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INVESTIGATION OF THE MAIN RADIOACTIVE INVENTORY IN FUGEN DECOMMISSIONING ENGINEERING CENTER

Haryo Seno[1], Yoshitsugu Morishita[2], Hiroki Iwai[2]

[1]National Nuclear Energy Agency (BATAN) – Indonesia
[2]Japan Atomic Energy Agency (JAEA) – Japan
Email: haryo@batan.go.id

ABSTRAK

STUDI INVENTORI RADIOAKTIF DALAM REAKTOR DI PUSAT REKAYASA DEKOMISIONING FUGEN. Studi tentang inventori radioaktif telah dilakukan di Pusat Rekayasa Dekomisioning FUGEN. Reaktor FUGEN memiliki struktur tabung bertekanan yang rumit, berbeda dari jenis reaktor berpendingin air ringan pada umumnya. Perilaku neutron di dalam reaktor sangat kompleks, oleh karena itu perlu penentuan densitas fluks neutron secara akurat sebelum menganalisis besaran nilai aktivasi neutron. Densitas fluks neutron ditentukan dengan kode kalkulasi DOT3.5 dan selanjutnya diverifikasi melalui pengukuran sampel di reaktor, yaitu dengan foil aktivasi dan detektor neutron. Densitas fluks neutron yang telah terevaluasi kemudian digunakan untuk menganalisis nilai aktivasi neutron, yang juga memerlukan data komposisi material dan data sejarah operasi reaktor. Perhitungan nilai aktivasi neutron ini menggunakan kode kalkulasi ORIGEN79. Hasil dari perhitungan ini harus diverifikasi dengan pengukuran aktivitas pada daerah yang spesifik di dalam reaktor. Verifikasi untuk inventori radioaktif dilakukan dengan cara mambagi reaktor menjadi tiga bagian, yaitu bagian dalam teras reaktor, bagian perisai, dan bagian di luar perisai. Namun, dalam studi ini lebih ditekankan pada bagian perisai. Bagian ini terdiri dari perisai beton biologis dan perisai besi-air. Inventori radioaktif utama yang ada di dalam perisai beton biologis adalah Co-60, Eu-152 dan Eu-154, sedangkan di dalam perisai besi-air adalah Fe-55, Ni-63, dan Co-60.

Kata kunci: dekomisioning, inventori radioaktif, kode perhitungan, perisai besi-air, perisai biologi.

ABSTRACT

INVESTIGATION OF THE MAIN RADIOACTIVE INVENTORY IN FUGEN DECOMMISSIONING ENGINEERING*. The study of radioactive inventory was carried out in Fugen DEC. Reactor of Fugen has a complicated pressure tube structure, differ from light water reactor type in generally. Behavior of neutron in the reactor is very complex, so it is necessary to determine neutron flux density accurately before analyzing neutron activation value. Neutron flux density is determined by DOT3.5 calculation code and then is verified by measurement of samples, such as activation foils and neutron detector. The evaluated neutron flux density then be used to analyze neutron activation value, including the material composition data and operating history of reactor. It was conducted by ORIGEN79 calculation code. The result of this calculation should be verified with measurement of activity in specified region. Verification of radioactive inventory is undertaken by dividing reactor into three parts i.e. inner core part, shield part, and outer shield part. However, this study is emphasized in the shield part. This part consists of biological concrete shield and iron-water shield. The main radioactive inventory in biological concrete shield are Co-60, Eu-152, and Eu-154, while in the iron-water shield are Fe-55, Ni-63, and also Co-60.*

Keywords:biological shield, calculation code, decommissioning, iron-water shield, radioactive inventory

INTRODUCTION

Decommissioning is the final phase in the life-cycle after siting, design, construction, commissioning, and operation. It is a complex process involving activities such as decontamination, dismantling, and demolition of equipment and structure, and management of resulting waste, while taking into account aspects of health and safety of the operating personnel and general public, as well as protection of the environment.The initial step in the decommissioning process is radiological characterization. It requires a logical approach in order to obtain the data necessary for planning program.The detailed data of radioactive inventory is necessary to make a decision leading to selection of decommissioning strategy. Radioactive inventory in nuclear reactor comes from neutron activation and contamination level which have arisen during operation and remain at shutdown.

Decommissioning work in Fugen has been conducted since 2008, but all of preparation regarding to it, has done before. One of the essential activities is evaluation of radioactive inventory that present in the reactor. Therefore, the method of evaluation is required to perform subsequent works. The study of radioactive inventory was carried out in Fugen DecommissioningEngineeringCenter (Fugen DEC), which the reactor has complicated pressure tube structure, different from light water reactor type in generally. To simplify this matter, the evaluation of radioactive inventory is undertaken by dividing reactor into three parts i.e. inner core part, shield part and outer shield part. This study is emphasized in the shield part of the reactor. Radioactive inventory in the shield part of reactor is determined by the neutron activation. Behavior of neutron in the reactor is very complex, so it is necessary to determine neutron flux density accurately before analyzing neutron activation value. To calculate the neutron flux density, DOT3.5 has been used as the neutron transport calculation code.The neutron flux density should be verified by measurement of sample, such as activation foils and neutron detector. The evaluated neutron flux density then be used to analyze neutron activation value. The use of calculation code that is ORIGEN79 is qualified to analyze neutron activation value. This code also needs the data of material composition and operating history of reactor.

Fugen reactor is composed of calandria tank and iron-water shield covering the tank, etc., and it is cylindrical facility (9m in diameter and 7 m in height). As the main characteristic, Fugen reactor has 448 tubes of pair of pressure and calandria tubes being arranged concentrically to consist of a vertical 224 double-tubes structure. The structure is so narrow segment and complicated. In addition, stainless steel (SUS) slab of 150 mm thickness is used as upper and lower tube plate of calandriatank[1]. It is necessary to examine the reactor dismantlement in consideration of these characteristics. Furthermore, structure of the Fugen reactor is shown in Figure 1.

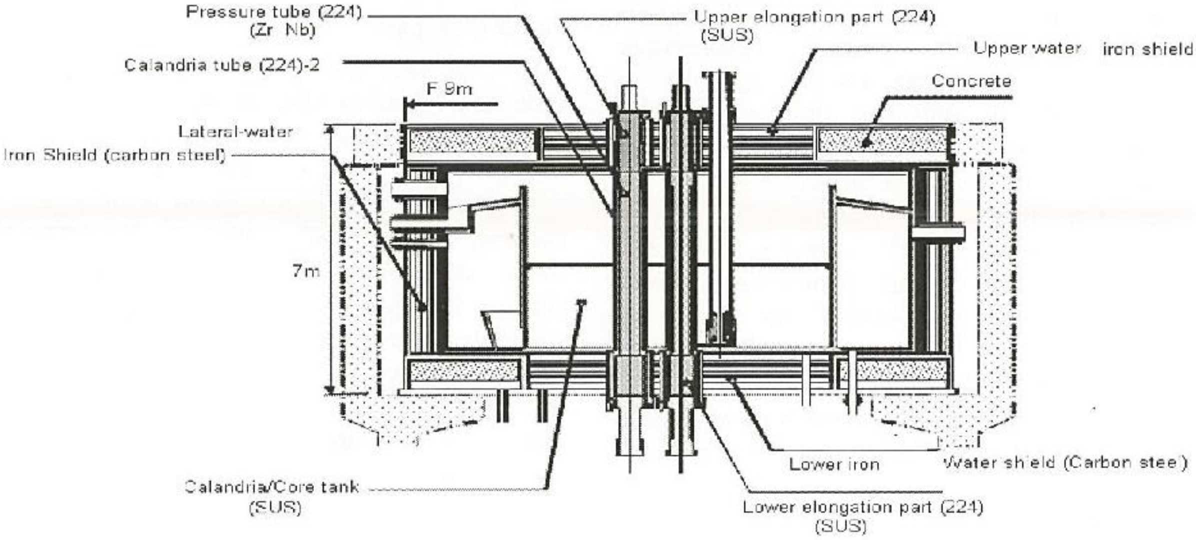

Figure 1. Structure of The Fugen Reactor

Table 1. Evaluation Method of Radioactivity by Neutron Activation in The Reactor Vessel[3]

Region of reactor		Calculation code	Measurement and Evaluation Method
Inner Core Part		Core management code (POLESTER) Lattice calculation code (WIMS-ATR)	Pressure tube specimen
Shield Part	Iron-Water	Neutron transportation calculation code (DOT3.5) Neutron activation calculation code (ORIGEN-79)	Activation foil
	Biological concrete		Concrete sample
Outer shield part		Measurement of Large Gold Foil; MCNP code Neutron activation calculation code (ORIGEN-79)	-Large gold activation foil -Concrete sample Bonner ball measurement

Evaluation Method of Neutron Activation

Calculating activated materials by neutron activation, it is essential to provide neutron flux density in overall part of reactor. Neutron flux density should be evaluated firstly through calculation and measurement, before it is used to calculate amount of radioactivity. The measurement of neutron flux can be undertaken by measuring the activation foil. Many activation foils set around the reactor core in the shielding area and irradiated for a certain period during the reactor operation were gathered and measured[2]. For neutron flux calculation, reactor of Fugen is divided into three parts, i.e. inner core part, shield part and outer shield part[1,3]. Appropriate calculation codes were chosen in order to calculate the neutron flux density properly. Table 1 shows evaluation method of neutron flux density in reactor vessel. Moreover, water content in the concrete was measured in the case of biological shield, because it affects the neutron flux density calculation results. The calculation result of neutron flux density is evaluated with the measurement of activation foils, and the result of radioactivity is evaluated through the direct activity measurement. Figure 2 shows the diagram flow of neutron activation evaluation method.

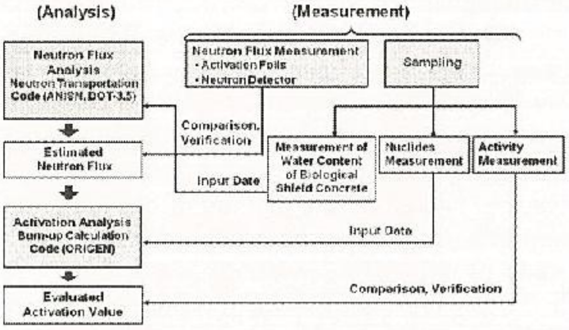

Figure 2. Flow Diagram of Neutron Activation Evaluation Method[4]

Object of Evaluation In The Reactor

Inner core part

Core structure materials, such as pressure tube and calandria tube are strongly activated and it is impossible to cut sample before the final reactor shutdown. Therefore, computer calculation method is usable for evaluation of radioactivity of these materials. POLESTAR (core management code) and WIMS-ATR (lattice calculation code) has been used to calculate neutron flux density[1]. Pressure tube monitoring specimens have been irradiated in the reactor since the first loading of fuel. Some material testing capsules with the pressure tube specimens are installed in the material irradiation testing fuel assembly (special fuel) and four special-fuels have been loaded in the core.

The special fuels are irradiated for about 3 to 4 years and then exchange to fresh fuels. However, the material testing capsules are installed again in the fresh special-fuels and those capsules are irradiated continuously. The aim of the irradiation of pressure tube specimen is to check the mechanical toughness periodically. Measurement of radioactivity in pressure tube specimen has been carried out for evaluation purpose.

Shield part

Shield part consists of two sub-parts, i.e. iron-water shield and biological concrete shield. Neutron flux distribution in this part is calculated by neutron transport calculation code, DOT3.5. The neutron flux level in iron-water shield is on the order of 10^{12} to 10^{6}nv. Neutrons are moderated and absorbed by the water in the concrete. The neutron spectrum becomes softer as it diffuse deep into the concrete. Because of the gap (1 meter) between iron-water shield and biological concrete shield, it is possible to access over there and set some irradiation foils during the reactor outage.

Many irradiation foils are also placed around the iron-water shield, such as above or beneath the core. Three kinds of activation foils, those are gold (with/without cadmium cover), cobalt (with/without cadmium cover) and nickel were used. After half year of irradiation, the radioactivity of retrieved foils was measured by gamma spectrometer (Ge detector). Calculation of neutron flux distribution in biological concrete shield is also performed by DOT3.5 code, whereupon radio-activation in concrete is calculated using radioactive calculation code, ORIGEN79. For evaluation purpose, concrete walls were bored and the radioactivity of samples is measured by gamma spectrometer. Three points of biological concrete shield, which are activated enough to measure, are selected. The boring was done from the outer surface of the biological concrete shield since the inner surface is so close to the core that is not easy work. A comparison of element composition in the concrete was performed between measured sample data and NUREG data. There is no big difference between it, especially cobalt and europium which are important nuclides for concrete activation. The result of this shows us that the NUREG data is usable for neutron activation calculation of concrete in Fugen[1]. 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 concrete ingredients[5].

Outer shield part

All of equipment in outer shield part has complicated shape and so the neutrons scattered randomly by these equipments or concrete also. Because of this reason, neutron flux distribution must be evaluated by measurement. Therefore, many large gold foils are installed to measure many points in outer shield part. The neutron spectrum is calculated by Monte Carlo code (MCNP) and the result of measurement of activation foils is used to calculate neutron flux density. It is important to measure gold foils as quickly and precisely since Au-198 has short physical half time and so do the lowness of its activation level. Verifying calculation accuracy, certain concrete is taken as sample. It is concrete which located near the heavy water dump tank and the value of neutron flux approximately 10^{4} n/cm2/sec at the surface. At this sample, Co-60 and Eu-152 are measured precisely with gamma spectrometer (Ge detector)[1].

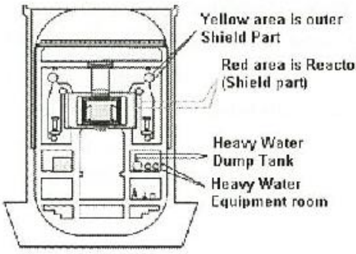

Figure 3. Reactor Vessel and its Region[4]

Activation products in the reactor

The principal activation products present in reactor materials at shutdown are Fe-55, Co-60, Ni-59, Ni-63, Ar-39, Nb-94 (in steels), H-3, C-14, Ca-41, Fe-55, Co-60, Eu-152, Eu-154 (in reinforced concretes) and H-3, C-14, Eu-152, and Eu-154 (in graphites). In terms of radiation levels, Co-60 is the most predominant radionuclide.[6]

METHODS

Materials

The materials used in this study are the data of source term, including the spectrum index of neutron. These data are provided in the library of neutron transport calculation code (DLC185/BUGLE-96). The other is the data of material composition and density that is used as the shield of Fugen reactor, the data of irradiation scheme during the reactor operation and the data of cooling time during reactor shutdown.

Instrumentation

The instrument used in this study is PC (Personal Computer) with the DOT3.5 and ORIGEN79 code installed.

Determining Neutron Flux Distribution in The Reactor

DOT3.5 code is used to calculate the neutron flux distribution. DOT3.5 is a two-dimensional empirical code using discrete ordinates method. It solves the Boltzmann transport equation in two-dimensional geometries. Searches on multiplication factor, time absorption, nuclide concentration, and zone thickness are available for reactor problems.

The input parameter of DOT3.5 code is the dimension of the geometry model, source term parameter, material parameter, and mesh size.3)Dimension of geometry for cylindrical shape model consists of radial distance (X axis) and axial distance (Y axis). The library used In the source term parameter is DLC185/BUGLE-96, with cross section data included.

Calculate Radionuclide Density by Neutron Activation

ORIGEN79 code is used for the quantification of activated nuclides by neutron. It is an isotope generation and depletion code system, matrix exponential method. It uses a matrix exponential method to solve a large system of coupled, linear, first-order ordinary differential equations with constant coefficients.ORGEN79 solves the equations of radioactive growth and decay allowing continuous first order chemical processing and a neutron flux described by a three-region spectrum. Complex decay and transmutation schemes can be treated.

The input parameter of ORIGEN79 code are spectrum index, thermal neutron flux, material composition, irradiation scheme, and cooling time of reactor operation[3]. The spectrum index consists of fast, resonance (epithermal) and thermal neutron flux. Determination of spectrum index in this calculation is the average value from each neutron flux category.

For neutron flux, the maximum value of thermal neutron flux is being used in this calculation. As the input parameter in this code, material composition is the list of elements of the material and the composition (percent of weight). Material composition data for carbon steel (SS41) is taken from NUREG data, while for concrete is based on sample measurement in the plant. The irradiation scheme and cooling time are related. The irradiation scheme is time schedule of the operating and stopping time of reactor in certain cycle during its operation term, while the cooling time is the time after completion of irradiation scheme. In this calculation, the irradiation scheme is divided into seven-step cycles, i.e. cycle 1-5, cycle 6-10, cycle 11-15, cycle 16-20, cycle 21-25, cycle 26-30, cycle 31-34, while the cooling time is divided into nine time scale, those are immediately after termination (initial), cooling for 2, 5, 10, 15, 20, 40, 100, 200 years after termination.

RESULT AND DISCUSSION

The following Figure 4, 5, 6 shows the result of DOT3.5 calculation. It represent the geometry of Fugen reactor (core and shield part), neutron flux contour and radial neutron flux distribution, respectively.

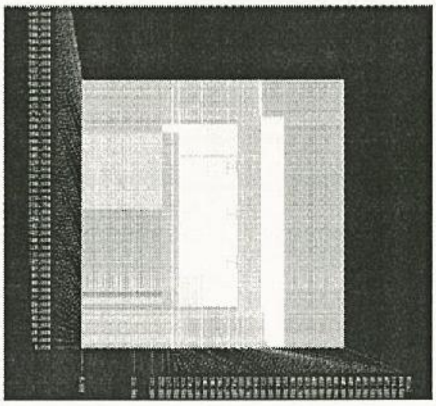

Figure 4. Geometry of Fugen Reactor (Core Part and Shield Part)

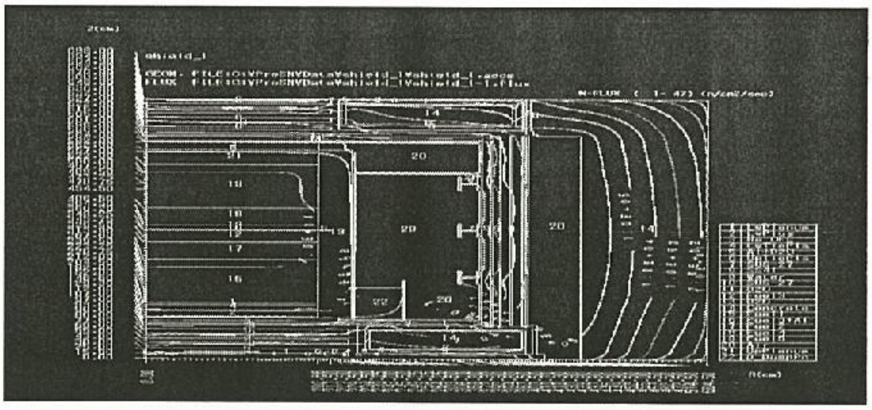

Figure 5. Contour of Neutron Flux in Overall Group of Neutron Energy (1 – 47)

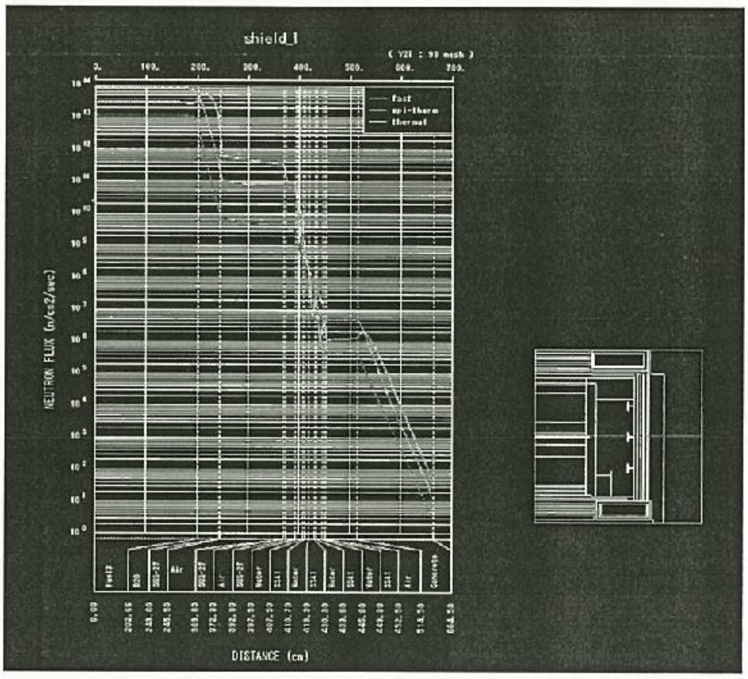

Figure 6. Radial Neutron Flux Distribution in The Axial Centerline of The Fuel Zone

There are too many nuclides have arisen by neutron activation in shield part. The main activation products present in concrete are H-3, C-14, Ca-41, Fe-55, Co-60, Eu-152, and Eu-154, while in steel are Ar-39, Fe-55, Ni-59, Co-60, Ni-63, and Nb-94. However, in this study, the investigation is focused to Co-60, Eu-152 and Eu-154 for biological concrete shield and Co-60, Fe-55 and Ni-63 for carbon-steel. As shown in Table 2, the highest activity of Co-60 found in region 3, as well as Eu-152 and Eu-154. It shows that the highest activity level of concrete shield is at the radial distance from the axial centerline of the fuel zone that is vertical height from 267.6 cm up to 406.5 cm of concrete shield, in details. Based on this, the subsequently observation is performed based on the radionuclide data of this region. The activities of Eu isotopes require consideration, owing to the presence of trace quantities of rare earth elements in source materials used in reactor graphite and bioshield concretes.[6]

Table 2. Activation Product of ConcreteShield in VariousRegions at InitialCoolingTime

Position	Radionuclide activity (Bq/g)		
	Co-60	Eu-152	Eu-154
Region 1	1.21E+00	2.10E+00	2.26E-01
Region 2	3.85E+00	6.59E+00	7.25E-01
Region 3	4.29E+00	7.29E+00	8.07E-01
Region 4	3.96E+00	6.81E+00	7.44E-01
Region 5	2.27E+00	3.89E+00	4.26E-01

Table 3 shows the amount of main radioactivity in the concrete shield in the cooling time period (after termination of Fugen). It shows the radioactivity in Bq/gram of Co-60, Eu-152 and Eu-154 from initial time to 200 years.

Table 3. Activation Product of ConcreteShield in PeriodAfterTermination

Time scale after termination	Radionuclide activity (Bq/g)		
	Co-60	Eu-152	Eu-154
initial	4.29	7.29	8.07E-01
2 years	3.29	6.48	6.77E-01
5 years	2.21	5.44	5.18E-01
10 years	1.14	4.07	3.32E-01
15 years	5.92E-01	3.06	2.13E-01
20 years	3.06E-01	2.29	1.37E-01
40 years	2.20E-02	7.22E-01	2.31E-02
100 years	8.10E-06	2.26E-02	1.11E-04
200 years	1.53E-11	6.99E-05	1.54E-08

As shown in Figure 7, Co-60 and Eu-152 is accounted for the major part of the inventory in the first of ten years after shutdown. Co-60 is negligible in more than 100 years, while Eu-152 and Eu-154 is still present though in slightly number.

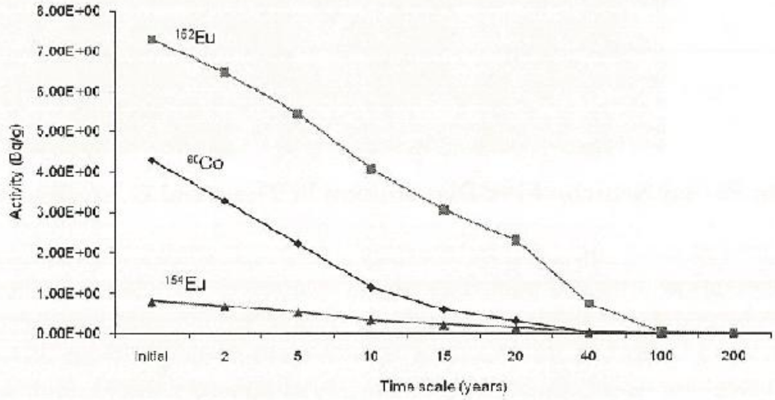

Figure 7. Radioactivity of Activation Product in Biological Concrete Shield in Various Regions at Initial Cooling Time

In the carbon-steel, the highest activity of Co-60, Fe-55, and Ni-63 are found in region1 because of the closest distance from the fuel zone, as shown in Table 4. It shows that the highest activity level of carbon-steel is innermost upright position of iron-water shield perimeter. Fe-55 and Co-60 are the major nuclide in the first fifteen years, while Ni-63 has the lowest amount than both of them. Ni-63 is by far the most abundant activation product expected to be present in a light water reactor (LWR)[6]. Because of the use of light water as reactor coolant, it caused by the abundance of Ni-63.

Table 4. Activation Product of Carbon-Steel

Position	Radionuclide activity (Bq/g)		
	Co-60	Fe-55	Ni-63
Region 1	9.66E+04	2.73E+06	1.33E+04
Region 2	7.29E+03	1.61E+05	7.77E+02
Region 3	7.33E+02	1.28E+04	6.11E+01
Region 4	1.10E+02	2.54E+03	1.23E+01

"Region" is used to define the carbon steel in upright position of iron water shield perimeter. The region number from 1 - 4 represent the sequence from innermost up to outermost gradually.

Table 5 shows the amount of main radioactivity in the carbon-steel in the cooling time period (after termination of Fugen). It shows the radioactivity in Bq/gram of Fe-55, Co-60, and Ni-63 from initial time to 200 years.

As shown in Figure 8, the most significant radioactivity level in carbon-steel during the early period is Fe-55, although it is negligible after 40 years.

Table 5. Activation Product of Carbon-Steel in PeriodAfterTermination

Time scale after termination	Radionuclide activity (Bq/g)		
	Fe-55	Co-60	Ni-63
initial	2.73E+06	9.66E+04	1.33E+04
2 years	1.60E+06	7.40E+04	1.31E+04
5 years	7.18E+05	5.00E+04	1.28E+04
10 years	1.90E+05	2.58E+04	1.24E+04
15 years	5.00E+04	1.34E+04	1.19E+04
20 years	1.32E+04	6.92E+03	1.15E+04
40 years	6.36E+01	4.96E+02	9.88E+03
100 years	7.22E-06	1.82E-01	6.29E+03
200 years	1.90E-17	3.45E-07	2.95E+03

The second significant radionuclide is Co-60, yet it is also negligible after 40 years. However, Ni-63 is still present in relatively fixed amount, so that it's become the most abundant nuclide after about twenty years until 200 years. The comparison result of these nuclides can be seen at Figure 8.

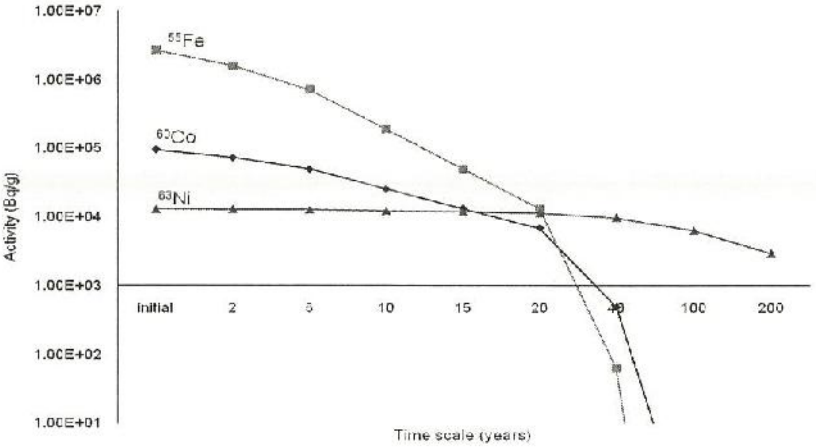

Figure 8. Radioactivity of Activation Product in Carbon-Steel (Iron-WaterShield)

CONCLUSION

Based on valid calculation code that be implemented in Fugen, the main radioactive inventory in shield part has been investigated. In the period around twenty years after termination, the maximum amount of radioactivity in biological concrete shield is contributed by Eu-152 and Co-60, while for carbon-steel is contributed by Fe-55 and Co-60.

In biological concrete shield, the abundance of Eu-154 after long time of termination (over 40 years) is significant enough as occupational radiation exposure, so it needs to be a concern for safety of decommissioning work in that period. However, in carbon-steel, the abundance of Ni-63 after long time of termination (over 40 years) is more significant than Co-60 and Fe-55. The amount of its activity is still high, so it is important to note, for safety of decommissioning work in that period.

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