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51

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Measurement of Fission Rate Ratios and Determination of Bucklings on PuO_2-UO_2 Lattices

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Abstract

Plutonium fission rate ratios and material bucklings in PuO_2-UO_2 lattices have been measured as a function of plutonium fuel enrichment by the use of the Deuterium Critical Assembly (DCA). The content of plutonium fissile is about 91 wt % and the plutonium fuel enrichment defined as the weight percent of PuO_2 to PuO_2-UO_2 are 0.54 wt % and 0.87 wt %.

Plutonium fission rate ratios were obtained by the foil activation method by the use of Pu-Al and ^{235}U-Al alloy foils. Material bucklings were obtained by progressive substitution method developed from the second order perturbation theory.

The experimental results were compared with calculational by the nuclear design code METHUSELAH II.

It is concluded from this study that the dependence of 0.3 eV resonance fission of ^{239}Pu on the thermal neutron spectrum is estimated rather weak in the METHUSELAH II code. This code, however, makes good predictions on material buckling. Consequently, the nuclear design code METHUSELAH II has the acceptable accuracy on plutonium fuel enrichment adopted in the FUGEN core.

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Contents

1. Introduction

FUGEN is a heavy water moderated, boil-ing light water cooled pressure tube type reactor being built in Japan. The main feature of FUGEN is that it will use plutonium mixed oxide fuel in the central region of the initial core, thus reducing coolant void reactivity and more effectively utilizing its fuel[(1)]. In order to reduce local peaking in the fuel assembly, fissile Pu concentration of the mixed oxide fuel rods is set at 0.55 wt % for the outer ring and 0.8 wt % for the inner rings.

Since the average enrichment of the plutonium fissile in the fuel cluster is about 0.7 wt %, more than half of the fission processes are caused by plutonium nuclei in the lattice of the central region. This necessitates the experimental clarification of the nuclear characteristics of the plutonium fueled lattice and confirmation of the calculational accuracy of the nuclear design codes. Heavy water critical experiments on cell thermal flux distributions and void reactivity in the plutonium lattice of the DCA have already been reported [(2)] [(3)].

^{239}Pu has a sharp resonance absorption peak at 0.3 eV. From the microscopic point of view, the thermal neutron spectrum around 0.3 eV is largely influenced by this absorption. As the result, macroscopic parameters such as material buckling must be also affected in a complicated manner. In order to find out how the nuclear characteristics are affected by resonance absorption at 0.3 eV, ^{239}Pu fission rate ratios and material bucklings were measured as a function of the enrichment of plutonium fissile. Examination was also made as whether the accuracy of the METHUSELAH II[(4)] code is appropriate even if the fuel is highly enriched with plutonium.

2. Measurements

2.1 Experimental conditions

Dimensions and compositions of the plutonium fuel pellet are shown in TABLE 1. The plutonium fuel enrichment is here defined as the weight percent of PuO_2 to PuO_2-UO_2 mixture. The content of plutonium fissile is about 91 wt %, while the content of ^{240}Pu is considerably lower. The average enrichment of plutonium fissile in the fuel cluster of FUGEN is set between 0.54 wt % Pu enrichment and 0.87 wt %.

The cross section of the fuel assembly is shown in FIG.1. The cluster consists of 28 rods separated from the D_2O moderator by an aluminium pressure tube, an air gap, and an aluminium calandria tube. The fuel rods are arranged with their centers on three concentric circles. The experiments were conducted with and without H_2O coolant in the pressure tube (0 % and 100 % coolant void fractions)

In FIG. 2, the plane view of the core configuration of the DCA is shown. As an example, the core configuration comprising 25 plutonium mixed oxide fuel cluster is shown in this figure. The central region was loaded with plutonium mixed oxide fuel clusters and in the surrounding region 1.2 wt % enriched UO_2 fuel clusters were used.

2.2 Fission rate ratios

The measurement of plutonium fission rate ratios was made by the use of 0.1 mm thick Pu-Al alloy and 89.9 wt % enriched ^{235}U-Al alloy foil. As shown in FIG. 3, these foils were set between PuO_2-UO_2 pellets in each ring of four asymmetrical positions 1, 2, 3 and 4 shown in FIG. 1. The foils were enveloped in 0.02 mm thick Al foil to avoid contamination caused by fission products and PuO_2-UO_2 powder(5). The measurements of the cadmium ratio of the plutonium fission rate in fuel were taken using a 0.5 mm thick cadmium ring, and the neutron flux depression caused by the cadmium was corrected experimentally.

Definition of plutonium fission rate ratios are as follows:

$$\delta^{49} = \frac{\int_{Ecd}^{\infty} \sigma_f^{239} (E)\ \phi(E)\ dE}{\int_{0}^{Ecd} \sigma_f^{239} (E)\ \phi(E)\ dE}$$

$$\delta = \frac{\int_{0}^{\infty} \sigma_f^{239} (E)\ \phi(E)\ dE}{\int_{0}^{\infty} \sigma_f^{235} (E)\ \phi(E)\ dE} .$$

δ^{49} is defined as the ratio of epicadmium fissions to the sub-cadmium fissions of ^{239}Pu, and derived from the plutonium foil fission cadmium ratios. δ is defiend as the ratio of ^{239}Pu fissions to ^{235}U fission, and derived from the ratio of specific activities of enriched plutonoium foils to enriched uranium foils.

A 2"ϕ×2" NaI scintillation counter was used to detect the γ-rays from the fission products. The detector was set with enough interval from the foil to avoid a pile-up effect caused by γ-rays from ^{241}Am decay. The counting of γ-rays was done several times about 20 hours after the end of irradiation.

2.3 Material buckling

The progressive substitution method was adopted to measure the material buckling, Bm^2, with the use of a small amount of test fuels in question and a well-known reference core. When part of the uranium fuel in the reference core is replaced with plutonium fuel, two problems are conceivable: a large disturbance in neutron flux distribution and a mismatching in the neutron spectrum. Therefore, a formula was developed from the second order perturbation theory to take into account the above two effects.

The reference core consists of 121 clusters of 1.2 wt % enriched UO_2 fuel. Material buckling of the reference core, B_m^2 ref, was measured by the Cu wire activation method: radial and axial neutron flux distributions were fitted respectively to Bessel and cosine functions. Data near the core boundary were omitted in this fitting.

The UO_2 fuel clusters in the central part of the reference core were progressively replaced with 1, 5, 9 up to 25 clusters of PuO_2-UO_2 fuel shown in FIG. 4. The substituted fuel clusters were arranged around the core axis with four-fold rotation symmetry. Critical D_2O heights were converted to axial buckling by using extrapolation length measured in the reference core in order to obtain differences in axial buckling from that of the reference core, δB_z^2.

The relation between δB_z^2 and the substituted number of the fuel clusters was derived from the second order perturbation theory. The formula is as follows

$$\delta B_z^2 = \alpha W_{00} + \beta \sum_{m,n\neq 0} \frac{W_{mn}^2}{\delta B_z^2 + \left(\frac{\lambda_{4m,n}}{R}\right)^2 - \left(\frac{\lambda_{00}}{R}\right)^2} ,$$

where W_{mn} is a function of the dimension of the substituted area. $\lambda_{4m,n}$ represents the n-th zero point of the 4m-th order Bessel function, R the effective radius of the reference core. Coefficients α and β were determined from the least square fitting of δB_z^2 to the equation. Since in a fully substituted core W_{00} becomes unity and W_{mn} $(m,n\neq 0)$ vanishes, the material buckling of the unknown PuO_2-UO_2 lattice is obtained from the following relation,

$$B_m^2 = B_{m\ ref}^2 + \alpha$$

3. Results and Discussion

In FIG. 5, experimental results of δ^{49} and δ measured for plutonium fuels at 0 and 100 % coolant void fractions are shown as a function of Pu enrichment, together with the results calculated by METHUSELAH II code.

Measurement errors for the δ^{49} and δ were estimated as each $\pm$4.0 % and $\pm$ 5.1

From the definition of δ^{49} it is understood that δ^{49} indicates the ratio of epi-cd to sub-cd fission reaction rates. Therefore the experimental results of δ^{49} mean that δ^{49} increases with the increase of Pu fuel enrichment and changes in proportion to changes in the absorption cross section. Changes in the void fraction from 100 % to 0 % cause the experimental values of δ^{49} to greatly decrease. This is readily understood from the additional slowing down effect of H_2O coolant.

From the definition of δ it is also understood that δ is a kind of spectral index of thermal neutron flux around 0.3 eV resonance of plutonium absorption. Experimental results of δ reveal that the relative number of neutrons in the region of the 0.3 eV resonance increases due to absorption hardening of the spectrum with an increase in Pu enrichment.

On the other hand, dependence of the calculated δ^{49} and δ on Pu enrichment is less than with the experimental resutls. Increasing of the coolant void fraction further emphsises this trend, and it is suggested that the calculation code underestimates plutonium absorption of the 0.3 eV resonance. However, agreement between the experiment and the calculation is fairly good, especially in the 0 % void fraction.

In FIG. 6, experimental values of material bucklings are compared with calculations by METHUSELAH II as a function of Pu enrichment and coolant void fractions. Measurement errors of material bucklings were estimated about $\pm$2 %. The values of material bucklings increase with Pu enrichment in both 0 % and 100 % void fractions due to the increased fissionable material. Changes in the void fraction from 100 % to 0 % cause the values of material bucklings to increase. It is thought mainly due to the increase in the slowing down power of the coolant material. Agreement between the experiment and the calculation is better in 0 % void fractions than in 100 % void fractions.

4. Concluding Remarks

Plutonium fission rate ratios and material bucklings in PuO_2-UO_2 lattices have been studied as a function of plutonium fuel enrichment.

It is concluded from this study that the dependence of 0.3 eV resonance fission of ^{239}Pu on the thermal neutron spectrum is estimated rather weak in the METHUSELAH II code. This code however, makes,good predictions on material buckling. Consequently, the nuclear design code METHUSELAH II has acceptable accuracy or plutonium fuel enrichment adopted in the FUGNE core.

The authors are indebted to Mr. T. Wakabayashi and the operating staff of DCA for their help in the experiments, and the discussions. We would like to thank Dr. Y. Hachiya for his critical reading of the manuscript. We would like also to thank managers Mr. Y. Miyawaki and Mr. H. Sakata (former) for their strong support given to this work.

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TABLE 1. DIMENSIONS AND COMPOSITIONS OF FUEL PELLET

Fuel Pellet	0.54w/o PuO_2-UO_2	0.87w/o PuO_2-UO_2
Outer Dia. (cm)	1.47	1.47
Density (g/cc)	10.17	10.17
Isotope (w/o)		
U-235	0.6214	0.6194
U-238	86.782	86.503
Pu-238	0.000102	0.000145
Pu-239	0.4304	0.6849
Pu-240	0.04115	0.06584
Pu-241	0.004359	0.006960
Pu-242	0.000303	0.000510
O-16	12.12	12.12

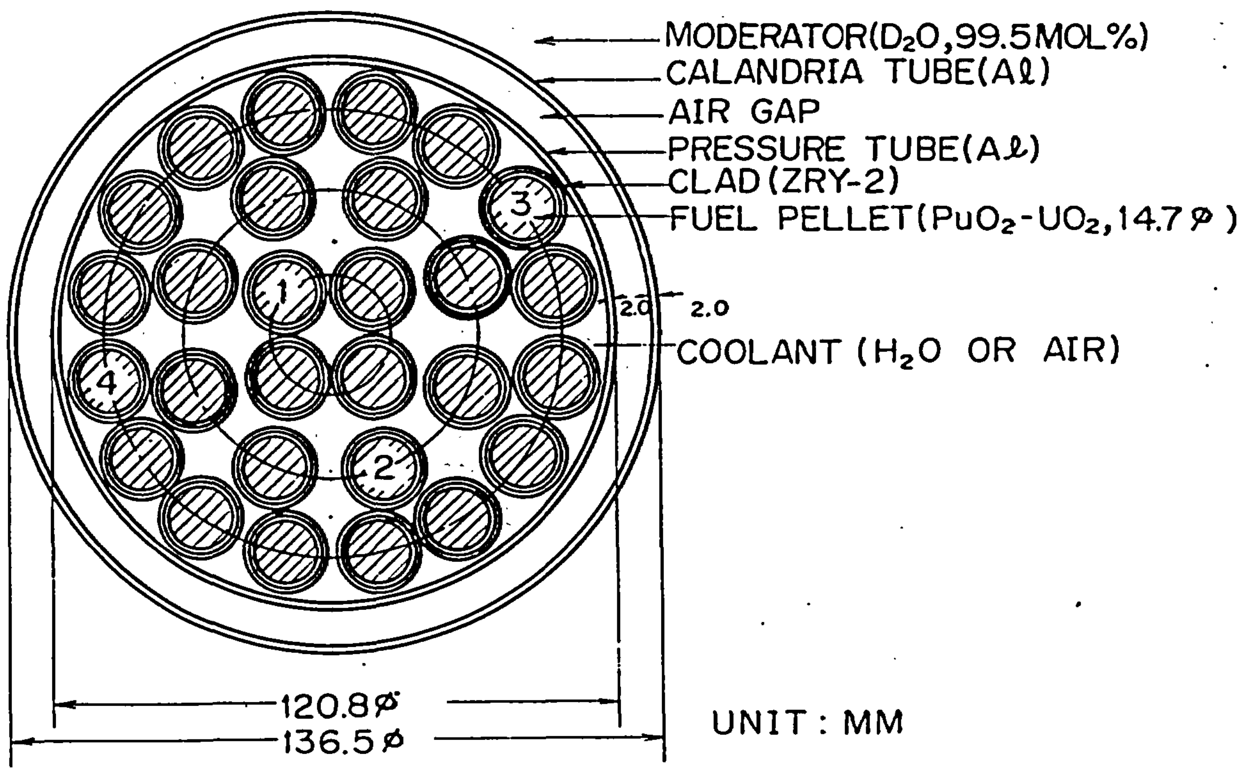

FIG.1. CONFIGURATION OF PLUTONIUM FUEL ASSEMBLY

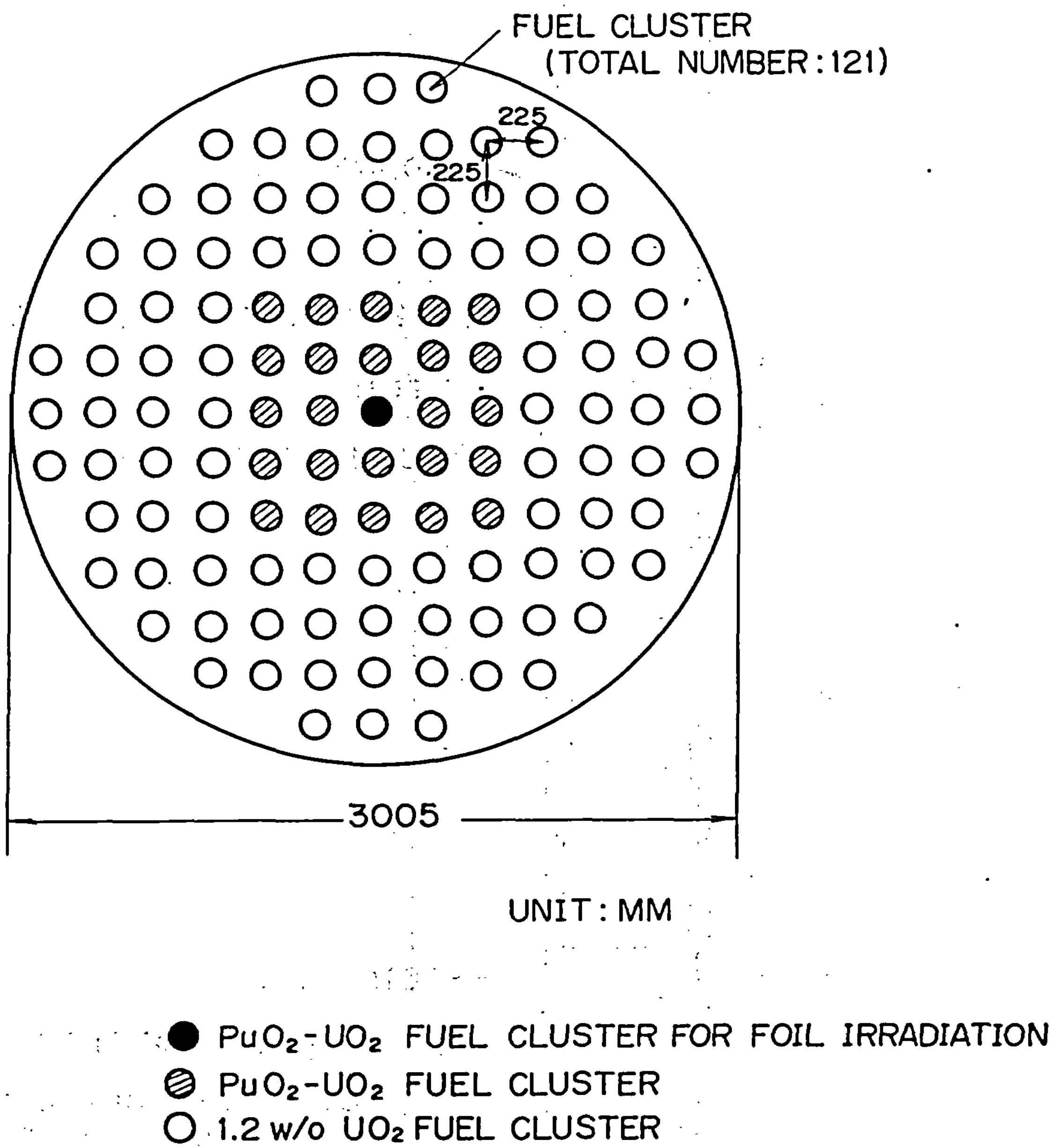

FIG.2. DCA CORE CONFIGURATION

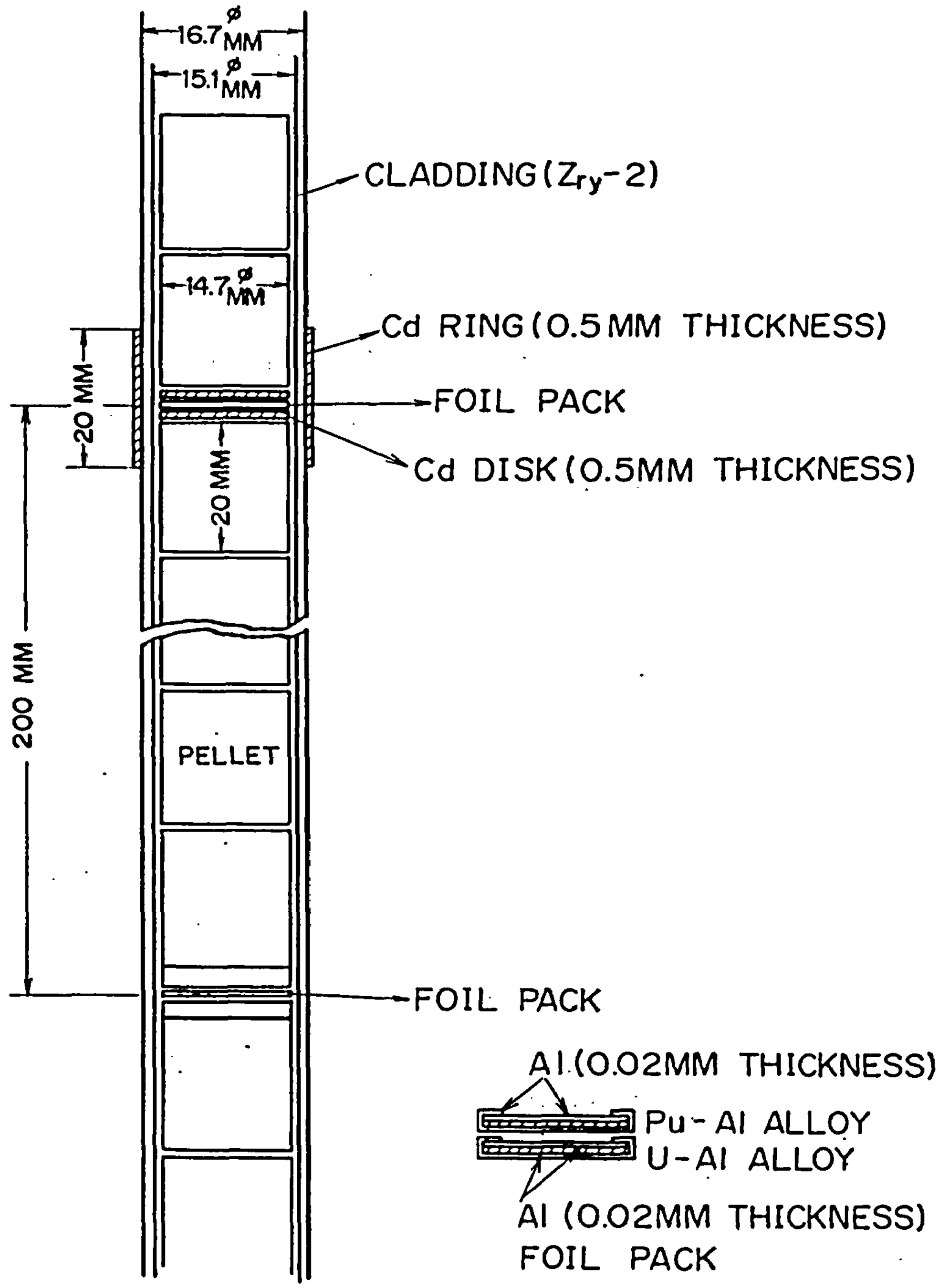

FIG.3. FOIL ARRANGEMENT IN PuO_2-UO_2 FUEL

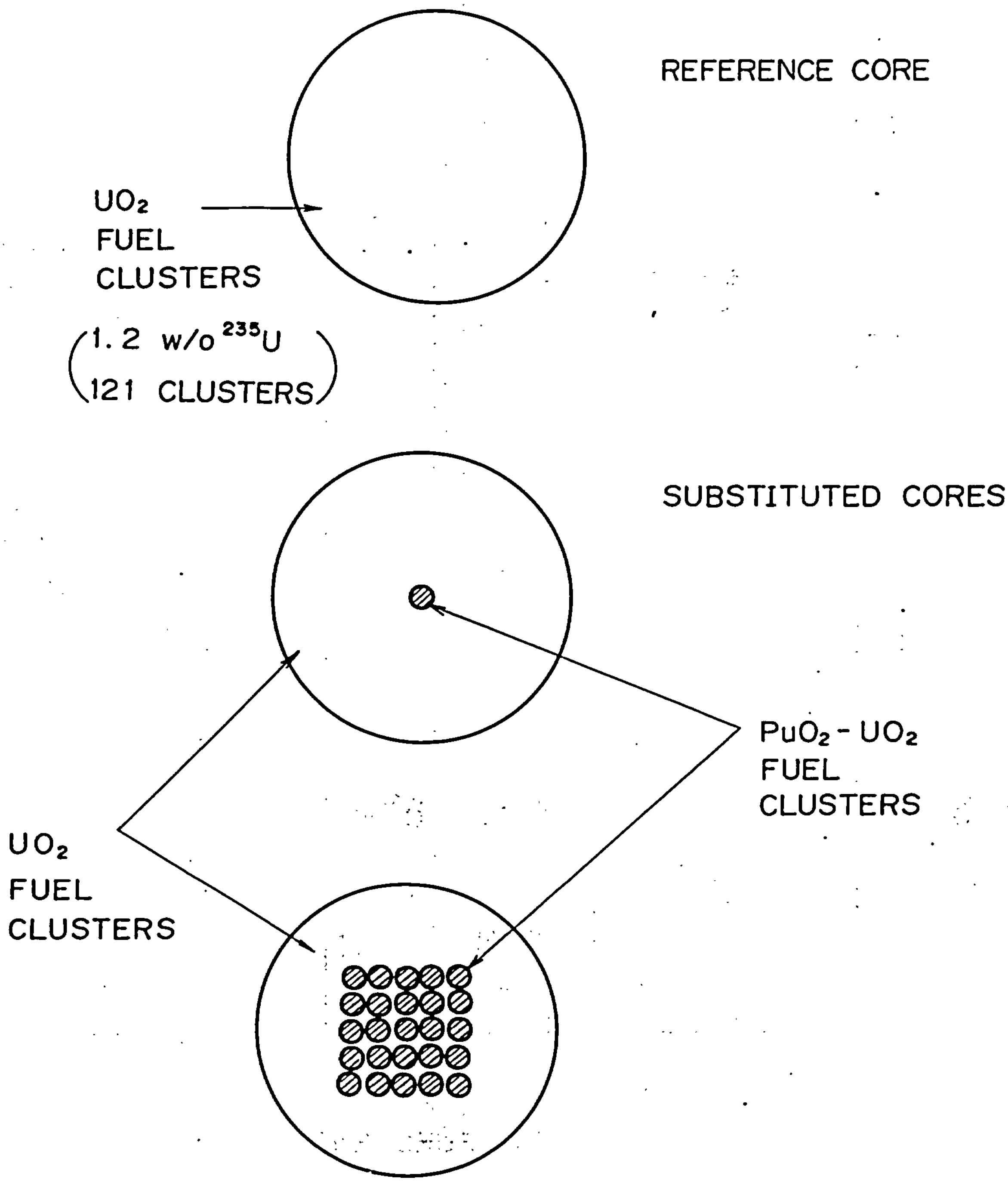

FIG.4. PROGRESSIVE SUBSTITUTION EXPERIMENT

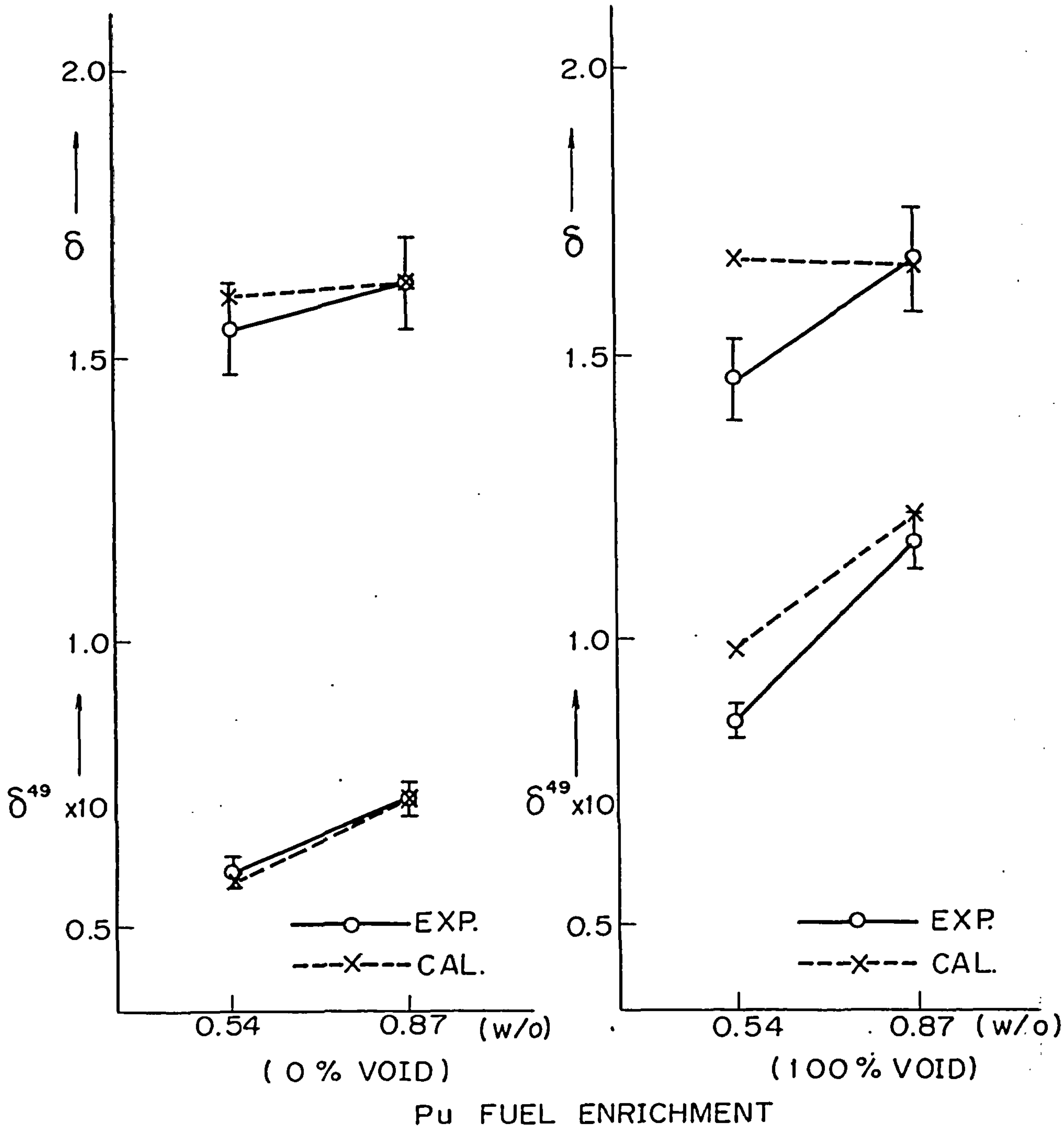

FIG.5. DEPENDENCE OF δ AND δ^{49} VS. Pu FUEL ENRICHMENT

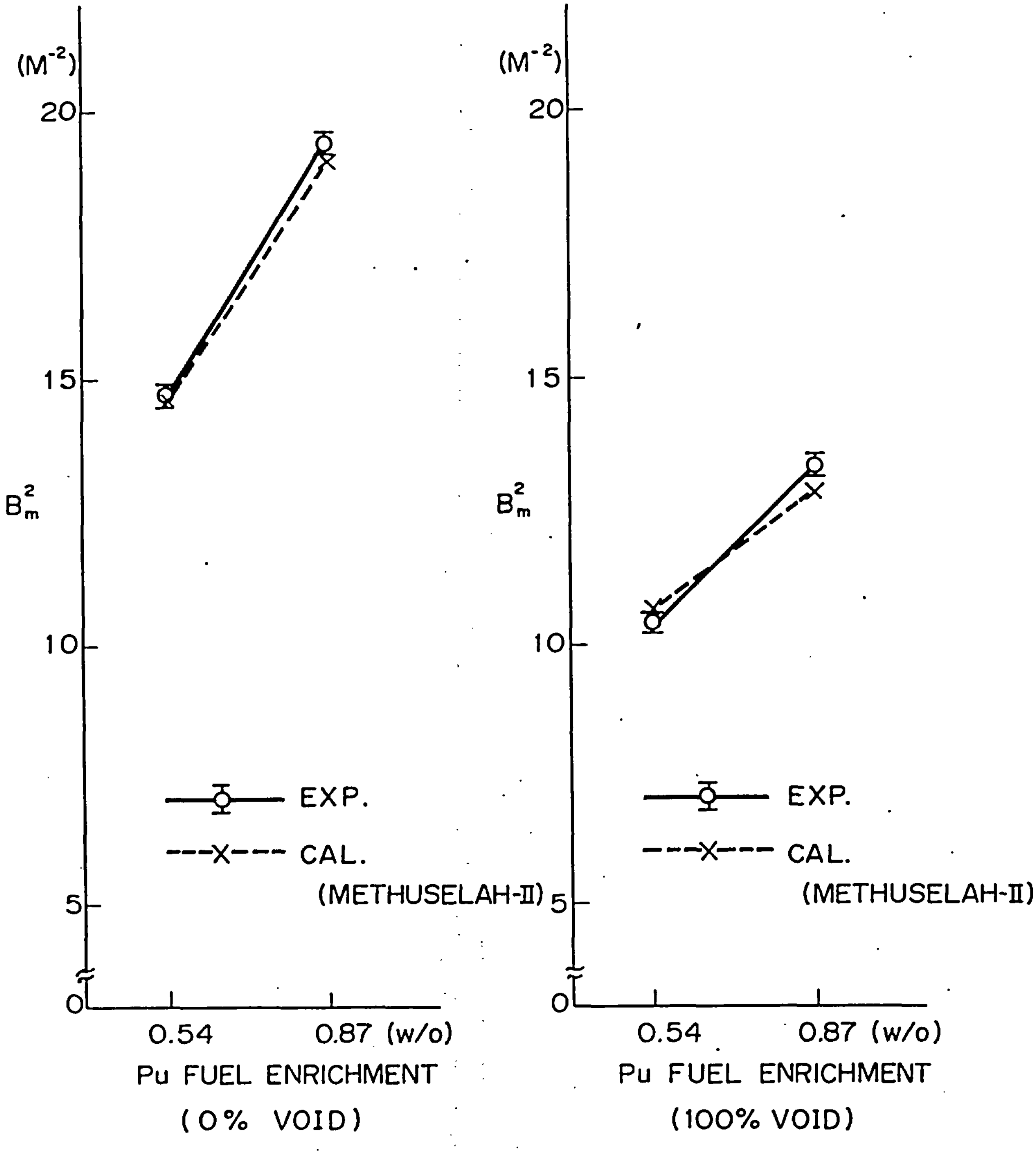

FIG.6. DEPENDENCE OF B_m^2 VS. Pu FUEL ENRICHMENT