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**CORRELATIONS BETWEEN HEAVY ISOTOPES IN IRRADIATED
FUELS OF LIGHT WATER POWER REACTORS**

by

C. FOGGI and P. FRANDOLI

1974



**Joint Nuclear Research Centre
Ispra Establishment - Italy**

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March 1974

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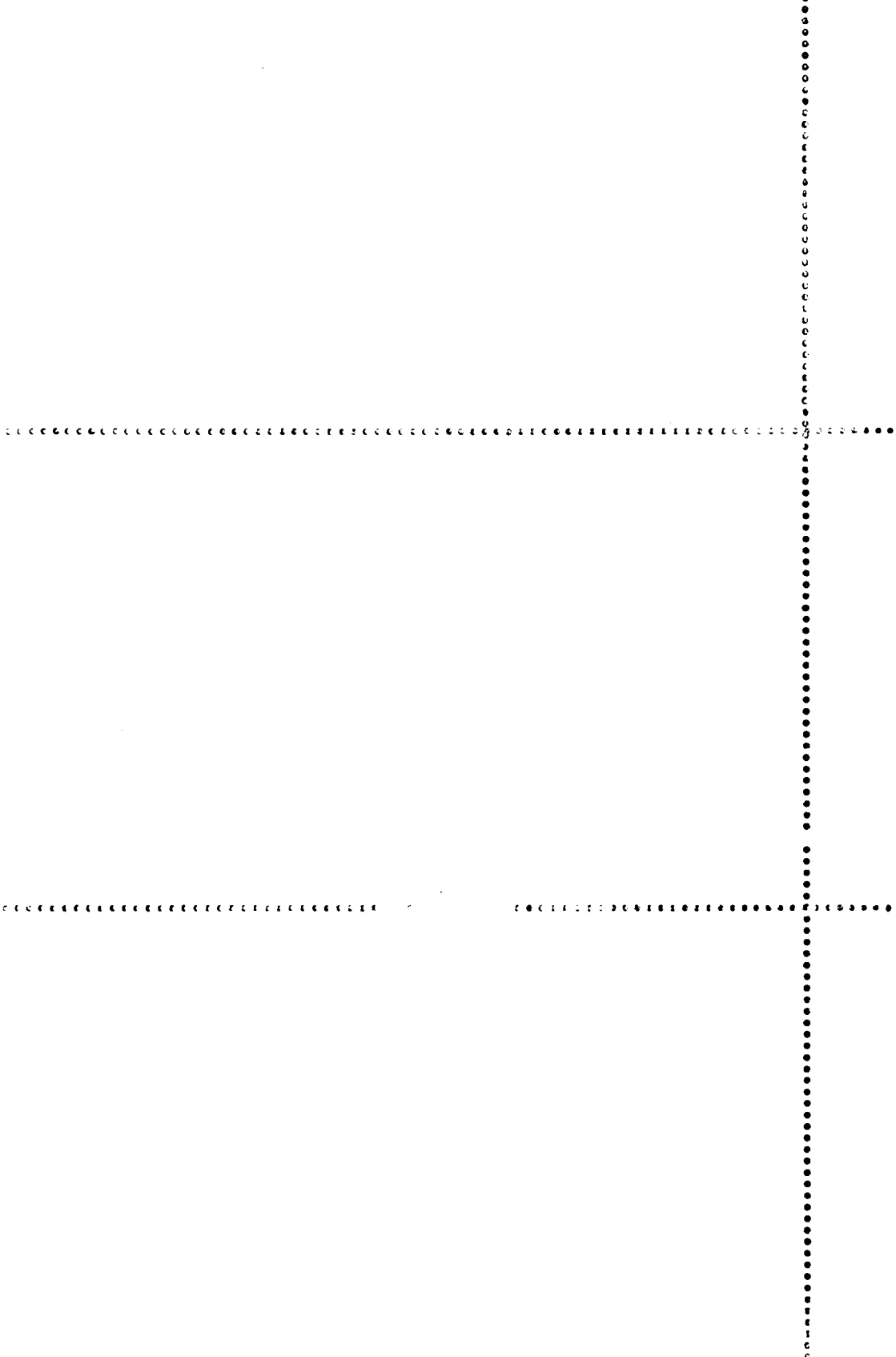
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ABSTRACT

A theoretical study of the correlations between isotopes of plutonium and uranium in irradiated fuels of light water power reactors has been performed. The dependence of the correlations upon several reactor parameters (fuel enrichment, moderator-to-fuel volume ratio, cladding material, reactivity control method) has been analysed; a wide range of possible lattices for pressurized reactors, and some specific lattices for boiling reactors have been considered. Analytical expressions for the correlations have been prepared in several cases. Comparison has been made with experimental data concerning the fuels of the reactors TRINO VERCELLI, YANKEE ROWE, DRESDEN 1, HUMBOLDT BAY.

KEYWORDS

POWER REACTORS
BOILING WATER REACTORS
PRESSURIZED WATER REACTORS
SAFEGUARDS
CORRELATIONS
ISOTOPE RATIO
PLUTONIUM ISOTOPES
URANIUM ISOTOPES
NUCLEAR FUELS
BURNUP
ANALYTICAL SOLUTION
ANALYTIC FUNCTIONS

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$$x(o) / MR$$

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1. INTRODUCTION

The irradiation of a nuclear fuel causes important and complex changes in its isotopic composition, namely:

- depletion of the isotopes of uranium initially present (and, where applicable, of plutonium);
- build-up of artificial isotopes of heavy elements (e.g. ^{236}U , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu);
- build-up of fission products.

These changes are often correlated to each other in very simple manner; in many instances, the relationship representing the correlation occurs to be a simple proportionality between depletion and build-up of selected groups of isotopes; in other cases, its regular form allows a simple analytical representation.

Normally, each correlation is influenced to a certain extent, by the main characteristics of the core and by the behaviour of the reactor. However, since a certain number of correlations can be expressed in very simple analytical forms, and depend on few reactor parameters through well known relationships, it is clear how they can constitute a valid alternative to the experimental determination of the quantity of some isotopes; in fact, whenever the direct measurement is difficult or impossible or unreliable, the use of a suitable correlation enables to deduce the quantity of these isotopes from the concentrations of other isotopes that are more easily measured.

The correlation technique was expressly developed to satisfy this requirement, particularly in the field of fissile material safeguards; at present this technique is also considered for application to problems concerning reactor operation and fuel reprocessing. Many applications can be cited, especially those made in the U.S.A. (ref. 4-5-7-8-9-10).

The post-irradiation analysis of pellet size samples of spent fuel from different reactors provided the first basis for the establishment of the correlation technique. Consequently, input accountability batch data at the reprocessing plant were the subject of detailed correlation studies.

After having gained experience on empirical correlations obtained by means of experimental data, the necessity was felt to make a theoretical analysis so as to better understand and value the influence of reactor parameters on the correlations, and to extrapolate them to wider irradiation ranges.

The scope of the present paper is to determine the influence of the various parameters of the reactor on the correlations between heavy isotopes; comparison with available experimental data has also been made.

2. GENERAL

The theoretical study of the correlations between isotopes of uranium and plutonium was performed on irradiated fuels of light water reactors; both PWR's and BWR's were considered. The study has been based on the analysis of the fuel history of a single cell; therefore, the correlations obtained refer to small portions of fuel, that is they are "point correlations"; in many cases these "point correlations" are also valid for the bulk of a fuel charge.

The calculations were carried out by the use of the cell burn-up code LASER (ref.1) which can take into account the neutron spectrum variations during the irradiation of the fuel.

The evolution of the isotopic composition of the fuel is influenced by various parameters, of which we have considered the following ones:

- initial enrichment of the fuel
- moderation ratio
- cladding material
- reactivity control method.

In the course of the study, these parameters were varied in a rather wide range in order to examine their influence on the correlations; the results thus obtained enable to understand the differences in the isotopic composition of fuel pellets taken from different positions of the assembly, or from different assemblies of fuel charge, similarly, these results enable to evaluate the changes in the correlations when fuel charges of different characteristics are considered.

As regards PW reactors, a series of burn-up calculations was made, by varying some of the above parameters in such a way as to cover a wide range of possible lattices; the lattices of the standard reactors now built are easily within the limits of the assumed variabilities.

The lattices studied have the following characteristics:

- fuel	UO ₂
- fuel pellet diameter	0.932 cm
- cladding thickness	0.035 cm (SS) / 0.065 cm (Zr)
- cladding material	Zircaloy/stainless steel
- fuel density	10.3 g/cm ³ (94% of theoretical value)
- initial enrichment	2 w/o, 3 w/o, 4 w/o
- moderation ratio	1.2, 1.64, 2.2
- fuel temperature	667°C
- moderator temperature	285°C
- moderator density	0.74 g/cm ³

For each type of cladding material nine cases were examined, obtained by coupling the three initial enrichment values and the three moderation ratio values in all possible ways; the set of nine cases referring to the Zircaloy clad fuel was calculated by simulating two different reactivity control methods, that is:

- a) neutron leakage from the cell
- b) poison (boron) diluted in the moderator.

The set of nine cases referring to the stainless steel clad fuel was calculated by simulating only the first type of reactivity control.

With the results of the calculations relating to the PW reactors, a thorough investigation was made on the way in which the correlations depend on the initial enrichment, the moderation ratio, the cladding material and the control method. A comparison was made between our results and the experimental data obtained from the analysis of the irradiated fuels of the TRINO VERCELLESE and YANKEE ROWE reactors (ref. 2-3).

The study of BW reactors has so far been limited to the examination of only five cases, the characteristics of which are described in Table 1;

Table 1:

Case	Initial Enrichment (w/o)	$\frac{\text{Vol. H}_2\text{O}}{\text{Vol. UO}_2}$	Cladding Material	Voids in the Moderator	Fuel Pellet Diameter (in.)
1	2.5	2.6	SS	20%	0.4
2	2.5	2.6	Zr	20%	0.4
3	2.5	2.6	SS	0	0.4
4	1.5	2.1	Zr	20%	0.498
5	1.5	2.1	Zr	0	0.498

the lattice parameters of these cases were chosen so as to reflect the characteristics of the DRESDEN I and HUMBOLDT BAY reactors, since the greatest part of the experimental data available to us for comparison, concerns the fuel coming from these two plants (ref. 4-5).

In both cases the study was extended to a wide irradiation range (0-30,000 Mwd/tU).

3. TYPES OF CORRELATIONS ANALYSED AND GENERAL CONCLUSIONS

In the course of our analysis we have considered especially those correlations which have already been evidenced by various authors; we have given particular attention to those presenting an accentuated linear behaviour; in fact, they are of great interest since they are simple in form and can be easily expressed analytically, with an evident advantage in their practical use; as a result of this, a more complete study has been made on the correlations having a linear behaviour.

The study of each correlation includes two phases:

- the first phase covers the determination of the form of the function; in the case of marked linearity, the calculated points were fitted with a straight line; errors caused by the substitution of the fitted line to the actual points, were calculated;
- the second phase covers the determination of the dependence of the correlation from the different reactor parameters; when this dependence is sensibly linear, analytical expression of the correlation as a function of the initial enrichment and the moderation ratio, has been given.

The correlations analysed were divided into three groups:

a) correlations among isotopes of uranium

$$\begin{array}{lll} {}^{236}\text{U}/{}^{238}\text{U} & \text{versus} & \text{depletion of } {}^{235}\text{U} \\ {}^{236}\text{U}/{}^{238}\text{U} & \text{versus} & {}^{235}\text{U}/{}^{238}\text{U} \\ {}^{236}\text{U}/{}^{235}\text{U} & \text{versus} & {}^{236}\text{U}/{}^{238}\text{U} \end{array}$$

All the quantities are expressed by weight.

The analyses have shown that the first two correlations are definitely linear in the whole irradiation range examined (figs.1, 7); this confirms the conclusions

of the experiments; the relative differences between the actual curve and the fitting straight line, are lower than 1.5%.

The third correlation is not linear (fig.5), but nonetheless has a very regular form which can be well represented by a parabola.

The cladding material and the reactivity control method have little influence on these correlations (see paragraph 4.1 and 4.2).

The initial enrichment and the moderation ratio greatly influence, especially the first, all three correlations, thorough investigation was made so as to determine the dependence from the cited factors. The results are set out in detail in paragraph 4.1 and 4.2.

All three calculated correlations agree with the experimental results; the comparison was made with the data obtained from the analysis of the irradiated fuel of the reactors : DRESDEN I, HUMBOLDT BAY, YANKEE ROWE and TRINO VERCELLESE; the differences between calculates results and experimental data are limited to a few percent (figs.4, 6, 9).

b) correlations between Pu/U ratio and the isotopes of uranium

One of the most important correlations, which has been taken into consideration by the studies so far made, is that which relates the plutonium accumulated during irradiation to the depletion of ^{235}U , that is:

Pu/U versus Depletion of ^{235}U

where:

Pu = quantity of plutonium (by weight)

U = quantity of uranium (by weight)

The theoretical analysis of the correlation confirmed the linearity (figs. 10, 11, 12) already ascertained by the use of experimental data; the errors made by substituting over a wide irradiation range (0 - 30,000 Mwd/tU) the actual curve with a straight line are lower than 1.5% - 2%; obviously, rectification of the curve over a more restricted range (in the usual discharge limits of the fuel, i.e. 15,000 - 20,000 Mwd/tU), causes the error to decrease down to few per mil.

The correlation greatly depends from the initial enrichment and the moderation ratio and also the reactivity control method has a remarkable influence on the correlation behaviour; cladding material has little influence on this correlation. The results are set out in detail in paragraph 4.3 and 5.

For the PW reactors the comparison with the experimental data gave satisfactory results. For the BW reactors the comparison presented some difficulties as in some cases appreciable differences were found; a possible explanation for this fact is given in paragraph 5.

c) correlations between Pu/U ratio and the isotopes of plutonium

Many correlations come within this category as there is a number of parameters which can be formed with the basic isotopes of plutonium (^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu). The investigation, so far made on a considerable number of correlations, led to the ascertainment of an accentuated and irregular variability with the different parameters of the reactor, especially with the reactivity control method; this has so far prevented us from describing with sufficient precision the dependence of the correlation from these parameters.

The results are set out in detail in paragraph 4.4.

4. DESCRIPTION OF THE CHARACTERISTICS OF THE CORRELATIONS (PWR's)

4.1. $^{236}\text{U}/^{238}\text{U}$ versus depletion of ^{235}U

4.1.1. General

The depletion of ^{235}U can be expressed in two different forms, that is:

$$D_5 = \frac{^{235}\text{U}(o) - ^{235}\text{U}}{^{235}\text{U}(o)}$$

$$d_5 = x(o) - x$$

where:

$^{235}\text{U}(o)$ = quantity of ^{235}U in non irradiated fuel (by weight)

^{235}U = quantity of ^{235}U in irradiated fuel (by weight)

^{236}U = quantity of ^{236}U accumulated during irradiation (by weight)

^{238}U = quantity of ^{238}U in irradiated fuel (by weight)

$x(o)$ = initial fuel enrichment (w/o)

x = fuel enrichment after irradiation (w/o)

4.1.2. Analysis of the correlation

The basic characteristic of the correlation is the linearity (fig.1), whether the depletion of ^{235}U is expressed with the parameter D_5 or with d_5 . The correlations can therefore be written as follows:

$$\frac{^{236}\text{U}}{^{238}\text{U}} = P D_5$$

$$\frac{^{236}\text{U}}{^{238}\text{U}} = p d_5$$

Coefficients P and p, represent the slope of the fitting straight line determined over an irradiation range from 0 to 30,000 Mwd/tU; the relative differences between the actual curve and the straight line are generally not higher than 2%. If the rectification is made over a more restricted range, the deviation can be as low as few per mill.

4.1.3. Analysis of slopes P and p

The slopes of the correlation, defined in paragraph 4.1.2., are expressed as follows:

$$P = \frac{^{236}\text{U}/^{238}\text{U}}{D_5}$$

$$p = \frac{^{236}\text{U}/^{238}\text{U}}{d_5}$$

They depend on several parameters of the lattice, of which the following ones have been analyzed:

- initial fuel enrichment
- moderation ratio (vol. H₂O/vol. UO₂)
- cladding material
- cell reactivity control method

The ratios P and p are mainly influenced by the moderation ratio and the initial enrichment (figs. 2, 3); the dependence from the two parameters is parabolic, but, in the second case the parabola has a very small curvature, so that it is possible to substitute it with a straight line, with errors not greater than 1% for P and 0.5% for p.

In both cases, it was found that the cell reactivity control method has not much influence on the correlation.

In fact, two series of calculations were made simulating two different control methods, one by diluting boron in the moderator and the other by allowing leakage of neutrons from the cell. The curves relating to the two control systems differs slightly; therefore the mean curve was adopted as being representative of every control situation.

In figs. 2 and 3 are shown ratios P and p for the two control methods mentioned; it can be observed that the mean curve differs from the two cases by about 1.5%. It should be born in mind that the two reactivity control methods considered, represent two extreme hypotheses that correspond to real situations only in a few very particular cases (for example, reactivity control by neutron leakage takes place only in those zones of boiling water reactors having high voids). In fact, the reactivity control of a cell corresponds to a combined action of neutron leakage and local absorption of neutrons; therefore it can be reasonably assumed that the correlation concerning the fuel of a reactor is represented, with a good precision, by the previous defined mean curve.

Concerning the influence of cladding material, it was found that the use of stainless steel in place of Zircaloy causes an increase in the values of P and p in the order of 1.5% - 1.8%.

The analytical expression of the correlations for the average control system (as defined above) and for Zircaloy cladding are:

$$\frac{^{236}\text{U}}{^{238}\text{U}} = [(0.14439 \cdot x - 0.10278) \text{MR}^2 - (0.78396 \cdot x - 0.63734) \text{MR} + (3.1488 x - 1.7368)] \cdot 10^{-3} \cdot D_5$$

$$\frac{^{236}\text{U}}{^{238}\text{U}} = \left[(0.11794 \cdot x + 0.78800) \text{MR}^2 - (0.8020 x + 3.5151) \text{MR} + (2.4056 x + 18.944) \right] \cdot 10^{-2} \cdot d_5$$

MR is the moderation ratio.

4.1.4. Comparison with experimental data

The correlation is based on the amount of ^{236}U formed during irradiation, to the exclusion, therefore, of the ^{236}U initially present in the fuel; practice has shown that the greater part of the fuel used in the LW reactors contains a certain amount of it. In order to be able to compare the experimental data with the calculations, it was necessary to subtract from the measured amount, the residual quantity of the initial ^{236}U : the result represents the quantity of ^{236}U formed during the cycle examined. The method used for the correction of the experimental data is set out in the appendix.

Comparisons were made with experimental data concerning the fuel of TRINO and YANKEE reactors, i.e.:

- a) data obtained from the analysis of fuel pellets from 3 assemblies of the first core of TRINO reactor, with enrichments of 2.72 w/o, 3.13 w/o and 3.9 w/o; stainless steel cladding; the initial content of ^{236}U (ref.2) is not known;
- b) data obtained from the input analyses at the reprocessing plant for the fuel of the YANKEE reactor, core V (enrichment 4.1 w/o), core VI (enrichment 4.9 w/o) and core VII (enrichment 4.93 w/o); stainless steel cladding; the initial content of ^{236}U is only known for core VII (0.0564 w/o) (ref.3,4).

When unknown, the initial quantity of ^{236}U was estimated as described in the appendix; the results are the following ones:

Reactor	Initial enrichment	$\frac{^{236}\text{U}}{^{238}\text{U}}$ estimated (w/oo)
TRINO VERCELLESE	2.72	0.43
	3.13	0.43
	3.9	0.53
YANKEE ROWE	4.1	0.43
	4.9	0.48

Fig.4 shows the calculated curves of P relative to stainless steel clad fuel, and the experimental values of the ratio $(\frac{^{236}\text{U}}{^{238}\text{U}})/D_5$; where several data are available for a single fuel type, only the variability range of this ratio is indicated (see also Table 2). The slopes P of the correlations obtained fitting the experimental points, core by core, are also reported in Table 2.

Reactor	Initial enrichment (w/o)	$(\frac{^{236}\text{U}}{^{238}\text{U}}) / D_5$ experimental		fit of experimental points
		Min.	Max.	
Trino	2.72	$4.60 \cdot 10^{-3}$	$5.20 \cdot 10^{-3}$	$4.84 \cdot 10^{-3}$
	3.13	$5.28 \cdot 10^{-3}$	$6.42 \cdot 10^{-3}$	$5.78 \cdot 10^{-3}$
	3.9	$7.61 \cdot 10^{-3}$	$8.03 \cdot 10^{-3}$	$7.81 \cdot 10^{-3}$
Yankee	4.1	$7.55 \cdot 10^{-3}$	$7.78 \cdot 10^{-3}$	$7.65 \cdot 10^{-3}$
	4.9	$9.97 \cdot 10^{-3}$	$10.13 \cdot 10^{-3}$	$10.04 \cdot 10^{-3}$
	4.93	$9.66 \cdot 10^{-3}$	$9.84 \cdot 10^{-3}$	$9.75 \cdot 10^{-3}$

Table 2

4.1.5. Conclusions

The correlations are linear and mainly dependent on the initial enrichment; they also depend on the moderation ratio and, in a lesser degree, on the cladding material and the reactivity control system.

This agreement with some measurements is due, most probably, to the fact that they refer to fuels irradiated in peripheral zones of the core and, therefore, subject to a perturbed neutron spectrum.

$$4.2. \quad \frac{^{236}\text{U}}{^{238}\text{U}} \quad \text{versus} \quad \frac{^{235}\text{U}}{^{238}\text{U}}$$

4.2.1. Analysis of the correlation

The correlation, shown in fig. 7, is linear and can be expressed as follows:

$$\frac{^{236}\text{U}}{^{238}\text{U}} = P \left[\frac{^{235}\text{U} (o)}{^{238}\text{U} (o)} - \frac{^{235}\text{U}}{^{238}\text{U}} \right] = P \Delta \left(\frac{^{235}\text{U}}{^{238}\text{U}} \right)$$

The symbols indicate the quantity by weight of each isotope; symbol (o) indicates that the quantity considered refers to non irradiated fuel.

Coefficient P represents the absolute value of the slope of the straight line fitting the calculated points of the correlation over an irradiation range from 0 to 30,000 Mwd/tU; maximum differences between the actual curve and the straight line are in the order of 2%.

All correlations relating to fuels having the same initial enrichment, intercept the axis of the abscissa at the same point, corresponding to the initial value of ratio $\frac{^{235}\text{U}}{^{238}\text{U}}$; the other parameters causes the straight line representing the correlation to rotate very slightly around this point; this limited influence of other parameters can be useful for certain applications of the correlation.

4.2.2. Analysis of slope P

The absolute value of the slope of the correlation can be expressed as follows:

$$P = \frac{^{236}\text{U} / ^{238}\text{U}}{\Delta(^{235}\text{U} / ^{238}\text{U})}$$

Slope P mainly depends on the initial enrichment and on the moderation ratio (fig.8); the cell reactivity control method does not have much influence on the ratio examined; also in this case, as already discussed in paragraph 4.1.3., the mean correlation amongst those relating to the two extreme control methods, can be considered as representative of each real situation. The cladding material does not have much influence; it was in effect found that the use of stainless steel cladding causes a 2.3% average increase in P, with respect to the use of a Zircaloy cladding.

The dependence of P from the initial enrichment can be well represented by a straight line; the expression is the following one (mean control system):

$$P = a x + b$$

Coefficient a depends in a linear manner on the moderation ratio, whereas b depends on the same parameter with a parabolic law.

The analytical expression of the correlation for a Zircaloy clad fuel and for the mean control system is therefore :

$$\frac{^{236}\text{U}}{^{238}\text{U}} = [(-0.003235 \text{ MR} + 0.016347) x + (0.012328 \text{ MR}^2 - 0.052125 \text{ MR} + 0.20131)] \cdot \Delta(^{235}\text{U} / ^{238}\text{U})$$

4.2.3. Comparison with experimental data

Values of the ratio $(^{236}\text{U} / ^{238}\text{U}) / \Delta(^{235}\text{U} / ^{238}\text{U})$ were derived from experimental data originated by the analysis of TRINO and YANKEE fuels; the experimental

values of ^{236}U were corrected to account for the initial quantity, according to the method described in appendix.

The following table shows the minimum and the maximum values of the ratio for each group of measurements referring to the same core.

For each core, also the correlation obtained by fitting the experimental points was determined; the corresponding values of P are reported in the following Table.

Reactor	Initial enrichment (w/o)	$\frac{^{236}\text{U}}{^{238}\text{U}} / \Delta(\frac{^{235}\text{U}}{^{238}\text{U}})$ experimental		Fit of experimental points
		Min.	Max.	
Trino	2.72	0.166	0.189	0.172
	3.13	0.167	0.202	0.181
	3.90	0.191	0.201	0.195
Yankee	4.1	0.190	0.196	0.193
	4.9	0.210	0.214	0.212
	4.93	0.189	0.193	0.191

Fig. 9 shows the dependence of P on the initial enrichment and the moderation ratio, for a stainless steel clad fuel; the variability range of the experimental values of the ratio $(^{236}\text{U}/^{238}\text{U}) / \Delta(^{235}\text{U}/^{238}\text{U})$ for each core, is also represented.

4.2.4. Conclusions

The correlation is linear on a wide irradiation range (0-30,000 Mwd/tU); it depends on the initial enrichment, the moderation ratio and, to a lesser degree, on the type of cladding used and the reactivity control method. A comparison with the experimental data gave satisfactory results.

4.3. $\frac{\text{Pu}}{\text{U}}$ versus ^{235}U depletion

4.3.1. General

The ^{235}U depletion has been expressed in two different forms, i.e. by parameters D_5 and d_5 previously defined in paragraph 4.1.1.; the other symbols indicate:

Pu = quantity of plutonium (sum of isotopes ^{239}Pu , ^{240}Pu ,
 ^{241}Pu , ^{242}Pu) accumulated during irradiation (by weight)

U = quantity of uranium present after irradiation (^{235}U plus
 ^{236}U plus ^{238}U), by weight.

4.3.2. Analysis of the correlation

The correlation is linear in either of the two forms in which the ^{235}U depletion is expressed (figs. 10, 11, 12); in fact, the differences between the actual calculated points and the fitting straight line do not generally exceed 2%; rectification was made on a wide irradiation range (0-30,000 Mwd/tU). It can be noted in figs. 10, 11 and 12 that the cell reactivity control method greatly influences the formation of plutonium, this fact is common to all the correlations in which isotopes of plutonium are represented, and causes some difficulties in the analysis of the correlations.

4.3.3. Analysis of ratios P and p (slope of the correlation)

The slopes of the correlations are represented by the following ratios:

$$P = \frac{\text{Pu}/\text{U}}{D_5}$$

$$p = \frac{\text{Pu}/\text{U}}{d_5}$$

The dependence on the initial enrichment, on the moderation ratio and on the reactivity control method is shown in figs. 13 and 14.

Ratio P is an increasing linear function of the initial enrichment whereas ratio p decreases according to a parabolic law; in both cases the dependence is strong. Also the moderation ratio greatly influences P and p.

The use of stainless steel cladding in place of Zircaloy cladding causes an increase of the slope of 4% in both cases.

The dependence of P and p on the cell reactivity control method was also analyzed. The differences caused by the use of either of the two control systems are great and depend on the initial enrichment (figs. 13, 14); the Pu/U ratio corresponding to control by poison in water is always higher than in case of control by neutron leakage; the relative difference increases from 4% to 18% when the enrichment increases from 2% to 4%. For this correlation, therefore, the control system is a parameter which must be taken into account; it is always possible to use the mean values of P and p between the two control methods (index m), bearing however in mind that they are rather strongly affected by the uncertainty A due to the effective control situation, as set out below:

$$\frac{Pu}{U} / D_5 = \left(\frac{Pu}{U} / D_5 \right)_m (1 \pm A)$$

$$\frac{Pu}{U} / d_5 = \left(\frac{Pu}{U} / d_5 \right)_m (1 \pm A)$$

The maximum value of the uncertainty A can be calculated as a function of the initial enrichment by means of the following expression:

$$A = 0.025 x - 0.025$$

As mentioned in § 4.1.3., the limits of uncertainty can be restricted when considering the effective reactivity control system of a reactor; in fact it is very improbable that, at least for the most common reactor types, the control completely coincides with only one of the two extreme situations examined.

Considering that it is not possible to specify the control method used by each type of reactor, our analysis was performed on the mean slopes P_m and p_m ; on the basis of the preceding considerations we have supposed that they can represent with sufficient approximation the real cases.

The mean slope P_m can be expressed as:

$$P_m = ax + b$$

where x is the initial enrichment (w/o) and a and b are two coefficients depending on the moderation ratio according to a parabolic law. The analytical expression of the correlation for Zircaloy clad fuel is:

$$Pu/U = [(0.023913 x + 0.32586) MR^2 - (0.19450 x + 1.3961)MR + (0.54877 x + 1.9872)] \cdot 10^{-2} \cdot D_5$$

4.3.4. Comparison with experimental data

Values of the ratio $(Pu/U)/D_5$ were derived from the experimental data originated by the analysis of TRINO and YANKEE fuels.

The following table shows the minimum and the maximum values of the ratio for each group of measurements referring to the same core.

For each core, also the correlation obtained by fitting the experimental points was determined; the corresponding values of P are reported in the following Table.

Table :

Reactor	Initial Enrichment (w/o)	$(Pu/U)/D_5$ experimental		fit of experimental points
		Min.	Max.	
Trino	2.72	0.0137	0.0165	0.0144
	3.13	0.0139	0.0168	0.0154
	3.90	0.0147	0.0179	0.0170
Yankee	4.1	0.0199	0.0205	0.0202
	4.9	0.0218	0.0230	0.0224
	4.93	0.0229	0.0235	0.0232

Fig. 15 shows the theoretical dependence of P on the initial enrichment and the moderating ratio, for a stainless steel clad fuel; the variability range of the experimental values of the ratio $(Pu/U)/D_5$ for each core, is also represented. The agreement is satisfactory.

4.3.5. Conclusions

The correlation is linear and greatly depends on the initial enrichment, on the moderation ratio and on the reactivity control method; the cladding material has a more modest influence. Agreement with experimental data can be considered as satisfactory.

4.4. Correlations between isotopes of plutonium

A partial investigation was made on a certain number of correlations concerning fuels of both PW and BW reactors. The correlations which have been considered are:

- Pu/U versus $^{240}\text{Pu}/^{239}\text{Pu}$
- Pu/U versus $^{242}\text{Pu}/^{239}\text{Pu}$

- Pu/U versus $^{241}\text{Pu}/^{239}\text{Pu}$
- Pu/U versus $^{242}\text{Pu}/^{240}\text{Pu}$
- $^{242}\text{Pu}/^{240}\text{Pu}$ versus $^{240}\text{Pu}/^{239}\text{Pu}$
- Pu/U versus $(^{241}\text{Pu}/^{238}\text{U}) / (^{240}\text{Pu}/^{239}\text{Pu})$
- Pu/U $(^{240}\text{Pu}/^{239}\text{Pu}) / (^{241}\text{Pu}/^{238}\text{U})$ versus $(^{242}\text{Pu}/^{240}\text{Pu}) / (^{241}\text{Pu}/^{239}\text{Pu})$
- Pu/U versus $\frac{^{242}\text{Pu} \cdot ^{239}\text{Pu}}{(^{240}\text{Pu})^2}$

Of all the correlations examined, the only one decidedly linear is

$$\frac{\text{Pu}}{\text{U}} \text{ versus } \frac{^{242}\text{Pu} \cdot ^{239}\text{Pu}}{(^{240}\text{Pu})^2}$$

previously considered by various authors (ref. 4 and 5).

The calculated points can be fitted by a straight line on an irradiation range from 5,000 to 30,000 Mwd/tU, with deviations in the order of 1.5%-3%; the correlation is slightly dependent on the moderation ratio (therefore on the quantity of voids in the case of BWR's) and on the cell reactivity control method used.

The other correlations, even though of a regular form, cannot be substituted by a straight line, and in addition, depend in a rather irregular manner on the various parameters of the reactor (figs. 18 through 27).

5. ANALYSIS OF THE CORRELATIONS FOR BW REACTORS

As already mentioned, six BWR lattices were examined, the characteristics of which are shown in Table 1. The results of the calculations were compared to the experimental data (ref. 4, 5) concerning the fuels irradiated in the HUMBOLDT BAY and DRESDEN I reactors (whose lattices are very similar to those studied). The analysis of linearity gave the same results as in the case of the PWR's.

The results of this investigation can be summed up as follows:

- in the case of correlations between the isotopes of uranium (fig. 15), the influence of the presence of voids in the moderator is very limited, the comparison with the experimental data can be considered as satisfactory; it is to be pointed out that the difference between the experimental results of HUMBOLDT BAY and the theoretical ones is due to a slight difference in the initial enrichments (2.6 w/o actual; 2.5 w/o in the calculations) which causes a strong variation in the slope of the correlation (see § 4.1.3); therefore, in order to interpret correctly the experimental results, a correction was made to the theoretical curve (fig. 16) in accordance with the formula shown in § 4.1.3;
- in the case of the correlation between the accumulation of plutonium and the depletion of ^{235}U (fig. 17) the interpretation of the experimental data is more difficult, due to the strong influence of the voids on the slope of the correlation and to the impossibility of ascertaining exactly the quantity of voids effectively present during the life of the core, in the zone where the fuel assembly has been irradiated. In fact we know only the quantity of voids averaged on all the core and on the whole cycle; it is possible that the effective void conditions be different from the average ones considered by us; therefore this correlation can be used with good results only when the voids history in the vicinity of the fuel assembly is known with a certain degree of precision.

APPENDIX 1

Modification of the experimental data relating to ^{236}U

The quantity of ^{236}U present in the fuel after irradiation has two distinct origins:

- a) quantity accumulated during irradiation as a result of reaction $^{235}\text{U}(n,\gamma)^{236}\text{U}$
- b) residue of the quantity initially present.

This latter part can be easily calculated by means of the procedure here described, when the initial quantity of ^{236}U and the depletion of ^{235}U are known. Let us call:

$$N_5(o) = \text{initial quantity of } ^{235}\text{U (at/cm}^3\text{)}$$

$$N_6(o) = \text{initial quantity of } ^{236}\text{U (at/cm}^3\text{)}$$

$$N_5 = \text{residue of } ^{235}\text{U after irradiation (at/cm}^3\text{)}$$

$$N_6 = \text{residue of initial } ^{236}\text{U after irradiation (at/cm}^3\text{)}$$

$$D_5 = \frac{N_5(o) - N_5}{N_5(o)} : \text{depletion of } ^{235}\text{U}$$

$$D_6 = \frac{N_6(o) - N_6}{N_6(o)} : \text{depletion of } ^{236}\text{U}$$

The differential equations that express the depletion of the initial ^{235}U and ^{236}U are:

$$\frac{dN_5}{dI} = -\sigma_{a5} N_5$$

$$\frac{dN_6}{dI} = -\sigma_{a6} N_6$$

On the assumption that the average absorption cross-sections are constant, the following expressions are obtained by integration:

$$N_5 = N_5(o) e^{-\sigma_{a5} I}$$

$$N_6 = N_6(o) e^{-\sigma_{a6} I}$$

where:

σ_{a5} = microscopic absorption cross-section of ^{235}U (barn)

σ_{a6} = microscopic absorption cross-section of ^{235}U (barn)

$$I = \int_0^t C(t) dt = \text{irradiation (n/cm}^2\text{)}$$

The two expressions can be written in the form:

$$1 - D_5 = e^{-\sigma_{a5} I}$$

$$1 - D_6 = e^{-\sigma_{a6} I}$$

from which it follows:

$$(1 - D_6) = (1 - D_5)^{\frac{\sigma_{a6}}{\sigma_{a5}}}$$

which enable us to calculate D_6 and therefore the residue of the initial quantity of ^{236}U , from the depletion of ^{235}U ; it should be noticed that, in general, D_6 is very small ($D_6 = 0.05-0.10$).

Generally, the initial quantity of ^{236}U is not known; it can be obtained with sufficient precision by plotting the experimental values of $^{236}\text{U}/^{238}\text{U}$ versus D_5 , and fitting to these points a straight line; the intersection point of this correlation with the ordinate axis represents the initial content of ^{236}U .

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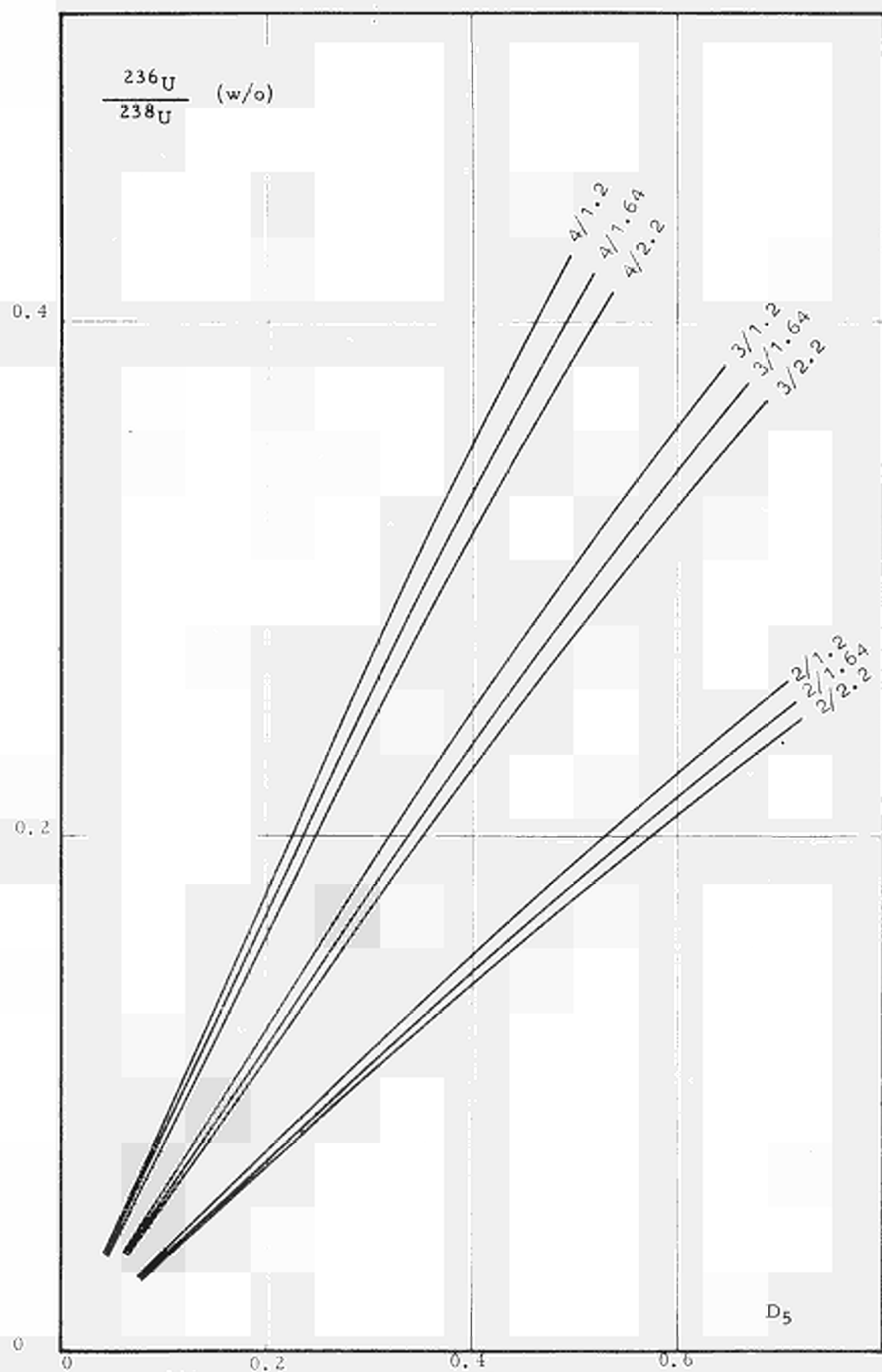


Fig. 1 - ^{236}U accumulation versus ^{235}U depletion (zircaloy cladding; reactivity control by soluble poison)

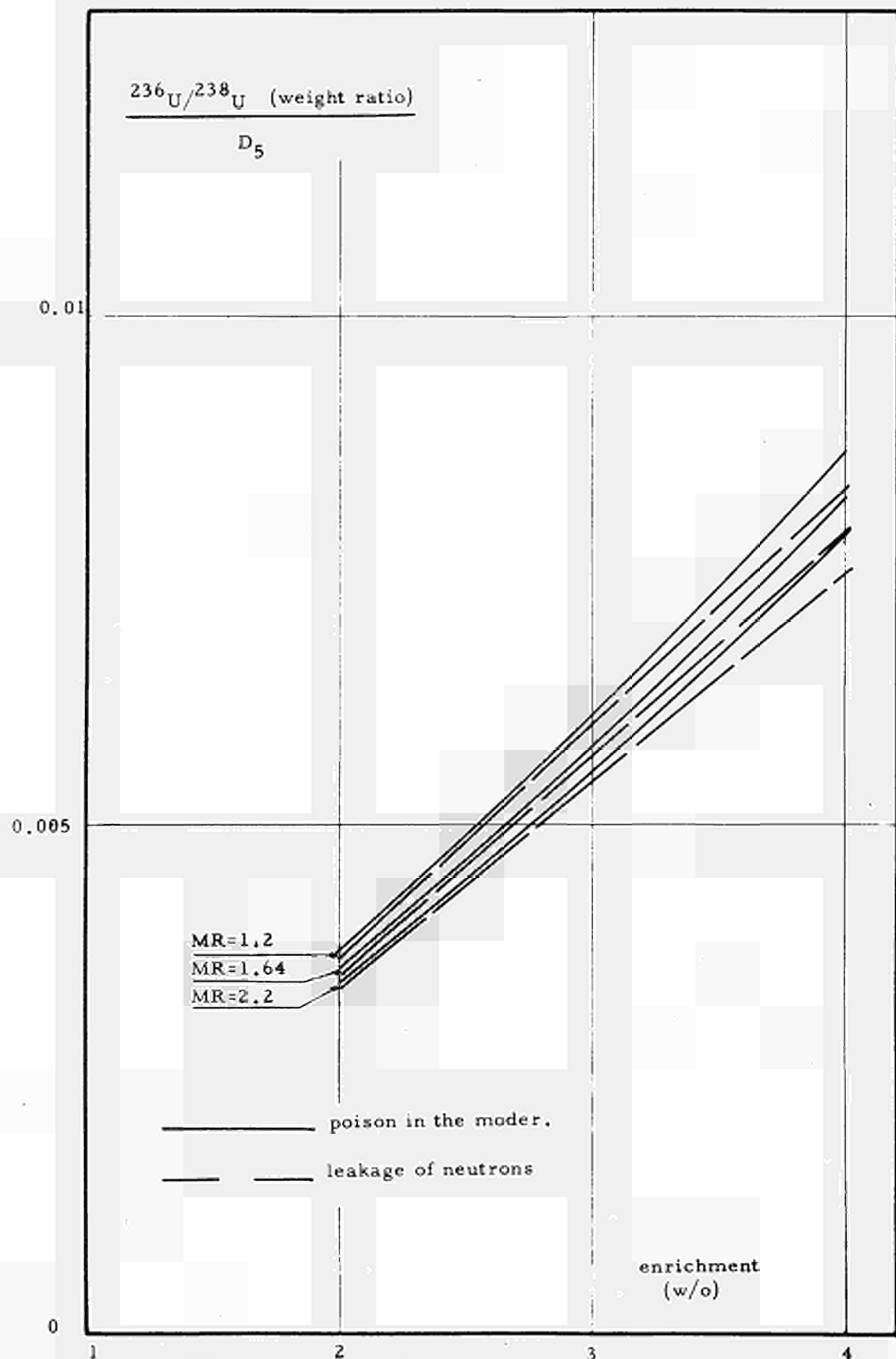


Fig. 2 - ^{236}U accumulation/ ^{235}U depletion ratio for different lattices and different reactivity control methods (Zircaloy cladding).

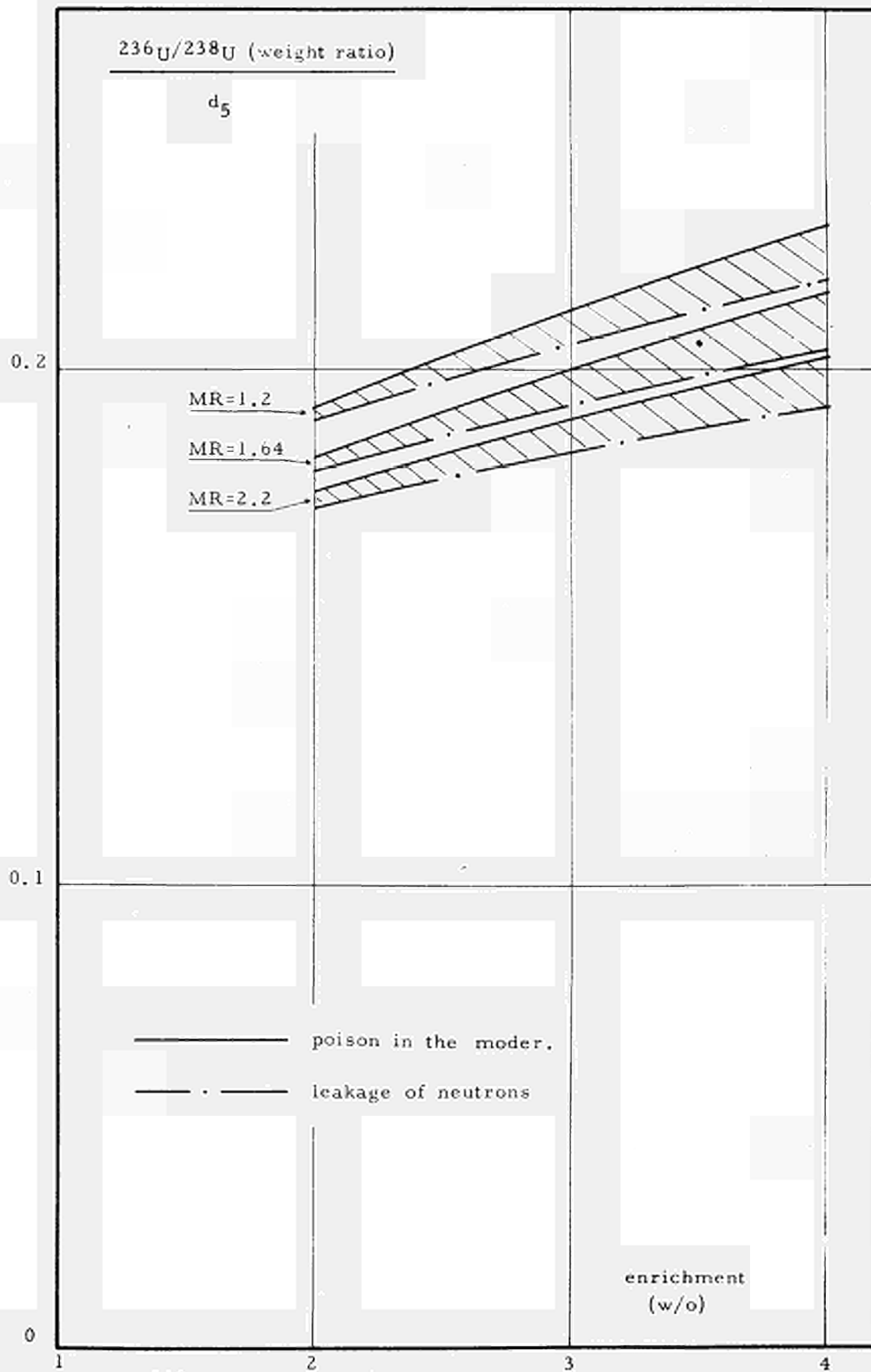


Fig. 3 - ^{236}U accumulation/ ^{235}U depletion ratio for different lattices and different reactivity control methods (Zircaloy cladding).

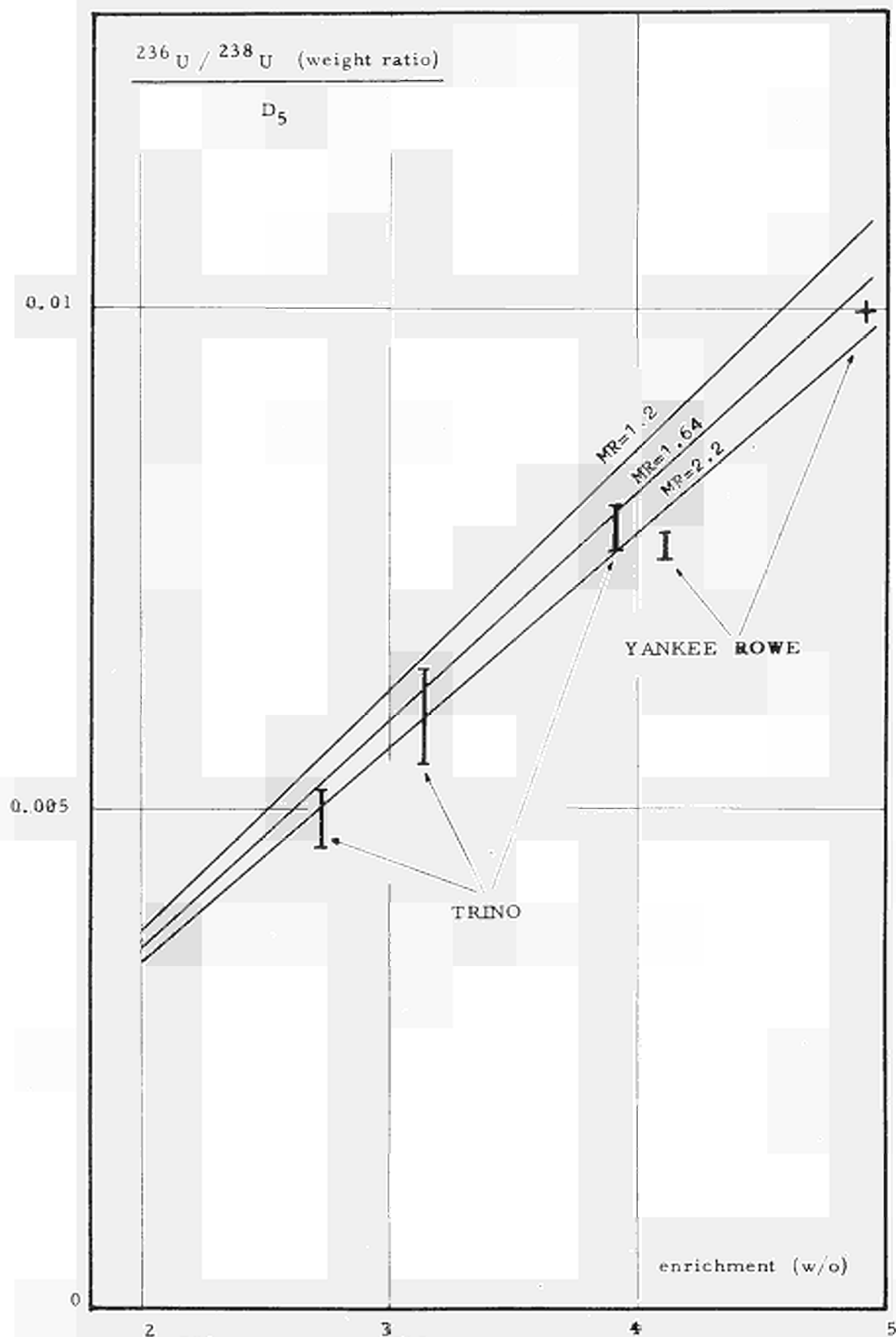


Fig. 4 - ^{236}U accumulation/ ^{235}U depletion ratio; comparison with experimental results from Trino and Yankee Rowe reactors

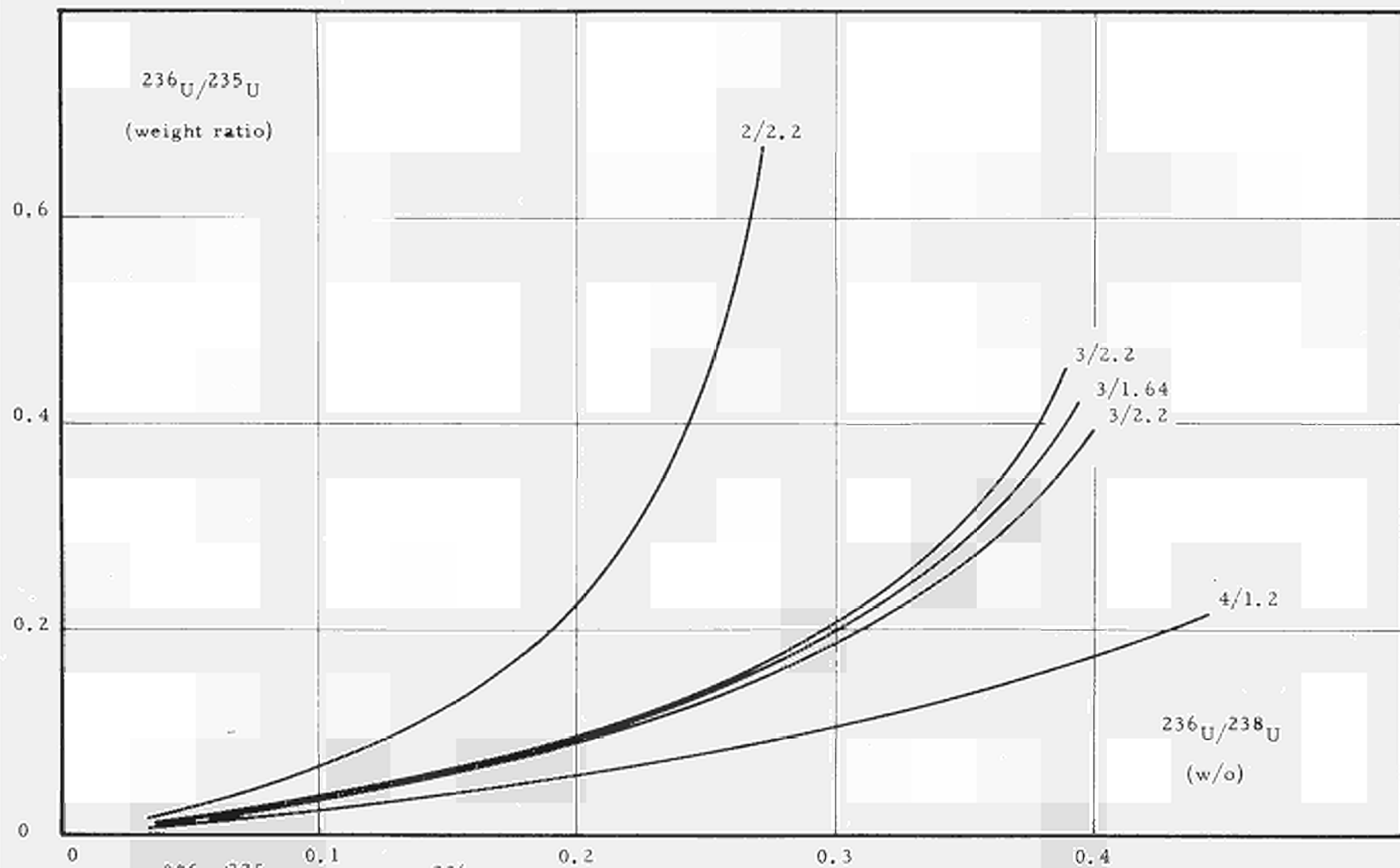


Fig. 5 - $^{236}\text{U}/^{235}\text{U}$ ratio versus ^{236}U accumulation (Zircaloy cladding; reactivity control by soluble poison).

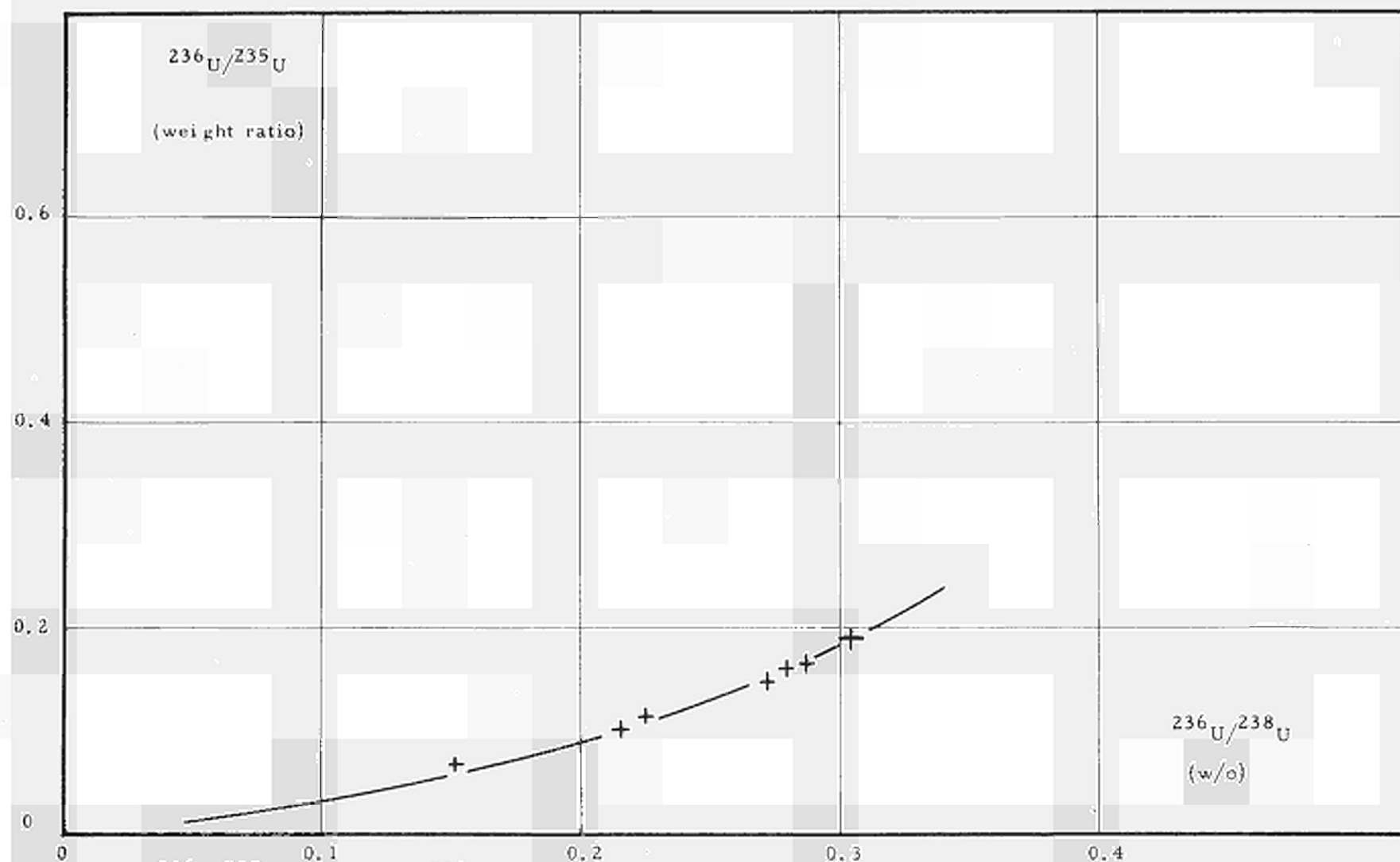


Fig. 6 - $^{236}\text{U}/^{235}\text{U}$ ratio versus ^{236}U accumulation; comparison with experimental results from Trino Reactor (fuel enrichment 3.13 w/o)

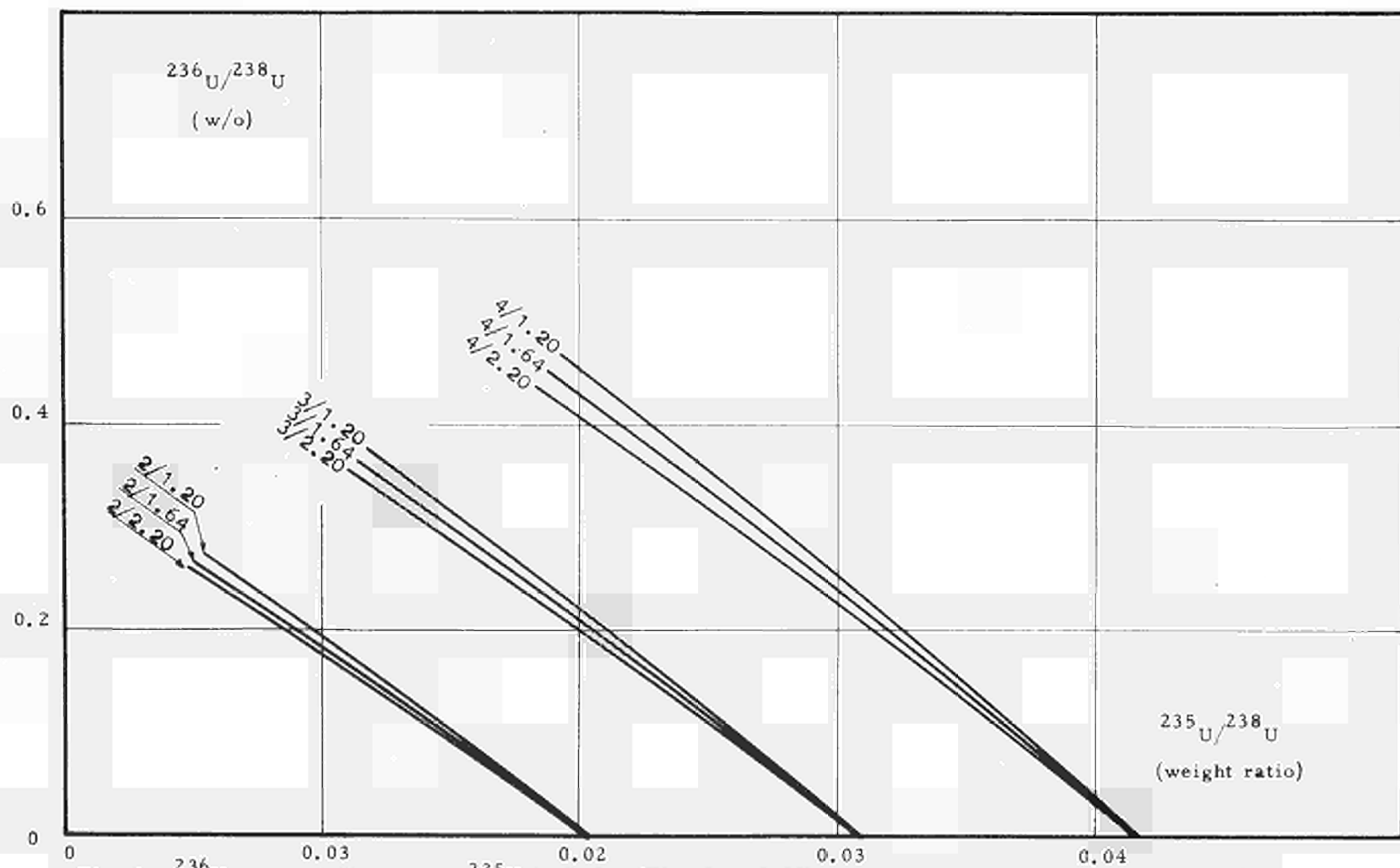


Fig. 7 - ^{236}U accumulation versus ^{235}U depletion (Zircaloy cladding; reactivity control by soluble poison).

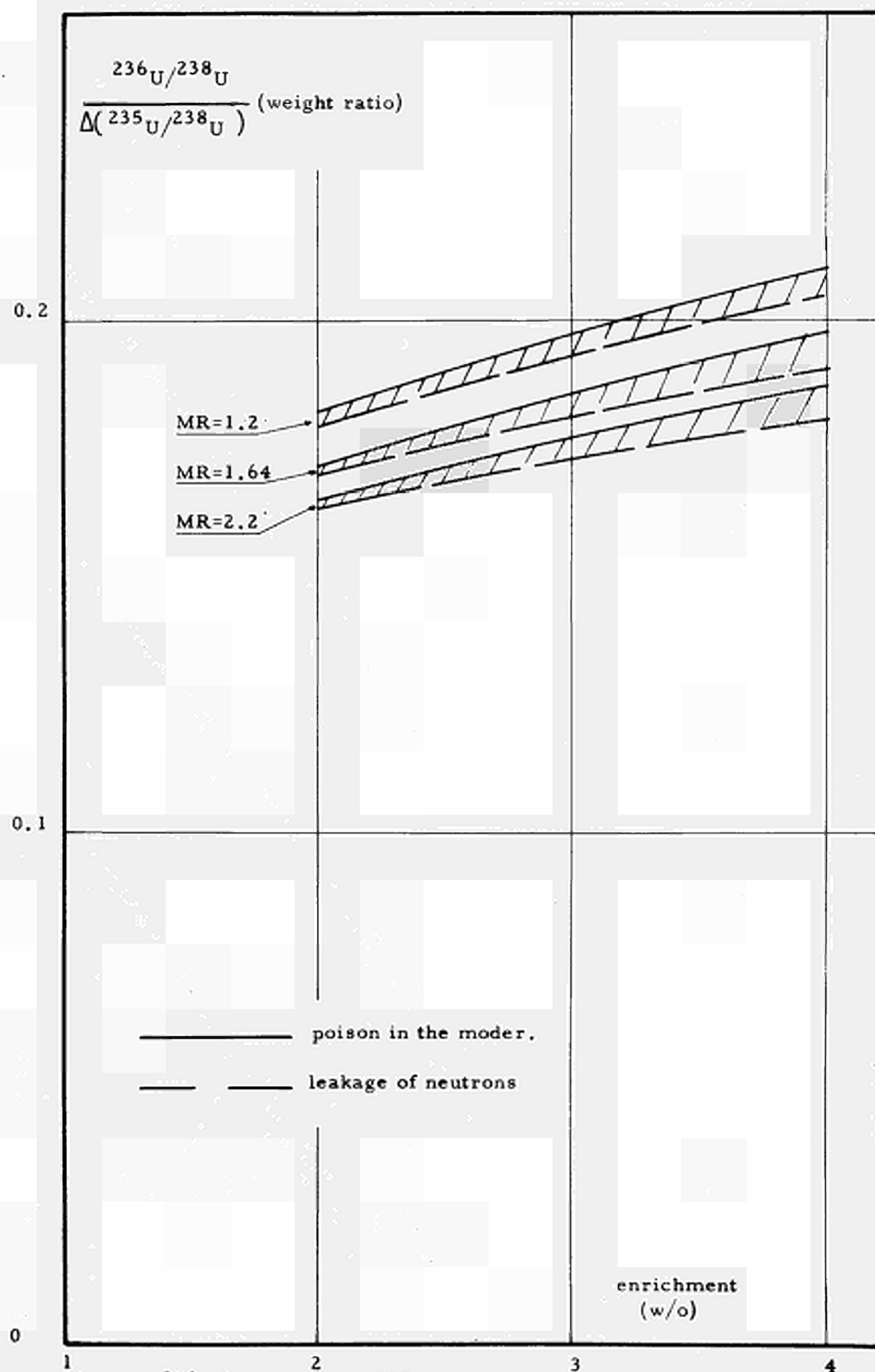


Fig. 8 - ^{236}U accumulation/ ^{235}U depletion ratio for different lattices and different reactivity control methods (Zircaloy cladding).

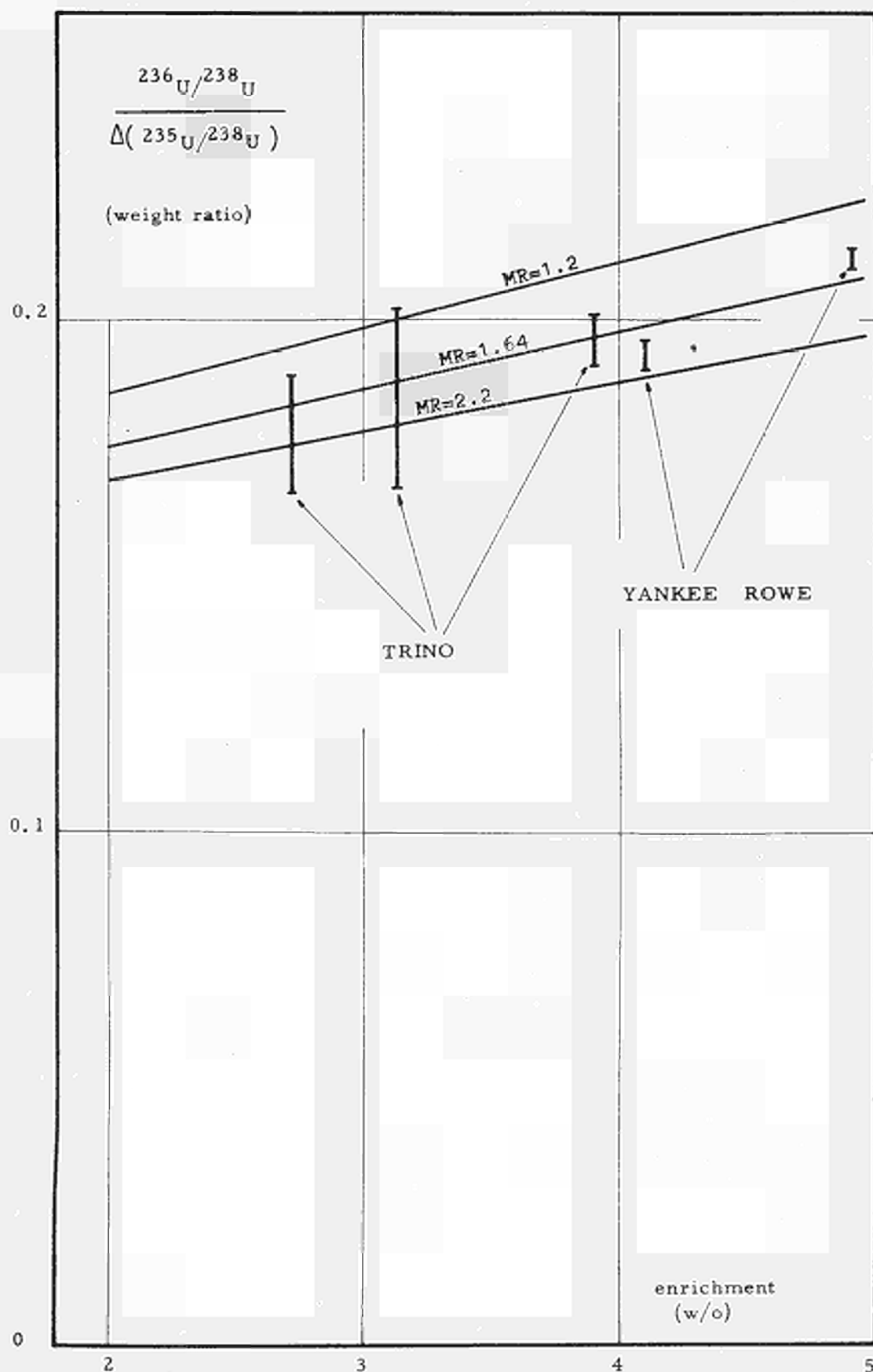


Fig. 9 - ^{236}U accumulation/ ^{235}U depletion ratio; comparison with experimental results from Trino and Yankee Rowe reactors.

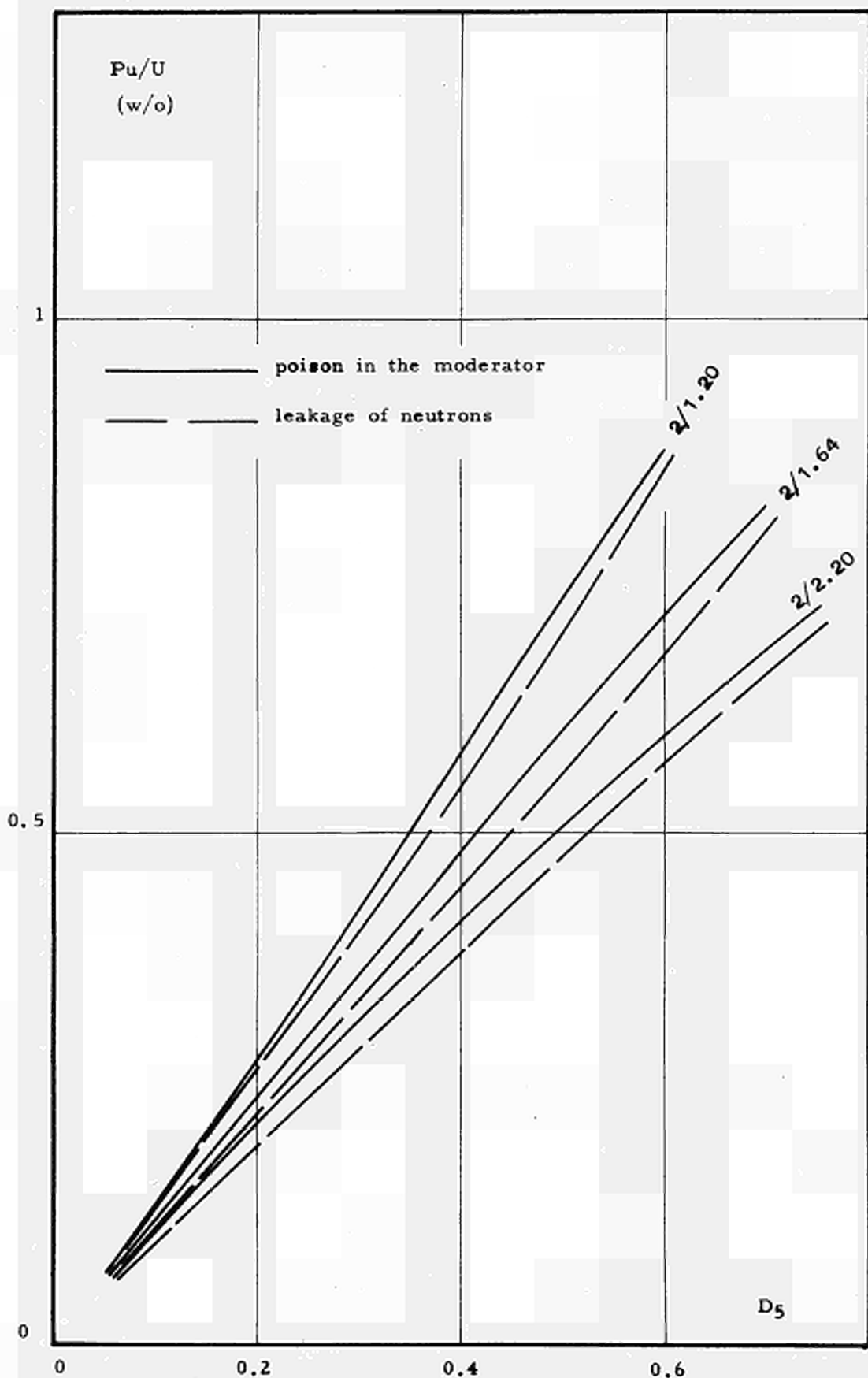


Fig. 10 - Pu/U ratio versus ^{235}U depletion (Zircaloy cladding); comparison between different reactivity control methods.



Fig. 11 - Pu/U ratio versus ^{235}U depletion (Zircaloy cladding); comparison between different reactivity control methods.

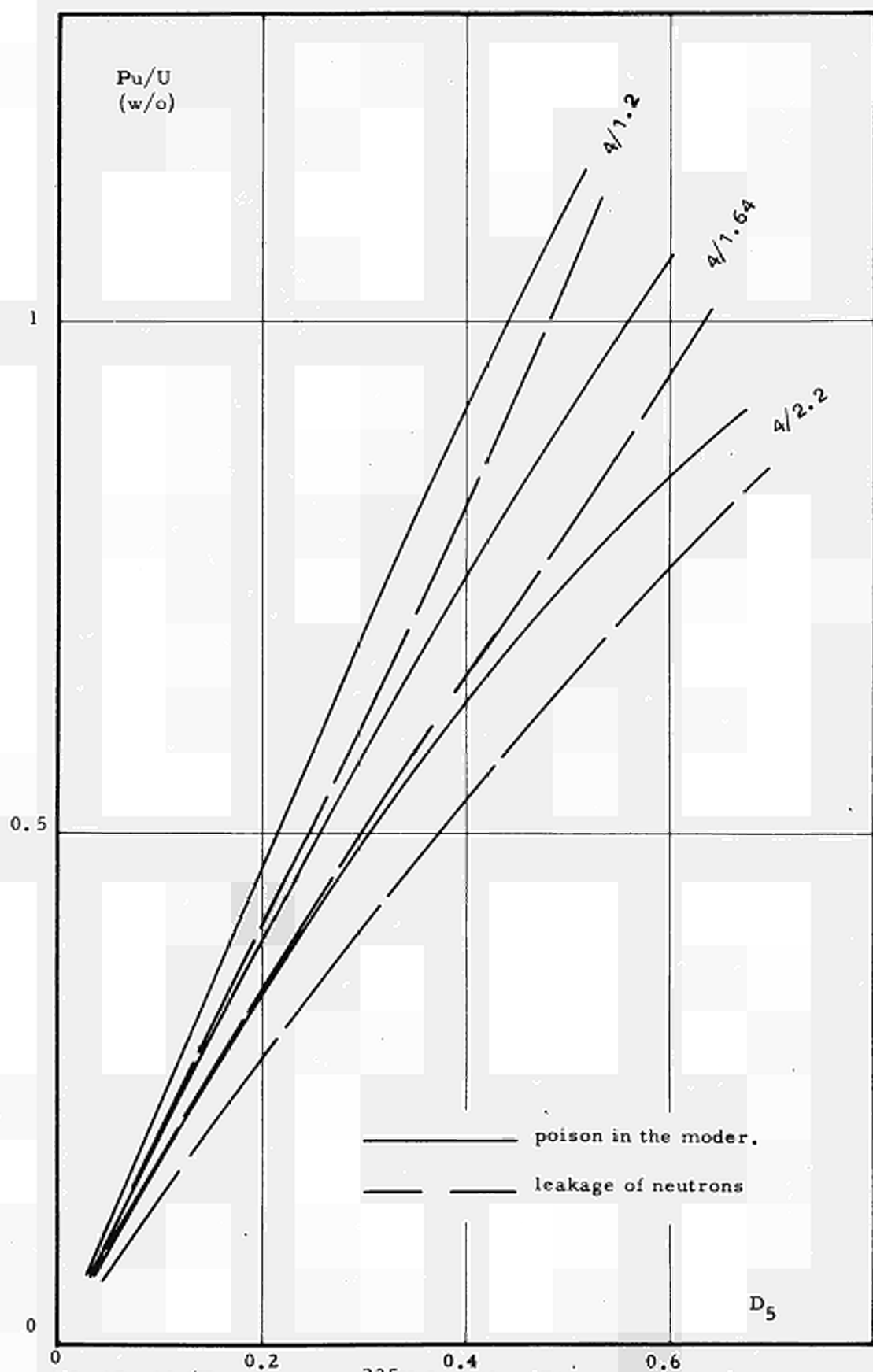


Fig. 12 - Pu/U ratio versus ^{235}U depletion (Zircaloy cladding); comparison between different reactivity control methods.

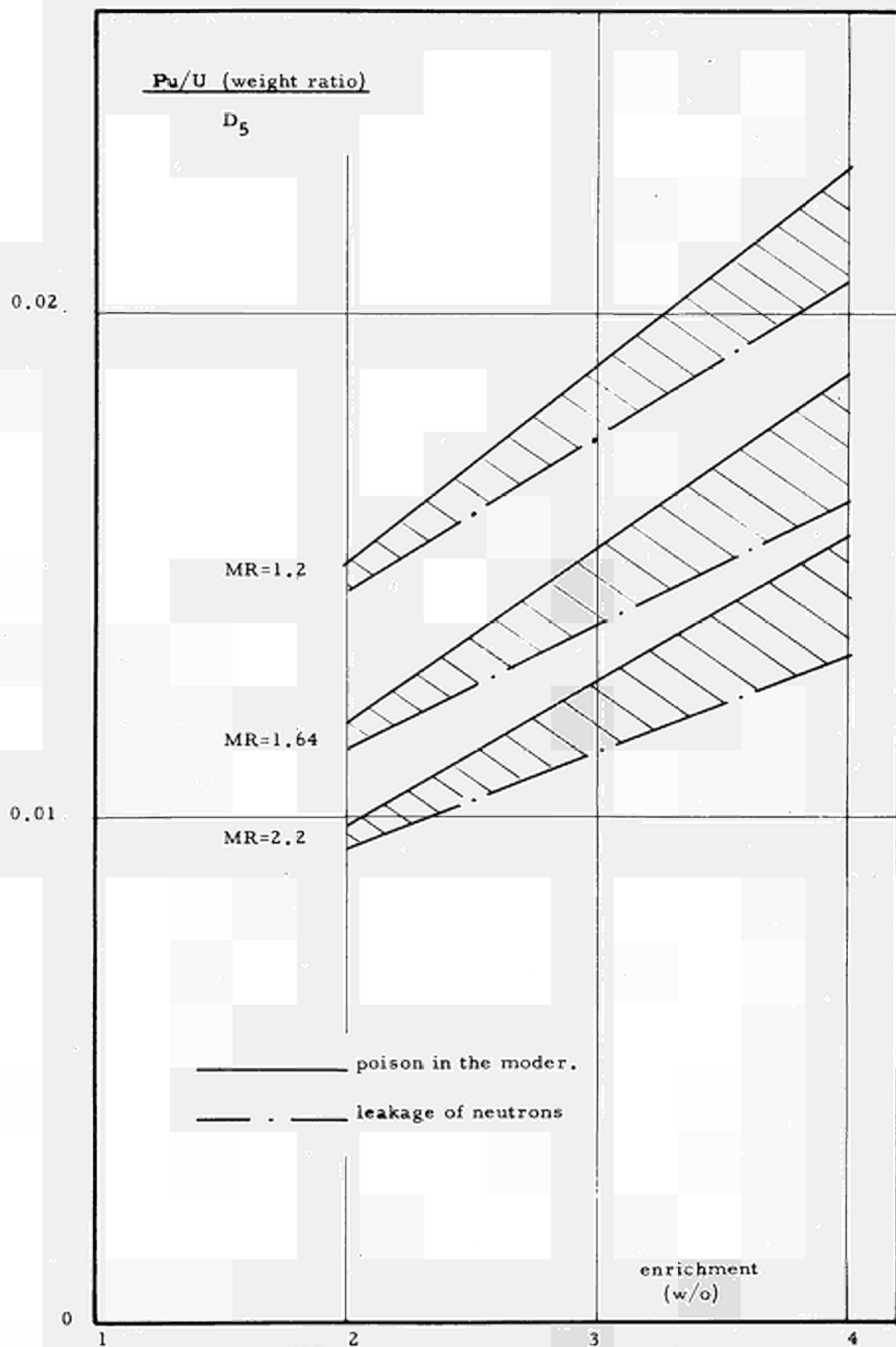


Fig. 13 - Pu/U to ²³⁵U depletion ratio for different lattices and different reactivity control methods (Zircaloy cladding).

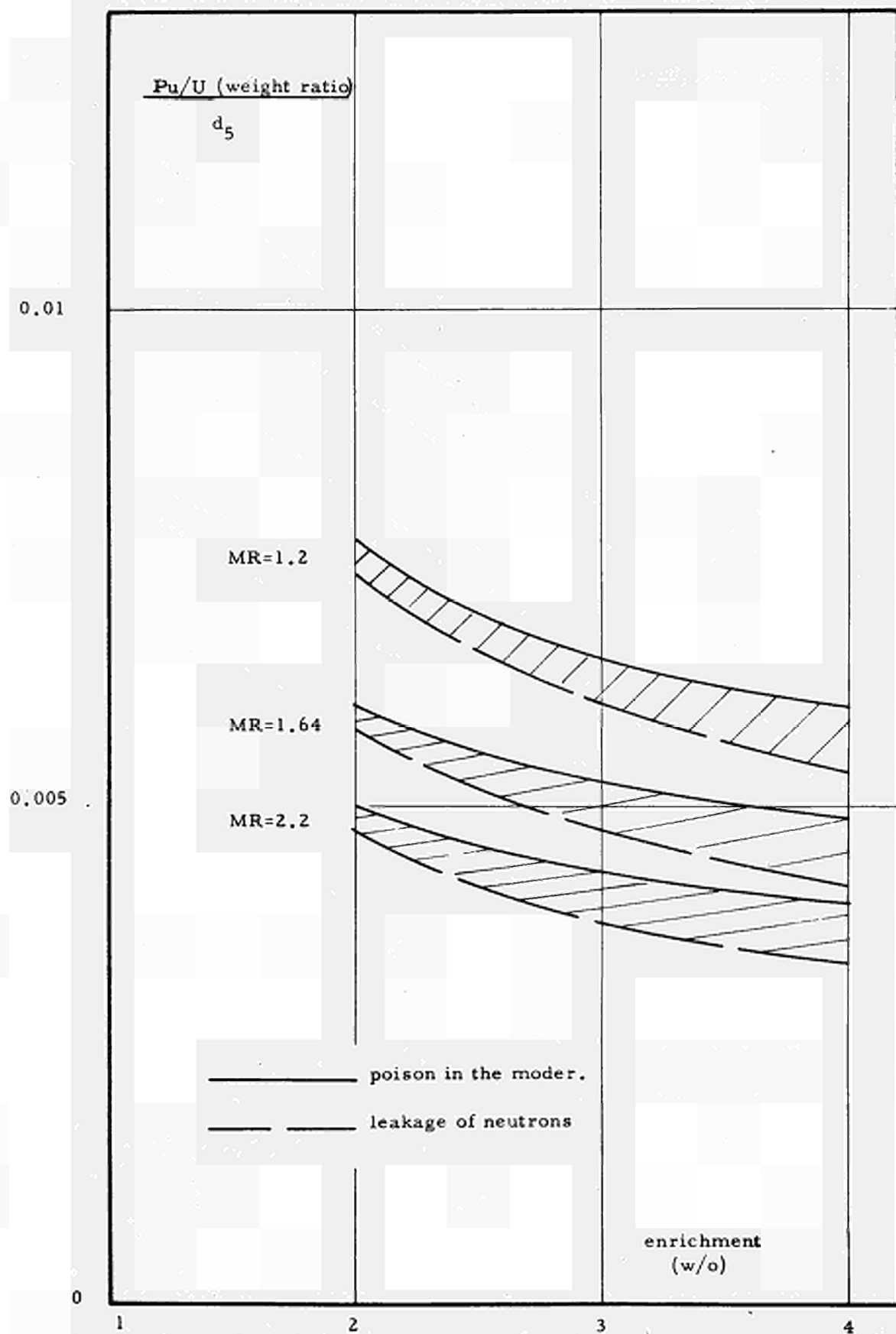


Fig. 14 - Pu/U to ^{235}U depletion ratio for different lattices and different reactivity control methods (Zircaloy cladding).

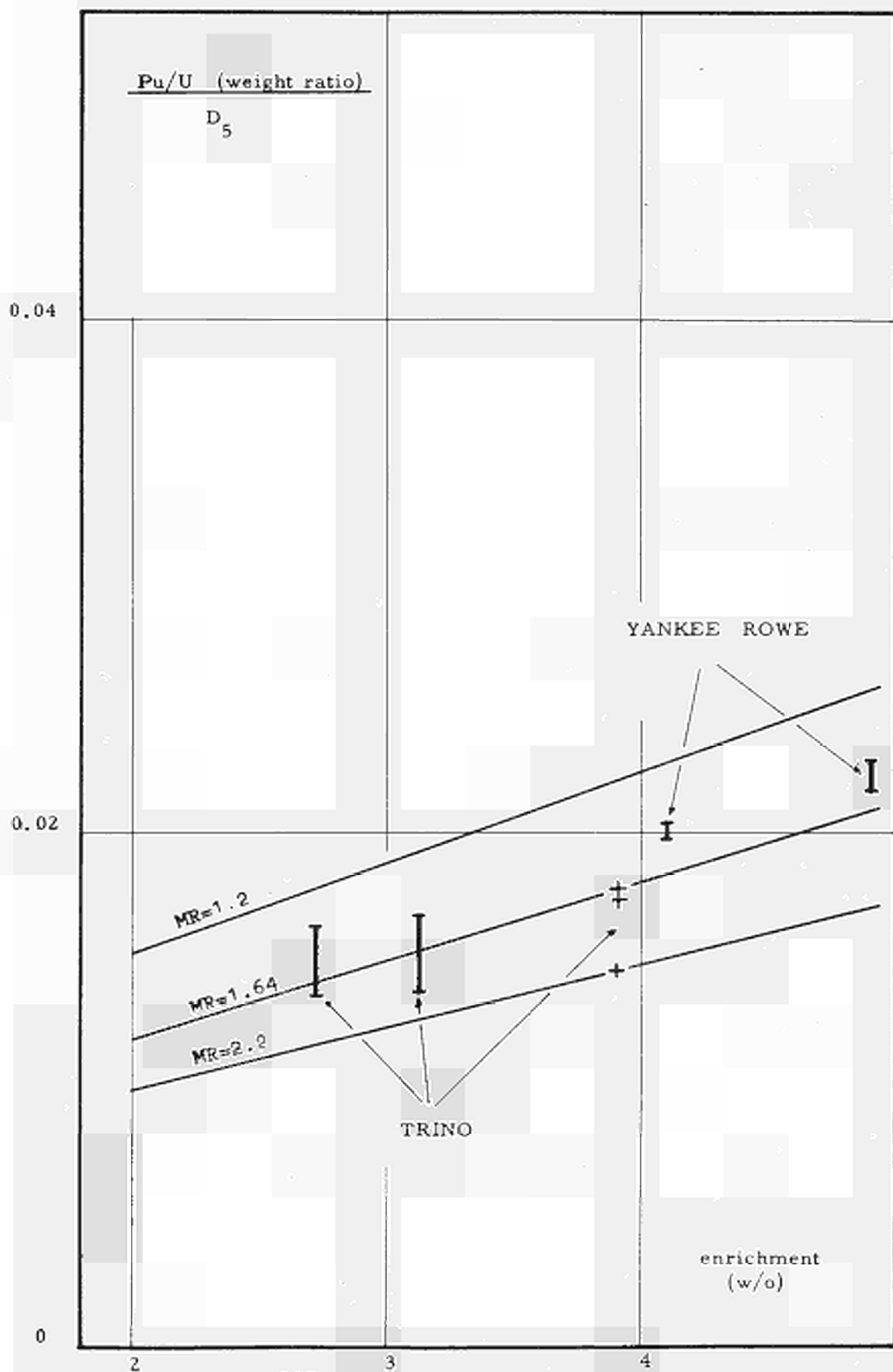


Fig. 15 - Pu/U to ^{235}U depletion ratio; comparison with experimental results from Trino and Yankee Rowe reactors.

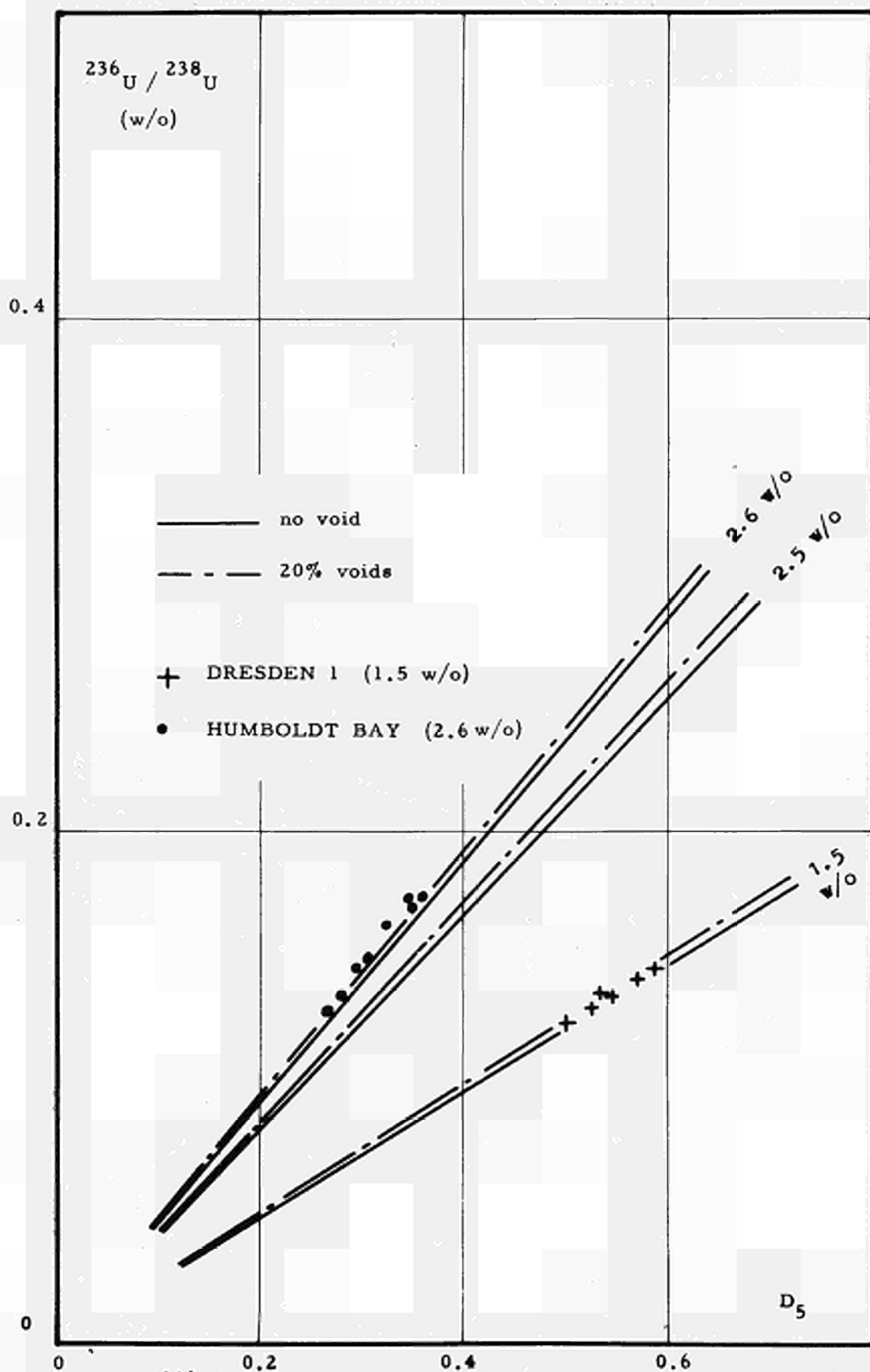


Fig. 16 - ^{236}U accumulation versus ^{235}U depletion (BWR reactors); comparison with experimental results.

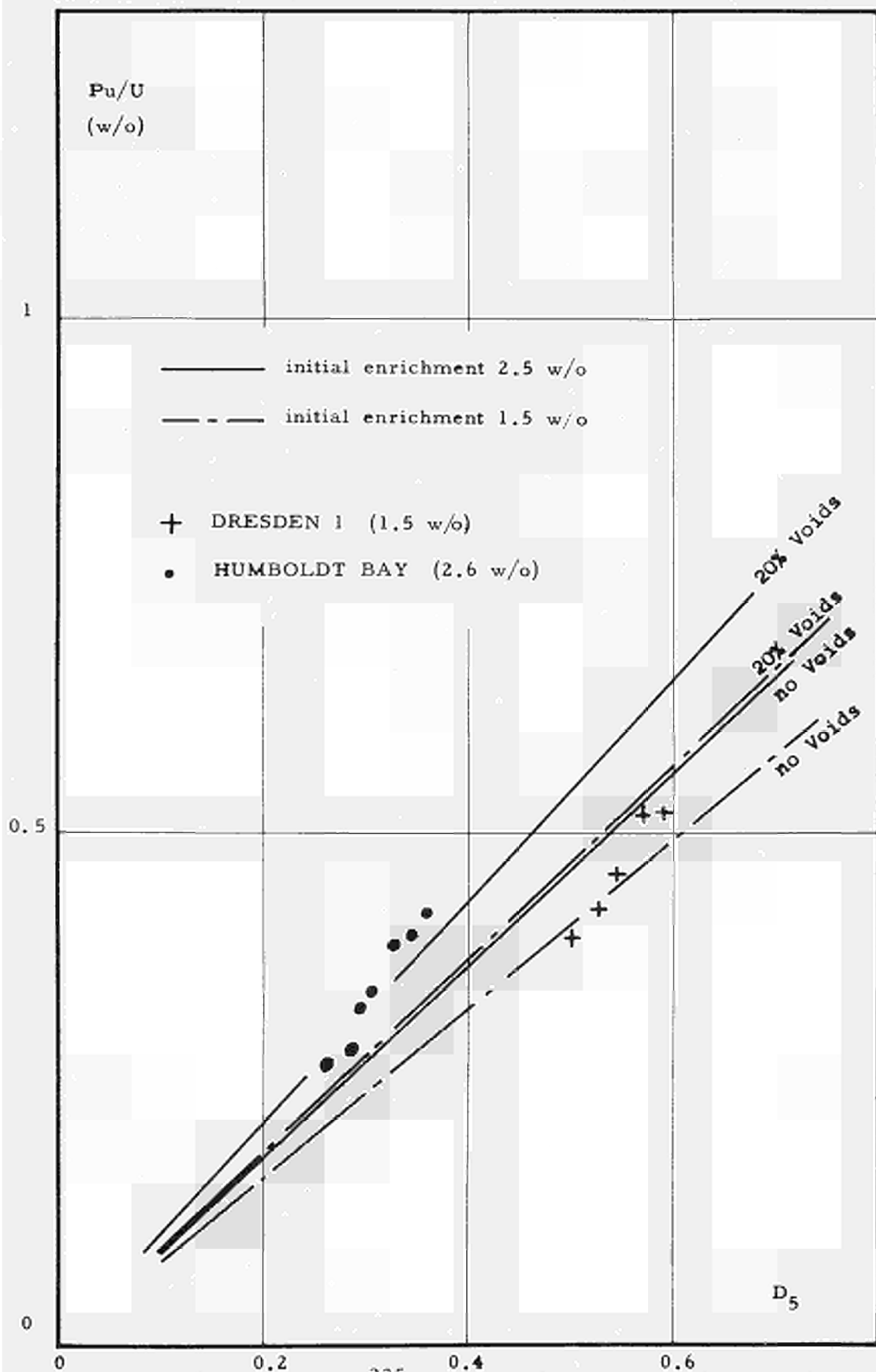


Fig. 17 - Pu/U ratio versus ^{235}U depletion (BWR reactors); comparison with experimental results.

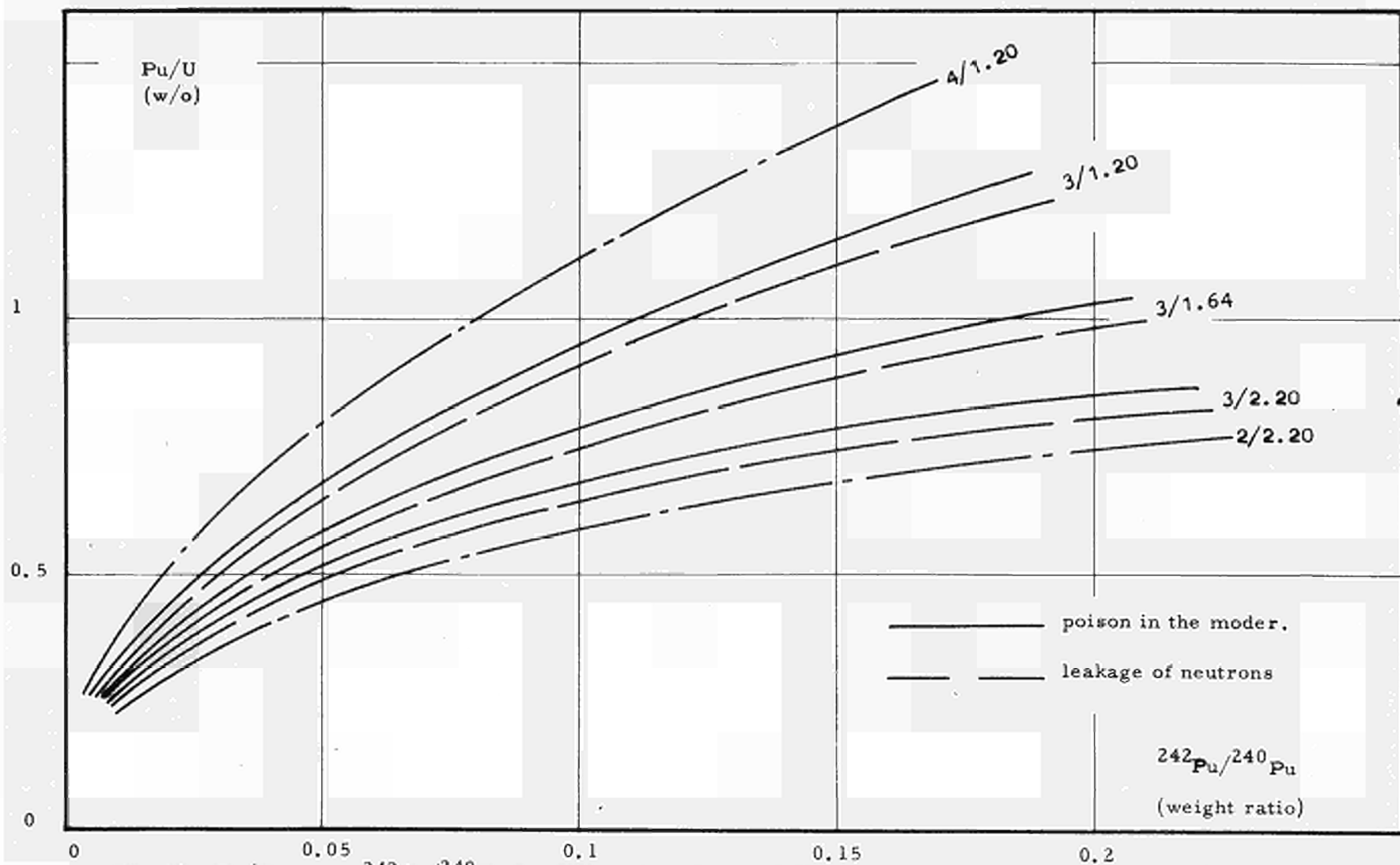


Fig. 18 - Pu/U versus $^{242}\text{Pu}/^{240}\text{Pu}$ for different lattices and different reactivity control methods (Zircaloy cladding).

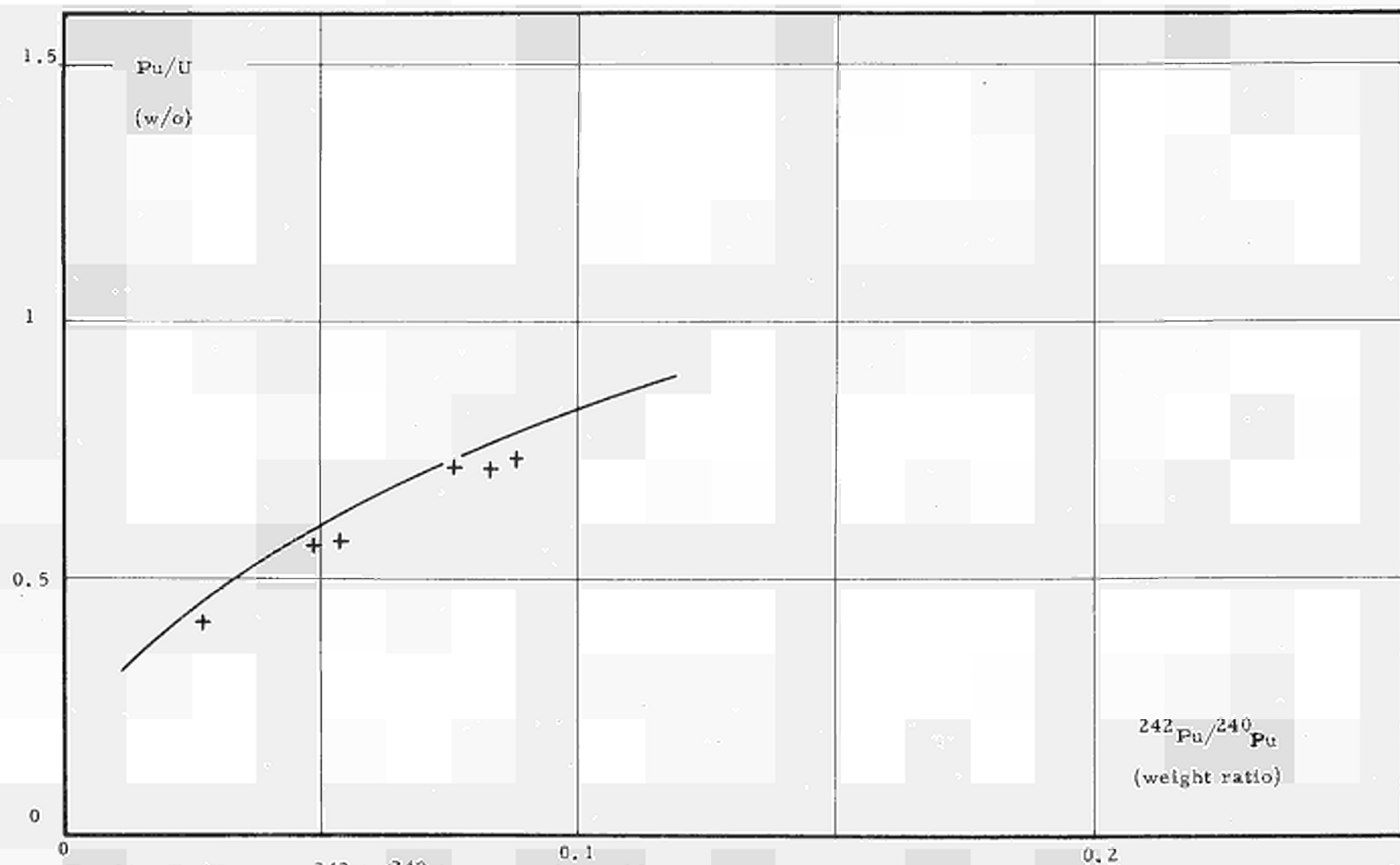


Fig. 19 - Pu/U versus $^{242}\text{Pu}/^{240}\text{Pu}$; comparison with experimental results from Trino reactor (fuel enrichment 3.13 w/o)

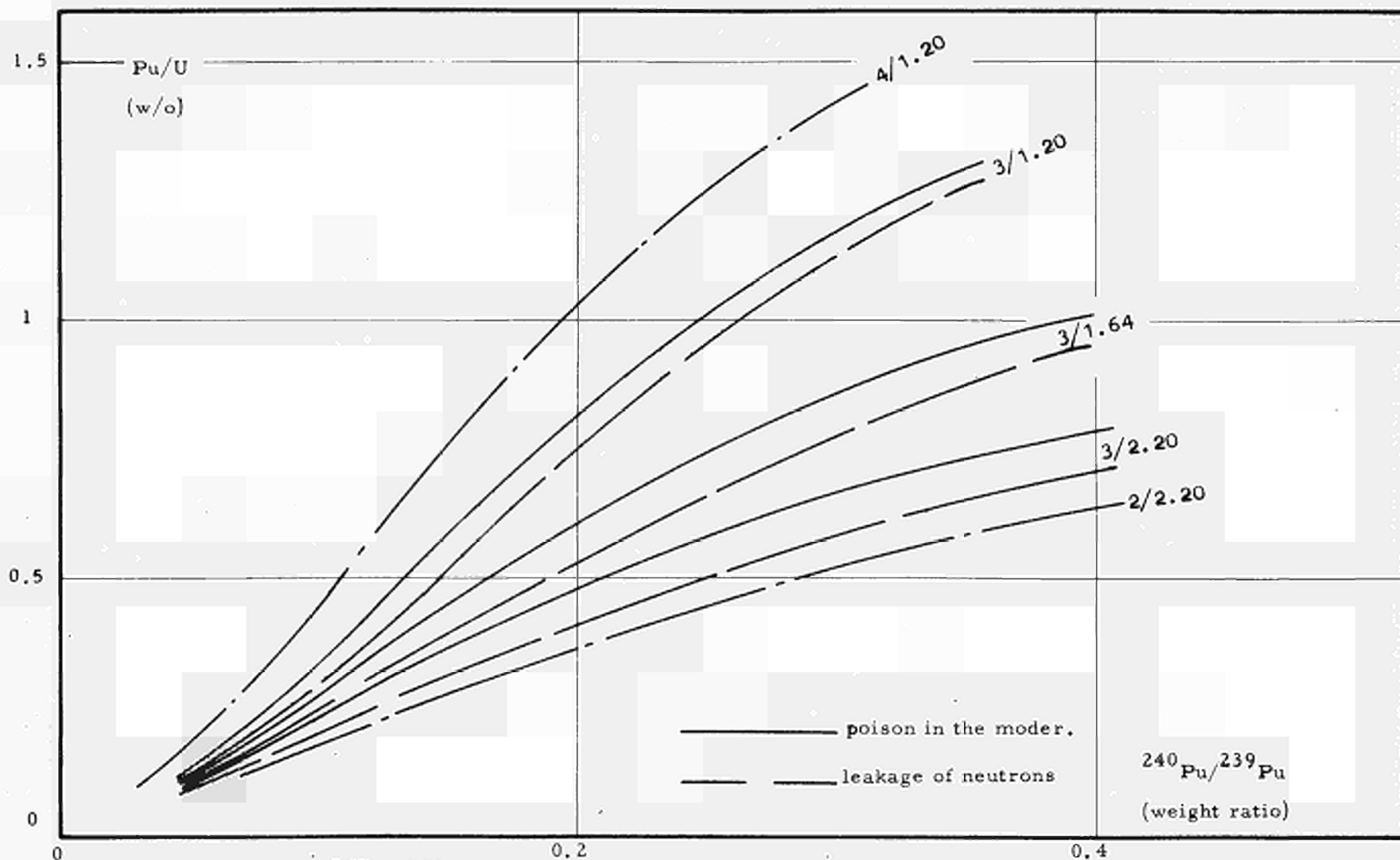


Fig. 20 - Pu/U versus $^{240}\text{Pu}/^{239}\text{Pu}$ for different lattices and different reactivity control methods (Zircaloy cladding).

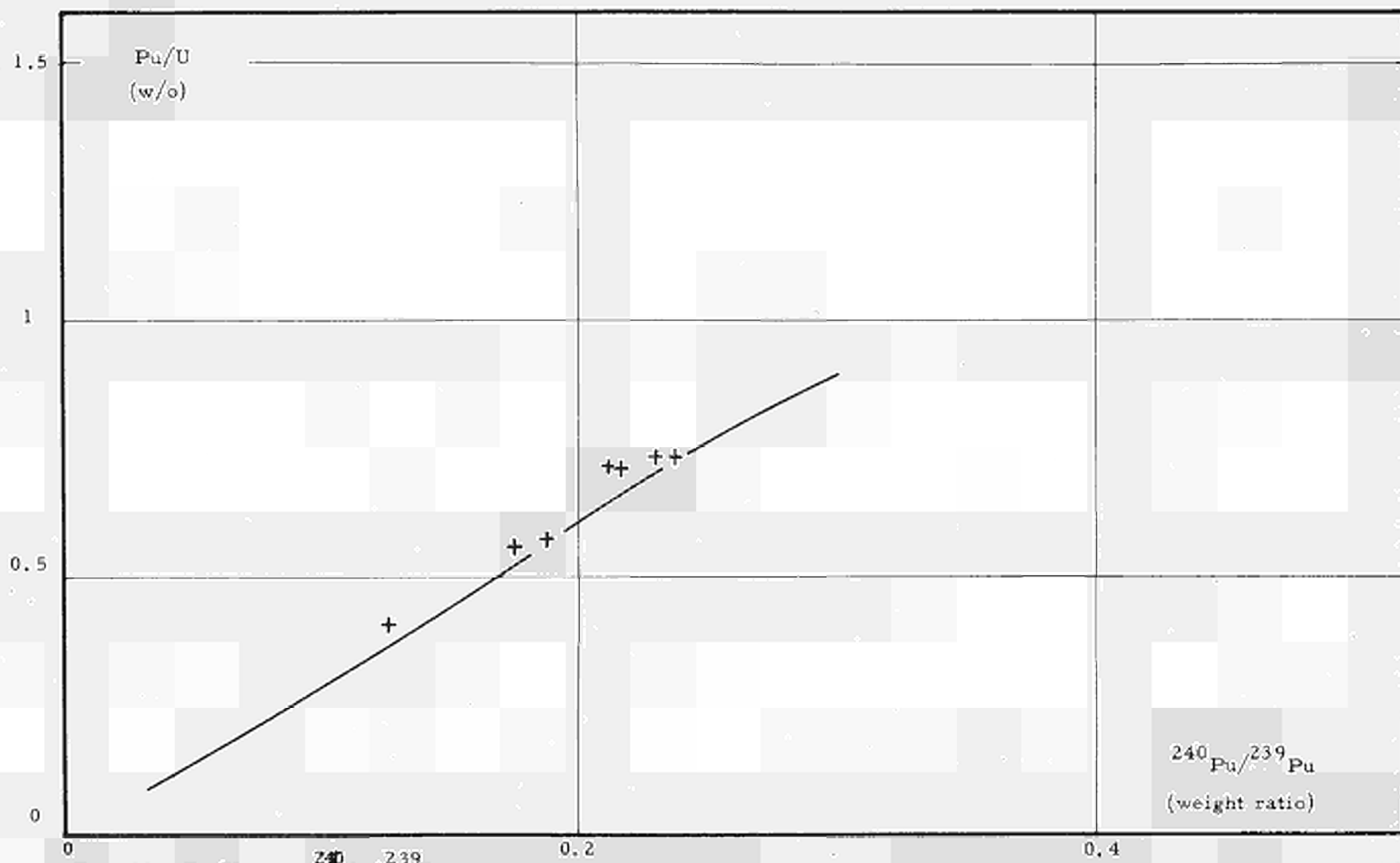


Fig. 21 - Pu/U versus $^{240}\text{Pu}/^{239}\text{Pu}$; comparison with experimental results from Trino reactor (fuel enrichment 3.13 w/o)

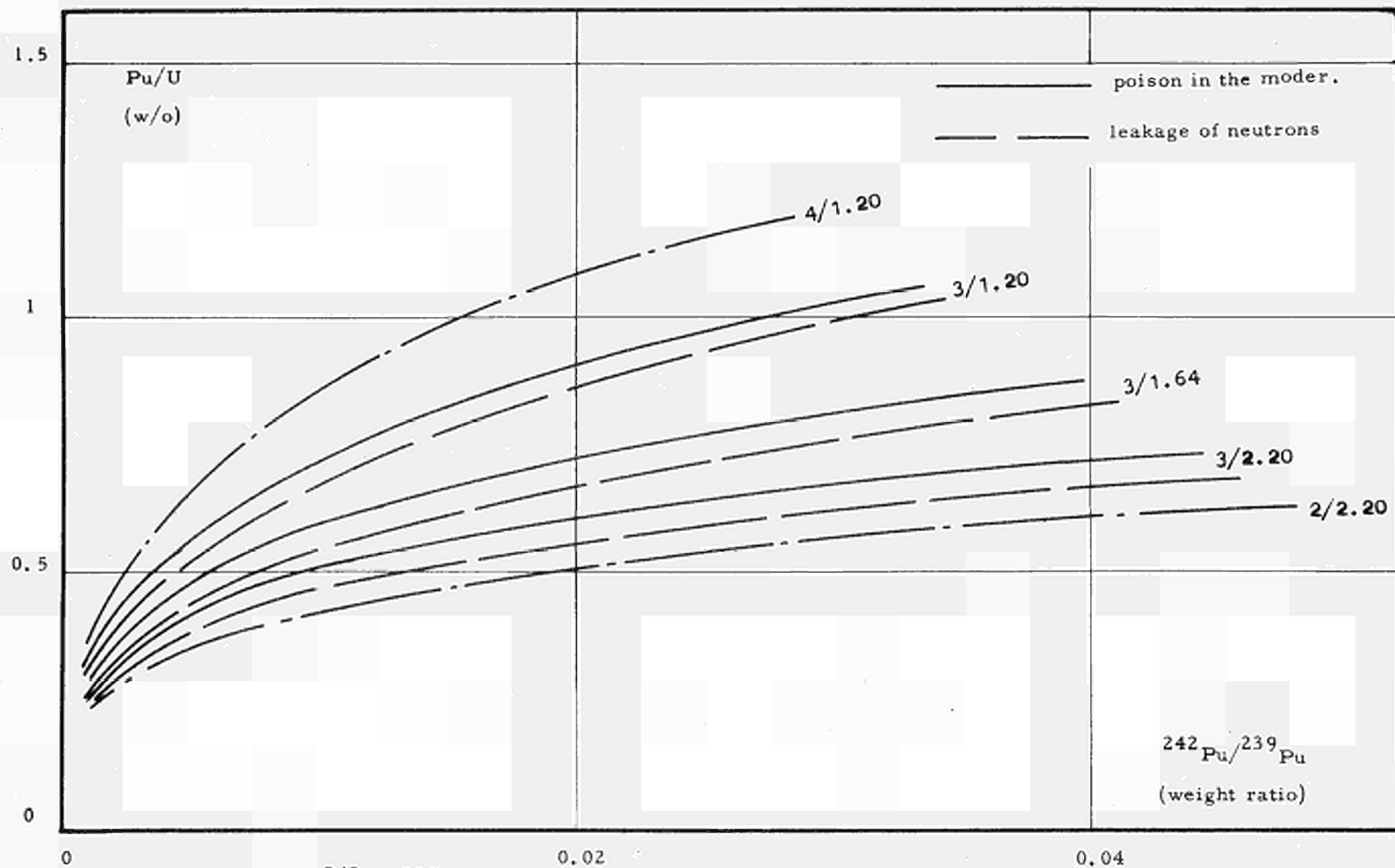


Fig. 22 - Pu/U versus $^{242}\text{Pu}/^{239}\text{Pu}$ for different lattices and different reactivity control methods (Zircaloy cladding).

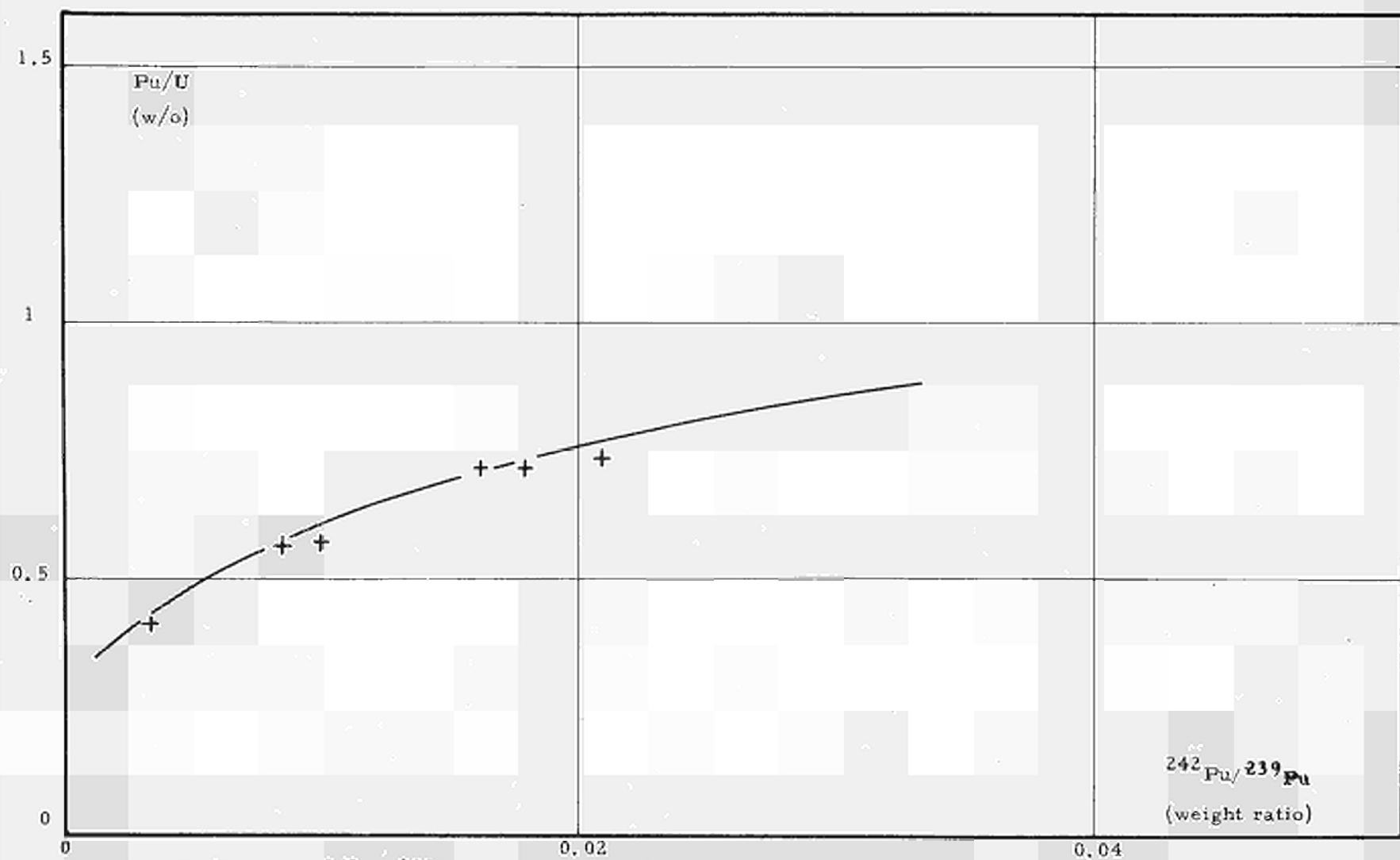


Fig. 23 - Pu/U versus $^{242}\text{Pu}/^{239}\text{Pu}$; comparison with experimental results from Trino reactor (fuel enrichment 3.13 w/o)

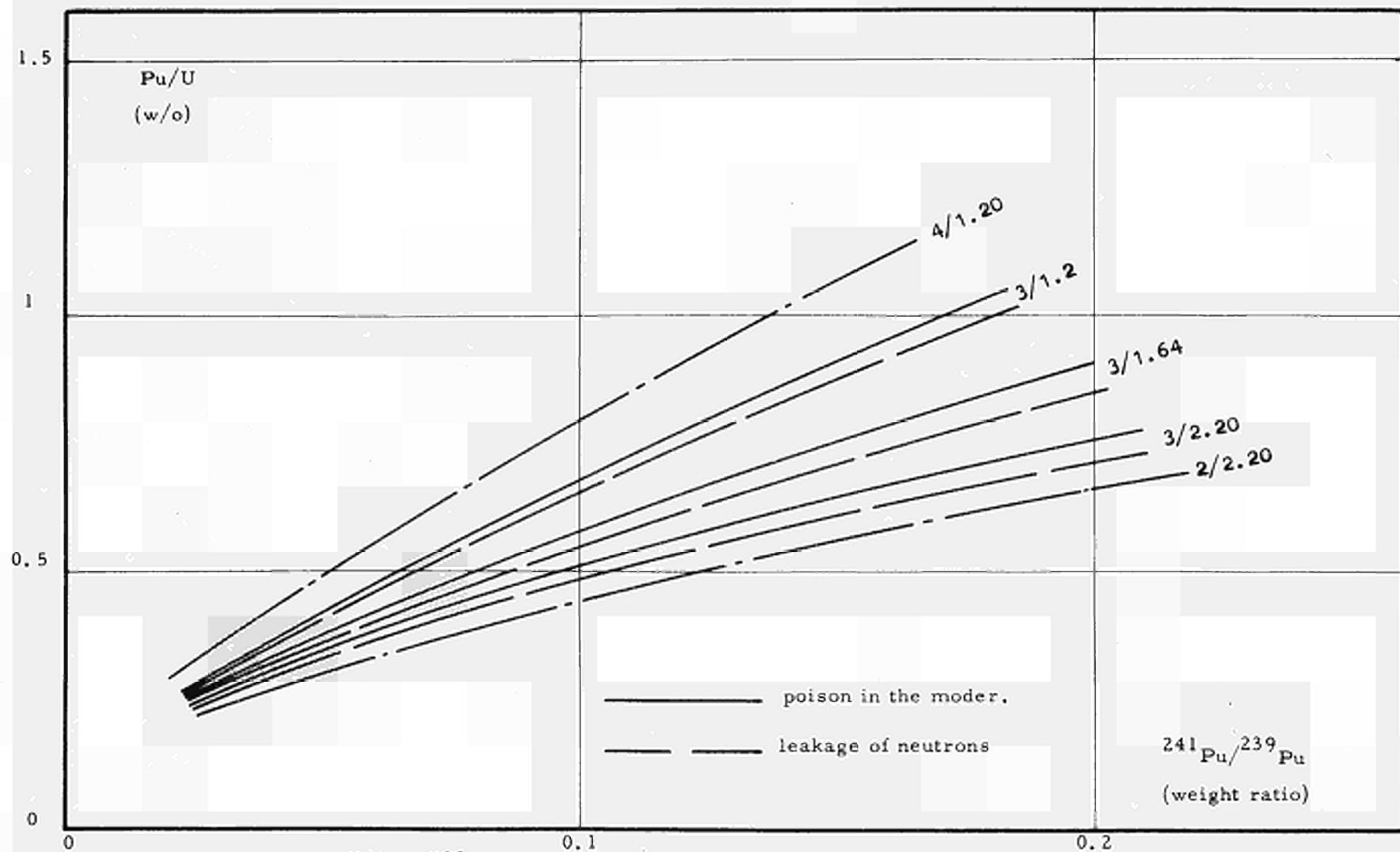


Fig. 24 - Pu/U versus $^{241}\text{Pu}/^{239}\text{Pu}$ for different lattices and different reactivity control methods.

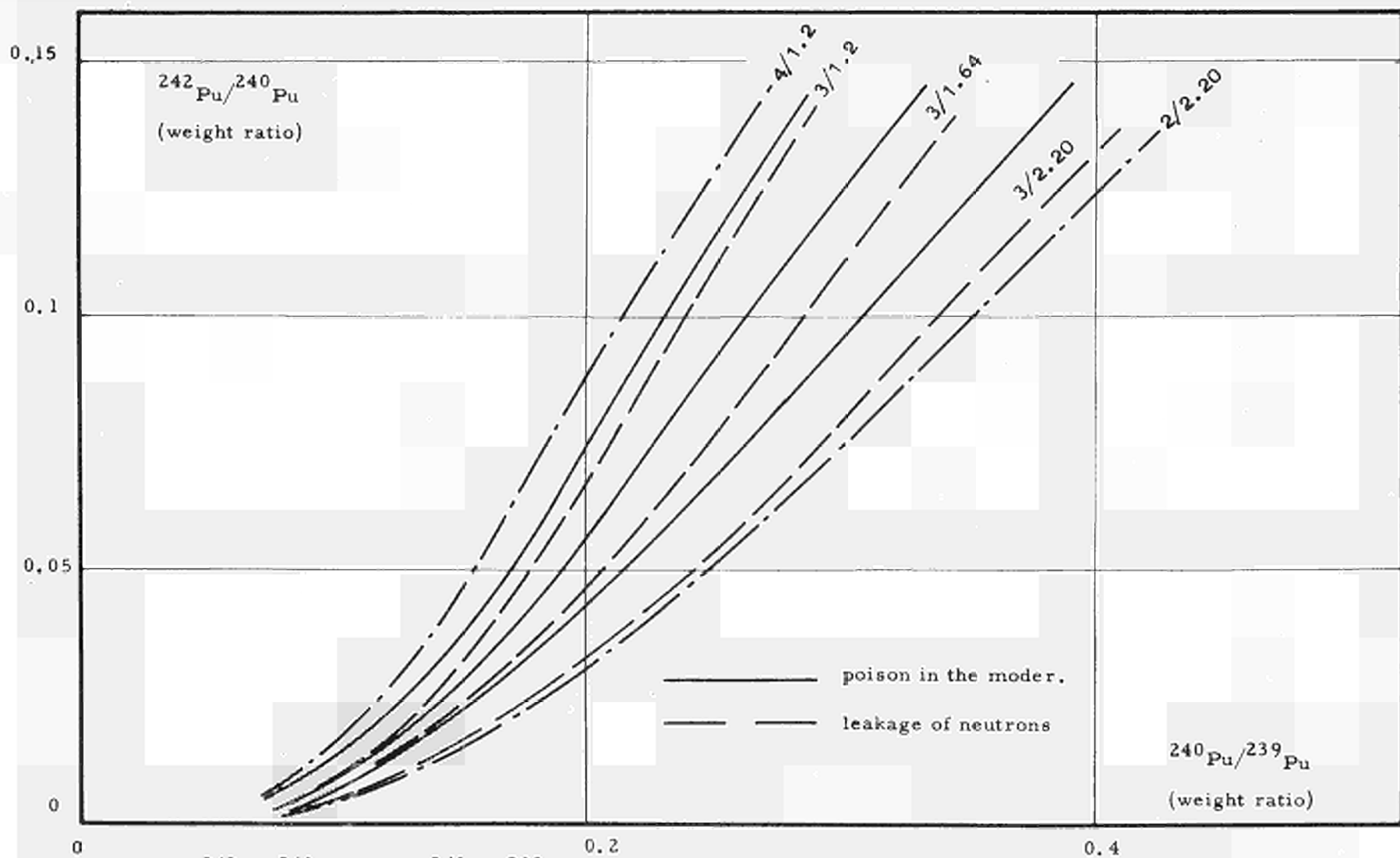


Fig. 25 - $^{242}\text{Pu}/^{240}\text{Pu}$ versus $^{240}\text{Pu}/^{239}\text{Pu}$ for different lattices and different reactivity control methods.

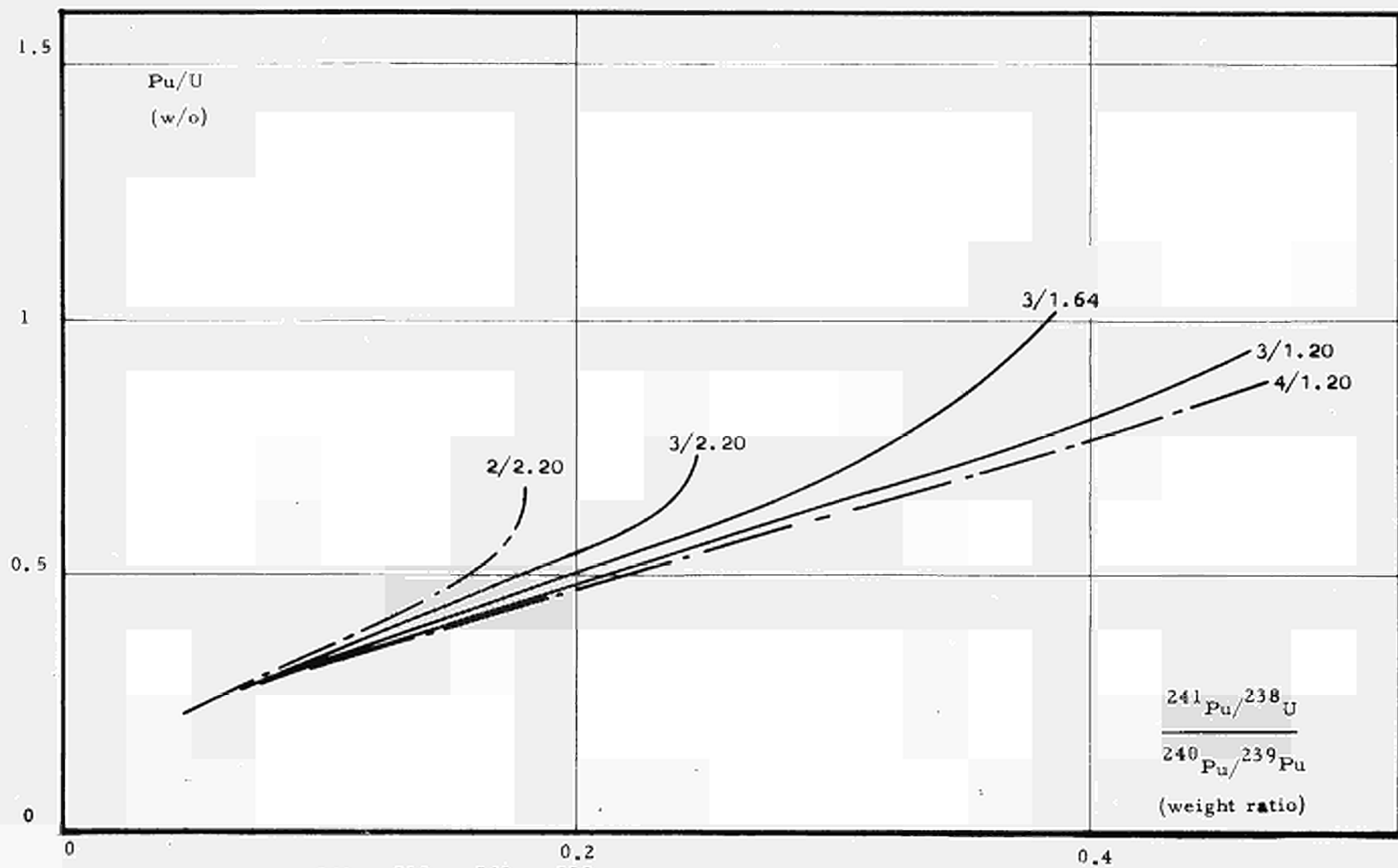


Fig. 26- Pu/U versus $({}^{241}\text{Pu}/{}^{238}\text{U})/({}^{240}\text{Pu}/{}^{239}\text{Pu})$ for different lattices

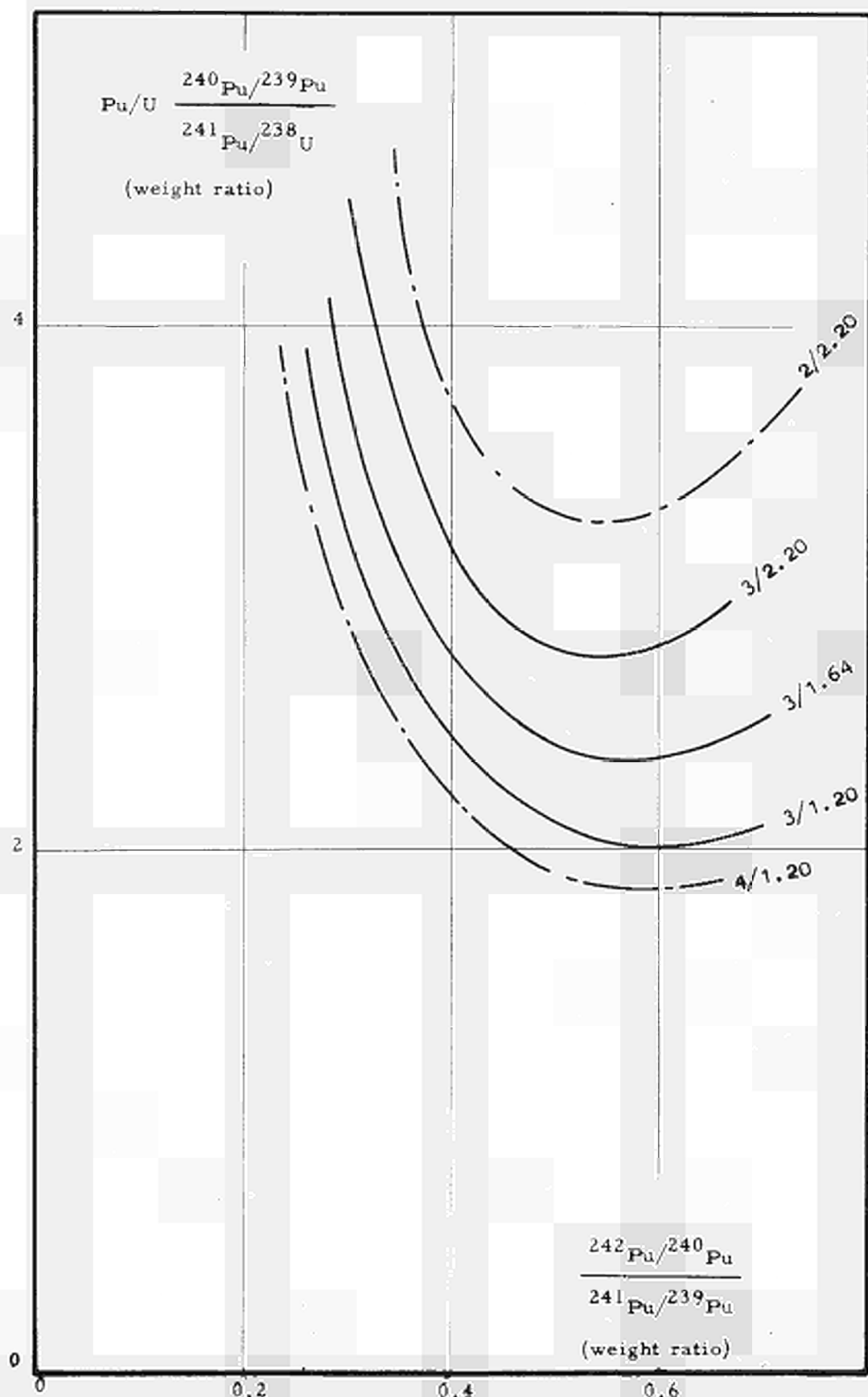


Fig. 27- $\text{Pu}/\text{U} \left[\frac{^{240}\text{Pu}/^{239}\text{Pu}}{^{241}\text{Pu}/^{238}\text{U}} \right]$ versus $\left(\frac{^{242}\text{Pu}/^{240}\text{Pu}}{^{241}\text{Pu}/^{239}\text{Pu}} \right)$ for different lattices



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