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IRRADIATED FUELS BY NON-DESTRUCTIVE

ISOTOPIC CORRELATION TECHNIQUE

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Determination of Pu Accumulated in Irradiated Fuels
by Non-Destructive Isotopic Correlation Technique[*]

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Division of JPDR, Tokai Research Establishment, JAERI

Non-destructive gamma-ray spectrometry was made of spent
fuel of average burnup 4,400 MWd/t discharged from JPDR-I (BWR).
The activity ratios of fission products in fuel rods of an
assembly were correlated with the burnup and the atom ratio
between accumulated plutonium and remaining uranium which were
obtained by destructive assay of the fuel rods. The $^{134}Cs/^{137}Cs$
activity ratio was found to be a good indicator of the Pu/U atom
ratio rather than of the burnup. The Pu/U atom ratio in respective
assemblies was obtained by using the average of the activity ratio
and the correlation curve from measurements of the fuel rods.
The estimated amount of plutonium agreed with the value measured
in the reprocessing plant within 2%.

非破壊同位体相関法による照射済燃料内のPu生成量の決定

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　平均 4,400 MWd／t の燃焼度を持つ JPDR-I の使用済燃料について，非破壊ガンマ線スペクトロメトリを行った。

　1 体の燃料集合体内の 8 本の燃料棒について，核分裂生成物のガンマ線強度比と，破壊測定によって得られている燃焼率および蓄積された Pu と残存 U の原子数比とについて相関関係を調べた。この結果，^{134}Cs／^{137}Cs 強度比が，燃焼率に対してよりもむしろ Pu／U 原子数比に対してのより良い指標であることを見出した。次いで，燃料棒に対して得た相関関係を使って，各々の燃料集合体中に含まれている Pu／U 原子数比を求めた。このようにして得た集合体中の Pu 量は再処理工場での値と 2 ％の差で一致した。

＋ 現所属：日本原子力研究所企画室

Contents

目　　　次

JAERI-M 8599

1. Introduction

Sufficiently accurate determination of fuel burnup and of accumulation of transuranium elements in power reactors is indispensable for optimizing fuel management in reactor and for understanding the changes observed in reactor characteristics. In the development of the techniques which are applied to the determination of the burnup, there can be little doubt that non-destructive measurement is desirable by virtue of the prompt availability for many samples. An application of non-destructive gamma-ray spectrometry to spent fuels will provide valuable information not only to investigate reactor characteristics and fuel performance but also to account for nuclear materials in a safeguards system. Especially, activity ratios of fission products are promising parameters for use as burnup monitors.

Gamma-ray spectra of spent fuel assemblies of JPDR-I boiling water reactor were measured non-destructively prior to shipment to a reprocessing plant. The isotopic correlation technique to the fuel exposure history determination were examined. The activity ratios of fission products formed in fuel rods of an assembly were correlated with the burnup and the atom ratio between accumulated plutonium and remaining uranium. Then, the amount of plutonium accumulated in the fuel assemblies was estimated.

2. Configuration and operation of JPDR-I core

The reactor is a natural circulation BWR of 45 MW thermal output, loaded with 2.6 % enriched UO_2 fuels. The core consisted of 72 fuel assemblies, 16 cruciform control rods and 24 burnable poison curtains. The plan of the core is shown in Fig.1.

A fuel assembly consisted of 36 fuel rods arrayed in 6 × 6 square lattice. Each rod was formed of two segments—upper and lower—having the same active fuel length as shown in Fig.2. The specifications of the fuel are given in Tables 1 and 2.

The core was operated from October 1963 to August 1969. There was a shutdown of considerable length from June 1968 to June 1969 for inspection of the pressure vessel of the reactor. The average burnup of the core was estimated to be 4,400 MWd/t[1].

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height of about 2/3 of full insertion, and the other was gradually with-
drawn from full to 2/5 insertion with increasing burnup. Their withdrawal
pattern was periodically altered every 500 MWd/t to flatten the burnup
distribution in the core.

3. Isotopic correlation of fission products

Non-destructive gamma-ray spectrometry was carried out on eight fuel
rods in an assembly A-20. The average burnup of the assembly is 5,570 MWd/t
and its cooling time is 31 months. The locations of the fuel rods in the
assembly are shown in Fig.3. Gamma-ray spectra were measured with a
planer-type Ge(Li) detector on 12 positions of each rod as indicated in
Fig.2.

Several fission products, such as ^{106}Ru-^{106}Rh, ^{134}Cs, ^{137}Cs, ^{144}Ce-^{144}Pr,
and ^{154}Eu were identified in the spectra. The nuclides ^{134}Cs and ^{154}Eu are
produced by the neutron capture of ^{133}Cs and ^{153}Eu, respectively, which
are the direct fission products as is likewise ^{137}Cs.

If we assume that the neutron flux is constant during irradiation, the
activity from a direct fission product N_D (such as ^{137}Cs), and that from a
nuclide formed by neutron capture of fission products N_I (such as ^{134}Cs)
are respectively proportional to

$$N_D \propto \Sigma_f \cdot (\phi T) \quad \text{and} \tag{1}$$

$$N_I \propto \Sigma_f \cdot \sigma(n,\gamma) \cdot (\phi T)^2 \quad , \tag{2}$$

where

ϕ = spectrum and time-averaged neutron flux,

Σ_f = spectrum and time-averaged fission cross section,

$\sigma(n,\gamma)$ = spectrum and time-averaged neutron capture cross section
of the fission product, and

T = irradiation time.

Equations (1) and (2) show that the N_I/N_D ratio is also proportional to
(ϕT) and, in principle, can be used as a burnup monitor.

The activity ratios of the fission products, ^{134}Cs/^{137}Cs and ^{154}Eu/^{137}Cs,
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plutonium and remaining uranium in the fuel rods. The burnup and the atom

ratios were determined by chemical analysis on 24 points chosen from 96 positions where gamma-ray spectra were non-destructively measured[2]. For burnup determination, two techniques, the ^{148}Nd and ^{137}Cs methods, were employed. The burnup values obtained by both methods agreed with an error of 2 %. This result shows that the migration of cesium is not detectable.

(1) Correlation between activity ratios and burnup[3]

The activity ratios, ^{134}Cs/^{137}Cs and ^{154}Eu/^{137}Cs, are correlated with the burnup on a log-log scale in Fig.4. A linear correlation was obtained with a common slope of about 1.0 for each segment of the fuel rod. The correlation lines of the upper and the lower segments, however, separated each other. The same behavior was found on other fuel rods as well. Moreover, the ratios obtained from different fuel rods at the same height fell along straight lines with a slope of 0.5 when plotted against ^{137}Cs as seen in Fig.5. This phenomenon is interpreted in terms of the variation of power distribution pattern during irradiation and the spatial dependence of the neutron spectrum.

When the power distribution in the core varies during irradiation, the short-lived and long-lived nuclides come to differ in their distribution. The long-lived nuclides such as ^{137}Cs and ^{154}Eu retain their whole irradiation history, while the short-lived ^{134}Cs and ^{144}Ce reflect in their behavior mostly their more recent history. During the operation of JPDR-I, it was observed in gross gamma-scanning data that the position of the peak in the axial power distribution shifted upward with burnup. Therefore, the accumulated amount of short-lived product is larger in the upper segment than in the lower one. The amount of the change in ^{134}Cs/^{137}Cs ratio was calculated as 5 % by using the operation data of reactor, while that of ^{154}Eu/^{137}Cs was less than 1 %.

A typical characteristic of the BWR is the spatial variation of neutron spectrum in the core. The neutron spectrum varies corresponding to the void distribution in the axial direction. In addition, the neutron spectrum in the assembly varies with the radial location due to the existence of the water gap between fuel assemblies. The formation of ^{134}Cs and ^{154}Eu depends on the neutron capture cross section of ^{133}Cs and ^{153}Eu, respectively. The thermal cross section at 2200 m/s and the resonance integral are 30 and 450 barns for ^{133}Cs and 450 and 1,500 barns for ^{153}Eu. In contrast, the amount of accumulated ^{137}Cs depends predominantly on the thermal neutron

flux. As the result, the activity ratios become larger in the upper part of the core and in the central part of the fuel assembly even under the same burnup. The amounts of the difference in the $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$ activity ratios were calculated as 15 and 8 %, respectively, in the axial direction and 25 and 10 % in the radial direction.

Thus, the variation of power distribution pattern and the spatial dependence of the neutron spectrum are concluded to be the main causes of the scattering of data in the correlation.

(2) Correlation between activity ratios and Pu/U atom ratio

As shown in the preceeding section, the activity ratios of $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$ are affected by the spatial variation of neutron spectrum in the core. This is due to the large fraction of the resonance capture to the thermal capture of neutrons in the transmutation from ^{133}Cs to ^{134}Cs and ^{153}Eu to ^{154}Eu.

The situation is almost the same in the transmutation from ^{238}U to ^{239}Pu, because the resonance capture by ^{238}U is dominant in the process. The correlation of Pu/U atom ratio with the burnup is shown in Fig.6. The correlation curve shows the similality with that of $^{134}Cs/^{137}Cs$ activity ratio shown in Fig.4. Actually, a simple correlation is recognized between the Pu/U atom ratio and the $^{134}Cs/^{137}Cs$ activity ratio as shown in Fig.7. A similar correlation was obtained for the $^{154}Eu/^{137}Cs$ activity ratio.

A correlation curve between the Pu/U atom ratio $R_{Pu/U}$ and the activity ratio N_I/N_D was obtained by fitting a function

$$R_{Pu/U} = C_1 \cdot \frac{N_I}{N_D} \cdot \exp(-C_2 \cdot \frac{N_I}{N_D}) \tag{3}$$

where, C_1 and C_2 are unknown constants to be determined. The function form was deduced by considering the transmutation process of fission products and plutonium. In the case of the $^{134}Cs/^{137}Cs$ activity ratio, C_1 and C_2 are 0.0372 ± 0.0013 and 2.50 ± 0.40, respectively.

4. Determination of plutonium accumulated in fuel assemblies

Gamma-ray spectrometry was carried out on 72 spent fuel assemblies prior to their shipment to a reprocessing plant. The amount of plutonium accumulated in the assemblies was estimated by using the $^{134}Cs/^{137}Cs$

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(1) Measurement

Gamma-ray spectra of the assemblies were measured with a gamma-scanning apparatus which was installed temporarily at the fuel storage pool of JPDR. The apparatus consisted of a reclining fuel bed, a collimator set, a two-dimensional scanning mechanism, and a gamma-ray spectrometer with a coaxial-type Ge(Li) detector. An assembly was held horizontally on the fuel bed during the measurement.

The positions for measurement were determined from the distribution of ^{137}Cs activity and shown in Fig.8. An example of the distribution of ^{137}Cs activity across the assembly is illustrated in Fig.9. The measurement of gamma-ray spectra was made at ten positions on every side of A-8 assembly and eleven assemblies located in the one-octant region of the core as shown in Fig.10. The A-8 assembly has the same irradiation history as A-20 assembly of which fuel rods were examined by both a destructive and a non-destructive techniques. The other assemblies were measured at four positions of No.2, 3, 4, and 5 on one side only.

(2) Estimation of plutonium in assemblies

In order to calculate the amount of plutonium in an assembly, the average of $^{134}Cs/^{137}Cs$ activity ratios over the assembly is required. The average was calculated by integrating the functions which were fitted to the distribution of the activity ratios. The functions have forms as

$$\frac{N_I}{N_D} = A_1 \cdot X^2 + A_2 \cdot X + A_3 \quad \text{for radial distribution, and}$$

$$\frac{N_I}{N_D} = A_1' \cos(B \cdot X) + A_2' \cdot \cos(2 \cdot B \cdot X) + A_3' \cdot \sin(2 \cdot B \cdot X) \quad \text{for axial distribution,}$$

where, A_i and A_i' are the unknown constants to be determined, B is an axial geometrical buckling of the core, and X is the position in the assembly. In the process of integration of the distribution, its separability in radial and axial directions are postulated. For the assemblies which were measured only on one side, the average was calculated by using the relative distribution of the other assembly which is located at a symmetric position in the one-octant region of the core, and by normalizing the activity ratio with the value of the measured side.

The amount of plutonium in each assembly can be estimated by using the correlation curve shown in Fig.7 and the amount of uranium remaining in the assembly after making corrections: for the differences in counting efficiency between two Ge(Li) detectors and in shielding factors due to the difference in thickness of water layor and the degree of overlap of fuel rods. A correction factor including above mentioned ones was obtained by normalizing the average of the activity ratio of A-8 assembly to that of A-20 assembly, as the both assemblies were irradiated at symmetric positions in the core as shown in Fig.10 and have the same irradiation history. The average of the Pu/U atom ratio of A-20 assembly was calculated as 0.24 % from the distribution of the activity ratio and the correlation curve.

By using the correction factor to the activity ratio, the correlation curve, and the remaining amount of uranium in the assembly, the amount of plutonium accumulated in each assembly was estimated. The remaining amount of uranium in the assembly was calculated with the initial value and the burnup obtained from the operation data. The burnup is less than 1 % and its error included in the operation data is negligible. The resulting amounts of plutonium ranges from 0.04 to 0.17 kg per assembly.

Up to now, 57 assemblies were reprocessed and the estimated total amount of plutonium in their assemblies is 5.5 kg. On the other hand, the value reported by the reprocessing plant is 5.4 kg and the predicted value by the operation data of the reactor is 5.6 kg.

5. Conclusion

Non-destructive gamma-ray spectrometry was carried out on the spent fuels of JPDR-I boiling water reactor, which were discharged after 4,400 MWd/t burnup on the average.

At first, the activity ratios of fission products in fuel rods of an assembly were correlated with the burnup and the atom ratio between accumulated plutonium and remaining uranium which were obtained by a destructive assay of the fuel rods. The $^{134}Cs/^{137}Cs$ activity ratio was found to be a better indicator of the Pu/U atom ratio rather than of the burnup.

Then, the profiles of the $^{134}Cs/^{137}Cs$ activity ratio were non-destructively measured on fuel assemblies and the average of the ratio was calculated for every fuel assembly. The Pu/U atom ratio in each assembly was obtained by using the average of the activity ratio and the correlation

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Thus, the total amount of plutonium in 57 assemblies which were re-processed at a reprocessing plant was estimated as 5.5 kg. On the other hand, the value reported by the reprocessing plant is 5.4 kg. The agreement is quite good for low burnup fuels. Further measurements are planned to prove the applicability of this isotopic correlation technique to higher burnup fuels.

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Table 1 Specification of fuel rod and assembly

Fuel material	UO₂ sintered pellet, solid
²³⁵U enrichment	2.60 w%
Pellet diameter	12.5 mm
Pellet height	12.7 mm
Pellet density	10.4 g/cm³
Cladding material	Zircaloy-2
Cladding thickness	0.76 mm
Cladding outer diameter	14.12 mm
Active fuel length in a segment	721 ± 3 mm
Weight of UO₂ in a segment	921 ± 25 g
Dy₂O₃ content in end pellet	0.42 w%
Number of rods in an assembly	36 (= 6 × 6) in square
Lattice pitch	19.56 mm
Volume ratio of moderator to fuel in lattice	1.84
Channel box material	Zircaloy-4

Table 2 Reactor characteristics and operating condition of JPDR I

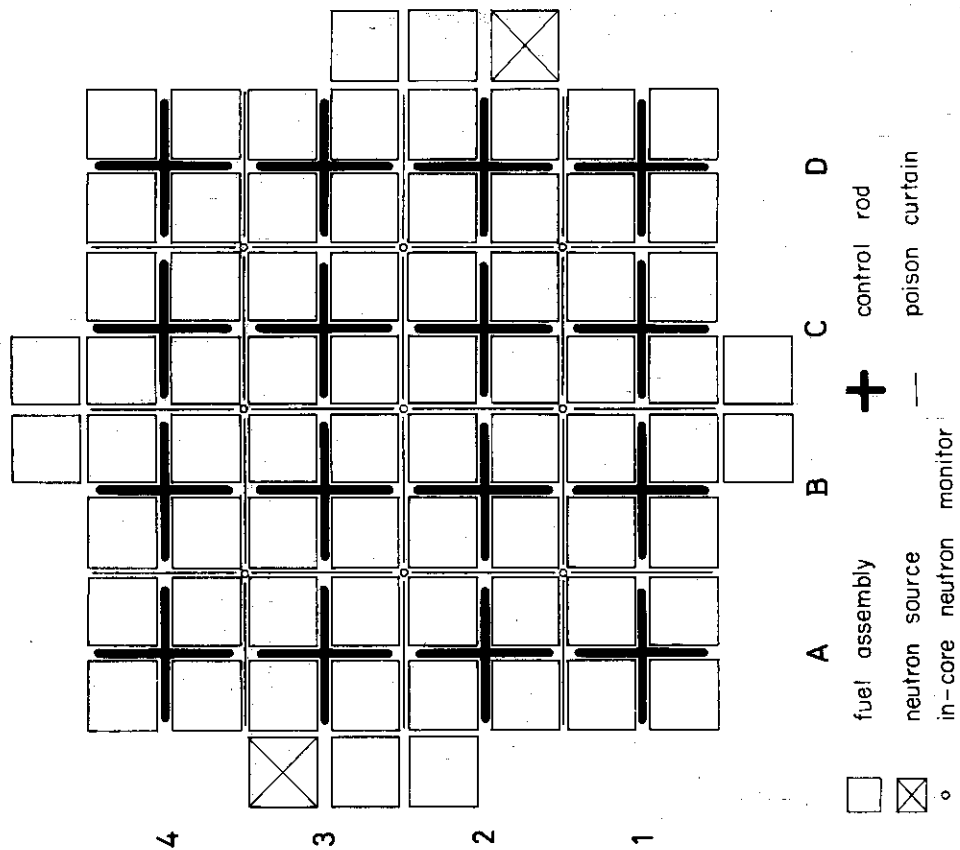| Reactor type	Naturally circulated BWR
Thermal power	45 MW
Equivalent diameter	127 cm
Effective height	147 cm
Number of fuel assemblies	72
Core-averaged volume ratio of moderator to fuel	2.7
Operating pressure	61.5 kg/cm², G
Temperature of coolant	277°C
Maximum temperature of fuel center	1,610°C
Average power density	22.5 kW/l
Radial power peaking factor	1.4
Axial power peaking factor	1.5
Average thermal neutron flux	1.42 × 10¹³ n/cm²·sec
Maximum heat flux	823,000 kcal/m²·hr
Average heat flux	226,100 kcal/m²·hr
Average void fraction	19%
Core-exit steam quality	4.7%
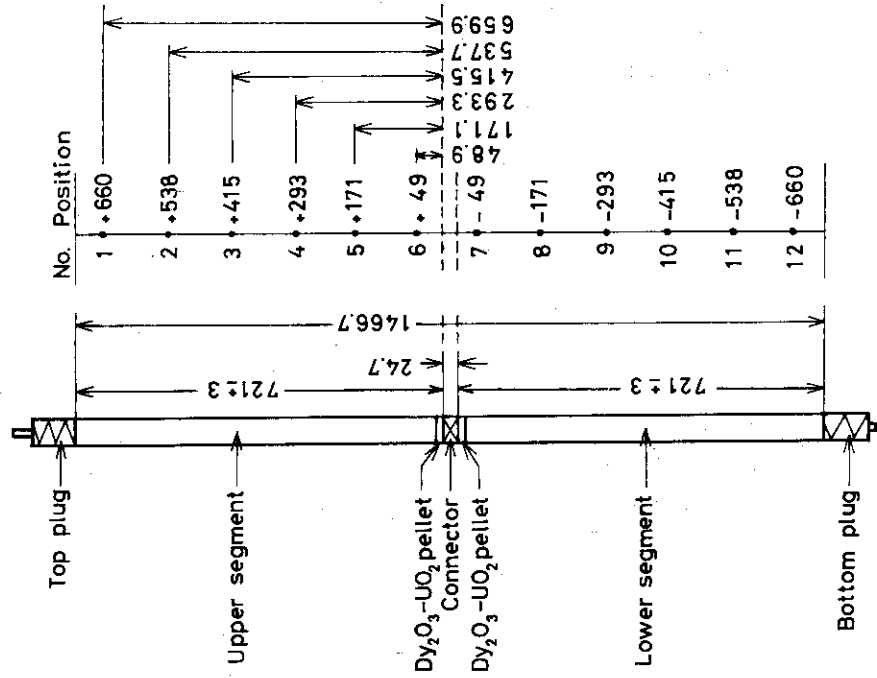
Fig. 2 Structure of a fuel rod and positions for measurement.

Fig. 1 Plan of JPDR-I core.

Fig. 4 $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$ activity ratios against burnup. Open and closed circles represent the values of the upper segment KA-1040 and the lower segment KC-1333, respectively.
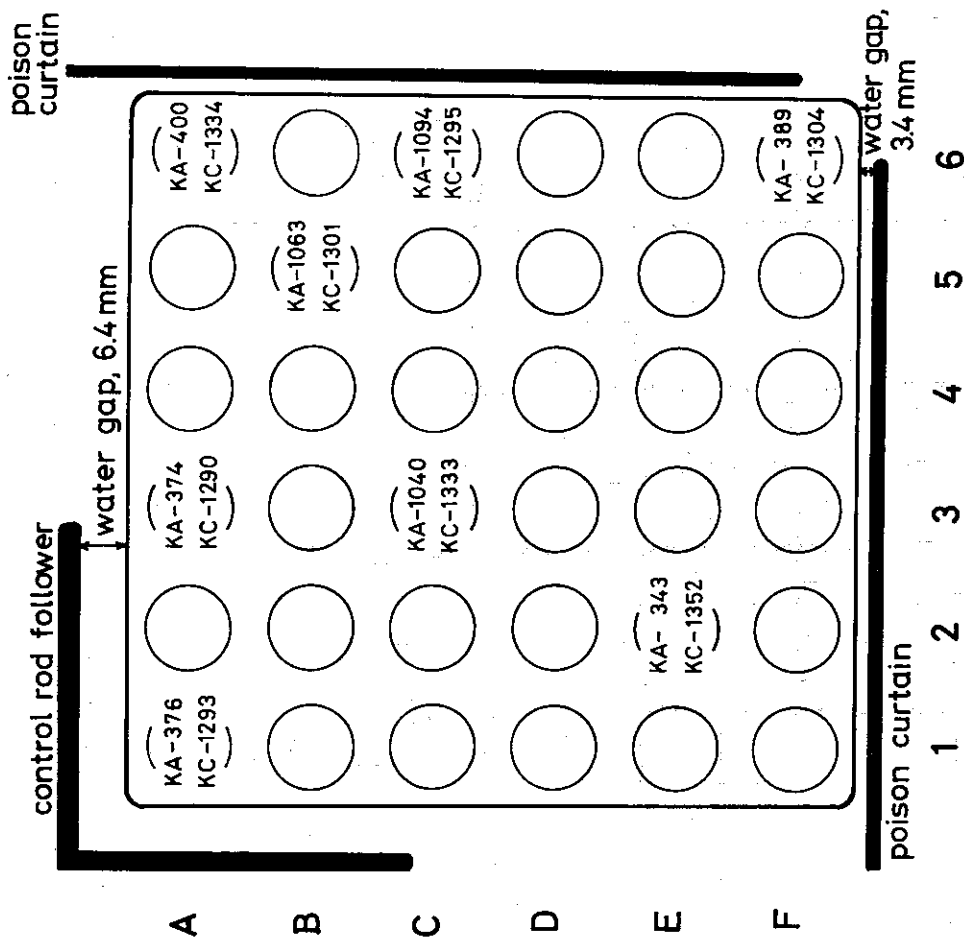

Fig. 3   Locations of fuel rods chosen for measurement in A-20 assembly.

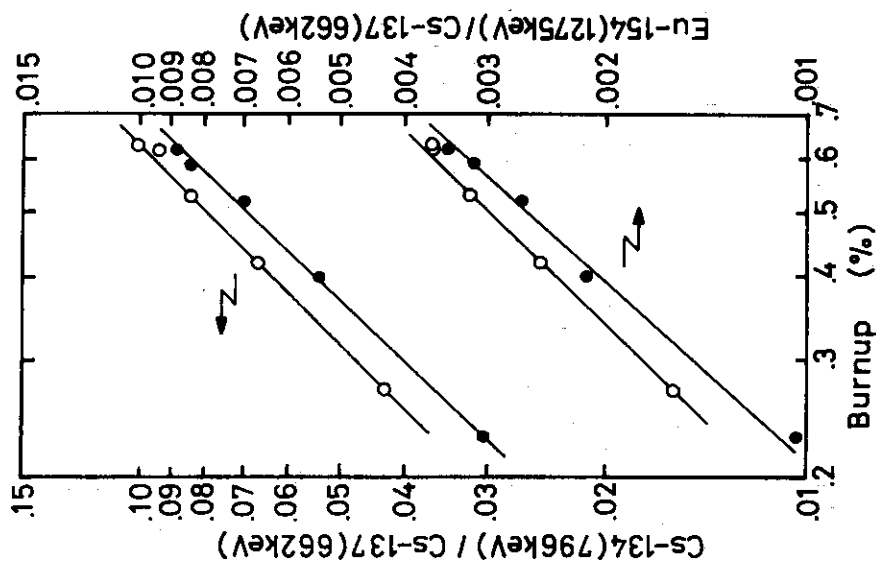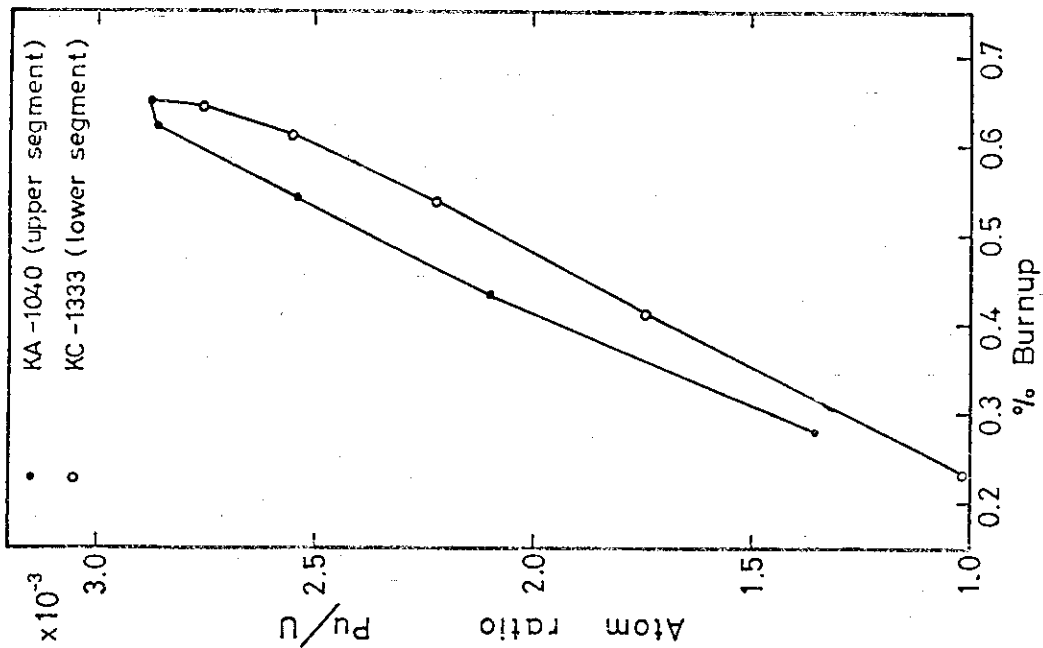KA- : number of upper segment,
KC- : number of lower segment.

Fig. 6 Pu/U atom ratio against burnup in atom % unit.
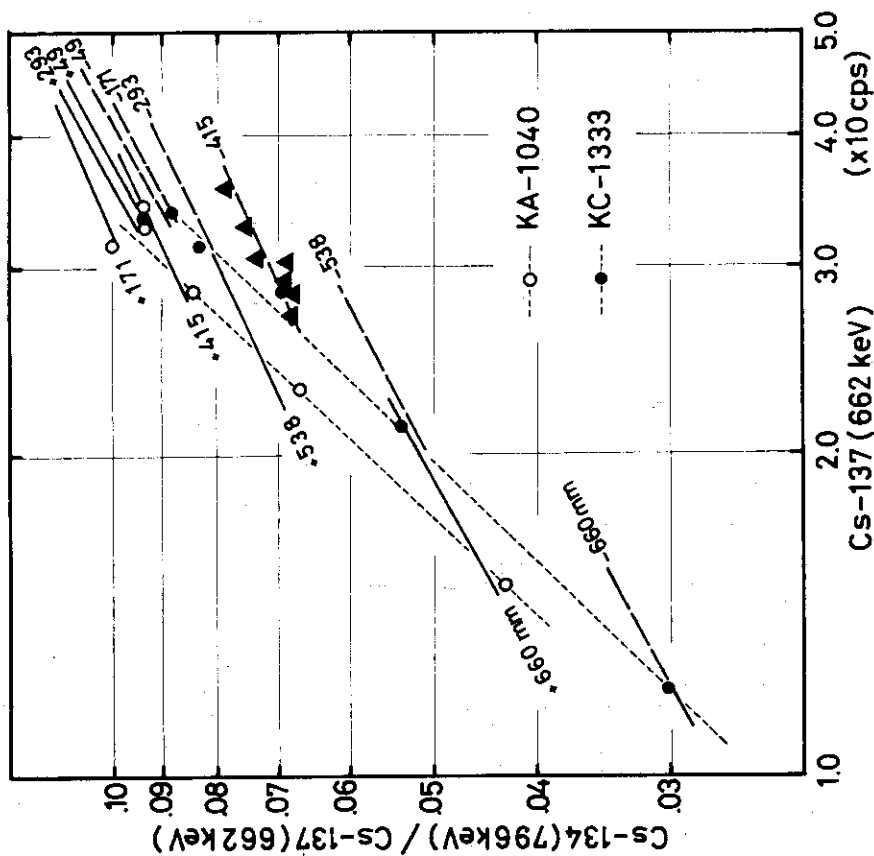
Fig. 5 $^{134}Cs/^{137}Cs$ activity ratio against ^{137}Cs activity at
various height in A-20 assembly. Open and closed
circles represent the values of specific segments of
a fuel rod, and triangles are the values of different
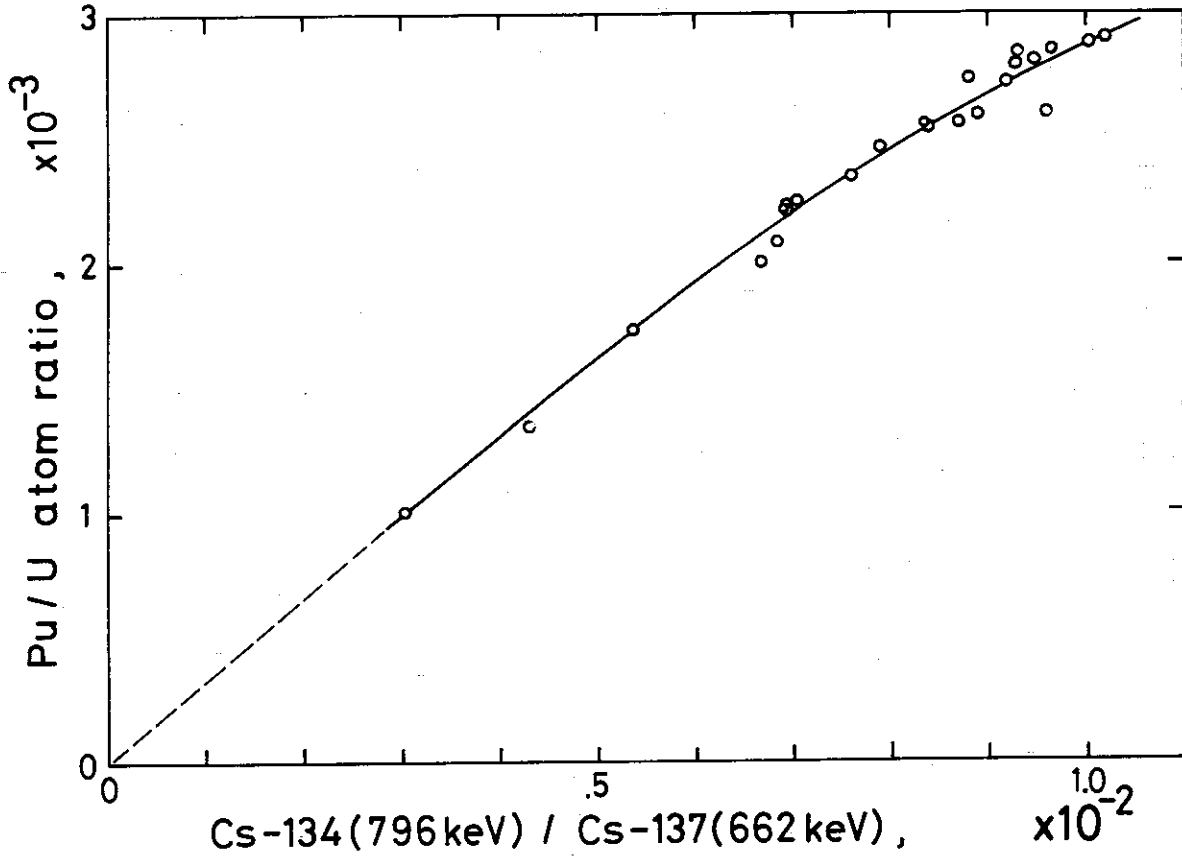segments at the same height (~415 mm).

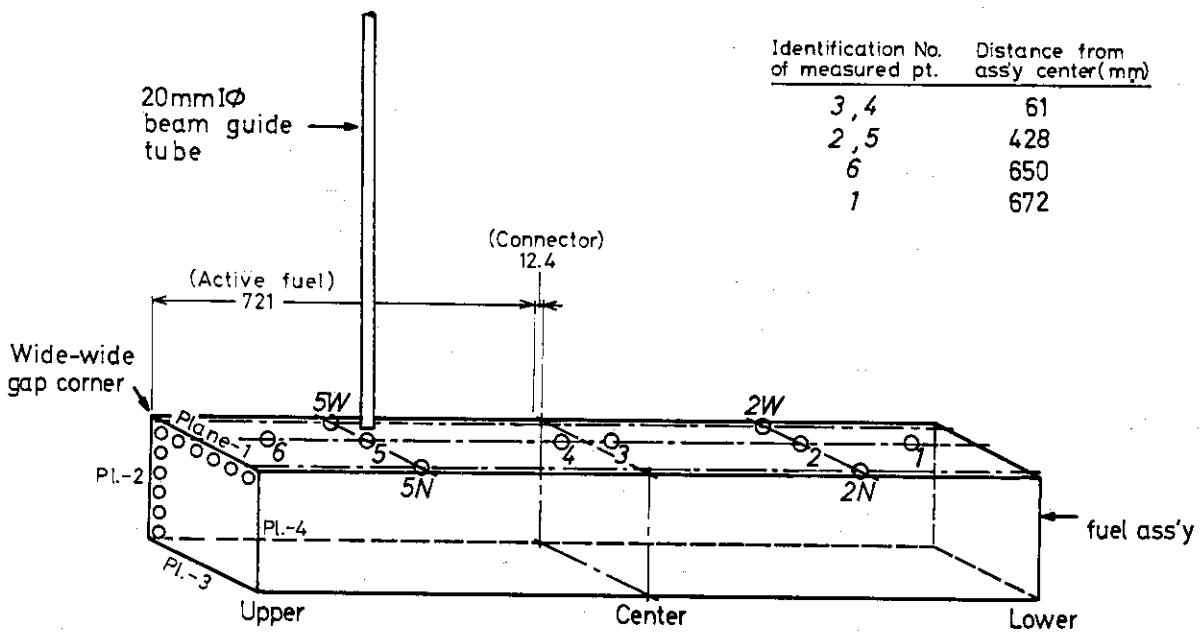Fig. 7 Correlation of Pu/U atom ratio with $^{134}Cs/^{137}Cs$ activity ratio.

Identification No. of measured pt.	Distance from ass'y center(mm)
3,4	61
2,5	428
6	650
1	672

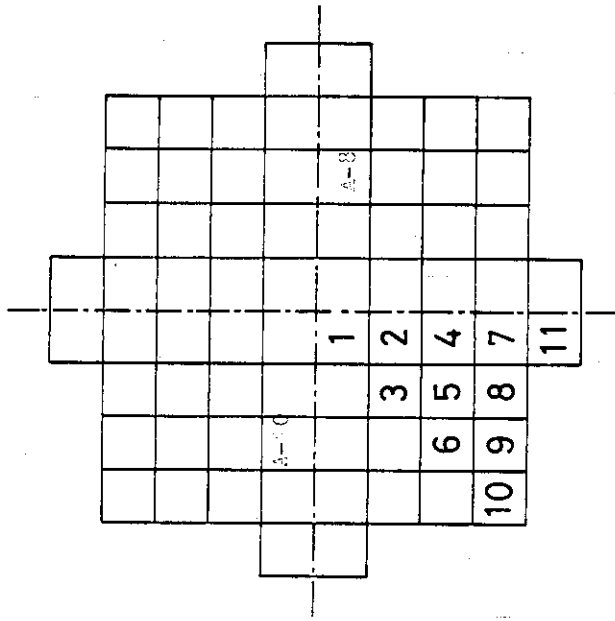Fig. 8 Positions for measurement on a side of assembly.

Fig. 10 Locations of A-8, A-20, and eleven assemblies in the one-octant region in core.
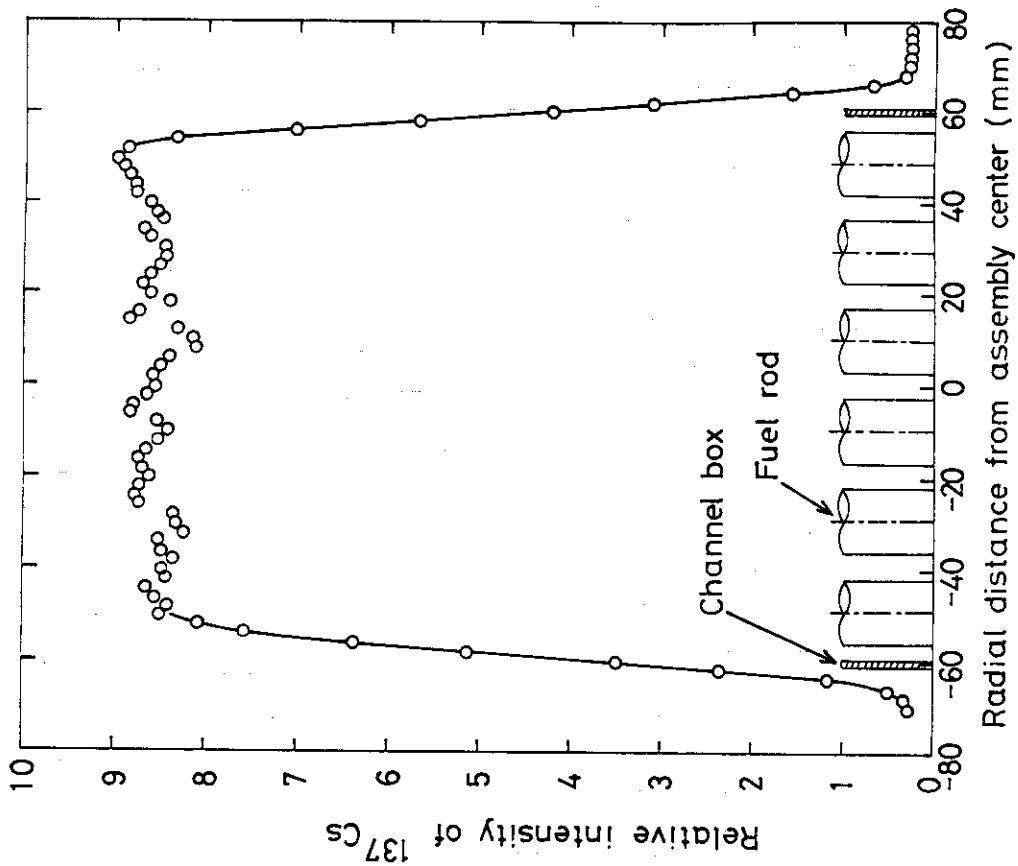
Fig. 9 Radial distribution of ^{137}Cs activity at +428 mm position on Plane 1 of A-8 assembly.