Development of construction material database for low-activation sheilding

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Abstract

After huge quantities of radioactive plume were dispersed over almost half of Japan, radioactive cesium has been detected across a large percentage of Japanese land. That causes some effects to the raw material for construction work and this influence will continue to expand because of ^{137}Cs. Due to this situation, the development of a radioactive database for construction materials after March 11th 2011 has been started to compare with a database from before the accident in Fukushima. In this paper, some natural radioactivities in fly ash were reported to understand the baseline level of radioactivity. Secondly, radioactive cesium in combustion ashes and slags, which were taken from all over the Japan after the accident, were measured to investigate the levels of contamination. Based on the measurements obtained, the trial investigation of the influence of the use of the construction materials with radioactive cesium on the shielding pod was conducted using a Monte Carlo calculation. The calculated dose rates from the shielding concrete itself with radioactive cesium were greater than those from concrete shielding pods containing contaminated soils in one case. These results confirmed the necessity of the development of radioactive database for the construction materials.

§1. Introduction

Concrete is one of the main materials used for constructing building, infrastructure, and facilities, which are a major portion of human public activities. In addition, these materials are typically used in reactor and accelerator facilities for shielding.

For the design of radiation shielding, detailed data for the material elements are required, and thus it is very important to establish a construction material database for radiation shielding. A lot of effort has been made to collect a variety of construction materials [1] across from a range of locations over different periods of time. The collection of these materials comprised 2000-3000 kinds of aggregates including crushed natural stones, by–products, and burning slags, approximately 1000 cement samples including various types of Portland cements, alumina cements, and clinkers which varied in the production year, and 1000-2000 kinds of additive including fly ashes, limestone powders, and combustion ashes. Among these materials, approximately 500 samples were analyzed for major and rare elements, including some radioactive nuclides.

On the other hand, it is also important to develop a radioactive database for the construction materials for shielding especially gamma-ray shielding. And that database

Keywords : database, construction material, clearance level, Monte Carlo, radioactive cesium

should include naturally occurring radioactive materials (NORM) and radioactive nuclides produced by the accident in Fukushima.

In this paper, some natural radioactivities in fly ashes were reported to understand the level of radioactivity for NORM. Radioactive cesium in combustion ashes and slags, which were taken from all over Japan after the accident, were also measured to investigate the levels of contamination. Based on these results, the trial investigation on the influence of the use of the construction materials with radioactive cesium on the shielding pod was conducted using a Monte Carlo calculation.

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§2. Radioactivity in raw materials for radiation shielding –typical example-

In this section, the quantities of NORM for the coal fired fly ashes and radioactive cesium for the furnace slags and coal fired fly ashes were estimated to recognize baseline levels.

2.1 Radioactivity in fly ash

Coal fired fly ash is one of the major additive materials for concrete. An advantage of the use of fly ash in the concrete is to reduce the initial stress of thermal heat by cement hydration reaction because of its small particle size. This property is particularly useful in thick concrete such as that used for radiation shielding.

On the other hand, some of the coal ashes are known as artificially concentrated materials of NORM. The level of activity is obviously higher than coal itself and could be around same level as Clearance Level (CL)[2]. Understanding the radioactivity level of the coal–fired ash is important for their use in concrete materials.

More than 60 fly ashes were gathered from all over the Japan. 20–30kg samples of fly ash were carefully taken from each plants site, and sent to us. These fly ashes were sampled in two ways – over a wide area at a single point in time, and at a single location over an extended period. Two plants were chosen for the investigation of changes over an extended period of more than 10 years. Samples from another 19 plants collected in a single year (2010) were examined to understand regionality.

Estimation of activities for fly ash samples were conducted by gamma ray measurement with an HP Ge detector. The nuclides measured were ^{214}Pb, ^{214}Bi (^{238}U series nuclides), ^{228}Ac, ^{212}Pb, ^{208}Tl (^{232}Th series nuclides), and ^{40}K, which are known as typical nuclides for NORM (naturally occurring radioactive materials). 30–50g of each fly ash was packed into a U-8 cylindrical plastic case, and were sealed by waterproof gel in order to ensure air-tightness and keep the radioactive equilibrium for nuclides of ^{232}Th series and ^{238}U series for more than one month.

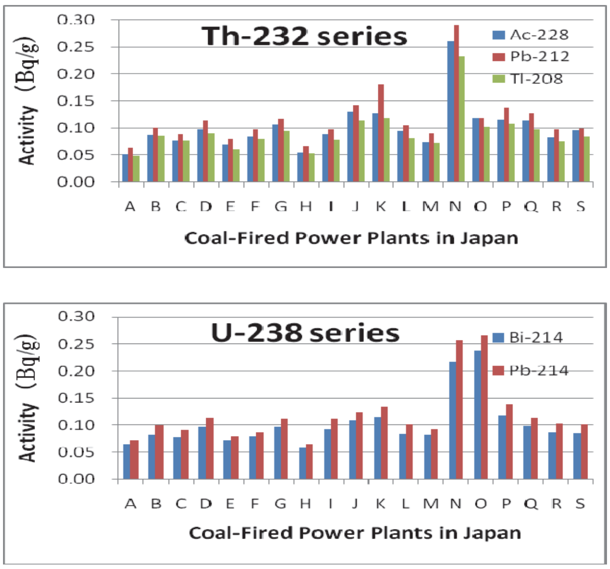

Figure 1 Activities of Th-232 &U-238series nuclides for 2010 gathered fly ashes.

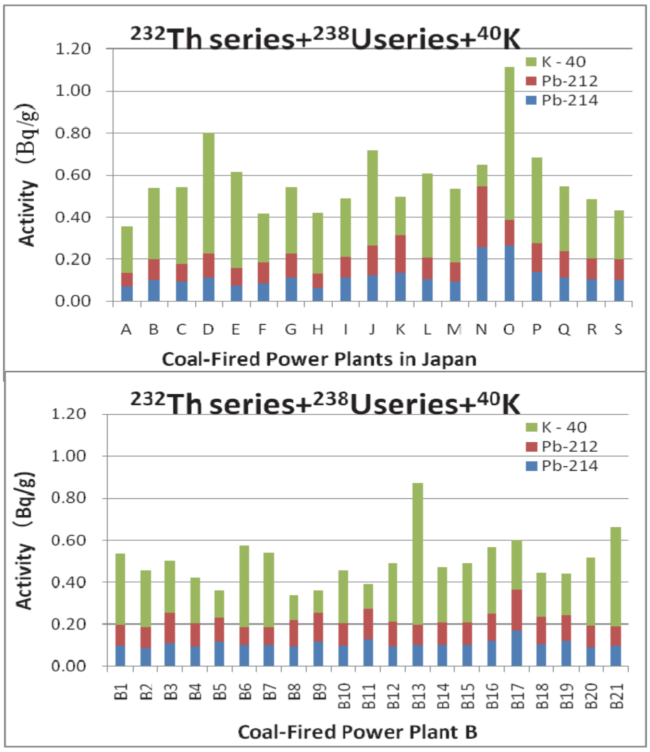

Figure 2 Activities of Th-232 &U-238series and K-40 for 2010 gathered fly ashes and Plant B.

Figure 1 shows the levels of radioactivity for ^{238}U series and ^{232}Th series for fly ashes gathered in 2010 physical year from 19 coal-fired power plants in Japan (named A to S shown in horizontal axis). These figures indicate ^{214}Pb in ^{238}U series and ^{212}Pb in ^{232}Th series are higher than the others, respectively. These tendencies are similar in other samples. ^{212}Pb and ^{214}Pb are defined as the representive nuclide for ^{232}Th series and ^{238}U series respectively in latter section for the estimation of the activity in fly ash, therefore.

The upper side of the **Figure 2** shows the levels of activities in summation of ^{212}Pb, ^{214}Pb and ^{40}K, for gathered from 19 coal-fired power plants in 2010 with each sample number at horizontal axis, which are distributed in a range of 0.7 Bq/g. The lower side in the figure 2 describes another variety of data gathered for more than ten years in one particular site. These figures indicate that ^{40}K is the largest contribution in total activities in most cases, and ^{232}Th series and ^{238}U series tend to behave similarly. The range of activity is from 0.4 to 1.1 Bq/g, and all but one of the 60 samples analysed are less than 1.0 Bq/g [3].

Table 1 Number of samples for measurement.

Materials (number of sample)	Sampling time	Measured nuclides		
		NORM	Radioactive cesium	
			Detracted	N.D.
Coal-fired fly ash (34)	Before March 2011	8	0	8
	After April 2011	26	0	26
Furnace slag (15)	Before March 2011	0	0	11
	After April 2011	0	2	2

2.2 Estimation of contaminated construction materials

Tremendous quantities of radioactivity were dispersed from Fukushima Daiichi nuclear power plant. That caused some effects to the raw material for construction work and this influence will increase in long term. Beside the accident in the Fukushima, other contamination sources, such as nuclear weapon experiments, or other accidents from nuclear facilities, should be also taken account. Therefore, estimation of the level of radioactivity for construction raw materials is important.

41 samples (15 for furnace slags and 26 for coal-fired fly ashes) were gathered after the accident in Fukushima and 49 samples, including eight fly ash samples from the sample storage in addition to the above, were analysed using an HP Ge detector. Samples of fly ash were only taken for two plants located in Tohoku area, because there were no reports of radioactive cesium for samples prior to 2010. 15 furnace slags were gathered from all over the Japan including Tohoku area, which were produced before and after the accident. **Table 1** summarizes the quantity and conditions of the samples analysed.

In 2 of the 15 furnace slag samples, ^{134}Cs and ^{137}Cs were detected as 0.987 and 1.20 Bq/g for first slag (S1) and 0.183 and 0.287 Bq/g for second slag (S2), compared with detection

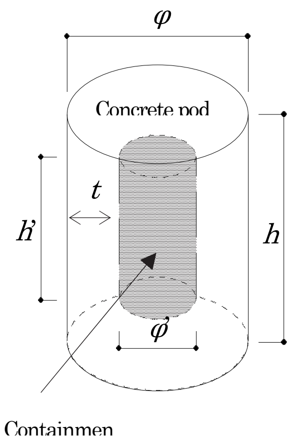

Table2 Size of shielding pods.

Size	Ordinary concrete	Middle density concrete
t	200	150
φ	700	600
h	800	700
φ'	300	
h'	400	

Unit : mm

Figure 3 Calculation geometry.

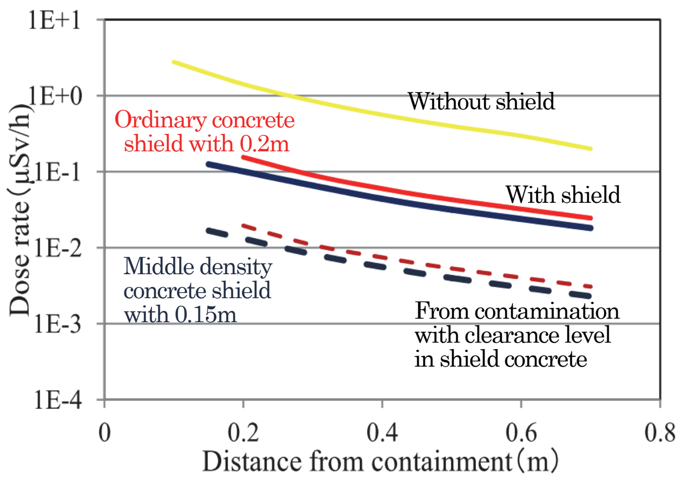

Figure 4 Calculated dose rates for the effect of contamination.

limits for ^{134}Cs and ^{137}Cs of 0.027 Bq/g and 0.030Bq/g respectively. These values were larger than the level of clearance level for ^{134}Cs and ^{137}Cs (0.1 Bq/g [2]). Radioactive cesium was not detected among samples produced before March 2011. The ratio of ^{134}Cs and ^{137}Cs is different in samples S1 and S2. The ratio of ^{134}Cs and ^{137}Cs for S2 (0.64) is about the same as that for the contaminated soil samples found in a separate study [4]. In the above analyses, no radioactive cesium was found in samples before or after Fukushima that originated in nuclear weapon experiments.

§3. Investigation of the influence on the effect of contaminated construction materials to use radiation shield

For the proper containment of contaminated soils in Fukushima, concrete shielding pods could be one idea. We therefore considered the effect of radioactive materials in the shielding concrete used in construction of the shielding pods.

3.1 Monte Carlo simulation for the investigation

The MCNP4C2 code [5] was employed for Monte Carlo simulation of photon in this work using MCNPLIB02 photo library [6] with "Radiation dose conversion coefficients for radiation shielding calculations: 2010" [7]. The geometry of the simulation is shown in **Figure 3** and **Table 2**, with a cylindrical volume source and cylindrical concrete shielding pod. Isotropic photon sources were uniformly placed in the region of the soil blocks in the case where the contaminated soil was contained in the concrete shielding pod. To estimate the influence of contaminated material use in the concrete, the photon sources were set only inside of the shielding concrete uniformly with single element (cesium or potassium) to evaluate the dose rate caused by that element. Cylindrical surface tallies were set along the line that lengthened perpendicularly from the cylindrical lateral central part as estimators. Dose rates were calculated at distances of 0.1 to 0.7 m from surface of the soil block for the case of soil block only (without shield), at distances of 0.2 to 0.7 m for ordinary concrete shield with 0.2 m thickness, and 0.15 to 0.7 m for middle density concrete shielding with 0.15 m thickness. The validity of this simulation with this geometry was confirmed by comparison with experimental data from another study [4].

Table 3 Calculated dose rates at 0.3m from the cylindrical containment side with various photon sources.

Shielding condition	Setting photon source			Calculated dose rate		Description
	Material	Nuclide	Activity (Bq/g)	(μSv/h)	Ratio to soils	
Without shield	Soils	^{134}Cs, ^{137}Cs	30.6,47.6	0.851		Bare
Ordinary concrete shield 0.2m thickness	Soils	^{134}Cs, ^{137}Cs	30.6,47.6	0.0896		Contaminated soils
	Concrete	^{134}Cs, ^{137}Cs	0.039,0.061	0.0108	12%	Clearance Level
	Concrete	^{40}K	0.738*	0.0138	15%	NORM
Middle density concrete shield 0.15m thickness	Soils	^{134}Cs, ^{137}Cs	30.6,47.6	0.0657		Contaminated soils
	Aggregate 1	^{134}Cs, ^{137}Cs	0.99,1.2	0.146	222%	Slag 1
	Aggregate 2	^{134}Cs, ^{137}Cs	0.18, 0.29	0.0313	48%	Slag 2
	concrete	^{134}Cs, ^{137}Cs	0.039,0.061	0.00818	12%	Clearance Level

* Max value in the reference [8]

3.2 Investigation result

Figure 4 shows the result of the calculation for a soil block only versus two types of concrete pods – ordinary concrete with 2.13 g/cm^3 (marked red in the figure) and middle density concrete with 3.09 g/cm^3 (marked blue in the figure). Solid lines were attenuation curves of dose rate for the experimental soil block (22.4 kg with a volume of 18.1*l*, 6.86E5 Bq of ^{134}Cs, and 1.07E6 Bq of ^{137}Cs) [4]. Dashed lines indicated the influence on the dose rates that the contaminated cesium radionuclide gave, with empty containment (no contaminated soil block inside). The activities of radioactive cesium nuclides in the shielding concrete were set equal to the clearance level (0.039 Bq/g for ^{134}Cs, and 0.061Bq/g for ^{137}Cs with the same ratio of ^{134}Cs and ^{137}Cs for existing contaminated soils [4]). This figure indicates that the levels of dose rates become smaller in 1/10 degree in the order of those for Bare (without shield), those for contaminated soil with concrete shield, and those for shielding concrete themselves with radioactive cesium of clearance level.

Table 3 shows the calculated dose rates by MCNP4C2 with same library, conversion factors, and geometry as described section 3.1, at 0.3 m from the cylindrical lateral central part for three shielding conditions, such as ordinary concrete pod with 0.2 m shielding thickness, middle density concrete pod with 0.15 m thickness, and without shield. The photon source was set in four different materials (soils, concrete, and two contaminated slags described section 2.2) uniformly with radioactive cesium or ^{40}K. Activities for radioactive cesium were set at the level of measured data in the experiment for "soils", clearance level for "concrete", and measured data in this work for "aggregate". Activity of ^{40}K was set to the maximum value in the concrete reported in the reference [8]. This table indicates that the influences of the radioactive materials are 10% to 200 % in the ratio to the containment (contaminated soils).

§4. Conclusion

As radioactive nuclides were widely spread all over the Japan, raw material for the construction has certain influence. In order to develop the radioactive database for construction materials, a radioactive survey of construction materials was conducted.

Firstly natural radioactivities in fly ashes were measured for more than 60 samples on two bases (area and period). Secondly radioactive cesium nuclides were measured for 49 samples gathered from all over the Japan, and two of these exceeded clearance level.

Based on the measured data, a trial investigation was conducted to estimate the effect for the use of the materials containing radioactive cesium and radioactive potassium. The calculated dose rates from the shielding concrete with radioactive cesium exceed those from the radioactive material in shielding concrete in some cases. These results indicated to the necessity of the development of radioactive database for the construction materials.

Acknowledgements

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Short comment R&D for low-activation concrete has long history. Obviously the phase was changed after 3.11. At that situation, maintenance of construction material database for low-activation shielding is very important. So, I try to proceed to upgrade of the database and make every effort to apply this material every possible market related radiation field.