# Low-Activation Reinforced Concrete Design Methodology -Fundamental Investigation for various Types of Low-Activation Concrete-

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#### Abstract

Concrete is very useful and inexpensive material. However after long-term use in a nuclear powerplant it can become both expensive and difficult. One possible solution to this problem is the use of low-activation concrete, which has much less residual radioactivity even after many years of operation. In this paper, we evaluated fifty raw materials by radiochemical analyses, which assessed the quantities of dominant trace elements for the activation under specific conditions. From these investigations, three kinds of aggregates (fused alumina ceramics, silica sand and limestone) and two kinds of cements (high alumina cement and white cement) were selected as raw materials for the low–activation concrete. Finally, six types of low-activation concrete and two mortars were proposed and their mix proportions described so as to make them suitable for different areas within nuclear plants. Areas of investigation for further improvements were also identified.

# §1. Introduction

Concrete enveloping a nuclear reactor retains residual radioactivity after decommissioning. Disposal of such radioactive concrete is very costly and requires strict supervision. From this point of view, we have developed a new concrete that retains little residual radioactivity -i.e. "low-activation" concrete [1].

A nationally-funded project for comprehensive development of the low-activation concrete has recently started in Japan[2]. The goal of this project is to reduce radioactive concrete by half and ensure the whole structure remains below the clearance level on decommissioning. This will contribute significantly towards solving safety and economic problems relating

Keyword: Low-Activation Concrete, Reactor, Clearance Level, raw materials to the decommissioning of nuclear power plants. The key areas of programme include; investigation of residual radionuclides in construction materials around a reactor, improvement and development of low-activation materials, and establishment of low-activation design method for the reduction of the radioactive waste below clearance level.

To achieve the above comprehensive project, a feasibility study [3] was performed for four tasks including two basic investigation into low-activation concrete as follows.

Proportioning of low-activation raw materials and improvements to low-activation cement: We proposed six kinds of mixes for the low-activation raw

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Criteria for waste	BWR (	1.1GW lev	rel)	PWR (1.1GW level)		GCR (1.1GW level)			
	meta	concrete	total	meta	concrete	total	meta	concrete	total
	1			1			1		
Low level radioactive	0.9	0.4	1.3	0.4	0.2	0.6	0.3	1.8	2.2
waste									
Not necessary to treat as	2.1	0.7	2.8	0.3	0.8	1.2	0.6	3.6	4.2
radioactive waste									
Not radioactive waste	0.8	48.7	49.5	3.4	44.3	47.7	1.0	11.9	12.9
total	3.8	49.8	53.6	4.1	45.4	49.5	1.9	17.3	19.2

Table 1. Estimated waste of the typical nuclear plants after operation

materials to make low-activation concrete, which reduction ratios to the andesite concrete were from 1/300 to 1/10 in  $\Sigma$ Di/Ci unit, where Di is the concentration of the radionuclide i and Ci was the clearance level of the radionuclide i. Three kinds of improvement plans for low-activation cement were also proposed.

Investigation of method for development of low-activation concrete: The methods for development of low-activation concrete and mortal which reduction ratio to the andesite concrete were from 1/300 to 1/10, were investigated to apply to the reactor shielding wall of the boiling water reactor (BWR) and to the shielding wall of the pressurized water reactor(PWR). The results indicated that application of low-activation concrete was economic, greater construction costs being more than outweighed by reduced quantity of radioactive waste requiring costly disposal

From the results of the feasibility study, we describe outline of the results relating to both the concrete itself the raw. Fundamental technical issues are discussed first, followed by investigation of the potential low-activation. Finally, six types of low-activation concrete are proposed. Their mix proportions are assessed against target characteristics, considering the application of the proposed concrete to the certain portion of the member in nuclear plants.

# §2. Criteria of clearance level for radioactive concrete waste

[Unit:  $10^7$  kg]

As well known, concrete is a very useful and inexpensive material, and is used in variety of applications, including buildings and infrastructure. Concrete is also used in nuclear plants as a structural member and shielding wall against radiation and radioactivity. Certain radiation, typically neutron radiation, may make concrete radioactive under certain conditions. This phenomenon is called "activation", and <del>which</del> concrete subjected to it can be called "activated concrete" [4].

In this case, the concrete can become expensive and hard to deal with. Activated concrete may need to be treated as radioactive waste at the end of the operation. In this case, there are major cost implications associated with specialist demolition, and underground storage. The costs of dealing with radioactive waste are estimated as between 65 times and 2800 times greater than construction costs for ordinary concrete (based on three differing levels of radioactivity for two type of reactors, as outlined in an intermediate report by Ministry of International Trade and Industry) [5].

However, most of the concrete used in the nuclear plant is not highly radioactive waste requiring strict supervision. Table 1 gives an idea of the quantity of the waste for the typical nuclear plant [5]. This table shows that most of the waste

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from nuclear plants is concrete and most of the not concrete waste is radioactive waste. Nevertheless, concrete radioactive waste is still 4000 ton for the BWR (1.1GW level). The remaining 7000 ton is categorized as the waste which is not necessary to treat as the radioactive waste. This waste is termed as "below clearance level". So the possibility exists, if sufficiently low-activation concrete could be developed, that all concrete used in the structure could be below clearance level on decommissioning.

Clearance level has been discussed to determine the line between regular waste and low-level radioactive waste (LLW). International Atomic Energy Agency (IAEA) has provided international guidelines for the LLW, specifying "basic safety standard" for LLW in the report "TECDOC-855" [6] in 1996, based on the principle of basic safety for the protection against the ionizing radiation and for the safety of radiation sources[7]. TECDOC-855 defines criteria to define trivial radiation level which poses negligible risk based on increased cancer risk of one per million per year, which translates to 100 µSv/y, comparing to 1000 uSv/v for individual radiation allowance recommended by International Committee for **TECDOC-855** Radiation Protection. So. recommended 10 µSv/y as clearance level criteria.

Japan Nuclear Safety Commission (JNSC) reported clearance level from exposure estimation performed using food-chain scenarios localized to the Japanese lifestyle [8], based on above IAEA 10  $\mu$ Sv/y criteria (describing "CL-Japan1999"). On the other hand, after above first report from IAEA, they reevaluated TECDOC-855 and issued RS-G-1.7 for clearance level ([9], describing "CL-IAEA2004"). JNSC also reevaluated exposure estimation using RS-G-1.7 and regulated in 2004(describing "CL-Japan2004").

# §3. Investigation for low-activation materials

Based on previous works [10], [11], [12], 45 raw

materials for concrete were selected as candidates for low-activation materials for low-activation concrete. Aggregates (30 kinds of limestone, 6 kinds of fused alumina aggregates, and 4 kinds of quartz aggregates including silica sand) and cements (a white cement and 5 kinds of high alumina cements) were investigated. Geo standard samples (JR-1, JA-1 and JB-1), standard ordinary concrete, and ordinary Portland cement were also selected as comparison samples for the above materials. Standard ordinary concrete was made from andesite aggregates, which was is the most common aggregate for the concrete in Japan, and ordinary Portland cement.

The term "low-activation" can have several meanings. For the purposes of this report, we take the term low-activation concrete to mean concrete that remains is below clearance level, even after long-term use in most portions of nuclear plants. We consider "long-term use" to comprise 40 years operation and 6 years cooling (maintaining after stop of the operation for 6 years).

Based on above conditions, radioactive nuclides in the concrete should be limited (typically  $^{60}$ Co,  $^{134}$ Cs and  $^{152}$ Eu), and therefore trace elements for the investigation were selected Co, Cs, Sc, Fe and Eu which is are representive of rare earth elements.

# 3.1 Radiochemical analysis

The trace elements predominantly responsible for the activation of concrete materials were evaluated by radiochemical analyses [4], as follows,

1. Collecting certain concrete materials



Figure 1 Configuration of the JRR-4 core

2. Crushing materials to certain size (typically under 1mm)

3. Packing above crushed samples for 0.1 to 1g with special treatment for irradiation

4. Irradiation by thermal neutron in the reactor core of JRR-4 (shown in Figure 1)

Thermal neutron flux:5.3E13 cm^2sec^1Irradiation time:20 minutes

5. Cooling for 66 to 87 days

6. Measurement of gamma spectrum for irradiated samples by a Ge detector

7. Evaluation of quantity of the trace elements for each sample.

# 3.2 Results for aggregates

From the above radiochemical analyses, quantities of the Co, Cs, Eu, Fe and Sc for 50 samples were evaluated. Figure 2 shows the distribution of the measured quantities of Eu and Co in typical aggregate samples (limestone aggregates, fused alumina aggregates and quartz aggregates) with those of other aggregates from previous works [10],[12]. This figure indicates above selected samples for this work have low levels of Eu

#### Table 2 $\Sigma^{5}$ Di/Ci ratio for evaluated aggregates

Aggregate	Σ <sup>5</sup> D/C ratio*
Fused alumina aggregate CA	0.00068
Fused alumina aggregate JA	0.0026
Fused alumina aggregate JB	0.0021
Fused alumina aggregate CF	0.0014
Fused alumina aggregate EA	0.00050
Quartz sand JT	0.0057
Quartz sand JA	0.65
Quartz sand AF	0.0049
Quartz aggregate IA	0.00076
Limestone aggregate FO	0.014
Limestone aggregate OK	0.038
Limestone aggregate HK	0.020
Limestone aggregate FA	0.0067
Limestone aggregate FK	0.023
Limestone aggregate TH	0.0085
Limestone aggregate AH	0.0054
Limestone aggregate SB	0.011
Limestone aggregate KT	0.011

<sup>\*</sup>The ratios is to the average of JR-1, JB-1, and JA-1, which is assumed the average aggregate.

and Co compared to those in ordinary aggregates (the distribution of Eu and Co for ordinary aggregates is located around the center of figure,



Figure 2 Distribution of quantities for Eu and Co in aggregates with enlargement of ordinary aggregates (Numbers in parentheses indicate the number of specimens )

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(Numbers in parentheses indicate the number of specimens)

and enlarged figure is also shown in Figure 3).

Figure 2 also has index curves of  $\Sigma^3$ Di/Ci=0.1 and  $\Sigma^3$ Di/Ci=0.1 for the activation. The  $\Sigma^3$ Di/Ci in this figure is defined by equation (1) as follows,  $\Sigma^3$ Di/Ci = D<sub>152Eu</sub>/C<sub>152Eu</sub> + D<sub>154Eu</sub>/C<sub>154Eu</sub>

 $+D_{60Co}/C_{60Co}$  (1)

Di:Concentration of radionuclide of  $^{152}Eu$ ,  $^{154}Eu$  and  $^{60}Co$  induced under  $2.0 \times 10^5$  n cm<sup>-2</sup>sec<sup>-1</sup> thermal

neutrons, 40 years of operation, 6 years of cooling. Ci:Clearance level referring CL-Japan2004 for this calculation, which are 0.1 for  $^{152}$ Eu,  $^{154}$ Eu and  $^{60}$ Co.

Measured limestone samples in the figure are located within <sup>3</sup>Di/Ci =1.0 line, and fused alumina ceramics are located within <sup>3</sup>Di/Ci =0.1, in comparison with other aggregates against  $\Sigma$ <sup>3</sup>Di/Ci curve. Therefore limestone, quartz, and fused alumina were selected as low-activation materials.

Table 2 shows the  $\Sigma^5$ Di/Ci ratio of the measured aggregates to the ordinary aggregate, which is average of

the Geo-standard samples JR-1 (Rhyolite), JB-1 (Basalt) and JA-1 (Andesite). <sup>5</sup>Di/Ci and is defined by equation (2), as follows,

$$\Sigma^{5}\text{Di/Ci} = D_{55\text{Fe}}/C_{55\text{Fe}} + D_{60\text{Co}}/C_{60\text{Co}} + D_{134\text{Cs}}/C_{134\text{Cs}} + D_{152\text{Eu}}/C_{152\text{Eu}} + D_{154\text{Eu}}/C_{154\text{Eu}} (2)$$

Di: Concentration of radionuclide of  ${}^{55}$ Fe,  ${}^{60}$ Co,  ${}^{134}$ Cs,  ${}^{152}$ Eu, and  ${}^{154}$ Eu induced under  $2.0 \times 10^5$  n cm<sup>-2</sup>sec<sup>-1</sup> thermal neutrons, 40 years of operation, 6 years of cooling.

Ci:Clearance level referring CL-Japan2004 for this calculation, which are 0.1 for  $^{152}$ Eu,  $^{154}$ Eu,  $^{60}$ Co,



Figure 4 Distribution of quantities for Eu and Co in cement

Cement	Σ <sup>5</sup> D/C ratio*
High alumina cement FR	0.045
High alumina cement EA	0.045
High alumina cement JA	0.0040
High alumina cement JB	0.056
High alumina cement JC	0.021
White cement S	0.35
Low heated Portland cement T	1.0

Table 3Σ⁵Di/Ci	ratio for evaluated cements
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<sup>\*</sup>The ratio is to the ordinary Portland cement, which is assumed the average cement

#### <sup>134</sup>Cs, and 1000 for <sup>55</sup>Fe.

Figure 2 and Table 2 shows that aggregates selected in this work are very useful for low-activation materials (the  $\Sigma$  Di/Ci ratio of fused alumina, quartz sand and limestone to the ordinary aggregates are about 1/1000, 1/200, and 1/20, respectively).

#### 3.3 Results for cements

Figure 4 compares the distribution of Eu and Co in the cements selected for this work with cements examined in previous works[10],[12].  $\Sigma$  <sup>3</sup>Di/Ci curves, which can be calculated by equation (1), are also set for the index of the activation in the figure.

Table 3 shows the ratio of Σ<sup>5</sup> Di/Ci, which is calculated by equation (2), for the evaluated some cements compared to the ordinary Portland cement manufactured by A factory in Japan.

Figure 4 and Table 3 indicate that the selected aggregates in this work are very useful for the low-activation materials (the $\Sigma^5$ Di/Ci ratio of high alumina cement, and white cement to the ordinary Portland cement are about 1/20 - 1/50 and 1/3, respectively).

#### §4. Proposal of low-activation concrete

The investigation in the last section is introduced possible low-activation materials for the low-activation concrete. Based on that investigation, various types of low-activation concrete are described below.

# 4..1 Supplement conditions for the low-activation aggregates and cements

The investigation assessed three aggregates and two cements as potential low-activation materials for low-activation concrete from the points of view of the activation by radiochemical analyses. Obviously, it is also very important to investigate other conditions in actual use of the

Material	ΣDi/Ci* ratio	Relative cost**	Ability for	Point characteristic
Fused alumina aggregate	1/400 — 1/1500	20- 40	Good	High density and hardness
Quartzite (Silica) aggregate	1/200 - 1/1200	5 - 10	Good	Hardness
Limestone aggregate	1/30 – 1/200	1.5 - 3	Good (partially N.G)	Require washing
High alumina cement	High alumina cement 1/20 – 1/50 30		Good	Long term durability, thixotropy
White cement	1/3	3	Good	Heat generation

Table 4 Supplement condition for low-activation materials

<sup>\*</sup>Di: Concentration of radionuclide i, Ci: Clearance level of radionuclide i, cited from IAEA-RS-G1.7, assuming the thermal neutron  $2.0 \times 10^5 n_{th}$  cm<sup>-2</sup>s<sup>-1</sup>, 40 years of operation, and 6 years of cooling.

\*\*The average aggregate is assumed 1.0 for aggregates, and the ordinary Portland cement is assumed 1.0 for cements.

above materials for concrete.

Table 4 shows the results of the investigation related to the supplement for the above materials

such as relative cost, ability of the supplement and so on. This investigation was performed to the company for each of the aggregates and cements investigated in the previous last section, by means of enquiries, site visits and orders of a specific quantity. The  $\Sigma$ Di/Ci ratios of the above materials to compared with the assumed average materials (the average of the geo standard samples for aggregates and ordinary Portland cement for cements) are also shown in the table. The  $\Sigma$ Di/Ci for each material in this table is recalculated under the new clearance level (CL-Japan2004) focusing on actual nuclear plants. The point characteristics for each material are also addressed in the table. Relative cost compares the price of low-activation materials to the ordinary materials (average aggregate and Portland cement).

This investigation confirmed realization of the use of low-activation aggregates and cements listed above for the low-activation concrete.

# 4.2 Mix proportion design

The above investigation reveals some potential low-activation concretes. Table 5 summarizes the mix designs for the six types of the proposed low-activation concrete together with the target of characteristics. In this table,  $\Sigma$ Di/Ci ratio, which is the ratio of  $\Sigma$ Di/Ci for each designed low-activation concrete is relative to that for the Andesite concrete (Andesite aggregate and ordinary Portland cement) In order to apply low-activation concrete to certain portions of the nuclear plants effectively, the aspects of concrete examined for this study, such as compressive strength, shrinkages, and fresh concrete properties are not on their own sufficient determinants of suitability. Other conditions, such as cost, durability, low-activation, and so on, should be taken into account. Therefore low-activation design could be very complex and difficult. Table 6 summarizes six types of proposed concrete including two types of mortar and compares their characteristics, target applications and assignments for improvement.

Based on above mix proportions, dozens of trial mixing and execution experiments have been performed for various types of concrete and mortar [13],[14]. Figure 5 shows typical results for  $\Sigma$ Di/Ci of concrete A, concrete C, and concrete D in table 5 and table 6 with the comparisons to of ordinary concrete (OC: Andesite concrete). The values of  $\Sigma$ Di/Ci were calculated under the conditions of 2.0  $\times 10^5$  n<sub>th</sub> cm<sup>-2</sup>s<sup>-1</sup>, only thermal neutron, 40 years of operation and 6 years of cooling, and assuming BSW in BWR and a clearance level as defined in CL-Japan2004. This figure shows the results for only four radionuclides, which occupied comprised more than 98 % to of the total  $\Sigma$ Di/Ci except concrete A, for which the four radionuclides

	Coarse	Eine e nome nete	Comont	Target of characteristics		
	aggregate		Cement	ΣDi/Ci ratio*	Density (g/cm <sup>3</sup> )	
А	Fused alumina ceramics	Fused alumina ceramics	High alumina cement	1/300 – 1/400	3.0	
В	Quartzite	Silica sand	High alumina cement	1/150 -1/200	2.3	
С	Limestone	Limestone	White cement	1/30-1/50	2.3	
D	Limestone	Limestone	Low heated cement	1/10-1/30	2.3	
Е	Silica sand		High alumina cement	1/150	2.3	
F	Silica sand + Limestone powder		White cement	1/25	2.1	

<sup>\*</sup> Andesite concrete is assumed 1.0.

	Characteristics	Target application	Assignment Areas for improvement
А	Ultra low-activation (2Di/Ci ratio is expected 1/300), high density, good property for heat resistance and high thermal conductivity	Reactor shielding wall for BWR, inner portion of shielding wall for PWR, and partial spot for high neutron yield	Expensive, high drying shrinkage, Long term durability, thixotropy, heat generation and difficulty for execution work
В	Very low-activation (ΣDi/Ci ratio is expected 1/150)	Reactor shielding wall for BWR, inner portion of shielding wall for PWR, and partial spot for high neutron yield	Expensive, high drying shrinkage, Long term durability, thixotropy, heat generation, difficulty for execution work and not for use in high temperature
C	Low-activation (ΣDi/Ci ratio is expected 1/30 to 1/50) and could be moderate type as balance of cost and performance.	Biological shielding wall for BWR, and outer portion of shielding wall for PWR	Further improvement of white cement for low-activation, and heat generation during execution work
D	Relatively low-activation ( $\Sigma$ Di/Ci ratio is expected 1/10 to 1/30), and inexpensive Limestone	Biological shielding wall for BWR, and outer portion of shielding wall for PWR	Further improvement of low heated cement for low-activation
Е	Very low-activation (2Di/Ci ratio is expected 1/300) and good property of filling	Reactor shielding wall for BWR, inner portion of shielding wall for PWR, partial spot for high neutron yield and infilled mortar	Expensive, high drying shrinkage, Long term durability, thixotropy, heat generation, difficulty for execution work and not for use in high temperature
F	Low-activation (ΣDi/Ci ratio is expected 1/25), and good property of filling	Biological shielding wall for BWR, outer portion of shielding wall for PWR, and infilled mortar	Further improvement of white cement for low-activation, and heat generation during execution work

#### Table 6 Characteristics of six types of Low-activation Concrete

comprised is about 90 %. By From the results of these calculations, the  $\Sigma$ Di/Ci ratio of concrete A to OC is 1/323, that of concrete C to OC is 1/31 and that of concrete D to OC is 1/10, respectively.

# 4.3 Areas for further improvement

Most of the problems described in table 6 for the six proposed low-activation concretes are caused by the cements. The problems for the concrete where high alumina cement is used are typically high drying shrinkage, long term durability, thixotropy, and heat generation. Those for the concrete where white cement is used are heat generation and further improvement for low-activation and the assignment for the concrete in use of low heated cement is further improvement of low-activation.

In order to achieve further low-activation cement for white cement and low heated cement, raw materials used in the manufacturing stage should be carefully selected, especially Co and Eu content of in the cement. As mentioned in section 3, Eu and Co are dominant materials for activation under the conditions described in this paper.

Heat generation during execution work may cause unacceptable cracks [15], so it is also very important for the decreasing the heat generation. We have developed a method to reduce the heat generation for the concrete with white cement by adding a low-activation admixture. This could be one of the ways to improve heat generation for the concrete with white cements and alumina cements.

Long term durability is another problem for use of high alumina cements, and may improve by the using a mix proportion with low W/C (the ratio of water to cement) under 40% [16].

#### §5. Conclusion

In 2004, a project began to design

low-activation concrete, in order to reduce radioactivity of nuclear plant decommissioning waste below clearance level The feasibility project has four main aims: investigation of major construction materials and low-activation materials, determination of representative part and establishment of calculation and evaluation methods, proportioning of low-activation raw materials to make low-activation concrete, and investigation of methods for low-activation concrete and mortar. In this paper, the results of the feasibility studies related to concrete and its raw materials were mainly discussed.

As fundamental issues, criteria of the clearance level for radioactive concrete waste were addressed as a background of the comprehensive development. Fifty kinds of raw materials potentially for the low-activation concrete were investigated by radiochemical analyses, in order to the quantities of trace estimate elements dominated to the activation in radioactive concrete waste in certain condition. Limestone aggregates, quartz including silica sand, and fused alumina ceramics were selected as low-activation aggregates, and high alumina cements and a white cement were selected as low-activation cements, which were similar results as previous works.

Based on the above investigations, six types of low-activation concrete were proposed, which were Concrete A (fused alumina ceramics aggregates and high alumina cement), Concrete B (quartz aggregates including silica sand and high alumina cement), Concrete C (limestone aggregates and white cement), Concrete D (limestone aggregates and low heated cement), Mortal E (fused alumina ceramics aggregates including silica sand and high alumina cement) and Mortal F (silica sand including limestone powder and white cement).

The mix proportion designs were listed with characteristics, assignments, and target application in the nuclear plants for each proposed concrete. These assignments of the proposed concrete also conducted to the necessary points of further improvement for cements, which were discussed in this paper.

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**short comment** Low-Activation Concrete is originated for more than twenty years, and recently we started national funded project with Tohoku University and seven companies, aiming to future replacement of nuclear reactor.

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