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著者	Nakanishi Takashi, Hosotani Risa, Komura Kazuhisa, Muroyama Toshiharu, Kofuji Hisaki, Murata Yoshimasa, Kimura Shinzo, Sahoo Sarata Kumar, Yonehara Hidenori, Watanabe Yoshito, Bannai Tadaaki
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Residual neutron-induced radionuclides in a soil sample collected in the vicinity of the criticality accident site in Tokai-mura, Japan: A progress report

Takashi NAKANISHI, Risa HOSOTANI

Department of Chemistry, Faculty of Science, Kanazawa University, Kakuma-machi, Kanazawa, Ishikawa 920-1192, Japan

Kazuhisa KOMURA, Toshiharu MUROYAMA, Hisaki KOFUJI, Yoshimasa MURATA

Low-Level Radioactivity Laboratory, Kanazawa University, Tatsunokuchi-machi, Ishikawa 923-1224, Japan

Shinzo KIMURA, Sarata Kumar SAHOO, Hidenori YONEHARA, Yoshito WATANABE, Tada-aki BAN-NAI

Division of Human Radiation Environment, National Institute of Radiological Sciences, 9-1, Anagawa 4-chome, Inage-ku, Chiba, Chiba 263-8555, Japan

Author for correspondence: Prof. Dr. Takashi NAKANISHI
Department of Chemistry, Faculty of Science, Kanazawa University, Kakuma-machi, Kanazawa, Ishikawa 920-1192, Japan
Tel.: +81-76-264-5690, Fax: +81-76-264-5742

E-mail: nakanisi@cacheibm.s.kanazawa-u.ac.jp

Abstract

Residual neutron-induced radionuclides were measured in a soil sample collected in the vicinity of the location where a criticality accident occurred (in Tokai-mura, from 30 September to 1 October 1999). Concentrations of ^{24}Na, ^{140}La, ^{122}Sb, ^{59}Fe, ^{124}Sb, ^{46}Sc, ^{65}Zn, ^{134}Cs and ^{60}Co in the soil sample were determined by γ-ray spectrometry, and neutron activation analysis was carried out for selected target elements in the sample. Tentative estimates of the apparent thermal and epithermal neutron fluences which reached to the sample were obtained through combined analysis of ^{59}Fe/^{58}Fe, ^{124}Sb/^{123}Sb, ^{46}Sc/^{45}Sc, ^{65}Zn/^{64}Zn, ^{134}Cs/^{133}Cs and ^{60}Co/^{59}Co.

Keywords: JCO criticality Accident; Tokai-mura;
Residual neutron-induced radionuclides;
Neutron fluence

1. Introduction

Residual neutron-induced radionuclides such as ^{152}Eu, ^{154}Eu and ^{60}Co in materials exposed to atomic bomb radiation at Hiroshima and Nagasaki have been measured at Radiochemistry Laboratory of Kanazawa University for the past 20 years (Nakanishi, Imura, Komura & Sakanoue, 1983; Nakanishi, Kobayashi, Yamamoto & Miyaji, 1987; Nakanishi, Ohtani, Mizuochi, Miyaji & Yamamoto, 1991; Nakanishi, Miwa & Ohki, 1998) partly in a US-Japan joint reassessment project for atomic bomb radiation dosimetry. The joint project has contributed greatly to improvement of the health care for the survivors in Hiroshima and Nagasaki as well as to the understanding of the biological effects of ionizing radiation on mankind, but the accuracy and precision of the data obtained on neutron fluences are too poor to permit a reliable assessment of dose-effect relationship. This is due to the fact that the radioactivity of the neutron-induced had reached an extremely low-level when the problem on the biological effects of neutrons was recognized. Hence, studies related to neutron dosimetry after the criticality accident occurred at Tokai-mura seemed to be important for a better understanding of the biological effects of neutrons on mankind. Furthermore, neutron dosimetry by the methodology developed in the US-Japan joint project was expected to contribute to the health care of the workers at the plant and the public around the plant who were exposed to neutrons.

In this work, soil samples were systematically collected in the grounds of the plant where the criticality accident occurred. The purpose of the sampling was to obtain data for reliable neutron dosimetry, *i.e.*, to determine residual neutron-induced radionuclides. To verify the validity of the neutron dosimetry in

Tokai-mura, the results of these measurements of residual neutron-induced radionuclides must be compared with the results based on an appropriate model and on computation; only a tentative result for the neutron-induced radionuclides in a selected soil sample which proved highly activated is reported here.

2. Experimental

On 7 October, 1999, surface soil samples were systematically collected at various locations around the site where the criticality accident occurred. The sampling was carried out using a corer (5 cm ϕ × 5 cm h). At each location three cores were collected for cross check measurements, and each core was sealed individually in a polyethylene bag. A series of core sample was then brought to the Low-Level Radioactivity Laboratory of Kanazawa University during the day of sampling and a screening measurement was carried out. As a result of the screening measurement, one of the samples collected at the nearest point to the vessel in which the criticality phenomenon occurred proved to be highly activated. The sampling location was coded as S-12L (Fig. 1). The S-12L sample was then well homogenized, and a known aliquot (on a wet weight basis) was packed tightly in a plastic container (6 cm ϕ × 3 cm h) and sealed.

On 8 October, the S-12L sample was brought in its closed container to the Radiochemistry Laboratory of Kanazawa University to subject it to accurate and precise γ-ray spectrometry. The sample, weighing 114 g on wet weight basis, was measured with an ORTEC high-purity Ge detector, of 2.3 keV resolution at 1333 keV and 40 % relative efficiency, coupled to a 4K-channel pulse height analyzer. Photo-peak assignment and evaluation were performed with a computer

program "SPEC ANAL", developed at the Radiochemistry Laboratory of Kanazawa University. In addition to the peak identification by photopeak energies, a check on the half-lives of all the photopeaks observed was also carried out by saving spectral data every 2 h. Two spectra are illustrated in Fig. 2; the decay of each photopeak should be followed further.

A mock-up volume sample was prepared as follows to measure the counting efficiency for determination of residual neutron-induced radionuclides specifically in the S-12L specimen (114 g-wet): 114 g of wet soil sample collected in Kanazawa and known amounts of ^{60}Co, ^{137}Cs and ^{152}Eu standard solutions (calibrated at LMRI, France) were placed in a plastic container (6 cm ϕ × 3 cm h).

Parallel to the γ-ray spectrometry for the residual neutron-induced radionuclides, the other portion of the S-12L sample was subjected to measurement of dry/wet weight ratio and to neutron activation analysis of relevant target elements of (n, γ) reactions. For neutron activation analysis, 4 known aliquots (86〜124 mg each) of dried S-12L sample were taken and heat-sealed in pre-cleaned polyethylene vials. Known amounts (87〜144 mg each) of Geochemical Reference Rock samples such as JB-1a and JG-1a (issued by the Geological Survey of Japan) and reagent reference samples of ZnO (16.72 mg), Sb (1.05 mg) and CsCl (3.75 mg) were also vialed. For the correction of the relative neutron fluence among the sample vials during irradiation in the nuclear reactor, a weighed stainless-steel wire ring (15.3〜17.4 mg) was attached around each sample vial as a flux monitor. Neutron irradiation was carried out using the TRIGA Mark-Ⅱ nuclear reactor (100 kW) at the Atomic Energy

Research Institute of Rikkyo University. After neutron irradiation on the rotary specimen rack in the reactor (ϕ_{th} ~5×10^{11} cm^{-2} s^{-1}) for 30 min. and appropriate cooling, γ-ray spectrometry was carried out at Kanazawa University in a similar manner as above.

3. Results and discussion

The measurement of dry/wet weight ratio of the S-12L sample resulted in 71 g/114 g, and no obvious volume change was observed on drying. The concentrations of radionuclides and elements will hereafter be described on dry weight basis.

In the γ-ray spectra shown in Fig. 2, 10 neutron-induced radionuclides (*i.e.*, ^{24}Na, ^{82}Br, ^{140}La, ^{122}Sb, ^{59}Fe, ^{124}Sb, ^{46}Sc, ^{65}Zn, ^{134}Cs and ^{60}Co) and 2 fission products (*i.e.*, ^{131}I and ^{137}Cs) were identified. Iodine-131 seems to have been liberated during and after the criticality accident, though the origin of the ^{137}Cs is attributable to global fallout. Since every photopeak listed in Table 1 was well confirmed by γ-energy and/or half-life, the concentrations of the identified radionuclides in the S-12L sample at the end of criticality (EOC, *i.e.* 06:15 of 1 October 1999) could be determined using all the γ-spectral data recorded for the check of half-lives. The counting data were corrected for radioactive decay and counting efficiency and the resulting data for residual neutron-induced radionuclides (except for ^{82}Br) are given in Table 1.

The results of neutron activation analysis for the elements of interest are given in Table 2. It was proved that the elemental

composition of sample S-12L is extraordinary for a soil.

Thermal and epithermal neutron fluences were then evaluated using a data set on $^{59}Fe/^{58}Fe$, $^{124}Sb/^{123}Sb$, $^{46}Sc/^{45}Sc$, $^{65}Zn/^{64}Zn$, $^{134}Cs/^{133}Cs$ and $^{60}Co/^{59}Co$. Although the principles of the evaluation method were described elsewhere (Nakanishi *et al.*, 1987), we briefly describe it here.

The radioactivity (A/Bq) of a nuclide induced by an (n, γ) reaction in unit mass of sample at the end of irradiation, for example ^{59}Fe in 1 g of the S-12L sample at EOC, is given by the following expression:

$$A(^{59}Fe) = N(^{58}Fe) \times \{\phi_{th}\,\sigma_{th}(^{58}Fe) + \phi_{epi}\,\sigma_{epi}(^{58}Fe)\} \times \{1 - e^{-\lambda t}\} \quad \cdots(1)$$

where, N is the number of atoms of target nuclide in unit mass of sample, ϕ_{th} is the thermal neutron fluence rate in $cm^{-2}\ s^{-1}$, σ_{th} is the thermal neutron cross section for the (n, γ) reaction on the target nuclide in cm^{2}, ϕ_{epi} is the epithermal neutron fluence rate in $cm^{-2}\ s^{-1}$, σ_{epi} is the resonance integral for the (n, γ) reaction on the target nuclide, λ (= *ln*2/half-life) is the decay constant of the neutron-induced radionuclide in s^{-1}, and t is the time of neutron irradiation in s. When the half-life of neutron-induced radionuclide is more than 10 times longer than the neutron irradiation time (t), Equation (1) can be reduced to

$$A(^{59}Fe) = N(^{58}Fe) \times \lambda(^{59}Fe) \times \{\Phi_{th}\,\sigma_{th}(^{58}Fe) + \Phi_{epi}\,\sigma_{epi}(^{58}Fe)\} \cdots(2)$$

where, Φ_{th} is the thermal neutron fluence in cm^{-2}, and Φ_{epi} is the epithermal neutron fluence in cm^{-2}. By substituting the measured

values and nuclear data (Erdtmann, 1976; Firestone, Shirley, Baglin, Frank Chu & Zipkin, 1996) into Eq. (2) for $^{59}Fe/^{58}Fe$, $^{124}Sb/^{123}Sb$, $^{46}Sc/^{45}Sc$, $^{65}Zn/^{64}Zn$, $^{134}Cs/^{133}Cs$ and $^{60}Co/^{59}Co$, respectively, similar equations are obtained. By solving the simultaneous equations for Φ_{th} and Φ_{epi}, the thermal and epithermal fluences that can explain the $^{59}Fe/^{58}Fe$, $^{124}Sb/^{123}Sb$, $^{46}Sc/^{45}Sc$, $^{65}Zn/^{64}Zn$, $^{134}Cs/^{133}Cs$ and $^{60}Co/^{59}Co$ quotients simultaneously can be evaluated. The solutions for Φ_{th} and Φ_{epi} which were found to apply to S-12L are $\sim 6\times10^{10}$ cm^{-2} and $\sim 1\times10^{9}$ cm^{-2}. These estimates must be compared with model calculations for the criticality and neutron transport.

Since the criticality continued for some 20 h during which neutron output changed momentarily, we omitted the data for ^{24}Na (half-life $\sim$15 h), ^{140}La ($\sim$40 h) and ^{122}Sb ($\sim$65 h) in the above mentioned analysis of the thermal and epithermal neutron fluences; the data set of $^{24}Na/^{23}Na$, $^{140}La/^{139}La$ and $^{122}Sb/^{121}Sb$ will be used for an analysis of the temporal change of neutron output.

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References

Erdtmann, G. (1976). *Neutron Activation Tables.* Verlag Chemie, Weinheim.

Firestone, R.B., Shirley, V.S., Baglin, C.M., Frank Chu, S.Y., & Zipkin, J. (1996). *Table of Isotopes, Eighth Edition*, Vol. 1. New York: Wiley.

Nakanishi, T., Imura, T., Komura, K., & Sakanoue, M. (1983). ^{152}Eu in samples exposed to the nuclear explosions at Hiroshima and Nagasaki. *Nature, 302(5904)*, 132-134.

Nakanishi, T., Kobayashi, K., Yamamoto, T., & Miyaji, K. (1987). Residual neutron-induced radioactivities in samples exposed to the nuclear explosion in Hiroshima. US-Japan Joint Reassessment of Atomic Bomb Radiation Dosimetry in Hiroshima and Nagasaki, Final Report, Vol. 2, *Radiation Effects Research Foundation*, *Hiroshima* (pp. 310-319).

Nakanishi, T., Ohtani, H., Mizuochi, R., Miyaji, K., Yamamoto, T., Kobayashi, K., & Imanaka, T. (1991). Residual neutron-induced radionuclides in samples exposed to the nuclear explosion over Hiroshima: Comparison of the measured values with the calculated values. *Journal of Radiation Research, 32(Suppl.)*, 69-82.

Nakanishi, T., Miwa, K., & Ohki, R. (1998). Specific radioactivity of europium-152 in roof tiles exposed to atomic bomb radiation in Nagasaki. *Journal of Radiation Research, 39*, 243-250.

Figure Captions:

Fig. 1. The sampling location of the S-12L soil sample. The criticality phenomenon occurred in the "U solution".

Fig. 2. γ-ray spectra for 114 g (wet; *i.e.* 71 g on a dry weight basis) of the S-12L soil sample collected at the point shown in Fig. 1. Most of the photopeaks not specified are those from natural radionuclides.

Table 1

Residual neutron-induced radionuclides in the S-12L soil sample. The nuclides are arranged by half-life starting with the shortest

Nuclide	Half-life[a]	E_γ(keV)[a]		Bq(at EOC[b])/g-sample(dry)	
^{24}Na	14.9590 h	1369		**(5.5±1.3)**	**×10^{1}**
^{140}La	1.6781 d	*487*		*(1.0 ±0.1)*	*×10^{0}*
		816		*(1.1 ±0.3)*	*×10^{0}*
		1596		*(0.94±0.09)*	*×10^{0}*
			mean	**(1.0±0.1)**	**×10^{0}**
^{122}Sb	2.70 d	564		**(6.1±0.5)**	**×10^{-1}**
^{59}Fe	44.503 d	*1099*		*(3.3±0.3)*	*×10^{-2}* [c]
		1292		*(3.3±0.3)*	*×10^{-2}* [c]
			mean	**(3.3±0.2)**	**×10^{-2}** [c]
^{124}Sb	60.20 d	*603*		*(1.4±0.1)*	*×10^{-2}* [c]
		1691		*(1.3±0.1)*	*×10^{-2}* [c]
			mean	**(1.4±0.1)**	**×10^{-2}** [c]
^{46}Sc	83.79 d	*889*		*(5.0±0.2)*	*×10^{-2}* [c]
		1121		*(5.2±0.3)*	*×10^{-2}* [c]
			mean	**(5.1±0.3)**	**×10^{-2}** [c]
^{65}Zn	244.26 d	1116		**(1.4±0.2)**	**×10^{-2}** [c]
^{134}Cs	2.062 y	*569*		*(2.6±0.5)*	*×10^{-2}* [c]
		605		*(2.5±0.1)*	*×10^{-2}* [c]
		796		*(2.4±0.2)*	*×10^{-2}* [c]
			mean	**(2.5±0.2)**	**×10^{-2}** [c]
^{60}Co	5.2714 y	*1173*		*(2.5±0.4)*	*×10^{-3}* [c]
		1333		*(2.4±0.4)*	*×10^{-3}* [c]
			mean	**(2.4±0.3)**	**×10^{-3}** [c]

[a] Firestone *et al.* (1996).

[b] EOC: end of criticality.

[c] The data are tentative ones, because a check of the half-lives of the photopeaks must be further continued.

Table 2

Selected elements in the S-12L soil sample determined by neutron activation analysis. The elements are arranged as in Table 1

Element	g/g-sample(dry)[a]
Na	$(9.7 \pm 0.5) \times 10^{-3}$
La	$(1.07 \pm 0.04) \times 10^{-4}$
Sb	$(9.1 \pm 1.0) \times 10^{-5}$
Fe	$(6.6 \pm 0.2) \times 10^{-2}$
Sc	$(2.4 \pm 0.1) \times 10^{-5}$
Zn	$(1.75 \pm 0.06) \times 10^{-3}$
Cs	$(2.1 \pm 0.1) \times 10^{-4}$
Co	$(2.4 \pm 0.2) \times 10^{-5}$

[a] The analytical data are tentative except for those of Na and La, because a check on the half-lives of the photopeaks used in the present neutron activation analysis must be further continued.

Fig. 1.

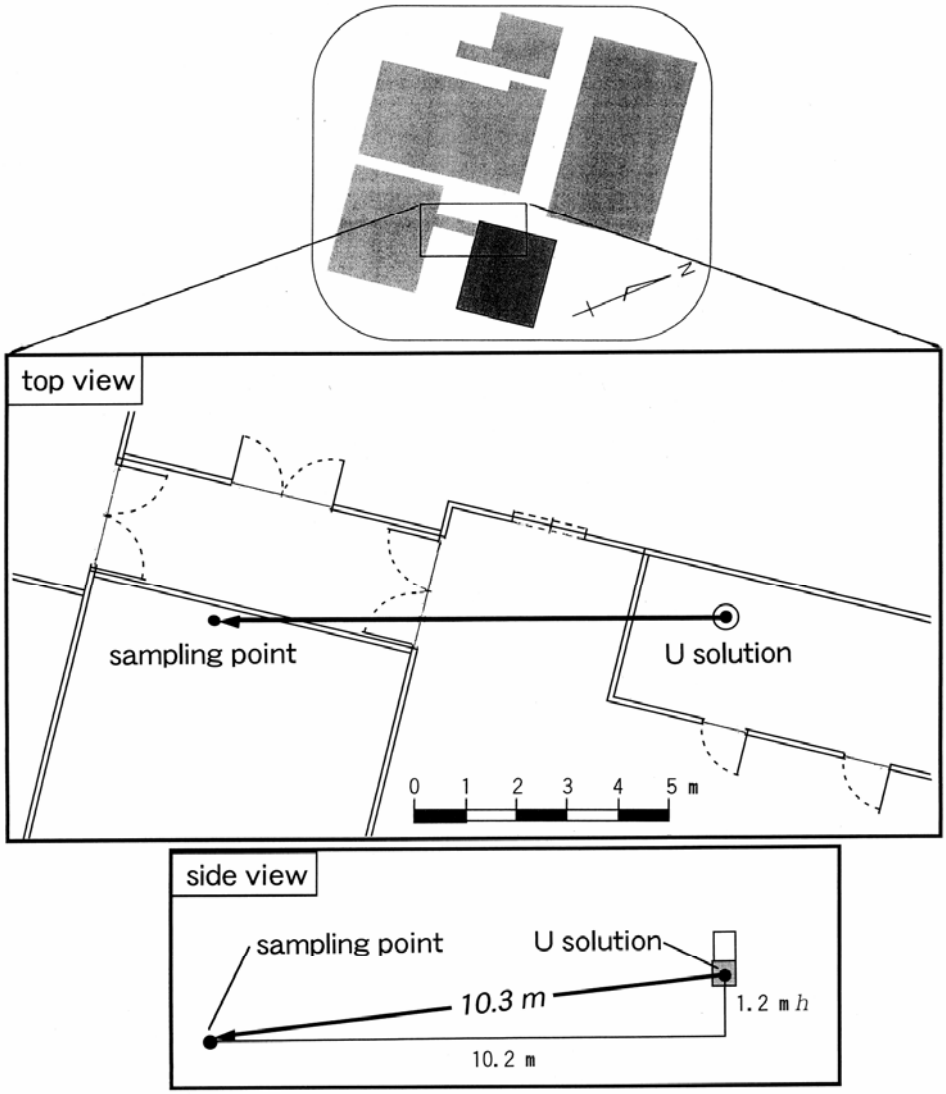
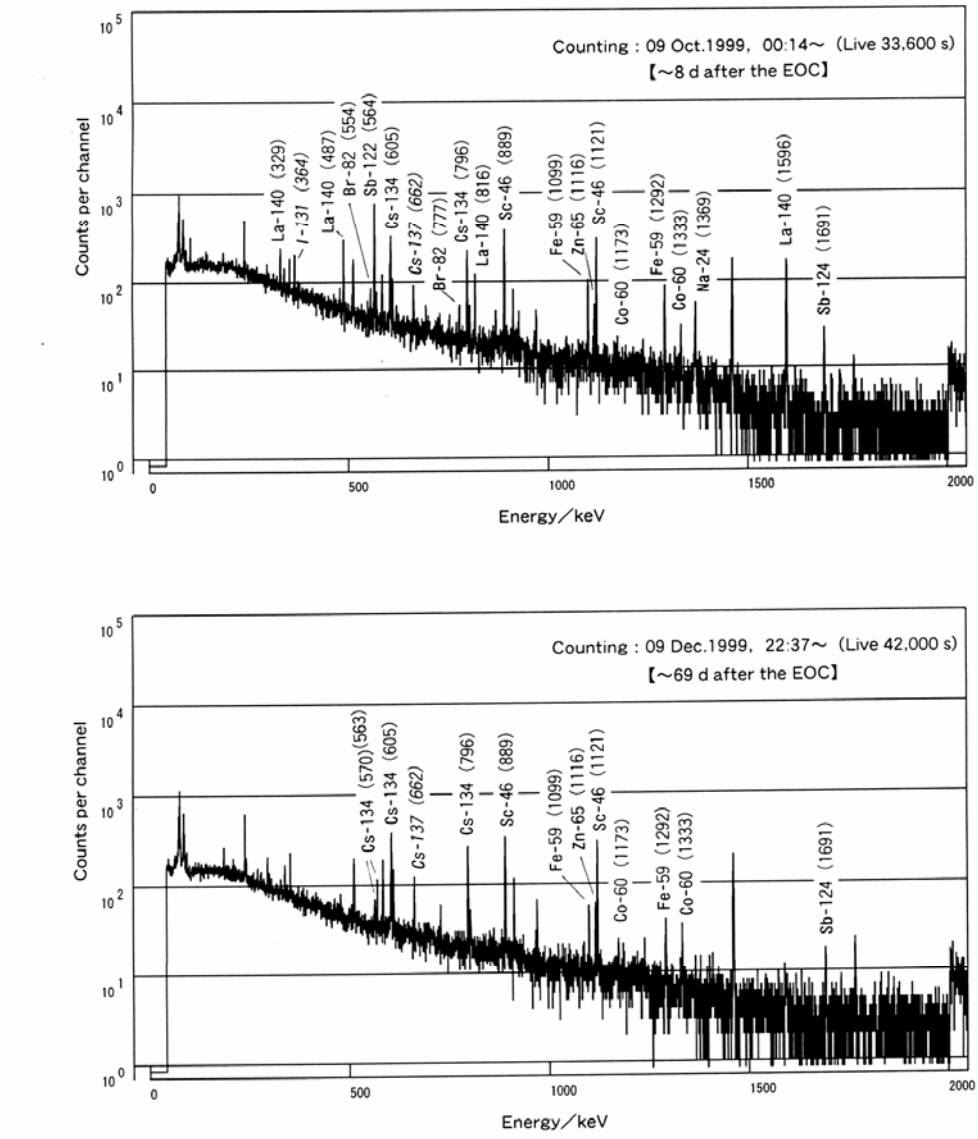

top view
sampling point
U solution
0 1 2 3 4 5 m
side view
sampling point
U solution
10.3 m
1.2 m h
10.2 m

Fig. 2.


Counting : 09 Oct.1999, 00:14～ (Live 33,600 s)
【～8 d after the EOC】
Counts per channel
Energy／keV
La-140 (329)
I-131 (364)
La-140 (487)
Br-82 (554)
Sb-122 (564)
Cs-134 (605)
Cs-137 (662)
Br-82 (777)
Cs-134 (796)
La-140 (816)
Sc-46 (889)
Fe-59 (1099)
Zn-65 (1116)
Sc-46 (1121)
Co-60 (1173)
Fe-59 (1292)
Co-60 (1333)
Na-24 (1369)
La-140 (1596)
Sb-124 (1691)
Counting : 09 Dec.1999, 22:37～ (Live 42,000 s)
【～69 d after the EOC】
Counts per channel
Energy／keV
Cs-134 (570)(563)
Cs-134 (605)
Cs-137 (662)
Cs-134 (796)
Sc-46 (889)
Fe-59 (1099)
Zn-65 (1116)
Sc-46 (1121)
Co-60 (1173)
Fe-59 (1292)
Co-60 (1333)
Sb-124 (1691)