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

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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.

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. The Pu/U atom ratio in each assembly was obtained by using the average of the activity ratio and the correlation curve determined from the measurements on the fuel rods. The estimated amount of plutonium coincided with the value measured at the reprocessing plant within 2 %.

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## I. 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 promissing 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.

## II. 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)].

The reactor power was regulated by the four central control rods, while the twelve peripheral control rods were fully withdrawn. The central rods were grouped in two pairs. One of the pairs was kept at a constant

height of about 2/3 of full insertion, and the other was gradually withdrawn 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.

## III. 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

$\phi$ = spectrum and time-averaged neutron flux,

$\Sigma_f$ = 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 $(\phi T)$ and, in principle, can be used as a burnup monitor.

The activity ratios of the fission products, $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$, were correlated with the burnup and the atom ratio between accumulated 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

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

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 $0.0372 \pm 0.0013$ and $2.50 \pm 0.40$, respectively.

IV. 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 $^{134}Cs/^{137}Cs$

activity ratio and the correlation curve obtained in the previous section.

(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.

## V. 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

curve determined from the measurements on the fuel rods.

Thus, the total amount of plutonium in 57 assemblies which were reprocessed 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.

References

1) Division of JPDR, JAERI : Unpublished results.
2) NATSUME, H., et al. : J. Nucl. Sci. Technol., 14(10), 745(1977).
3) MATSUURA, S., et al. : ibid., 12(1), 24(1975).

**Table 1** Specification of fuel rod and assembly

| | |
|---|---|
| Fuel material | $UO_2$ sintered pellet, solid |
| $^{235}U$ enrichment | 2.60% |
| 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 $UO_2$ in a segment | 921±25 g |
| $Dy_2O_3$ content in end pellet | 0.42% |
| 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 | $1.42\times10^{13}$ 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% |

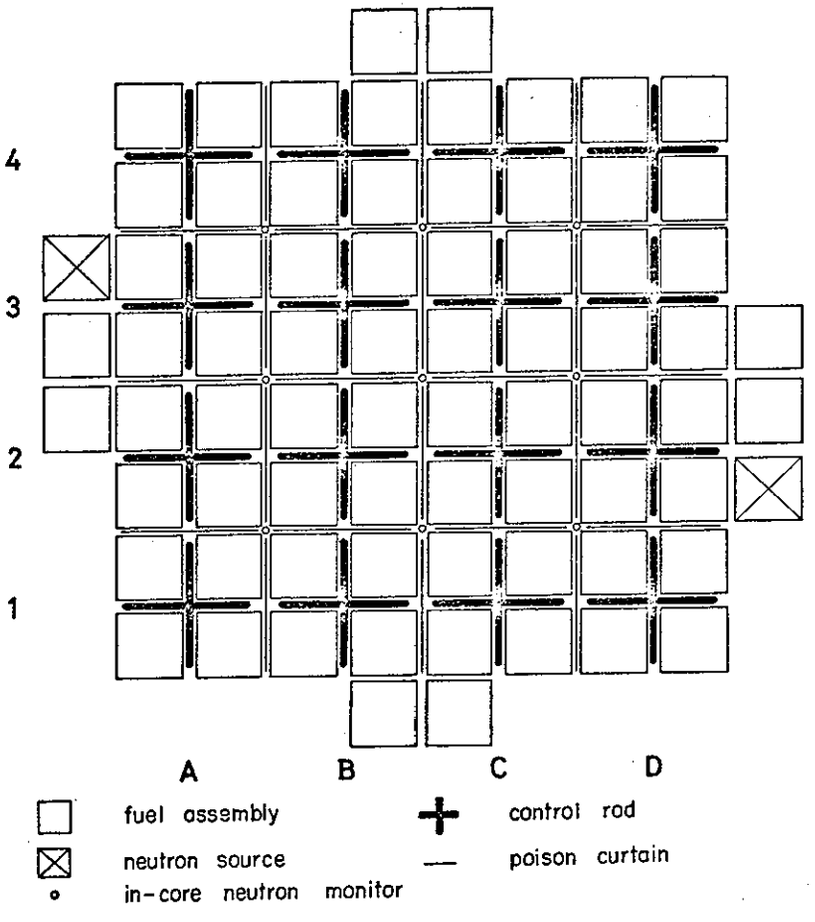



Fig. 1 Plan of JPDR-I core.

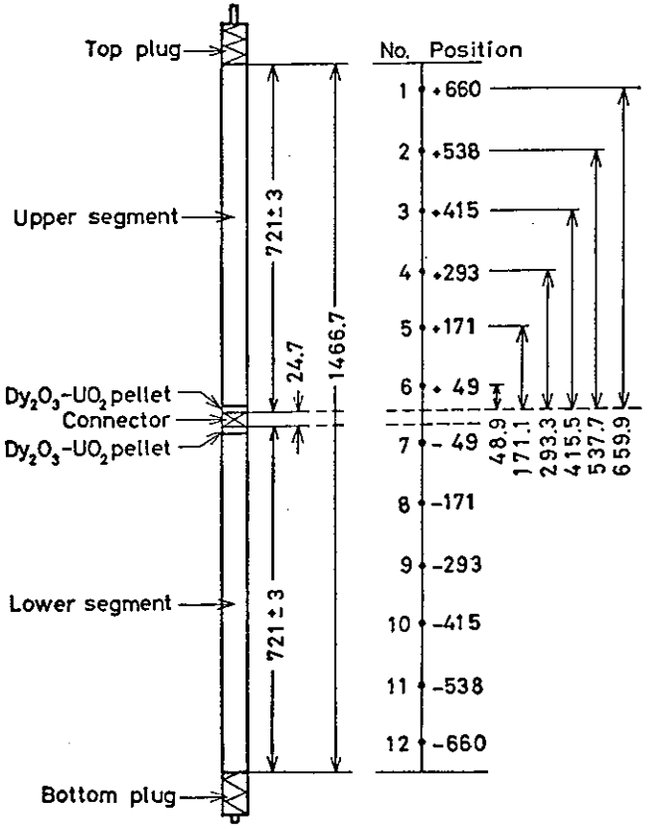



Fig. 2 Structure of a fuel rod and positions for measurement.

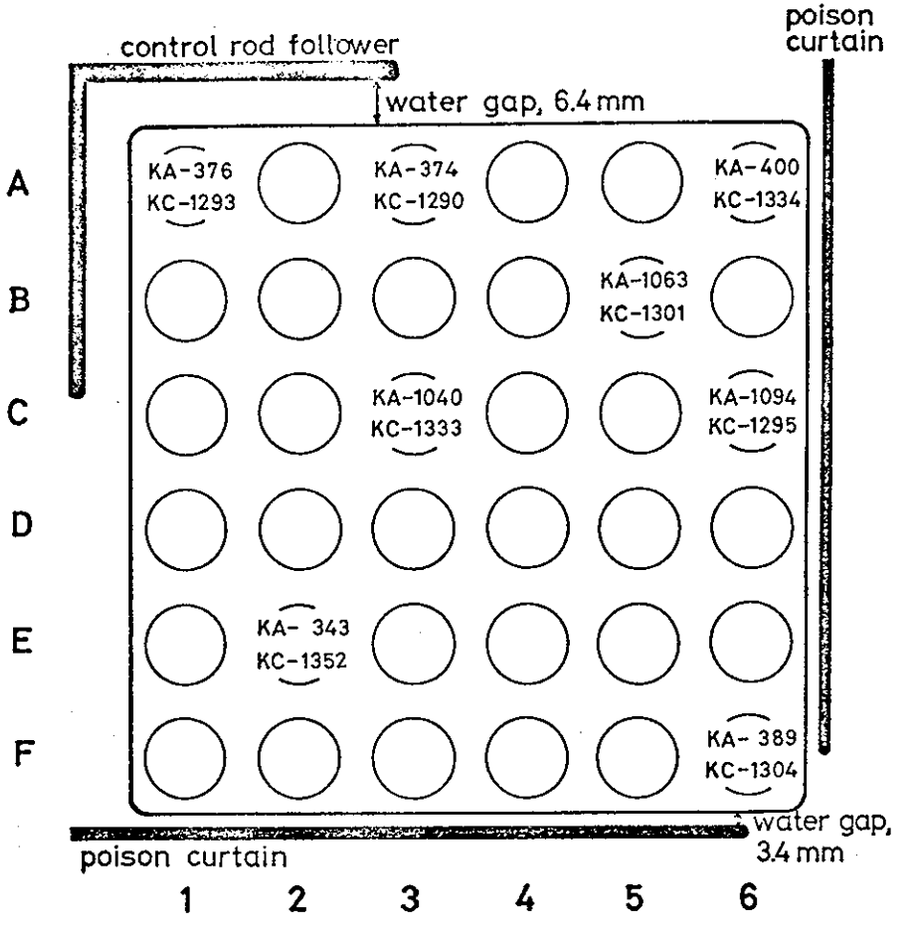



Fig. 3 Locations of fuel rods chosen for measurement in A-20 assembly.
KA- : number of upper segment,
KC- : number of lower segment.

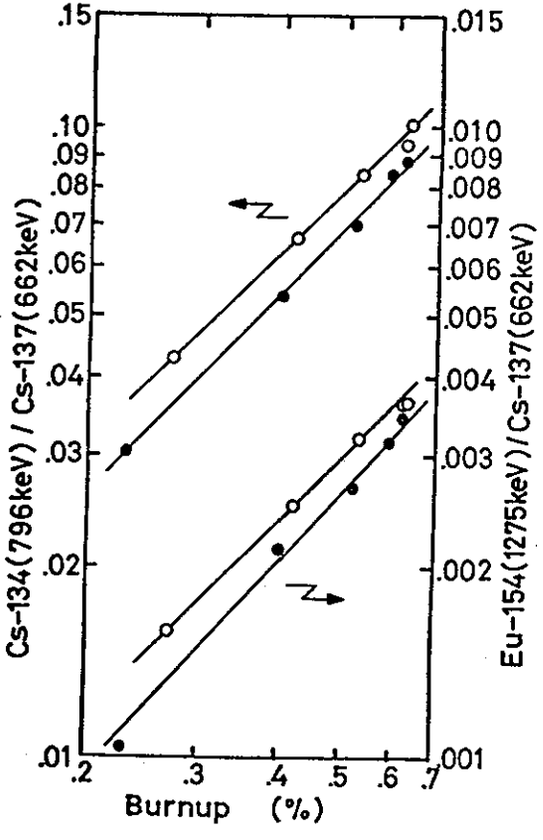



Fig. 4 $^{134}Cs/^{137}Cs$ and $^{154}Eu/^{137}Cs$ activity ratios against burnup. Open and closed circles represent the values of the upper segment KA-1040 and the lower segment KC-1333, respectively.

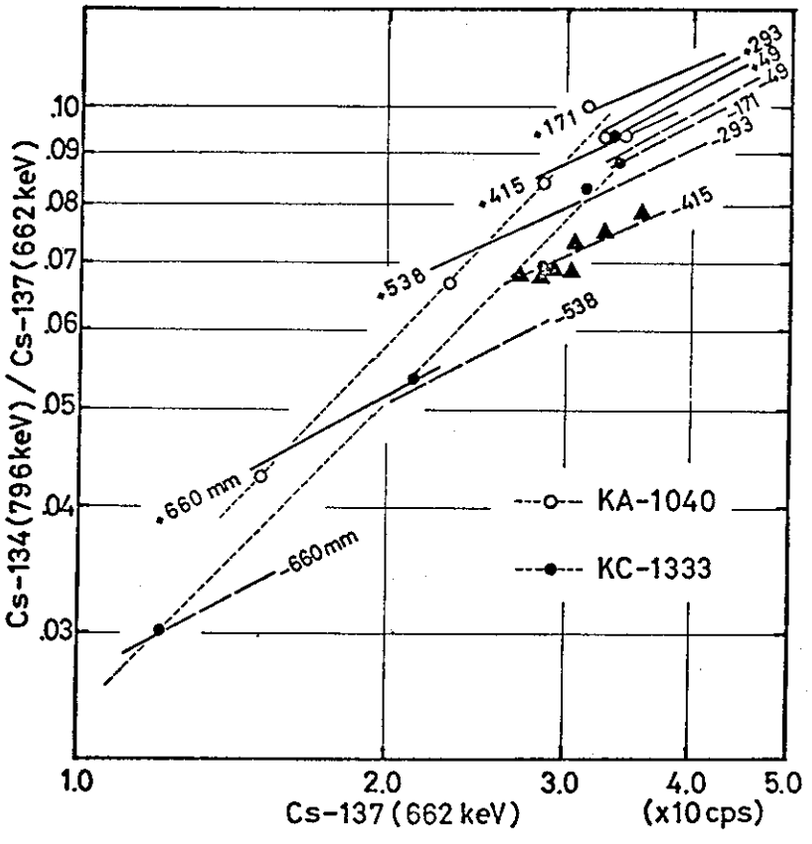



Fig. 5 $^{134}Cs/^{137}Cs$ activity ratio against $^{137}Cs$ activity at various height in A-20 assembly. Open and closed circles represent the values of specific segments of a fuel rod, and triangles are the values of different segments at the same height (-415 mm).

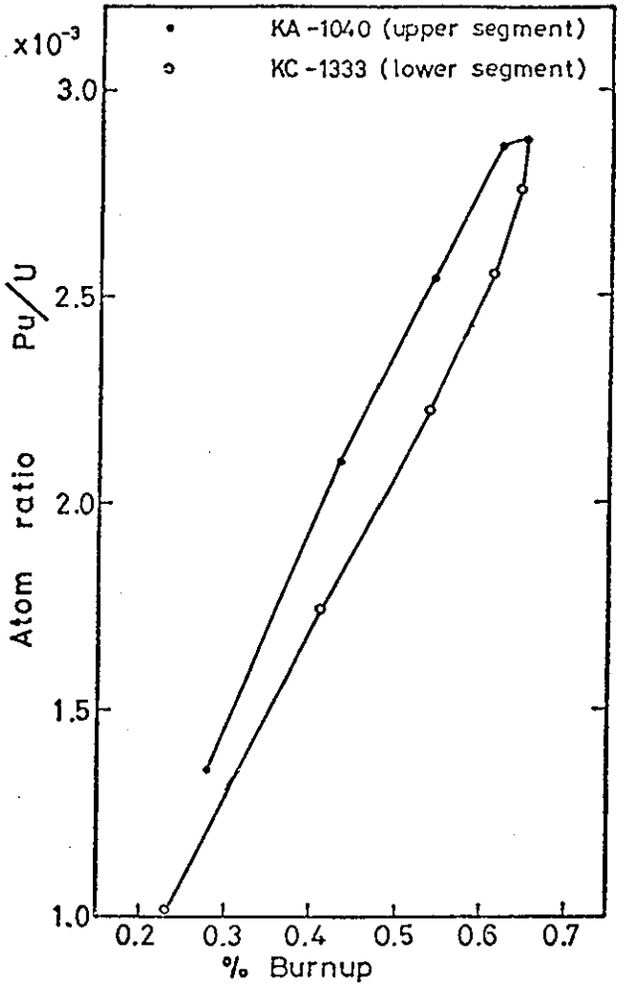



Fig. 6 Pu/U atom ratio against burnup in atom % unit.

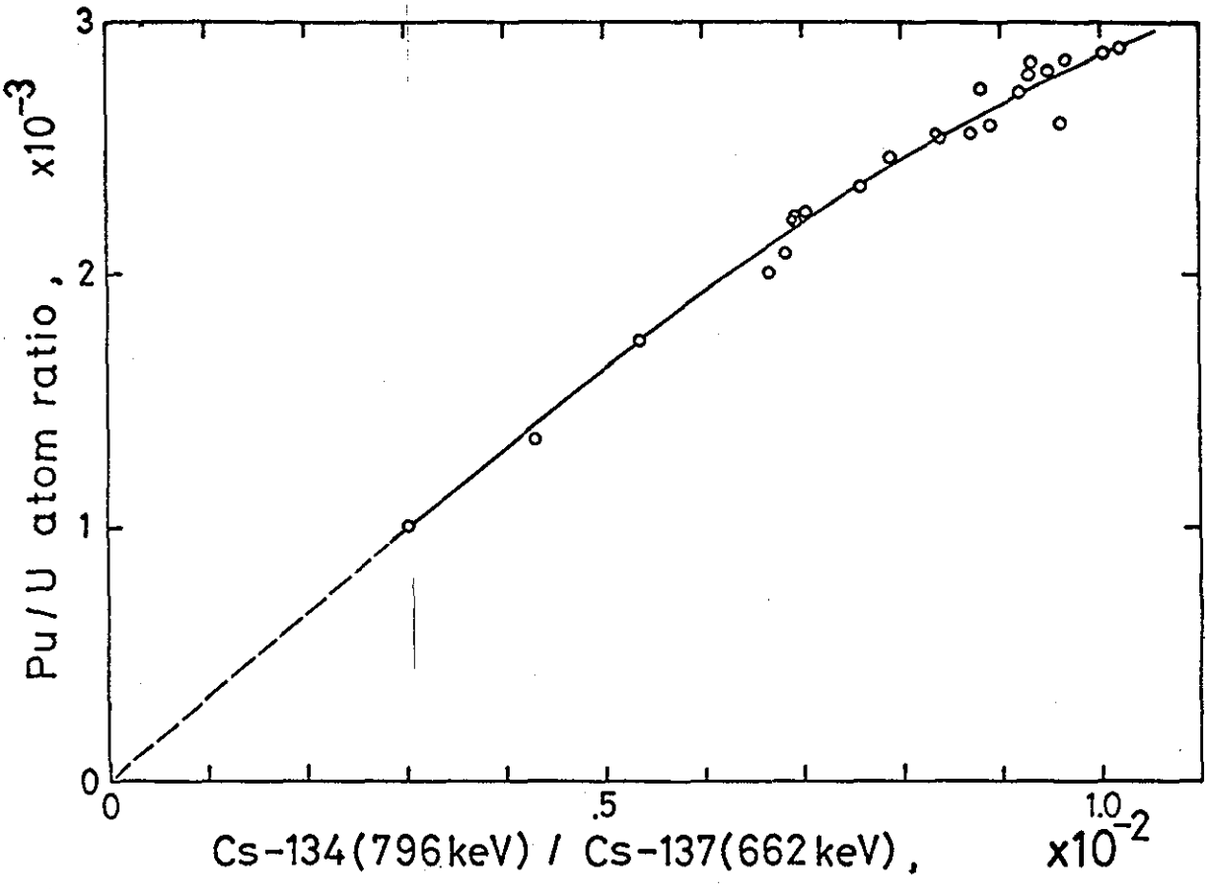
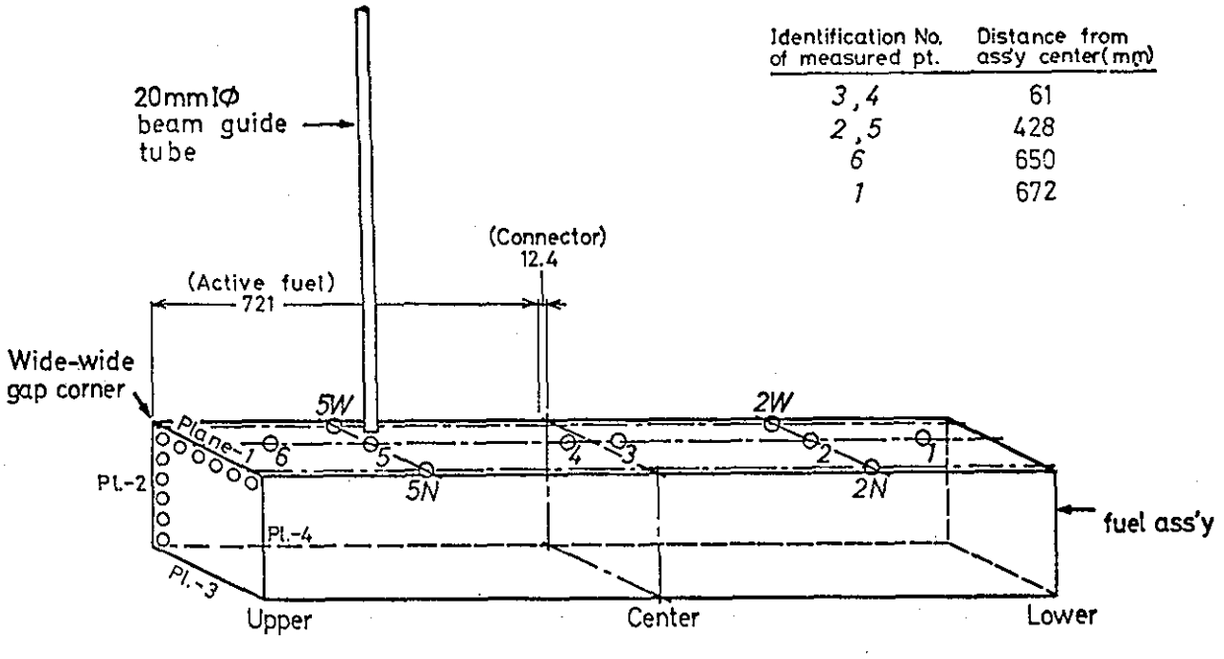



Fig. 7 Correlation of Pu/U atom ratio with $^{134}Cs/^{137}Cs$ activity ratio.




| 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.

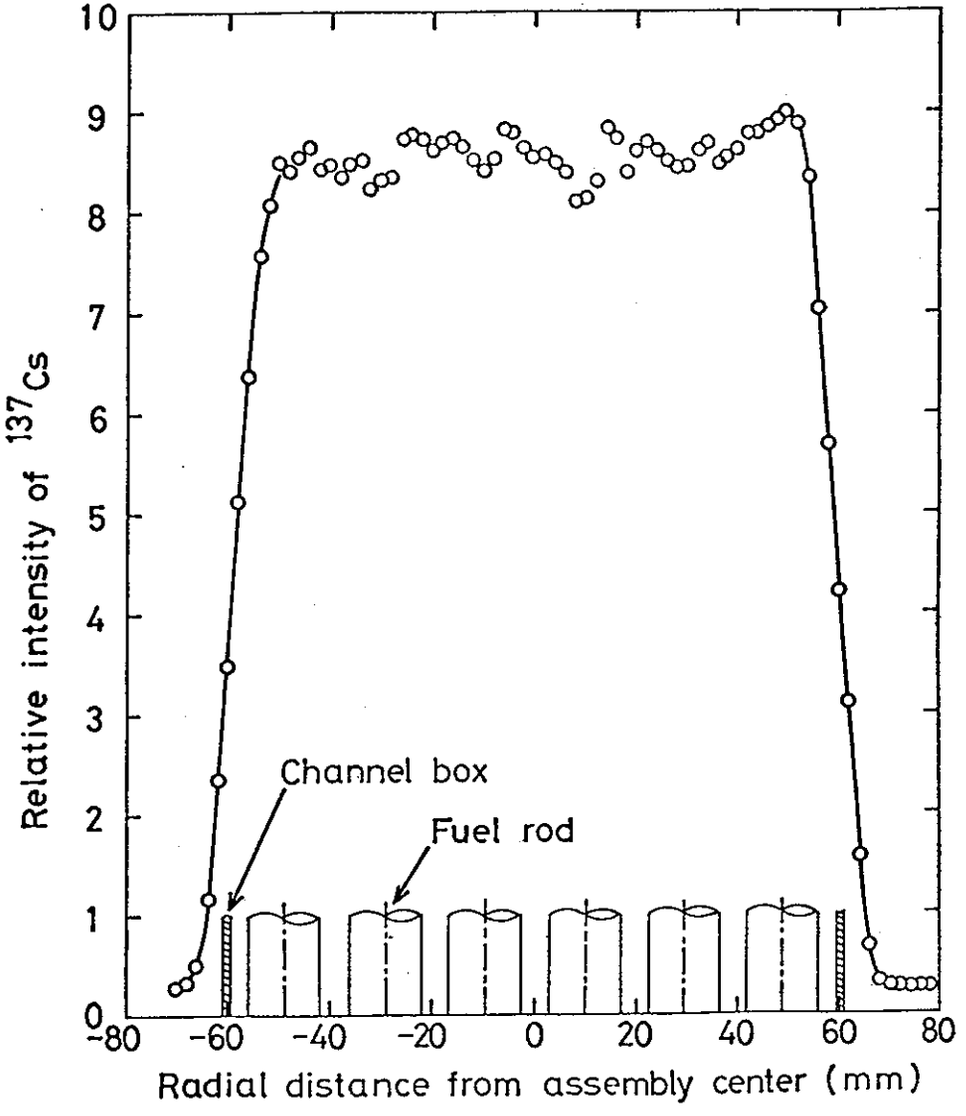



Fig. 9 Radial distribution of $^{137}Cs$ activity at +428 mm position on Plane 1 of A-8 assembly.

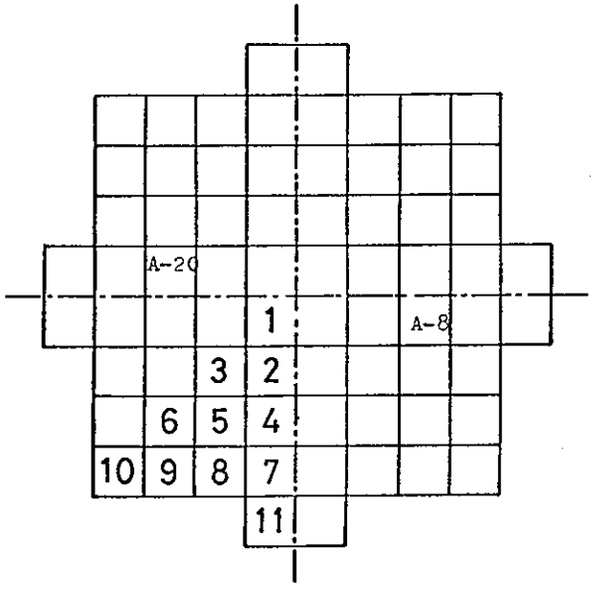



Fig. 10 Locations of A-8, A-20, and eleven assemblies in the one-octant region in core.