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NEUTRON ACTIVATION STUDY OF RADIATION SHIELDING CONCRETE

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ABSTRACT

By iradiation of neutrons for the period longer than one month, activation experiment of concrete constituents are performed ,and activated nuclides are analyzed with gamma ray counting. The elements which has very small density (trace element)of ppm order, such as Co, Sc, Cs, Eu, Ce, etc. are successfully identified and their contents are also obtained quantitatively. Activation calculation for concrete constituents was performed with ORIGEN2-82 computer code. It was concluded that radioactivity from the trace elements in reactor shielding concrete plays the important role after a long cooling time.

1. INTRODUCTION

Concrete is widely used to keep leakage radiation, neutrons and gamma rays, from nuclear facilities as low as possible. Most importantly, a reactor core is surrounded by a large amount of heavy and ordinary concrete which is called as biological shield. As this concrete is irradiated directly from the reactor core for a reactor life, various artificial radioactive nuclides are produced by the neutron activation. This concrete activation becomes a serious problem, when reactors are decommissioned or when nuclear facilities are maintained. In the case of reactor decommissioning, radioactive nuclides which have a long life time longer than one year, such as Co-60 ,Eu-152, etc. cause difficulty for waste management of activated concrete.

The amount of activation for structural material in and surrounding a reactor core can easily be calculated, when both ingredients of material and the histroy of neutron irradiation (strength of neutron flux and its time variation) are known. However, the most radio isotopes in concrete which have long life are generated from nuclides which have very small densities. To obtain precise density for the nuclides having the very small densities in concrete by the chemical analysys method is usually very difficult. Therefore, activation from isotopes which have the very small densities is not accurately predicted by calculation.

In the present paper, the activation analysis of concrete samples were conducted to obtain density of trace elements quantitatively. The activation analysis is usually performed with research reactors and the irradiation time of samples is longer than one hour. However, analysis of nuclides having a long life time is difficult in case of irradiation of several hours. We conducted the activation analysis by irradiating concrete samples for about one month. By analysing specific gamma rays, identification of activated nuclides in concrete samples was performed and their parent nuclides were also identified.

The method employed in the present paper for identification and quantitative analysis was a comparison method. That is, the irradiations of reference and concrete samples were conducted at the same condition. After irradiation ,gamma rays from both samples were analyzed and compared for the positions of the gamma ray peaks and the area under the peaks. As the ingredients of reference samples were known by rock analysis, we could obtain densities of trace elements in concrete samples by comparison between both samples.

2. EXPERIMENT

2.1 Reference and concrete samples

As the reference sample, G-2 of the U.S. Geological Survey Standard was used. The activation samples were aggregate , river sand and cement for concrete. Table 1 and table 2 shows the ingradients of the reference and samples. The weight of irradiated samples were 0.0269g for G-2, 0.1020g for aggregate, 0.104g for river sands and 0.1090g for cement.

2.2 Irradiation experiment

The irradiation was conducted at the vertical irradiation hole of the research reactor of RIKKYO University (TRIGA, 100 kW). The all samples were sealed in polyethylene bags and contained in polyethylene cylinder of 10 cm length and 3 cm diameter. This cylinder was put at the vertical irradiation hole and irradiated with neutron flux of $0.5x10^{12}$ n/cm2/sec. The irradiation period was about one month,and EFPH(effective full power hour) was 90 hour.

2.3 Measurement of gamma ray spectrum

The gamma-ray measurements were conducted for samples at 10 days and one month after irradiation. The gamma ray spectra were measured with 4K channel pulse height analyser with a high resolution Ge detector. Identification of gamma ray spectra and area analysis under peaks were performaed by a analysis code.

3. ANALYSIS

3.1 Spectrum analysis

In Fig.1, the gamma-ray spectrum is shown for the reference sample(G2). The identified peaks for life time longer than one month is also shown in the same figure. Also, Fig.2 showes the gamma-ray spectrum of the river sand and the identified peaks. It was shown that every sample contained Co-60, Sc-46, Fe-59 and Cs-134, and Eu-152, Eu-154 and Ce-141 were found in the river sands and the cement. The quantity of the identified elements was evaluated by comparing the each peak between the reference and the samples. The following relations were used for this evaluation.

$$t = t_R \cdot \frac{S}{S_R} \cdot \frac{M}{M_R}$$

where, t : element density in the sample,
S : the area of under the peak of gamma ray,
M : mass of the samples,
and the suffix R means data of the reference.

The results are shown in Table 3. It can be seen that except Fe, the trace element contents have the order of ppm. The partial content of Fe by the present analysis is 66.6% for aggregates, which can be compared with 60.9% by the chemical analysis previously performed.

3.2 Effect of trace element to total activity

The radioactivities for the aggregates, the river sands and the cement which were studied in the present paper was calculated under the power reactor condition. The calculation was performed with the ORIGEN2-82 computer code.

The conditions of calculation are shown in Table 4 and the results are shown in Table 5. This table shows the tendency that, as the cooling time becomes longer, the effect of the trace element to total activity becoms more serious.

4. CONCLUSIONS

Followings are conclusions that are obtained in the present paper.

- By the activation experiment, radioactive nuclides which had a half life longer than one month, Co-60, Sc-46, Fe-59, Cs-134, Eu-152, Eu-154 and Ce-141 were identified in the concrete ingredients. The partial contents of parent elements of these radioactive nuclide, Co, Sc, Fe, Cs, Ce and Eu were obtained quantitatively.

- By the calculation with ORIGEN2-82 computer code performed for the samples, residual radioactivity mainly originates from isotopes generated from tarce elements of very small partial densities, as the cooling time becomes longer. Important radioactive nuclides are found to be Co-60, Eu-152, etc.

REFERENCES

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Table 1. Dominant composition by weight(percent)

	G-2	Aggregate	River sand	Cement
SiO_2	69.1	5.4	72.6	21.7
Al_2O_3	15.3	1.0	11.5	5.3
Fe_3O_4	1.1	84.2	3.7	3.2
CaO	2.0	2.6	1.6	64.7
Na_2O	4.2	—	3.2	0.3
K_2O	4.5	—	2.3	0.6

Table 2. Average dominant trace element content of G-2(ppm)

Element	ppm	Element	ppm
Ba	1950	Pb	28.7
Ce	166	Pr	20
Co	4.9	Rb	234
Cr	9.0	Sc	3.9
Cu	10.7	Sm	8.3
Ga	20.2	Sr	463
Hf	7.5	V	37.0
La	112	Zn	74.9
Mn	265	Zr	316
Nb	16.1		

Table 3. Trace element contents(ppm)

Element	Aggregate	River sand	Cement
Sc	5.6	4.9	9.6
Co	63.5	8.0	4.5
Cs	17.1	3.9	3.2
Ce	——	28.9	42.8
Eu	——	0.7	1.0
Fe	66.6%	1.4%	2.1%

Table 4. Conditions of Calculation

Total neutron flux	1×10^8 n/s/cm^2
Irradiation time	20year(EFPY)
ORIGEN2-82 library	PWR 3.3GWD/TM

Table 5. Total radioactivity and tarce element activity(Bq/g)

Sample		Cooling time			
		Discharge	One month	One year	Ten year
Aggregate	Total activity	1.1×10^4	1.0×10^4	7.1×10^3	6.6×10^2
	Trace element	5.9×10^2	4.0×10^2	3.2×10^2	8.1×10^1
River sand	Total activity	6.7×10^3	3.8×10^3	6.7×10^2	8.6×10^1
	Trace element	2.1×10^2	1.5×10^2	1.2×10^2	5.2×10^1
Cement	Total activity	1.5×10^4	2.5×10^4	4.6×10^2	1.2×10^2
	Trace element	2.5×10^2	1.8×10^2	1.3×10^2	6.5×10^1

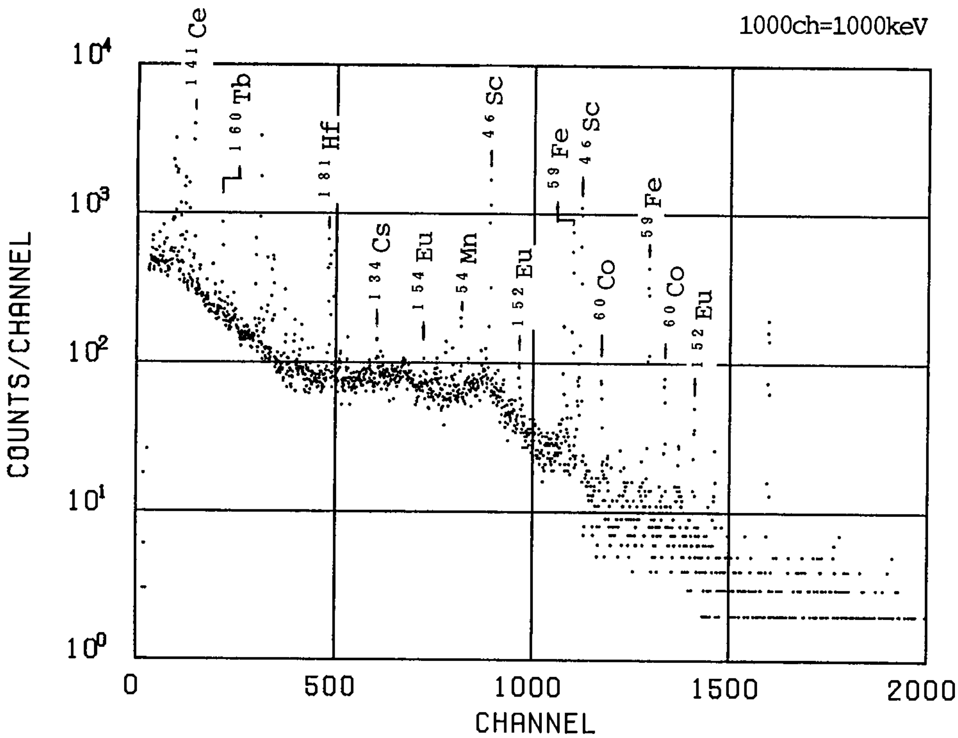

Fig.1 Gamma ray spectrum from U.S.geological survey standard (G-2) with Ge-detector (10day cooling)

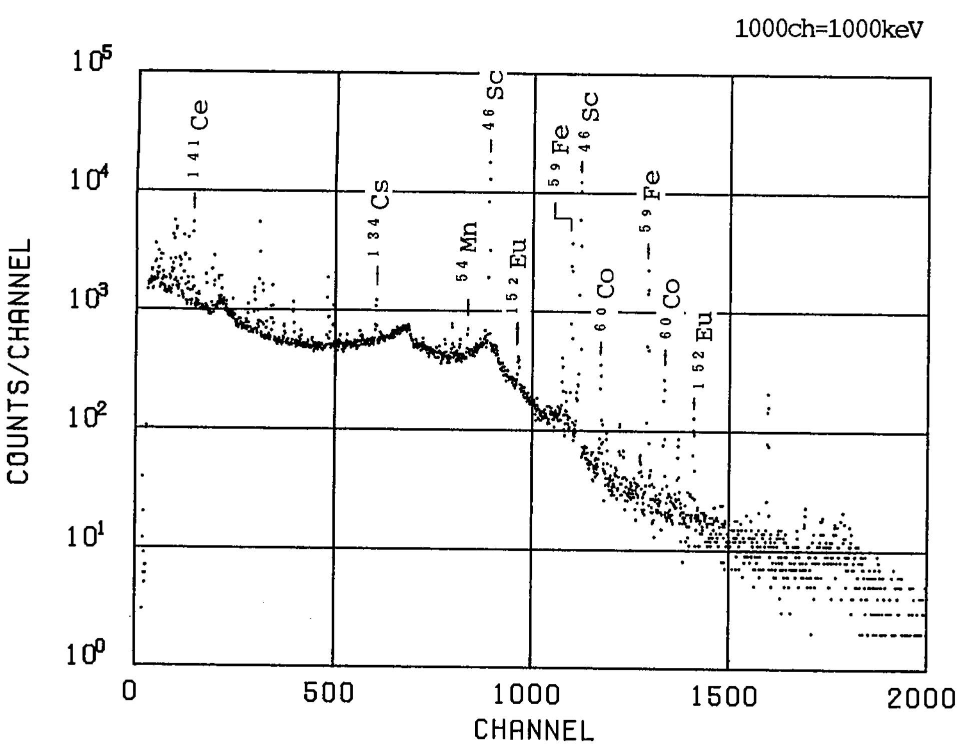

Fig.2 Gamma ray spectrum from river sand with Ge-detector (10day cooling)