactive fallout related to nuclear explosion testings. Occasional monitoring of sea water in the port of call of USA's nuclear-powered warships

is one of the missions. Now the activity has extended to the cross-check programme among prefectural institute to keep the consistency and to improve the reliability of the monitoring results for environmental radioactivity and radiation dosimetry.

The ecology related to the environmental radioactivity especially of marine biota are studying in NIRS as a part of the extensive research programmes on radiology, and routine surveilance of environmental radiation and radioactivity was carried out.

Some of prefectural institutes are very active in this field, and have very good facilities and experiences. and play important roles to get so called public acceptance from the inhabitants. Because such regional institute always perform their tasks from the standpoints of the inhabitants. Among them the activities and achievements of the Fukui Prefectural Institute of Public Health and the Ibaraki Prefectural Institute should be emphasized.

The Japan Atomic Energy Research Institute (JAERI) and the Power Reactor and Nuclear Fuel Development Cooperation(PNC) are the big two among the research organizations in the field of nuclear engineering in Japan. They always monitor the radioactivities in the effluents, air and environmental samples and the exposures in the site and the neibouring places. These situations are all the same in the other nuclear power stations and nuclear facilities.

For such measurements, The Science and Technology Agency(STA) established a series of deatailed standard procedures and guidances to make reliable monitorings. These are sometimes called as the STA manuals. So far 16 guidances have been established,and published in the form of booklets as shown in Table 1.

In Japan, all the informations for the nuclear safety should open to the public. If any irregular or abnormal results were found, this must be informed to STA, and anounced to the public.

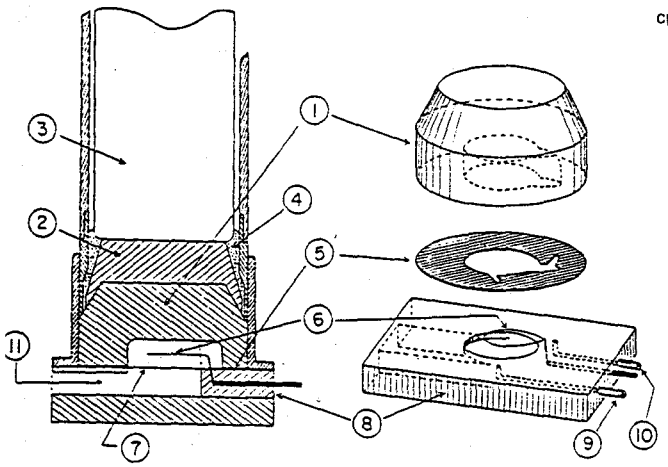
Table 1. List of standard procedures for environmental radioactivity monitoring established by STA

1. Measurements of radioactivity based on the total β-counting
2. Analysis method of radioactive strontium
3. Analysis method of radioactive cesium
4. Analytical method of radioactive iodine
5. Analytical method of radioactive cobalt
6. Instrumental radioactivity analysis procedures with NaI(Tl) scintillation spectrometers
7. Instrumental radioactive analysis procedures with germanium semiconductor detectors
8. Analytical method of radiactive zirconium
9. Analytical method of tritium
10. Analytical method of Radioactive Ruthenium
11. Analytical method of radioactive cerium
12. Analytical method of Plutonium
13. Pre-treatment of samples for instrumental analysis with Ge semiconductor detecors
14. Analytical method of Uranium
15. Measurement of radioactive iodine in emergency
16. Sampling techniques of environmental substances
17. Environmental γ-ray measurements with continuously recording monitors
18. Environmental γ-ray measurements with thermo-luminescence dosimeters

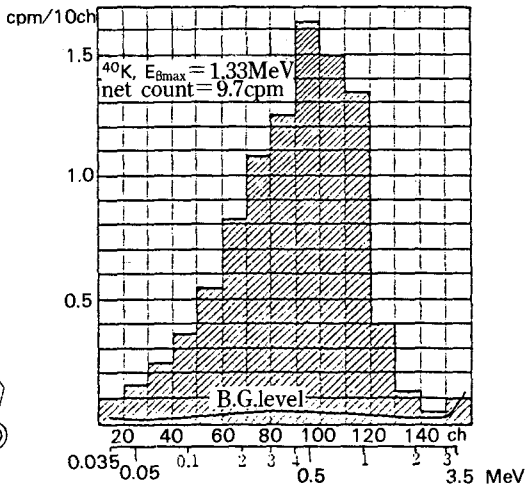
3. Techniques for Radioactivity Monitoring

3.1 Chemical analysis

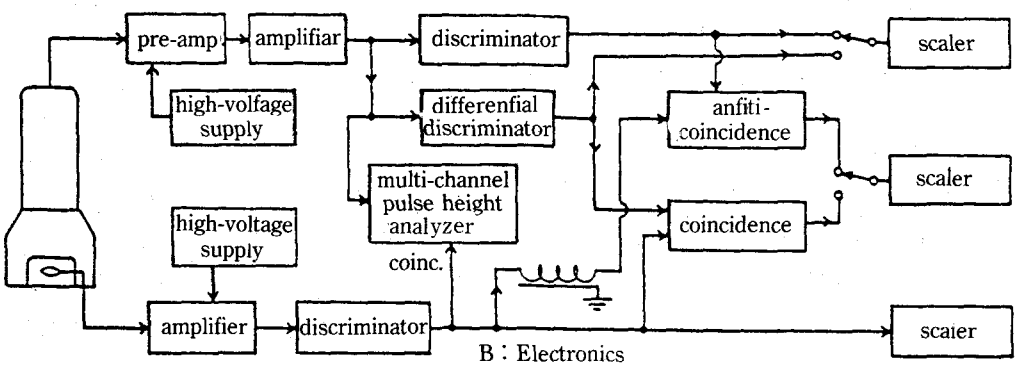
In the early stage of radioactitvity assay in environmental samples, chemical analisis was the leading player. Because in that time only a low background counting system with GM or gas flow counter tube and/or a scintillation counter were available to use, and the exact chemical sepration of each nuclides from the bulk samples were most essential. This was indeed tedious and labour-and time-consuming. This is true even now in the case of measurements of some pure β–emitters such as ^{90}Sr-^{90}Y and of α-emitters. Most of the STA series describe the radioachemical analysis methods of some important nuclides such as ^{90}Sr-^{90}Y, ^{137}Cs, ^{131}I, ^{60}Co, ^{95}Z, ^{106}Ru($+^{103}$Ru), ^{144}Ce($+^{141}$Ce), ^{239}Pu+^{240}Pu, U and ^{3}H from various kinds of balk environmental samples. A low background β-ray spectrometer with coincidence type was developed in Japan in the early stage of 1960's resulting much improvement of the techniques for weak β-source. By the use of such spectrometer, the analysis and identification of the nuclides bacame much easier, and the sensitivity was considerably improved. The complete set of such counting system are produced in the Fuji Electric Co. As the name of "pico bata". Fig. 1 is the basic diagram and some example of the β-spectrum for 1 gram of ash of a cigarette.



A : Detector part



C : Example of measured spectrum (ash of 3cm cigarette)



B : Electronics

Fig. 1. Schematic diagrame of a coincidence type low background β-ray spectrometer (pico beta) and an example of measured spectrum.

3.2 Photon spectrometry with semiconductor detectors

Now the chemical analysis has retired from the leading players in the stage of the environmental radioactivity measurements, and modern γ-and α-ray spectrometer with semicondutor detectors have been increasingly employed for quantitative analysis of environmental samples such as rain water, vegitables marine products and the like without tedious chemical separation.

For γ-ray analysis, semiconductor detectors made of highly pure germanium, Ge(Hp), are favourably employed. Nowadays wide varieties of Ge(Hp) photon detectors with defferent shapes and effective volumes of detector element and with various configulations of cryostat are available from some manufacturers as a commercial basis. As example of such a lineup is shown in Table 2.

Table 2. Typical specifications of commercially available germanium detectors.

Material	Geometry	Window Thickness	Energy Range	Efficiency (Size)	Energy Resolution	P/C
p-type HpGe	Closed-End C.	600μm	40keV-10MeV	10-40%	1.75-2.2keV	37-60
n-type HpGe	Closed-End C.	0.3μm	3keV-10MeV	10-30%	1.8-2.2KeV	36-50
p-type HpGe	Well	0.3μm	10keV-10MeV	70-120cm^3	2.1-2.4keV	
p-type HpGe	Planar	0.3μm	3keV-0.6MeV	6-36mm dia.		

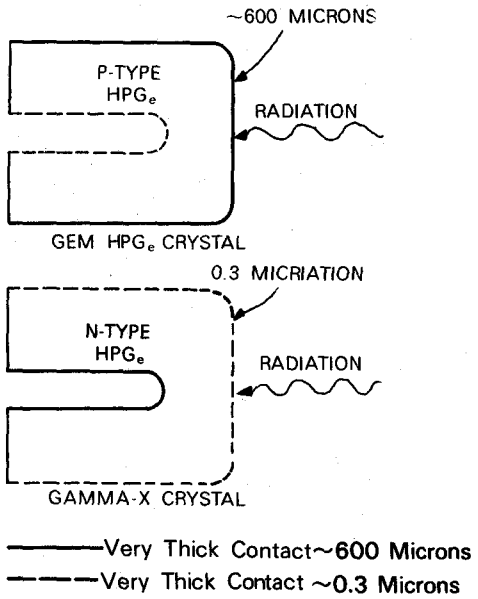


Fig. 2. Two different configurations of coaxial germanium detectors. From ref.(1).

As for the detector crystal configuration, a closed end coaxial type and a planar type are in popular use. For some of special purposes, a well type detector is also available from some of manufactuers. Among them, a closed end coaxial type is preferably used for the radioactivity measurements of environmental samples. The detector crystal can be fabricated from either n- or p-type high pure germanium. In a case of p-type material, the entrance dead layer can be made thin enough(less than 1μm) with the ion-implantation technique as illustrated in Fig. 2, and this type of detectors can be used for the analysis of low energy photons down to 3keV as well as of high energy photons, whereas the use of the p-type coaxial detectors is limited in the energy region above several ten keV due to a thick dead layer of up to 600μm. However, kin practice, both types of detectors can be employed for most of environmental radioactivity measurements, since the photon energies emitted from the important nuclides range mostly from several ten keV to a few MeV.

Better energy resolutions and higher peak-to-Compton ratios are profitable to separate and to ana-

lyse small peaks from continuum background. However,considering the balance to the economical situation, detectors with the relative efficiencies(250cm efficiencies) of 20~30% (75~130cm^3) and with the energy resolutions of 1.8~2.0keV are considered to be a practical choice.

Among several variations about the cryostat configulations, the vertical type and the J type are considered to be suitable for the measurement of environmental samples from the requirement for setting of liquid or powder samples and for the use of the Marinelli beakers.

For low level radioactivity measurements, shielding of detector from external background radiations is an essential requirement. Practicall 10cm thick lead is necessary to reduce the background to an acceptable level. As the material, use of lead is preferable to steel, since materials of lower atomic number may deteriorate the pulse height spectrum due to their more dominant backscattering property.

A small amount of ^{210}Pb(RaD) is inevitably contained in lead. The daughter nuclide ^{210}Bi(RaE) of this isotope emits energetic β-rays($E\beta max = 1.17$ MeV) resulting Bremsstrahlung which maycontribute to the increase of the background, and hence material with less ^{210}Pb should be chosen.

In order to eliminate the effect of characteristic x-rays from lead, it is desirable to line the inner surface with cadmium and copper plates. Larger inner volume is preferable to reduce the scatter effect. It is recommended to ensure an inner dimentions of 30×30×30×cm^3 at the minumum.

Figure 3 shows an example of background spectrum obtained with a 20% efficiency detector installed

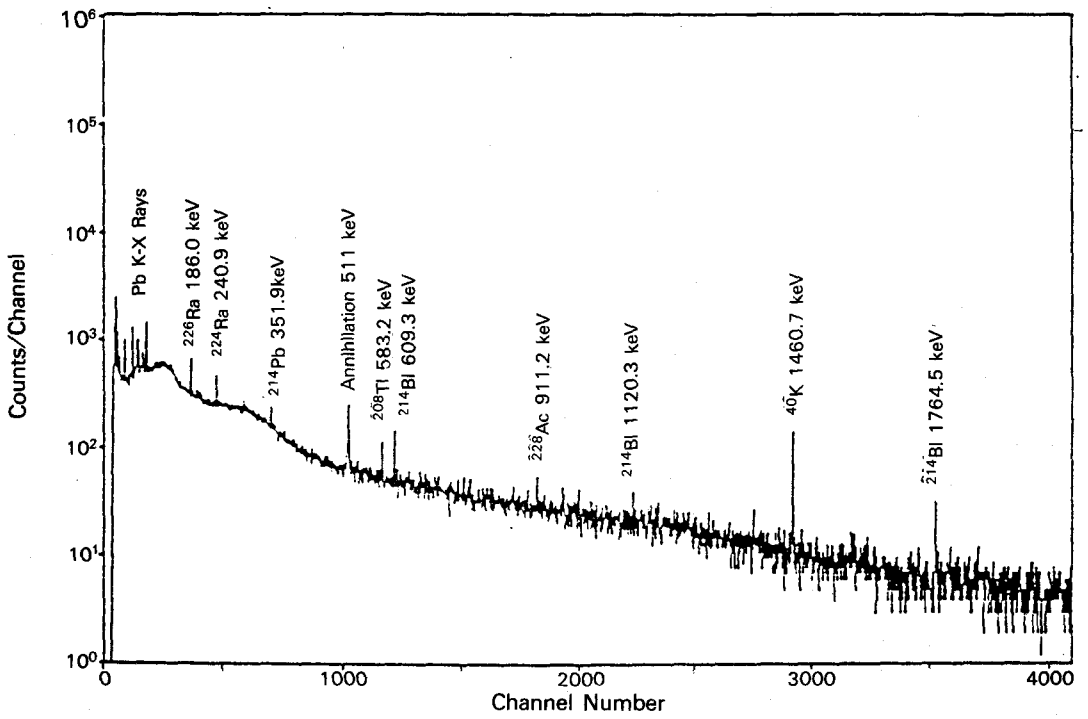


Fig. 3. An example of background spectrum. Detector : 20% efficiency closed end coaxial type. Shield : 10cm lead, 1mm Cadmium, 1cm steel. Measuring time : 100,000s. Channel width : 0.5 keV/ch.

in a shielding box of 10cm lead, 1mm cadmium and 1cm steel.

The multichannel pulse-height analyzer(PHA) essentially important tool in the quantative nuclear spectrometry. As a result of the recent development and mass production of excellent electronic devices such as integrated circuits, memories, microprocessors etc., the performance such as the memory capacity and the utility of PHAs have been improved remarkably with the cost remaining constant or decreasing. Nowadays, PHAs with a memouy capacity of 4K, 8K or more become quite popular, and use of 8K channel analyzers is minimum requirement for routine environmental radioactivity measurements.

In the analysis of the specturm, the areas of the full energy peaks and the peak positions are major concerns. Owing to the superior energy resolution of Ge detectors, the peak region spreads over only in a narrow part of the specturm, and is mostly separated from other peaks. This permits a simple way for peak area analisis. Usually a simple subtraction of the base line continuum from the total counts involving in the peak region which is usually defined as (2.5~3.0) times of FWHM. Several methods have been demonstrated to determine the contribution of the base line continuum. Among them most simple and practical one is a linear interplation refering the continuum parts at nerbouring channels of both sides of the peak as shown in Fig. 4. in the case of separated independent peaks, the final results are not dependent seriously on the methods. However, if two peaks are overlapped, more complicated analysis such as the least-squares fitting is required. In the STA manual, several methods to meet such problems are shown.

(Use of computers)

The peak location is made by the method of the first derivative. These peak-area determinations and peak-position locatings are not complicated in procedures, and can be done manually if numbers of peaks

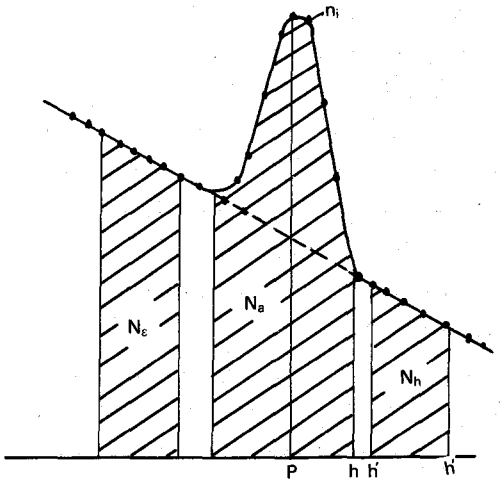


Fig. 4. Subtraction of base line continuum by linear interpolation.

of interest are not many. However, usually many peaks exsist in the obserbed spectrum of environmental samples, and the spectrum analysis over the entire region are very time- and labour-consuming. This situation has led to the introduction of computers in the interpretation and evaluation of vast spectrul data.

In most of software commercially available in Japan, the methods of spectrum analysis are mostly consistent with the guidance established by STA, and a library on the decay data such as the energies and the intensities of each γ-rays and the half lives, multiplicity etc. are also provided. The automatic identification of the nuclide and the calculation of the radioactivity at the reference date etc. are carried out. The advent of modern powerful microcomputers makes such computerized analysis easy.

There are many choices of the micro-computers. Among them NEC(Nippon Electric Co.)PC9800 and DEC PC350 are very popular in Japan. The softwares also available on a commercial basis from several company.

(Calibration)

The spectrometry is the relative method, and the instruments should be calibrated in order to get the radioactivity. Since the efficiency of a Ge detector is a smoothly varying function with γ-ray energy, the calibration is made at some energy points with appropriate energy intervals. For standard sources, ^{109}Cd(88 keV), ^{57}Co(122keV), ^{51}Cr(320ieV), ^{85}Sr(512keV), ^{134}Cs(606, 796keV), ^{137}Cs(662keV), ^{60}Co(1173, 1332 keV) and ^{88}Y(898, 1836keV) are preferably used, since these nuclides have simple decay schemes, proper energies and acceptably long half lives. In the calibration, the coincidence sum effects should be corrdcted for ^{134}Cs, ^{60}Co and ^{88}Y, since these nuclides emit plural γ-rays in cascade, and the γ-rays are not registered in the full energy peak when two or more γ-rays interact simultaneously in the detector.

A known amount of mixture of these standard solution is spiked and mixed to the matrix materials such as water, agar, aluminum oxide and manganese dioxide in sample containers to simulate the souce conditions. In the preparation of such standard sample, the amount of radioactivity should be minimized to order of nCi(37 Bq) to avoid possible contamination of thr detector. Such voluminous standard samples can be obtainable from some of isotope suppliers such as the Amerciam International(UK). However, the variety of the size of commercially available standard sources is very limited. In Japan, the Japan Chemical Analysis Center distributes such kinds of voluminous standard samples suitable for environmental radioactivity measurements by the requests of some specified users such as regional centers for environmental radioactivity surveilance. Normalization of the size and configuration of sample containers is also desirable from the standpoint of the effective use of the standard samples.

Multi-gamma emitting nuclides such as ^{154}Eu can be used as the standards for the calibration of γ-ray spectrometers when the source-detector distance is large. However, the use of such multi-gamma sources is not recommended because of the possible large sum effects when the souce is located closed to the detector.

The graphic representation of the efficiency curves are made mostly on log-log section papers. In this way the efficiency curve can be approximated to a linear line in the region above 100keV.

(Sample preparation)

In order to keep a maximum sensitivity in the γ-ray spectrometry for low level radioactivity concentration measurements, preparation of the measured samples are important.

First, much sample mass is advantageous, since total activity is proportional to the mass of original sample. Second, the sample volume should be minimized, since the efficiency decreases rapidly with increasing the distance from the detector. To satisfy these two inconsistent conditios, one can adopt the following treatments :

(1) Drying of aqueous solutions,
(2) Drying and powdering(leaves, vegitables),
(3) Precipitation of all or particular elements containing in aqeous solutions,
(4) Adosorption of particular elements containing in an aqueous solution to adsorbents or ion exchage columns, and
(5) Ashing(marine biota, fishes, animals)

Use of Marinelli beakers(re-entrant type beakers) is advantageous for the measurements of large amount of samples with keeping a high sensitivity.

In environmental samples, a considerable amount of naturally origined radioactivities such as ^{40}K are sometimes contained, disturbing high-sensitive radioactivity measurements. Removal of such ele-

ments by some apporopriate chemical procedures can improve the sensitivity.

To keep the measuring condition constant and to avoid the possible contamination, source containers should be used. In Japan, several kinds of commercially available small plastic containers with volumes of 100-300cm^3 are often used for routine environmental radioactivity measurements. For large volume samples, 1 liter marinelli beakers are employed.

(Detection limit in the γ-ray spectrometry)

Detection limit of radioactivity measurements depends on the efficiency and resolution of the detectors, the shielding conditions, the volume and geometrical configuration of samples, the propertics of the nuclides etc. A rough estimation of the limit of detection in term of the radioactivity concentration is 5 pCi/kg for ^{137}Cs in the case of the use of a 20% detectorand 100g sample, and 1 day measuring time.

3.3 α-ray spectrometry with surface barrier Si detector

Alpha ray spectrometry is a useful mean fo quantitative radioactivity measurements of α-emitters such as transuranium elements. As the α-ray detector silicon surface barrier detectors(SSB) are mostly used. The specifications for commercially available SSBs extend from 7mm^2 to 2,000mm^2 in effective area and from 13keV to 50keV in energy resolution(for 5.477 MeV α-rays) and from 100μm to 2,000μm in deplation depth. Since the energy of α-rays from isotopes is 8 MeV at the maximum, detectors with a 100μm deplation layer is suitable. A larger effective area is desirable to use for effective measurements. However, it is noted that larger detectors tends to have poorer energy resolutions. Considering these situations, the use of detectors with 450mm^2 area, 100μm depth and 20 keV resolution is practical choice for α-ray spectrometry for environmental radioactivity measurements.

This type of detector is mostly operated at room temperature, but a vacuum chamber is necessary to avoid the energy loss in air between the source and the detector. For evacuating the chamber, a simple rotary pump can be used.

The energy of α-rays emitted from radioisotopes ranges in a limited energy region from 3.5MeV to 8 MeV, If the pulse-height spectrum of this region was enlarged by the use of the digital offset in PHA or by a biased amplifier, the memory capacity of PHAs can be saved remarkbly, and simultaneous data acquisition from several differnent detectors can be established with a single 4K or 8K analyzer by the aid of a multiplexer.

The range of α-rays in the materials is very short, i.e. the maximum flight path of 5.3MeV α-rays in air is only 3.8cm. This means requests the critical sample preparation to get very thin sources. The quality of the observed spectrum strongly depends on the source conditions. This is quite different aspect of α-ray spectrometry compared with the γ-ray spectrometry. It is difficult to obtain line spectra for thick or bulky samples, and the electrical deposition of the trace amount of the element after chemical extractions are inevitably required. In the STA series, the detailed procedures are shown for uranium and plutonium analysis. Shielding from external radiations is not necessary in the case of α-ray spectrometry.

The counting efficiency of SSBs for α-rays are mostly determined by the source-to-detector geometry, and practically independent on the α-ray energy. So, calibrations performed by a particular nuclide such as ^{241}Am can be adopted for other nuclides.

In the case of a bulky source, the obserbed spectra become stepwise rather than lines. Such spectra also can be used for the rapid assay of radioactivity concentration of α-emitters.

4. Monitoring of external radiation

Monitoring of external radiations in the external field is also important to keep safety the public health from unexpected and undesirable exposure.

The STA also established some guidances to observe the external radiations in the neibourhoods of nuclear power stations/facilities.

The exposure rate is continuously recorded with (1)a pressurized ionization chamber, (2)a discrimination-bias-modulation(DMB) type NaI(Tl) scintillation counter and/or (3)a special NaI(Tl) scintillation counter with special cover. The techniques of DMB and special covering were developed to get a flat energy response in terms of exposure.

The long term integral dose is obserbed with TLDs. Usually these dosimeters are installed in a small cabin. Many such monitoring cabins are located in the sarrounding places of nuclear power stations. Some of the observed data are transmitted by telemeter.

References

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