Neutron activation analysis for soils of Hiroshima City and plaster under roof-tiles of Old Hiroshima House

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Abstract

For the early entrance survivors in Hiroshima and Nagasaki atomic bomb (A-bomb), radiation doses from activated materials induced by the A-bomb neutrons are dominant. For estimation of such doses, element concentrations of surrounding materials such as soil and rubbles are necessary. Especially Sc density in soil is important for estimating radiation doses at the time 20 – 30 days after explosion because ^{46}Sc, which has the half-life of 84 days, is considered to be the major gamma-ray emitting nuclide induced by A-bomb neutrons. However, few data of Sc density in soil are available in both of Hiroshima and Nagasaki cities. Purpose of this study is evaluation of Sc density in soil and its uneveness using neutron activation analysis.

Soil samples were taken from 10 locations within 4 km from the A-bomb hypocenter in Hiroshima City. And also, one plaster sample under roof-tiles of old Hiroshima house was taken. These samples together with a reference rock sample of JA-1 were activated in Kyoto University Reactor (KUR). Element concentrations were relatively obtained comparing counting rates of gamma-ray peaks of identified radionuclides from soil samples with those from the reference rock by Ge-detectors.

Twenty three element concentrations including Al, Mn, Na and Sc were obtained by the activation analysis. The obtained element concentrations were compared with values used in Dosimetric System 1986 (DS86) and found to be roughly the same as those in DS86. Sc density in Hiroshima soil was estimated to be 5.12 ± 0.59 (ppm). It was found the unevenness of Sc density in soils for 10 location of Hiroshima city was about 12%.

Introduction

In dosimetric system 1986 (DS86), cumulative radiation exposure due to activated soil around hypocenter of Hiroshima atomic bomb (A-bomb) had been estimated to be 0.8 Gy at maximum[1]. The DS86 was reevaluated and Dosimetric system 2002 (DS02) was establised in 2002 [2]. While the exposure from activated soil had not been updated in the DS02 [2], Imanaka et al. evaluated and updated the exposure from activated materials based on DS02 neutron fluence using thermal neutron fluence ratio (DS02/DS86) and ^{60}Co activation based on the results by

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Gritzner et al. in DS86 [3]. It was concluded that the exposure rate at 1 m height after 1 minute after from the Hiroshima bombing was estimated to be 4 Gy/h [4]. Tanaka et al. also evaluated that the gamma- and beta-ray exposure in air and on skin from induced radionuclides of ^{24}Na, ^{56}Mn, ^{42}K and ^{46}Sc in soil using the DS02 neutron fluence and Monte Carlo transport calculation [5]. It shows that exposure rates from activated soil are estimated to be ~50 mGy/h by Imanaka et al.[3] and ~ 40 mGy/h by Tanaka et al [5] at 1 hour after the bombing, respectively. In these estimation, the same element composition in soil as DS86 was used.

Element composition of Hiroshima soil was obtained for only two locations (Hiroshima Castle and A-bomb Dome) in DS86. Variation of the composition over Hiroshima City is unknown. Before DS86, element composition analysis for 16 locations in Hiroshima City were carried out by Hashizume et al. in 1967 [6]. However, their efforts were concentrated on Na and Mn density, and measured just one location for Sc density [6]. Sc is activated to ^{46}Sc which has a relatively long half life of 83.79 d. The radiation exposure of the early entrance survivors at a few 10 days or later after bombing is dominated by ^{46}Sc. Therefore the Sc concentration is important parameter for estimating radiation dose to early entrants. In this report, the element concentration, especially Sc concentration in soil of Hiroshima City have been investigated by neutron activation analysis.

Sample preparatrion

Eleven soil and one plaster samples were collected from 10 locations including Hiroshima Castle and Atomic bomb dome where the element concentration analysis were performed in DS86. The soil core (diameter 5 cm, length 20 cm) were taken using stainless steel pipes. The sampling locations are shown in Fig. 1.

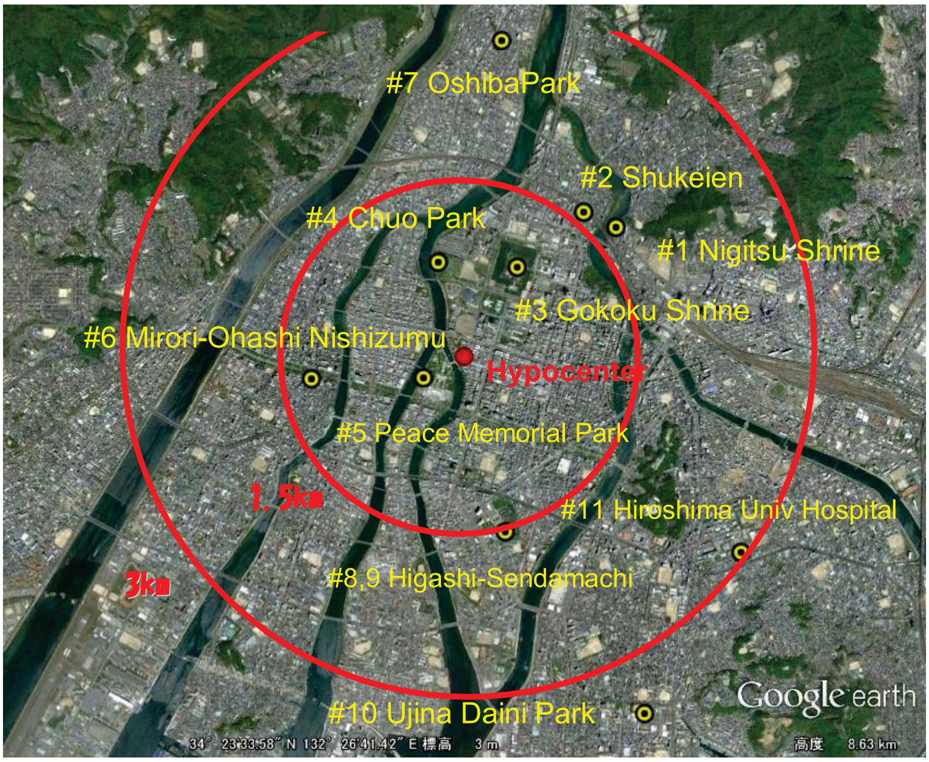

Figure 1 Sampling locations. Eleven samples were collected from 10 locations.

The plaster sample was supplied from Heiwa Kensetsu Co. Ltd. taken from a traditional Japanese house built more than 60 years in the Hiroshima region.

These samples were dried by an oven at 120 degrees more than one night. The dried samples were sieved through a 2-mm mesh to remove small rocks and big plant remains. The sample for neutron activation analysis of 10 g soil was grained with mortar for uniformity. About 0.1 g grained soil was packed into about 5 mm square with polyethylene film.

Activation analysis

The neutron irradiation has been carried out at Kyoto University Reactor (KUR) operatiing 1 MW. The nominal flux of thermal, epithermal and fast neutrons are 2.75×10^{13}, 1.09×10^{12} and 6.0×10^{12} ($n\ cm^{-2}\ s^{-1}$) at 5MW operation, respectively [7]. The soil samples and reference rock sample of JA-1[8] were activated in Kyoto University Reactor (KUR). The soils were neutron-irradiated for 30 sec for identifying short-life radionuclides. The gamma-ray spectrometry was carried out at about 2 min and 1 hour after the irradiation using Ge-detector (EG&G ORTEC, GEM-25185) at KUR. The first measurement is mainly for ^{28}Al-concentration determination. The second measurement is mainly for ^{56}Mn and ^{24}Na concentration determination. The soils were neutron-irradiated for 20 min for identifying medium and long-life radionuclides. The gamma-ray spectrometry was carried out at about 40 days after the irradiation using Ge-detector (EG&G ORTEC, GEM-30200-P) at Hiroshima University.

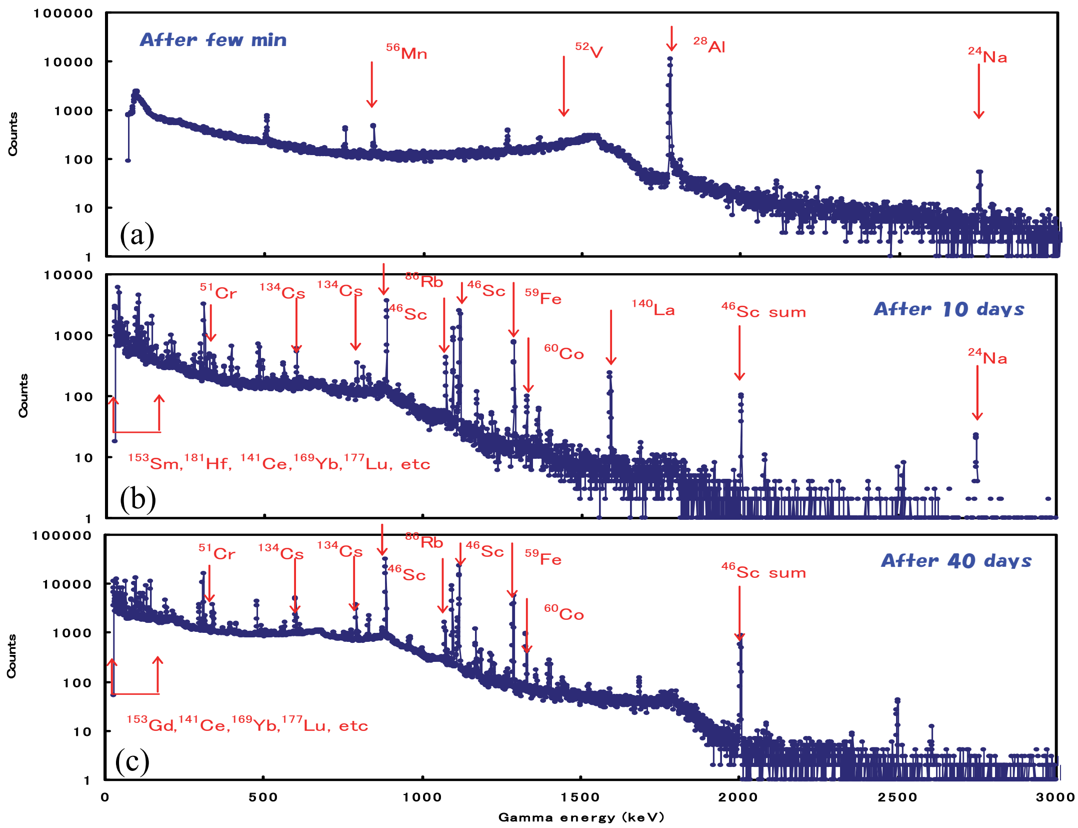

Figure 2 Example of gamma-ray spectrum

Results

The gamma-ray spectra for #3 (Gokoku Shrine) are shown in Fig.2. Short-life radionuclides of ^{28}Al, ^{56}Mn, ^{52}V and ^{24}Na are identified in Fig 2 (a). Medium- and long-life radionuclides of ^{153}Sm, ^{181}Hf, ^{141}Ce, ^{169}Yb, ^{177}Lu, ^{85}Sr, ^{51}Cr, ^{140}La, ^{134}Cs, ^{95}Zr, ^{134}Cs, ^{46}Sc, ^{86}Rb, ^{233}Pa, ^{152}Eu, ^{153}Gd, ^{60}Co and ^{59}Fe are identified in Fig. 2 (b) and (c). In order to get element concentration, counting rate ratio of soil samples to reference samples that are decay-corrected to the time at the finish of the irradiation are multiplied to the known element concentrations.

Twenty three element concentrations for soils and 19 element concentrations for plaster including Al, Mn, Na and Sc are obtained by the neutron activation analysis. The averaged values of concentrations over 11 soils and element concentrations in plaster are listed in Table 1.

Table 1 Averaged values of element concentration in soil and DS86 values[1)]

Element	Z	Concentration (ppm)		DS86 value (ppm)	
		Soil in average*	Plaster**	Castle	Dome
Na	11	19300 ± 20 %	11500 ± 1 %	16000	12400
Al	13	63300 ± 14 %	66400 ± 2 %	71000	64900
K	19	- ± -	17100 ± 16 %	31100	35600
Sc	21	5.12 ± 12 %	11.5 ± 1 %	5	5
Ti	22	- ± -	3240 ± 74 %	1520	1570
V	23	21.4 ± 26 %	67 ± 17 %	22.3	25.3
Cr	24	20 ± 65 %	55 ± 2 %	20.5	27.3
Mn	25	517 ± 13 %	1050 ± 4 %	467	587
Fe	26	17100 ± 16 %	31200 ± 1 %	17700	20600
Co	27	4.13 ± 23 %	14.8 ± 1 %	3.7	3.8
Rb	37	137 ± 12 %	89 ± 18 %	230	225
Sr	38	45 ± 38 %	- ± -	88	70
Zr	40	105 ± 40 %	133 ± 62 %	41	35
Nb	41	2 ± 60 %	- ± -	7	5
Sb	51	1.87 ± 11 %	- ± -	1.4	0.8
Cs	55	4.4 ± 14 %	3.3 ± 10 %	5	5
La	57	27.4 ± 23 %	- ± -	23	21
Ce	58	88 ± 24 %	43 ± 3 %	40	36
Sm	62	3.7 ± 27 %	- ± -	3.4	3
Eu	63	0.81 ± 15 %	0.94 ± 3 %	0.9	0.9
Gd	64	2.23 ± 44 %	- ± -	-	-
Yb	70	- ± -	1.9 ± 20 %	-	-
Lu	71	0.133 ± 33 %	- ± -	-	-
Hf	72	4.17 ± 19 %	4.7 ± 6 %	4	4
Ta	73	- ± -	0.84 ± 11 %	0.8	0.8
Th	90	14.3 ± 30 %	7.3 ± 6 %	13.3	9.9
U	92	0.85 ± 18 %	- ± -	2.8	2.6

*Concentration averaged over 11 soil samples. Errors are standard deviation for 11 samples.

**Concentrations are for one plaster sample. Errors are only statistics.

^{24}Na is not only produced by the ^{23}Na(n,g)^{24}Na reaction, also activated through the ^{27}Al(n,a)^{24}Na reaction. However, the fluence ratio of thermal neutron to 15 MeV-neutron in

KUR is less than 1/10000, then the yield from the $^{27}Al(n,a)^{24}Na$ reaction can be neglected comparing with the $^{23}Na(n,g)^{24}Na$ reaction. The aluminum concentration is estimated here neglecting with the $^{27}Al(n,a)^{24}Na$ reaction.

The obtained element concentrations in soils are compared with values used in Dosimetric System 1986 (DS86) [1] and found roughly the same as the reported values in DS86. Sc density in Hiroshima soil is estimated to be 5.12 ± 0.59 (ppm). It is found the unevenness of Sc density in soils for 10 location of Hiroshima city is about 12%.

The element composition in plaster under roof-tiles of old Hiroshima house is slightly different from soils. Especially, K-, Sc- and Mn-density are a half, twice and twice in compared with those of soil, respectively.

In case of simple condition which the activated materials uniformly distributed on ground, it is simply calculated using Tanaka's calculation results [5]. The time variation of dose in air for soil and plaster are shown in Fig. 3. The estimated dose in air from activated soil is to be 5.3 Gy/h for a few second and 0.6 Gy/h for one month after explosion at hypocente, respectively. On the other hand, dose in air from activated plaster is to be 5.3 Gy/h for a few second and 1.1 Gy/h for one month after explosion at hypocenter.

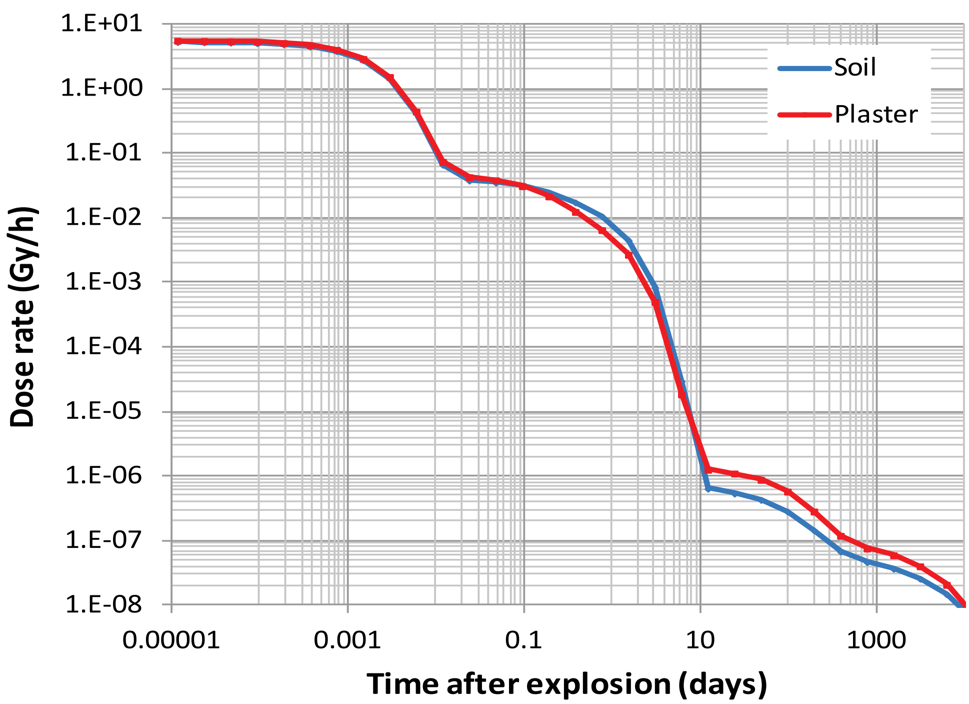

Figure 3 The time variation of dose in air for soil and plaster

Conclusion

In order to estimate exposure from activated soil by Hiroshima atomic bomb, element concentration in soil of Hiroshima City are analyzed by neutron activation analysis at Kyoto University Reactor. Eleven soil samples were collected from Hiroshima City within 4 km from the hypocenter.

As a result of the activation analysis, 23 element concentrations including Al, Mn, Na and Sc are obtained. Sc concentration is important for gamma-ray exposure to early entrants at about a few 10 days after bombing. Sc concentration and unevenness are obtained to be 5.12 ± 0.59 (± 12 %) ppm.

Mn concentration is a half of value by Hashizume et al [6], while Mn and Na concentrations are consistent with DS86 values. The averaged values of Mn and Na concentrations over 11 soil samples are Mn: 517 ± 68 (± 13 %) ppm and Na: 19300 ± 3900 (± 20 %) ppm, respectively.

The element composition in plaster is slightly different from soils. Especially, Na-, Sc- and Mn-density are a half, twice and twice, respectively.

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