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Cooling Time Determination of Spent Nuclear Fuel by Detection of Activity Ratio $^{144}Ce/^{137}Cs$

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방사능비 $^{144}Ce/^{137}Cs$ 검출에 의한 사용후핵연료 냉각기간 결정

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Abstract

Activity ratio of two radioactive primary fission products which had sufficiently different half-lives was expressed as functions of cooling time and irradiation histories in which average burnup, irradiation time, cycle interval time and the dominant fissile material of the spent fuel were included. The gamma-ray spectra of 36 samples from 6 spent PWR fuel assemblies irradiated in Kori unit-1 reactor were obtained by a spectrometric system equipped with a high purity germanium gamma-ray detector. Activity ratio $^{144}Ce/^{137}Cs$, analyzed from each spectrum, was used for the calculation of cooling time.

The results show that the radioactive fission products ^{144}Ce and ^{137}Cs are considered as useful monitors for cooling time determination because the estimated cooling time by detection of activity ratio $^{144}Ce/^{137}Cs$ agreed well with the operator declared cooling time within relative difference of ± 5 % despite the low counting rate of the gamma-ray of ^{144}Ce (about 10^{-3} count per second). For the samples with several different irradiation histories, the determined cooling time by modeled irradiation history showed good agreement with that by known irradiation history within time difference of ± 0.5 year. From this result, it would be expected to be possible to estimate reliably the cooling time of spent nuclear fuel without the exact information about irradiation history.

The feasibility study on identification of and/or sorting out spent nuclear fuel by applying the technique for cooling time determination was also performed and the result shows that the detection of activity ratio $^{144}Ce/^{137}Cs$ by gamma-ray spectrometry would be usefully applicable to certify spent nuclear fuel for the purpose of safeguards and management in a facility in which the samples dismantled or cut from spent fuel assemblies are treated, such as the post irradiation examination facility.

요 약

반감기가 서로 크게 다른 두 종류의 방사성 직접핵분열생성물의 방사능비(activity ratio)를 핵연

료의 냉각기간 및 연소이력의 함수로 표현하였으며 연소이력에는 사용후핵연료의 평균연소도, 연소기간, 연소주기간의 간격 그리고 주 핵분열물질등이 포함되었다. 고리 1호기에서 연소된 6개의 사용후 가압경수로(PWR) 핵연료 집합체로 부터 36개의 시료를 제작하여 이들 시료에 대한 감마선 스펙트럼을 고순도(HP) Ge 검출기를 사용하여 수집한 후, 각 스펙트럼을 분석하여 얻은 방사능비 $^{144}Ce/^{137}Cs$를 이용하여 냉각기간을 계산하였다.

그 결과 ^{144}Ce 감마선 검출계수율이 10^{-3} cps(count per second) 정도로 아주 낮았음에도 불구하고 검출된 방사능비 $^{144}Ce/^{137}Cs$를 사용하여 구한 냉각기간은 원자로 운전기록에 의한 냉각기간(operator declared cooling time)과 상대적인 차이가 ±5 % 이내로 잘 일치한 것으로 부터 핵분열생성물 ^{144}Ce 및 ^{137}Cs은 냉각기간 결정을 위한 좋은 모니터가 됨을 확인하였다. 여러가지의 연소이력을 갖는 핵연료를 대상으로 한 본 실험의 경우, 단순하게 모델화한 연소이력을 대입하여 얻은 냉각기간은 실제 연소이력을 대입하여 얻은 냉각기간과 시간차이가 ±0.5년 이내에서 잘 맞았으며 이로부터 연소이력에 대한 정확한 정보 없이도 신뢰할 수 있는 정도의 냉각기간을 추정하는 것이 가능할 것으로 생각되었다.

아울러 냉각기간 결정을 위한 본 기술을 활용한 사용후핵연료의 증명 및/또는 분류에 대한 타당성 연구를 한 결과 감마선 분광분석 방법으로 검출한 방사능비 $^{144}Ce/^{137}Cs$에 의해서 결정된 냉각기간은, 조사후시험시설(post irradiation examination facility)등과 같이 사용후핵연료 집합체를 해체 또는 절단하여 만든 시료를 취급하는 시설등에서, 사용후핵연료에 대한 안전관리 및 계량관리를 위하여 유용하게 활용될 수 있을 것으로 사료되었다.

1. Introduction

The information on cooling time is one of important parameters for safeguards of spent nuclear material and it is used as the input data for gamma-ray spectrometric burnup determination of spent fuel[1~4]. Activity ratio of two radioactive primary fission products which have quite different half-lives can be used as a monitor for cooling time determination because it can be expressed as functions of cooling time and a factor(or a parameter) concerned with the irradiation history of examined spent fuel. The relationship among these terms is described in the studies of H. Graber et al.[5] and J.R. Phillips et al.[6, 7].

If it is possible to get the detailed information on irradiation history, the equation for cooling time estimation contains only the term of activity ratio which is able to be measured by means of gamma-ray spectroscopy and thus the main purpose of cooling time estimation under this condition would be to verify the consistency between the experimentally estimated cooling time and the operator declared cooling time based on the operational log. When the detailed irradiation history is obscure or unknown, an acceptable irradiation history model must be applied or the monitor activity ratio which is practically independent of irradiation history must be chosen if possible.

According to I. Ursu et al.[8]'s study on cooling time determination, the cooling time measurements can be performed with errors between 2 and 15 % if activity ratio ^{144}Ce (696 keV)/^{137}Cs (662 keV) is used as a monitor for cooling time from 1.5 to 3 years and if activity ratio ^{144}Ce(2,186 keV)/^{137}Cs(662 keV) is used as a monitor for cooling time from 3 to 7 years. In the study of H. Graber et al.[5], activity ratio $^{144}Ce/^{137}Cs$, following correction for the irradiation history, appears to be a suitable monitor for determining cooling time between 0.5 and 5 years with a mean error of ±5 % and this uncertainty becomes greater when approximated irradiation history is used. Detailed requirements for monitor

isotope selection are described in the study of G.L. Hanna[9].

The interests focused in this study are first to confirm the accuracy of estimated cooling time by applying known irradiation history in comparison with the operator declared cooling time for the PWR fuels cooled from 5 up to 13 years, secondly to compare the measured cooling time by modeled irradiation history with that by known irradiation history in order to find the uncertainty caused by the simplification of irradiation history, and finally to study the applicability of the cooling time estimation techniques for safeguards and management of spent nuclear materials.

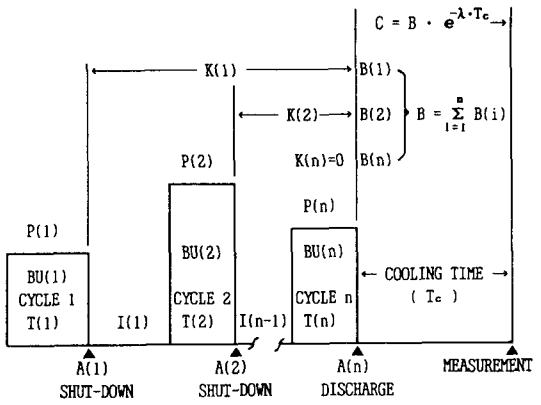

Fig. 1. Schematic Diagram of Irradiation History for Cooling Time Determination

2. Theory

The cooling time determination of spent nuclear fuel irradiated in a commercial power reactor was performed under the following assumptions (see Fig. 1):

1) Fuel was irradiated under a constant power during a cycle.

$P(i) \equiv k \cdot (BU(i)/T(i))$

where

k : constant

P(i) : power of cycle i

BU(i) : average burnup of cycle i

T(i) : irradiation time of cycle i

2) Monitor isotopes were generated only by fissioning of one type fissile material such as ^{235}U or ^{239}Pu, and they were depleted only by radioactive decay.

$$A_a(i) \equiv k \cdot (BU(i)/T(i)) \cdot Y_a \cdot (1-e^{-\lambda_a \cdot T(i)}) \quad (1)$$

where

$A_a(i)$: activity of isotope a at the end of cycle i

Y_a : fission yield of isotope a

λ_a : decay constant of isotope a

Based on the above assumptions, the residual activity of $A_a(i)$ at discharged date, $B_a(i)$, can be expressed by the following equation :

$$B_a(i) = A_a(i) \cdot e^{-\lambda_a \cdot K(i)}$$

where

$K(i) = \sum_{j=i+1}^{n} T(j) + \sum_{j=i}^{n-1} I(j)$ and

I(j) : interval time between two cycles j and j+1

Total activity of isotope a at discharged date, B_a, is given by the summation of $B_a(i)$.

$$B_a = \sum_{i=1}^{n} B_a(i)$$

Activity of isotope a at measured date, C_a, is given by multiplying B_a by exponential decay term in which cooling time is included.

$$C_a = B_a \cdot e^{-\lambda_a \cdot T_c} \quad (2)$$

Here, the time difference between 'measurement' and 'discharge' is defined as cooling time T_c which is the very one to be determined by detection of monitor activity ratio in this paper.

The equation for isotope b can be obtained by inserting subscript 'b' instead of 'a'.

$$C_b = B_b \cdot e^{-\lambda_b \cdot T_c} \quad (3)$$

Activity ratio of two isotopes a and b can be given by dividing Eq.(2) with Eq.(3).

$$A_b^a \equiv \frac{C_a}{C_b} = \frac{B_a}{B_b} \cdot e^{(\lambda_b - \lambda_a) \cdot T_c} = H_b^a \cdot e^{(\lambda_b - \lambda_a) \cdot T_c} \quad (4)$$

where $H_b^a = \frac{B_a}{B_b}$

Now, the arbitrary constant k in Eq.(1) is eliminated by Eq.(4). The parameter H_b^a can be evaluated by the information about irradiation history of the examined spent fuel in which dominant fissile material, irradiation time, interval time between cycles and relative cycle burnup are included. If the irradiation history is known, that's to say if the value of H_b^a is given, activity ratio A_b^a depends only on cooling time T_c.

To obtain cooling time T_c by means of gamma-ray spectrometry, the activity ratio should be represented by using the following experimental terms :

$$A_b^a=\frac{C_a}{C_b}=\frac{N_a^u\cdot[\epsilon^u]^{-1}\cdot[BR_a^u]^{-1}}{N_b^v\cdot[\epsilon^v]^{-1}\cdot[BR_b^v]^{-1}} \quad (5)$$

where

N_a^u, N_b^v : detection intensities of isotope a's energy u and isotope b's energy v, respectively

ϵ^u, ϵ^v : detection efficiencies of energy u and v, respectively

BR_a^u, BR_b^v : branching ratios of isotope a's energy u and isotope b's energy v, respectively

Substituting Eq.(5) into Eq.(4) results in the equation for cooling time calculation.

$$T_c=\frac{1}{\lambda_b-\lambda_a}\cdot\ell n\left(\frac{N_a^u}{N_b^v}\cdot\frac{\epsilon^v}{\epsilon^u}\cdot\frac{1}{H_b^a}\cdot\frac{BR_b^v}{BR_a^u}\right) \quad (6)$$

If the irradiation history of spent fuel is known, the cooling time can be directly calculated by inserting the experimentally measured data into Eq.(6). In this case the experimentally measured cooling time(MCT) can be compared with the operator declared cooling time(DCT).

If the irradiation history is ambiguous or unknown, the cooling time can be estimated by modeling the irradiation history to evaluate H_b^a in Eq.(6). Once the cooling time of a sample is given by Eq.(6), the following relationship can be used to calculate the cooling time of different model easily :

$$T_c(n)=T_c(m)+\frac{1}{\lambda_b-\lambda_a}\cdot\ell n\frac{H_b^a(m)}{H_b^a(n)} \quad (7)$$

where

$T_c(m)$, $T_c(n)$: cooling time by model m and n, respectively

$H_b^a(m)$, $H_b^a(n)$: H_b^a of model m and n, respectively

The MCT by modeling of irradiation history can be compared with the DCT and with the MCT by known irradiation history if the detailed information about the irradiation history of the examined spent fuel is available.

3. Experimental

Fig. 2 shows the gamma-ray spectrum of the spent nuclear fuel cooled of about 8 years after discharge from a commercial PWR[10]. The radioisotopes with intensive gamma-ray peaks in the spectrum are identified as ^{134}Cs, ^{137}Cs, ^{144}Ce and ^{154}Eu. ^{134}Cs is primarily produced by the neutron activation of ^{133}Cs that comes from ^{133}I through ^{133}Xe, and ^{154}Eu is essentially a product not from the fissioning of ^{235}U and ^{239}Pu but from the multiple neutron absorption of lower mass fission products[7]. The radioisotopes having relatively short half-life, such as ^{95}Zr and ^{106}Ru, are

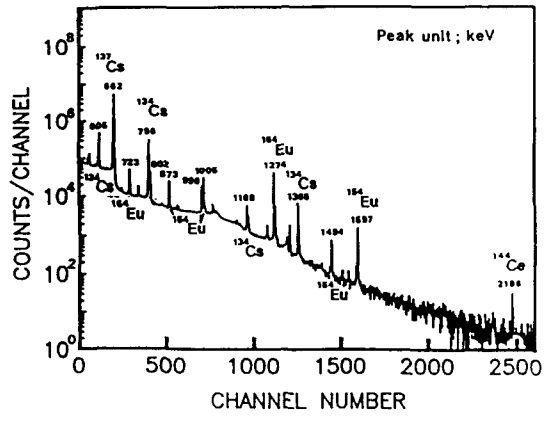

Fig. 2. Gamma-ray Spectrum Obtained from Spent PWR Fuel with Cooling Time of about 8 Years

not observed in the spectrum. The full energy peak of ^{144}Ce shown conspicuously in the spectrum is only 2,186 keV because its 696 keV which has the highest branching ratio is immersed in the background due to the Compton continuum of the energies higher than 696 keV. Therefore, the only available monitor activity ratio for cooling time estimation is ^{144}Ce(2,186 keV)/^{137}Cs(662 keV) for the spent fuel cooled over several years.

Table 1 shows the nuclear data used for cooling time determination of two direct fission products ^{144}Ce with half-life of 285 days and ^{137}Cs with that of 30.2 years. In this table, decay constant, gamma-ray energy and branching ratio were taken from Refs. 11 and 12, and fission yield was taken from Refs. 13 and 14. Decay constant of ^{144}Ce is about 39 times higher than that of ^{137}Cs due to their half-lives and the branching ratio of ^{144}Ce's 2,186 keV is only 1/123 of that of ^{137}Cs's 662 keV.

The fission yield ratio of two monitor isotopes, Y_{Ce-144}/Y_{Cs-137}, which is implied in parameter H^{Ce-144}_{Cs-137} of Eq.(6) can be evaluated when the information denoting dominant fissile material is available. Thus, cooling time uncertainty from 0 up to 0.47 year will be added when the information about dominant fissile material is incorrectly adopted because the fission yield of ^{144}Ce considerably depends on ^{235}U and ^{239}Pu. Here, 0.47 year was calculated on the basis of $1/(0.0230-0.8879)\cdot \ell n((6.26/5.39)/(6.65/3.80))=0.47$.

Table 1. Two Direct Fission Products and Their Nuclear Data Used for Cooling Time Determination

Nuclear Data	^{144}Ce	^{137}Cs
Decay Constant(1/year), λ	0.8879	0.0230
Gamm-ray Energy(keV)	2,185.6	661.6
Branching Ratio(%),BR	0.694	85.21
Fission Yield(%),Y(^{235}U)	5.39	6.26
Y(^{239}Pu)	3.80	6.65

In Table 2, the irradiation histories of 6 spent PWR fuel assemblies denoted as A,B,C,D,E and F which were irradiated in Kori unit-1 reactor and 17 spent fuel rods extracted from these assemblies are listed. One to three positions in a rod were selected and then sample of about 3 mm thick per position was prepared by cutting the rod cross-sectionally.

For the calculation of parameter H^{Ce-144}_{Cs-137}, the fission yield of ^{235}U was used because the fuel of the assemblies listed in the table had the initial ^{235}U enrichments of 2.122, 3.199 and 3.210 weight % and the dominant fissioning of the fuel took place in ^{235}U. Irradiation time T(i) and interval time I(i) of assembly, and relative cycle-burnup BU(i) of rod in the table were also used for the calculation of parameter H^{Ce-144}_{Cs-137}.

For the convenience of no requirement of relative cycle burnup, continuous irradiation without interval time between cycles under a constant power during irradiation period was taken as the modeled irradiation history in this study, and irradiation time of 3 years was chosen for this model because the spent fuel normally irradiated in a commercial PWR had its irradiation time in the neighborhood of 3 years. The calculated value of parameter H^{Ce-144}_{Cs-137} based on this model becomes 12.002 by putting irradiation time T(1)=3 years.

The activity ratio A^{Ce-144}_{Cs-137} given in Fig. 3 as a function of cooling time was theoretically calculated by means of Eq.(4). In this figure, activity ratio decreases exponentially as the cooling time T_c increases. The intersection at vertical axis is equal to the value of the irradiation history parameter H^{Ce-144}_{Cs-137}. The slope is related with the decay constant difference of two monitor isotopes, $\lambda_{Cs-137}-\lambda_{Ce-144}$.The calculated activity ratio for the fuel irradiated in a reactor during 10 days is marked by dot-dashed line and is also denoted as 'maximum limit' in the figure because the activity ratio for the fuels irradiated normally in a commercial PWR, denoted as 'normal irradiation' in the

Table 2. Known and Modeled Irradiation Histories for Cooling Time Determination

ASSEMBLY			ROD			SAMPLE
ID.	Irradia. Time,day T(i)	Interval Time,day I(i)	ID.	Cycle-BU (relative) BU(i)	H_b^a	The No. of Samples
A	T(1)=565	–	A1	–	18.371	2
			A2	–	〃	2
			A3	–	〃	1
B	T(1)=565	I(1)=74	B1	BU(1)=17.4,BU(2)=9.1	11.677	3
	T(2)=385		B2	=15.7, =7.9	11.526	3
			B3	=15.7, =7.9	11.526	3
C	T(1)=385	I(1)=96	C1	BU(1)=7.0, BU(2)=11.8	17.191	2
	T(2)=345		C2	=6.8, =11.8	17.317	2
D	T(1)=285	I(1)=54	D1	=10.0, =11.2	15.256	2
	T(2)=403					
E	T(1)=565	I(1)=74	E1	BU(1)=4.3, BU(2)=11.7, BU(3)=17.6	15.050	2
	T(2)=385	I(2)=96	E2	=11.4, =10.9, =6.4	9.009	3
	T(3)=345		E3	=14.1, =11.8, =7.7	9.002	1
			E4	=14.1, =11.8, =7.7	9.002	3
F	T(1)=285	I(1)=54	F1	BU(1)=1, BU(2)=1, BU(3)=1, BU(4)=1	8.859	2
	T(2)=403	I(2)=54	F2	=1, =1, =1, =1	〃	2
	T(3)=375	I(3)=41	F3	=1, =1, =1, =1	〃	1
	T(4)=370		F4	=1, =1, =1, =1	〃	2
M*	T(1)=3years	–	–	–	12.002	–

Note : Burnup of rods F1~F4 are arbitrary data.

* M stands for "Modeled irradiation history"

figure, can not exceed this limit. In the case of maximum limit, the parameter H_{Cs-137}^{Ce-144} becomes around 33 by putting irradiation time T(1)=10 days. Meanwhile, those of normal irradiation calculated by applying twenty irradiation history models expected in the spent fuel irradiated in a commercial PWR are in the range of 7~22. For example, the fuels used in this experiment actually have these values between 9 and 18. The calculated activity ratio by using the modeled irradiation history listed in Table 2 is represented as 'model' and marked by solid line in the figure. The activity ratio $^{144}Ce/^{137}Cs$ has the order of 10^{-1} when T_c is 5 years and 10^{-3} when T_c is 10 years.

Gamma-ray spectra of 36 samples listed in

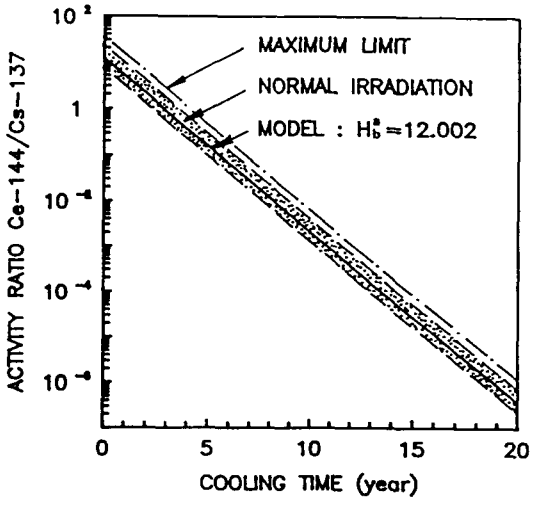

Fig. 3. Variation of Activity Ratio $^{144}Ce/^{137}Cs$ after Discharge from Reactor

Table 2 were collected by putting each sample on a table located in a radiation shielding lead hot cell and detecting the gamma-rays emitted from the sample by means of the gamma-ray spectrometric system equipped with a HPGe detector, signal shaping electronics, a 4,096 channel multichannel analyzer and a personal computer. The gamma-ray intensities of interest were derived from analyzing the full energy peak net area by means of the direct summation method with two channel background[15]. Thus the following two intensities were used to evaluate the intensity ratio N_a^u/N_b^v in Eq.(6) :

$N_{Ce}^{2,186}$ = full energy peak intensity of ^{144}Ce's 2,186 keV,

N_{Cs}^{662} = full energy peak intensity of ^{137}Cs's 662 keV.

The peak shape of ^{137}Cs's 662 keV was almost coincident with the Gaussian and the percent standard deviation of the net area was only about 0.05 %, and the analyzed intensity of N_{Cs}^{662} had the order of 10^2 cps(count per second). However in case of ^{144}Ce's 2,186 keV, the peak was statistically unstable and the percent standard deviation of the net area, which deeply depended on the sample burnup and declared cooling time, was in the range of 2~20%. The analyzed intensity of $N_{Ce}^{2,186}$ had merely the order of 10^{-3} cps.

The relative detection efficiency as a function of gamma-ray energy was derived from the multiple gamma-ray energies of fission product ^{154}Eu which remained in the examined spent fuel. Fig. 4 shows one of the relative detection efficiencies of the gamma-ray spectrometric system. In this figure, the net area intensity of 1,274 keV peak was put as unity and then the intensities of other peaks between 592 and 1,597 keV were normalized to that of basis peak. The relevant efficiency equation representing these normalized data was obtained by means of the least squares fitting, and then the following two detection efficiencies were calculated by using this equation in order to evaluate the detection efficiency ratio $\varepsilon^v/\varepsilon^u$ in Eq.(6) :

$\varepsilon^{2,186}$ = relative full energy peak efficiency of 2,186 keV,

ε^{662} = relative full energy peak efficiency of 662 keV.

The cooling time by known irradiation history and that by modeled irradiation history were estimated by substituting experimentally measured intensity ratio and efficiency ratio, and irradiation history parameter H_{Cs-137}^{Ce-144} into Eq.(6). The results of these were compared each other and compared with the operator declared cooling time.

4. Results and Discussion

Fig. 5 shows the MCT by known irradiation history against the DCT. It seems that the MCT agrees well with the DCT regardless of the cooling time length which is approximately between 5 and 13 years. Assembly A in the figure was irradiated during 1 cycle, assemblies B, C and D were 2 cycles, assembly E was 3 cycles, and assembly F was 4 cycles.

Fig. 6 is given to see the dispersion of the data from the dashed diagonal in Fig. 5. In this figure, the relative difference between the MCT by

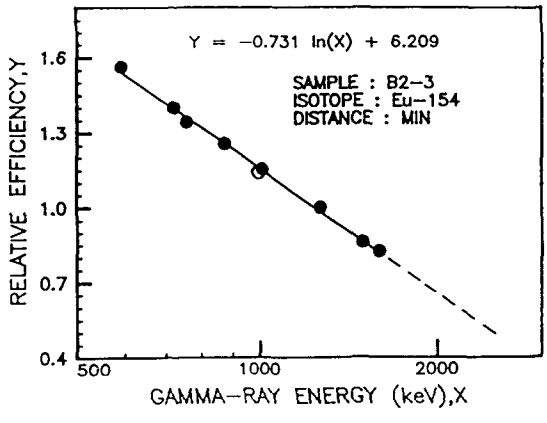

Fig. 4. Relative Detection Efficiency of the Spectrometric System

known irradiation history and the DCT is shown. 7 samples taken from assembly F have the differences only in the range of 0~2% in spite of the arbitrarily given burnup data as well as their 4 cycle irradiation. Despite the low counting rate of ^{144}Ce's 2,186 keV, the MCT of long cooled assemblies A, B, C and E show good agreement with the DCT within relative difference of ±5 % denoted by dot-dashed line in the figure. Thus, under the condition of well arranged gamma-ray spectrometric system, it seems that cooling time using known irradiation history can be determined with relative difference of ±5 % from the cooling time based on operational log when the detected activity ratio ^{144}Ce(2,186 keV)/^{137}Cs(662 keV) is used as a monitor for the fuel cooled up to around 13 years.

Fig. 7 is given in order to see how well the MCT by simplified irradiation model agrees with that by known irradiation history. The difference between both results lies within the dot-dashed line which indicates ±0.5 year limits of the dashed diagonal in the figure. Although the irradiation histories of samples taken from assembly B are somewhat different from modeled irradiation history which contains only continuous and constant 3 year irradiation, the resultant differences of their cooling time are merely between 0.03 and 0.05 year.

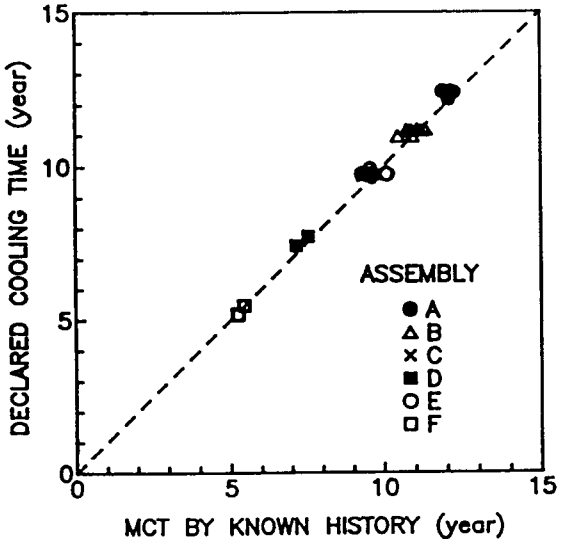

Fig. 5. Comparison of the Measured Cooling Time by Known Irradiation History with the Declared Cooling Time

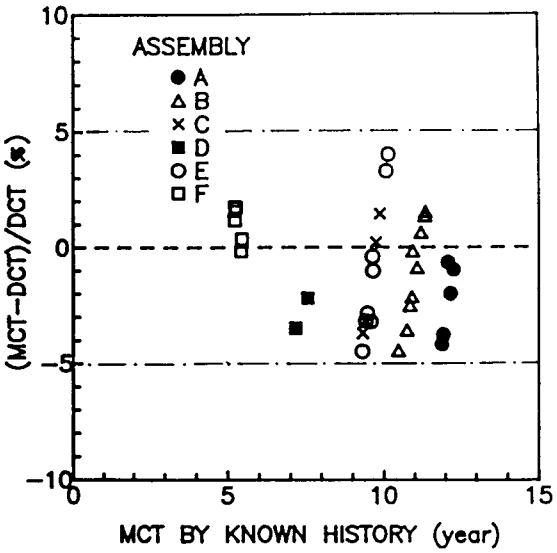

Fig. 6. Distribution of Relative Difference between the Declared Cooling Time and the Measured Cooling Time

From these results, the model adopted in this experiment or other suitable models representative of the samples could be usefully applied for the estimation of cooling time when the information of spent fuel irradiated normally in a commercial power reactor is obscure or not known obviously.

Contrary to the simplicity in identification of spent nuclear fuel mounted in an assembly by confirming the identification number marked on the assembly, the identification of spent fuel rods dismantled from assemblies or samples cut from spent fuel rods would be somewhat difficult when the amount of them are increased or when the movement and deformation of them happen very often. Thus, the periodic confirmation of rods or samples would be necessary for safeguards and management of spent nuclear materials.

Fig. 8 shows the schematic diagram for the sorting out and/or identification of spent nuclear fuel by means of cooling time estimation mentioned in this study on the basis of the satisfiable results shown in Fig. 7. In the figure, the grouping of spent fuel is based on the assemblies which have different cooling time.

In order to study the feasibility of identification of spent nuclear fuel by above technique, time differences caused by long experimental period due to a lot of samples as well as the relatively long spectrum collection time were compensated in the results of DCT and MCT by modeled irradiation history by putting the last experimental date as the comparison date.

The modified MCT by modeled irradiation history against the modified DCT is shown in Fig. 9. In the figure, the samples taken from assemblies D and F are sorted out perfectly among the samples from 6 assemblies. It might be easy to distinguish between the samples of assembly A and those of assembly E unless the samples of assembly B exist. The sorting out the samples of assemblies B and C will be possible if no sample of assembly E exists. The samples of assemblies C and E, which have the same declared cooling time but different irradiation history, are sorted out partially and from this result it is expected to be possible to distinguish totally those samples with quitely different irradiation histories.

Therefore, it would be possible to sort out spent nuclear fuel when the fuel with the difference in DCT over 1 year and/or the fuel with quite different irradiation history are grouped together.

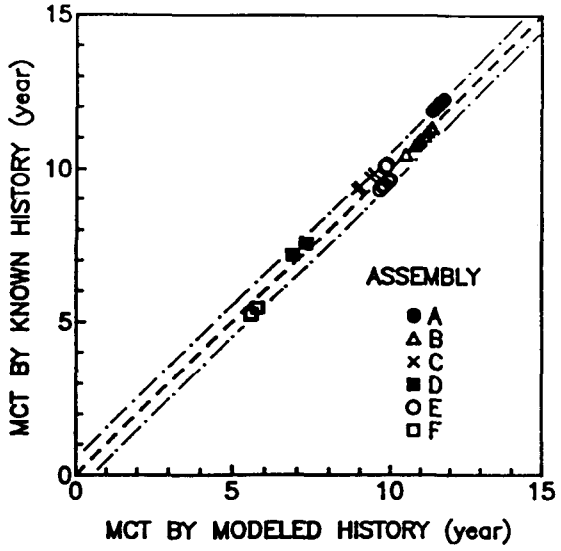

Fig. 7. Comparison between the Measured Cooling Time by Modeled Irradiation History and That by Known Irradiation History

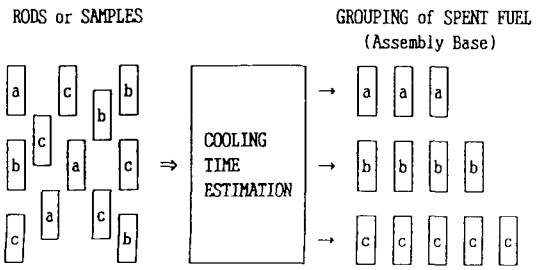

Fig. 8. Schematic Diagram for the Sorting-out and/or Identification of Spent Nuclear Fuel

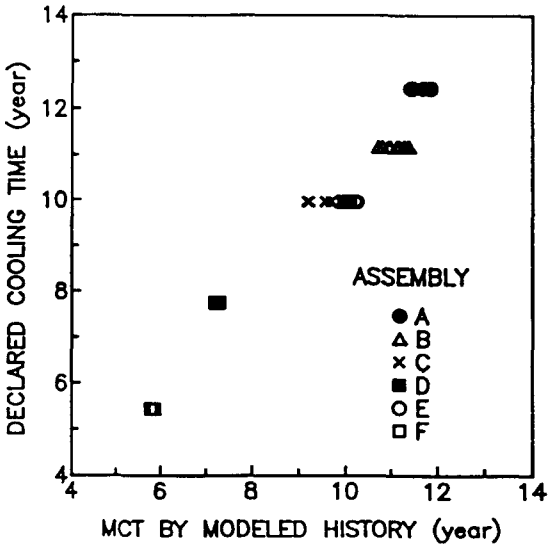

Fig. 9. Comparison of the Measured Cooling Time by Modeled Irradiation History with the Declared Cooling Time

Because the time difference between the calculated cooling time by the fissioning of ^{239}Pu and that by the fissioning of ^{235}U is 0.47 year, the spent fuel fissioned by compounded fissile and/or fissionable materials has it in the range of 0~0.47 year. Thus, the difference in DCT over about 1.5 year would be necessary to sort out UO_2, PuO_2 and MOX(mixed oxide) spent fuels.

5. Conclusions

The conclusions reached by cooling time determination through the detection of activity ratio $^{144}Ce/^{137}Cs$ by gamma-ray spectrometry are as follows :

1) Activity ratio $^{144}Ce/^{137}Cs$ can be used as a useful monitor for cooling time determination regardless of the unstable gamma-ray spectrometric peak with high statistical uncertainty due to low detection intensity of ^{144}Ce's gamma-ray energy.
2) By choosing suitable model for irradiation history of examined spent nuclear fuel, the measured cooling time by model could be agreeable with that by known irradiation history within time difference of ± 0.5 year. It would be expected to be possible to estimate reliably the cooling time of spent nuclear fuel without the exact information of irradiation history.
3) The technique for cooling time determination by gamma-ray spectrometry is considered to be a useful method to identify and to sort out spent nuclear fuel dismantled or cut from assemblies for safeguards and management of spent nuclear materials.

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