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THE BORIS KIDRIČ INSTITUTE OF NUCLEAR SCIENCES - VINČA
CHEMICAL DYNAMICS LABORATORY
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ABSTRACT

Possibility of determination of the neutron flux in the fuel of a heavy water reactor has been examined.

In determination of the flux an iterative procedure was used to compare calculated and measured contents of several fission products. The former contents were determined by calculation of the burning process balance and the latter - by non-destructive gamma-spectrometric analysis of fuel.

The obtained results prove the possibility of such determination of not only the average value of the flux but also of the change of its intensity during utilization of fuel.

СПОСОБ ОПРЕДЕЛЕНИЯ ПОТОКА НЕЙТРОНОВ В ТОПЛИВЕ ЯДЕРНОГО РЕАКТОРА

АННОТАЦИЯ

В работе исследовалась возможность определения потока нейтронов в топливе тяжёловодородного ядерного реактора.

При расчёте потока использовался способ подгонки, в котором сравнивались вычисленные и измеренные содержания нескольких гамма-радиоактивных продуктов деления. Первые содержания определялись с помощью вычисления баланса процесса выгорания, а вторые гамма-спектрометрическим анализом топлива без его разрушения.

Полученные результаты подтверждают возможность такого определения не только среднего значения потока, но и перемены его силы в течение использования топлива.

JEDAN POSTUPAK ODREDJIVANJA FLUKSA NEUTRONA U GORIVU NUKLEARNOG REAKTORA

ABSTRAKT

U radu je ispitivana mogućnost odredjivanja fluksa neutrona u gorivu teškovodnog nuklearnog reaktora.

Pri odredjivanju fluksa korišćen je iterativni postupak, u kome se porede izračunati i izmereni sadržaji nekoliko gama-radioaktivnih fisionih produkata. Prvi sadržaji su odredjivani preko proračuna bilansa procesa sagorevanja, a drugi - nedestruktivnom gama-spektrometrijskom analizom goriva.

Dobijeni rezultati potvrdjuju mogućnost ovakvog odredjivanja ne samo srednje vrednosti fluksa, već i promene njegovog intenziteta u toku korišćenja goriva.

CONTENTS

INTRODUCTION*

Dynamics of the nuclear fuel burning process depends on the flux intensity as a function of time. The balance of this process, and therefore also the burn up, depend on the integral neutron flux.

Radioactive fission product in the fuel: (1) is formed by the fission of fissile nuclides - in the amount which depends on the fuel burn up and on the yield of the fission product, and (2) disappears due to radioactive decay - in the amount which depends also on its half-life, as well as on the duration and dynamics of fuel burning.

The content of some fission product, whose half-life is approximately equal to or longer than duration of the fuel burning process, and whose losses due to radioactive decay for this reason are not great, is determined (in the time observed) by the whole burn out process. The content of such a fission product provides reliable evidence of the intensity of the whole observed burning process /1-3/ and is a suitable means of determination of the integral neutron flux intensity. On the contrary, the content of some short-lived fission product is practically determined only by some final phase of the process, is reliable evidence of the intensity of only that final phase of burning /2,3/ and is suitable means of determination of the neutron flux intensity in that period of time.

Dynamics and balance of the burning process also depend on a series of other quantities. When the burning process is defined in this respect, and when intensity of the neutron flux as a function of time can be expressed in a suitable form, it should be possible to determine both

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intensities on the basis of measured contents of one short-lived and one long-lived fission product.

The aim of the present work was to develop a procedure which would enable this, and to test it experimentally, In achieving this aim, some other, practical, reasons have also been taken into account. Namely, from the standpoint of economy, the methods of non-destructive analysis of spent nuclear fuel are much more suitable than these of destructive analysis which involve high cost of both fuel destruction and storage of radioactive materials. For this reason we based our procedure on utilization of gamma radioactive fission products and determination of their contents on non-destructive gamma spectrometric analysis of fuel.

CONDITIONS

The procedure is developed and tested on the fuel of the research, heavy water reactor RA in Vinča.

Filling of this reactor with fuel is determined by the content of 84 technological channels; eleven fuel elements are placed one above the other in each of these channels /4/.

Seventeen fuel elements, used under different conditions, in two technological channels, have been observed. Six of these elements were introduced into the reactor and placed at central positions of the channel 0305 on September 1, 1968; on September 1, 1969 they were moved to peripheral positions of the same channel and - exactly one year later, they were removed from the reactor. The remaining 11 elements were used in the channel 0808, without replacement, from February 1, 1969 to February 1, 1971.

Running of the reactor within this period of time

is shown in Fig. 1, where the ordinates of the graph represent the operator's records resulting from the measurements of the neutron flux.

The observed elements were analyzed at the beginning of 1972. A spectrometer with a Ge(Li) detector was used. The fission products: ^{95}Zr, ^{106}Ru, ^{134}Cs, ^{137}Cs and ^{144}Ce are determined with high degree of precision /5-7/. Later found changes of the data on the decay schemes of some of these isotopes /8,9/ have been introduced.

ASSUMPTIONS

The observed unirradiated fuel is uranium, containing the isotopes 234, 235 and 238, with the uranium 235 abundance of 2% /10/. Plutonium isotopes and fission products are formed later during fuel burning. Thereby considerable amounts of plutonium are formed only from uranium-238, while the fission products result from the fission of several fissile isotopes.

More than 99% of the consumption of fissile nuclides through the fission reaction, in this fuel, is carried out by two complex reactions from Fig. 2. Numerous fissile products are formed by the fission of fissile nuclides. However, we wish to base our determination on the measurements of ^{95}Zr, ^{134}Cs and ^{137}Cs and we include into the model of the burning process only the transformations which considerably affect the contents of these three isotopes (Fig. 3).

During non-destructive analysis /7/ only a part of the fuel - of small volume ($\sim$0.1 cm^3) - was observed. We assume that it is homogeneous, with respect to both its composition and the neutron flux. In this case the change of the contents of isotopes from the defined process, per

volume unit of fuel and time unit, is given by a system of differential equations (1).

$$\frac{d^{234}U}{dt} = Z_1; \quad Z_1 = \sigma_{234}\,\phi\,{}^{234}U$$

$$\frac{d^{235}U}{dt} = Z_1 - a_1 F_{235}; \quad a_1 = \frac{\sigma_{235}}{\sigma^f_{235}}; \quad F_{235} = \sigma^f_{235}\,\phi\,{}^{235}U$$

$$\frac{d^{238}U}{dt} = -Z_2 - F_{238}; \quad Z_2 + \sigma^{act}_{238}\,\phi\,{}^{238}U; \quad F_{238} = \sigma^f_{238}\,\phi\,{}^{238}U$$

$$\frac{d^{239}Np}{dt} = Z_2 - Z_3; \quad Z_3 = \lambda^{Np}_{239}\,{}^{239}Np$$

$$\frac{d^{239}Pu}{dt} = Z_3 - a_3 F_{239}; \quad a_3 = \frac{\sigma_{239}}{\sigma^f_{239}}; \quad F_{239} = \sigma^f_{239}\,\phi\,{}^{239}Pu$$

$$\frac{d^{240}Pu}{dt} = b\,F_{239} - Z_4; \quad Z_4 = \sigma_{240}\,\phi\,{}^{240}Pu; \quad b = \frac{\sigma_{239} - \sigma^f_{239}}{\sigma^f_{239}}$$

$$\frac{d^{241}Pu}{dt} = Z_4 - a_4 F_{241}; \quad a_4 = \frac{\sigma_{241}}{\sigma^f_{241}}; \quad F_{241} = \sigma^f_{241}\,\phi\,{}^{241}Pu \qquad (1a)$$

$$\frac{d^{95}Zr}{dt} = S^{*}_{95} - \sigma_{95}\,\phi\,{}^{95}Zr - \lambda_{95}\,{}^{95}Zr$$

$$\frac{d^{133}J}{dt} = S_{133}{}^{*} - Z_5; \quad Z_5 = \lambda^{J}_{133}\,{}^{133}J$$

$$\frac{d^{133}Xe}{dt} = Z_5 - Z_6; \quad Z_6 = \lambda^{Xe}_{133}\,{}^{133}Xe$$

$$\frac{d^{133}Cs}{dt} = Z_6 - Z_7; \quad Z_7 = \sigma_{133}\,\phi\,{}^{133}Cs$$

$$\frac{d^{134}Cs}{dt} = Z_7 - \sigma_{134}\,\phi\,{}^{134}Cs - \lambda_{134}\,{}^{134}Cs$$

$$\frac{d^{137}Cs}{dt} = S_{137}{}^{*} - \sigma_{137}\,\phi\,{}^{137}Cs - \lambda_{137}\,{}^{137}Cs \qquad (1b)$$

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* $S_i = \sum_j y_{i,j}\,F_j$

In this system the isotope symbol of an element also denotes the number of its atoms per volume unit of fuel (at/cm^3). Time (in sec) and intensity of the neutron flux (in $n/cm^2/sec^1$) are denoted as t and ϕ, respectively. Suffixes of the cross section (σ) and of the decay constant (λ) symbols correspond to the prefixes (mass numbers) of isotope symbols. Three kinds of the fissile nuclide cross section symbols (upper suffixes) denote: 1) σ^f - cross section for fission reaction, 2) σ^{act} - cross section for (n,γ) reaction and 3) σ - the total cross section for both reactions. Finally, the total S_i sums direct formation of an i^{th} fission product from fission reaction. Thereby, the probability of the formation of the i^{th} fission product from the fissile nuclide, denoted by j is $y_{i,j}$ and it has the dimensions: atoms/fission.

According to the system of equations (1) the change of the contents of isotopes depends on the neutron flux intensity and effective cross sections of reactions with reactor neutrons.

The flux intensity is correlated with the reactor power (W_R). We assume that:

$$\phi(\Theta) = f(\Theta)W_R(\Theta) \qquad (2)$$

where Θ is the time of the reactor operation and f is the proportionality factor whose dependence on Θ will be defined later.

Effective cross sections depend on neutrons' energy. First, we assume that the energy spectrum of neutrons in the observed part of fuel does not change with time. Second, the heavy water reactor RA is a reactor with well

thermalized neutrons, therefore, we conclude that the effective cross sections in this fuel can be determined by applying the Westcott`s formalism /11,12/.

CALCULATION PROCEDURES

The balance of the burning process is determined by the initial composition of fuel and by the neutron flux as a function of time. The initial composition is known from the manufacturer`s certificate and from performed control analyses /10/. Roughly speaking, approximate values of the neutron flux intensities, at the (nominal) reactor power of 6.5 MW, and of the r-factor from the Westcott`s formalism are known on the basis of the reactor core limiting values. Flux intensity as a function of time is given by eq. (2) and the reactor power as a function of time is known from the reactor operator`s records (Fig. 1).

The balance of the burn out process was determined by solving the system of equations (1) while the flux intensity and r-factor approximate values were improved in iterative procedure, by comparing calculated and measured contents of fission products*. Thereby we improved:

1.- Average flux intensity value for the whole period of time of fuel burning (at the nominal reactor power of 6.5 MW), on the basis of ^{137}Cs,

2.- Average r-factor value on the basis of ^{134}Cs and,

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*In the manner described earlier /13/.

3.- Neutron flux intensity in the last period of the fuel burning process (at the reactor power of 6.5 MW), on the basis of ^{95}Zr.

RESULTS

The obtained results are systematized in Tables 1-3. Thereby the average flux intensity value for the whole period of time of fuel use is denoted by $\bar{\phi}$, and the suffixes of other two intensities are in accordance with the suffixes of f-factor in Fig. 4.

The values from Table 1 refer to the elements from the channel 0305. Approximately half a time of their use they were at central positions (first positions) and later they were moved to peripheral positions (second positions). By such removal the flux intensity in a fuel element suddenly changed. This fact is taken into account by sudden change of the f-factor from eq. 2 (Fig. 4,a).

We do not know the manner of change of the flux intensity in the elements from the channel 0808. For this reason we have studied the two cases which seemed most simple and acceptable from the calculation standpoint. The results from Table 2 refer to the sudden change of the flux intensity, approximately in the middle of the interval of fuel use (Fig. 4,a) and the results from Table 3 are obtained for the case of continual and linear change of the neutron flux intensity (Fig. 4,b).

ESTIMATION OF ERRORS

Errors of determined intensities are due to the errors of a series of quantities.

The quantity $\bar{\phi}$ is determined on the basis of ^{137}Cs, and $\sim 99\%$ of the content of this isotope is from fission of

^{235}U and ^{239}Pu. Therefore, the error of $\bar{\phi}$ is caused by the error of measurement of ^{137}Cs, by the errors of its fission yields, the errors of ^{235}U, ^{238}U and ^{239}Pu cross sections as well as by the errors of quantities determining initial fuel composition. Since the errors of cited quantities most often are not greater than 1-2%, their contribution to the probable error of $\bar{\phi}$ usually is not greater than 5-10%. Losses of ^{137}Cs due to radioactive decay are small since its half-life (30 years) is considerably longer than the time of fuel use (1-3 years); at the same burn up the content of ^{137}Cs slightly depends on the dynamics of fuel burning so that the contribution of the errors of these quantities is practically negligible.

As in the case of the previous intensity $\bar{\phi}$, the error of the flux intensity in the last period of the burning process is determined by the errors of similar and the same quantities (^{95}Zr, ^{235}U, ^{238}U, ^{239}Pu etc.). However, the dynamics of the burning process is very important here from two reasons:

- first, the expression "in the last period of the burning process" is not unambigueous and in the two cases observed it denotes two different quantities (ϕ_2 and ϕ_1), and

- second, the loss of ^{95}Zr due to radioactive decay is very high and considerably depends on the dynamics of fuel burning, which itself depends on the manner of flux intensity change as a function of time; the measured content of ^{95}Zr is one and the same so that the error in determination of the manner of flux intensity change prominently increases the error in determination of its intensity near the end of the burning process (Tables 2 and 3).

Intensity ϕ_1 (or ϕ_i) results from $\bar{\phi}$ and ϕ_2 (or ϕ_1). Therefore its error is determined by the errors of $\bar{\phi}$ and ϕ_2 (or ϕ_1).

In addition, the observed intensities refer to some definite reactor power which in our case is obtained on the basis of the operator`s records. As such determinations can be erroneous up to 25% /14/ the obtained results might be erroneous over 25%.

DISCUSSION

Distributions in Figs. 5 and 6 are obtained by using the data from Tables 1 and 2. Dispersion of points around curves drawn by hand is not great and is considerably smaller than it should be expected on the basis of estimated errors of used data (25%).

This small dispersion could be due to two different reasons: namely, it is either accidental or some important facts are neglected, what has actually been done.

First, contributions of errors of quantities referring to ^{137}Cs, ^{95}Zr, ^{235}U, ^{238}U, ^{239}Pu etc. are systematic in character and do not affect the observed dispersion. However, except for the above quantities, this dispersion is conditioned by unrepeatability of measurement of the contents of ^{137}Cs and ^{95}Zr which amounts to about 3%.

Second, errors in determination of the reactor power affect and do not affect the observed dispersion.

Namely, in burning of the nuclear reactor fuel the amounts of formed radioactive fission products practically* depend "only" on the integral neutron flux and their losses due to radioactive decay depend, to more (^{95}Zr) or less (^{137}Cs) extent, on the flux intensity as a function of time. This function is expressed by eq. 2 which, when introduced

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*Most important cause of the limitation of this conclusion is the existence of ^{239}Np in the reaction b, in Fig. 2.

into the employed procedure, led to the correlation of the flux intensity with the reactor power.

In performed determinations it was assumed that the reactor during operation, when it was in operation, had practically the same power (Fig. 1). Times of starting and termination of the operation are not debatable so that only the data on the reactor power could be erroneous.

As first, let us assume that the errors of all these data (%) are of the same sign and amount. In that case performed calculations and determinations would be correct and only that part of the results which indicates the reactor power would be incorrect.

And, as the second, let us assume that the error of the reactor power data continually changes during utilization of fuel - what is more real /14/ and more unsuitable for us. In such a case the obtained intensity values are correct, and the data on the reactor power changes and completely looses its initial meaning.

In the discussion of this conclusion let us analyze separately each of the three observed intensities.

Average flux intensity is determined on the basis of the content of ^{137}Cs. Its losses due to radioactive decay are small, and small differences in the manner of change of the neutron flux intensity as a function of time are not noticeable (Tables 2 and 3). For this reason the obtained value is correct and represents the average value of the flux intensity in the observed fuel, for the whole period of its use.

Flux intensity in the fuel in the last period of its use is determined from the content of ^{95}Zr. Almost all the measured content of this isotope is formed in this period of time. Its losses due to radioactive decay are very high and they essentially depend on the manner of change of the

flux intensity in that period of time; therefore, it is quite irrelevant whether this change is due to the change of the reactor power or to some other circumstances. For these reasons the obtained intensity relates to the observed fuel and period of time and it is correct to the extent to which the assumption of the manner of change of flux intensity during that period of time is correct.

Flux intensity near the beginning of the burning process (ϕ_i or ϕ_1) is determined by $\bar{\phi}$ and $\bar{\phi}_1$, or ϕ_2. Therefore, it corresponds to the observed fuel and period of time and its error is practically only due to the error of ϕ_1 or ϕ_2, $\bar{\phi}$ being correct.

From this standpoint slight dispersions of points around the curves in Fig. 6 are not accidental since all the points around any of the observed curves correspond to the same period of time. On the contrary, slight dispersion of points around the curve in Fig. 5 is accidental one, pointing out, in the final instance, that the errors of reactor power determination in the two observed periods of time were of the same sign and approximately the same in amount.

CONCLUSION

A procedure has been developed for determination of the neutron flux in the fuel of a heavy water reactor. Its main advantage is that it uses economical, non-destructive gamma spectrometric analyses and its basic shortcoming lies in the use of numerous required data and arbitrary choice of the manner of flux intensity change with time.

Its applicability has been tested on the spent fuel of the RA reactor in Vinča.

The error of the obtained result is caused by the errors of a series of quantities; usually it need not be higher than ~10% but in some (discussed) cases it might reach considerably higher value (> 25%).

Repeatability of the obtained results is fairly high (~ 3%). It points out to the fact that this procedure could first of all be used for those investigations in technology of nuclear reactors in which the neutron flux can be expressed in relative form.

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TABLE I. Neutron flux intensities in the fuel from the channel 0305, at the reactor power of 6.5 MW.

Positions of the fuel element in the channel						
First position	6	7	8	9	4	5
Second position	1	2	3	4	10	11
$\bar{\phi} \times 10^{13}$ (a)	2.06 (1) (b)	2.48 (16)	2.46 (6)	2.46 (1)	2.27 (5)	2.39 (2)
$\phi_1 \times 10^{13}$	2.96 (5)	3.33 (17)	3.02 (8)	2.69 (3)	2.31 (3)	2.82 (5)
$\phi_2 \times 10^{13}$	1.00 (5)	1.48 (11)	1.80 (4)	2.19 (3)	2.22 (9)	1.89 (2)

a) In $n/cm^2/sec$.

b) Mean value, determined from 3-6 spectra, and mean square error (in parentheses) on the final figures of the result.

TABLE II. Neutron flux intensities in the fuel from the channel 0808, at the reactor power of 6.5 MW[a].

$\phi \times 10^{13}$, $n/cm^2/sec$	Position of the fuel element in the channel										
	1	2	3	4	5	6	7	8	9	10	11
$\bar{\phi}$	0.98[b] (3)	1.36 (3)	1.88 (2)	2.35 (3)	2.62 (7)	2.83 (1)	2.97 (8)	2.72 (4)	2.61 (9)	2.33 (6)	1.97 (1)
ϕ_1	1.14 (4)	1.57 (4)	2.12 (3)	2.55 (3)	2.80 (7)	2.99 (2)	3.09 (7)	2.87 (3)	2.74 (8)	2.38 (8)	1.96 (1)
ϕ_2	0.85 (2)	1.17 (2)	1.66 (1)	2.17 (5)	2.45 (7)	2.69 (1)	2.85 (8)	2.57 (5)	2.48 (12)	2.30 (5)	1.98 (1)

a) In the case of use of the dependence in Fig. 4,a.

b) See the explanation for Table I.

TABLE III. Neutron flux intensities in the fuel from the channel 0808, at the reactor power of 6.5 MW[a].

$\phi \times 10^{13}$, in n/cm²/sec	Position of the fuel element in the channel										
	1	2	3	4	5	6	7	8	9	10	11
$\bar{\phi}$	0.98[b] (3)	1.36 (3)	1.88 (2)	2.35 (3)	2.62 (7)	2.83 (1)	2.97 (8)	2.72 (4)	2.61 (9)	2.33 (6)	1.97 (1)
ϕ_i	1.15 (3)	1.59 (4)	2.14 (3)	2.57 (3)	2.82 (6)	3.01 (2)	3.10 (7)	2.89 (3)	2.75 (8)	2.38 (8)	1.96 (1)
ϕ_1	0.81 (2)	1.12 (2)	1.60 (1)	2.12 (4)	2.41 (7)	2.65 (1)	2.83 (9)	2.54 (5)	2.47 (11)	2.29 (5)	1.98 (2)

a) In the case of use of the dependence in Fig. 4,b.

b) See the explanation for Table I.

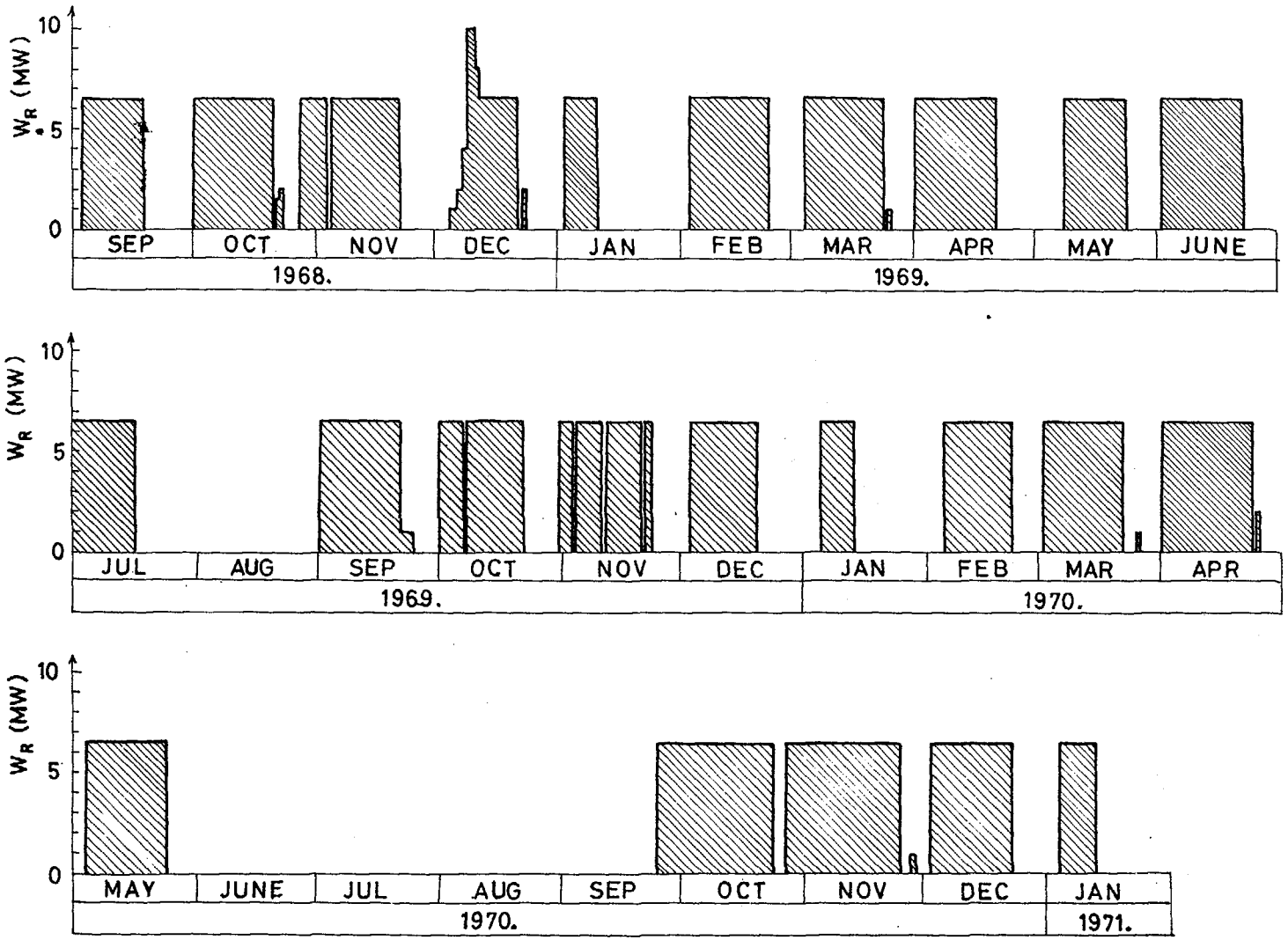

Fig. 1. Histogram of the RA reactor operation in the period of time of utilization of analyzed fuel.

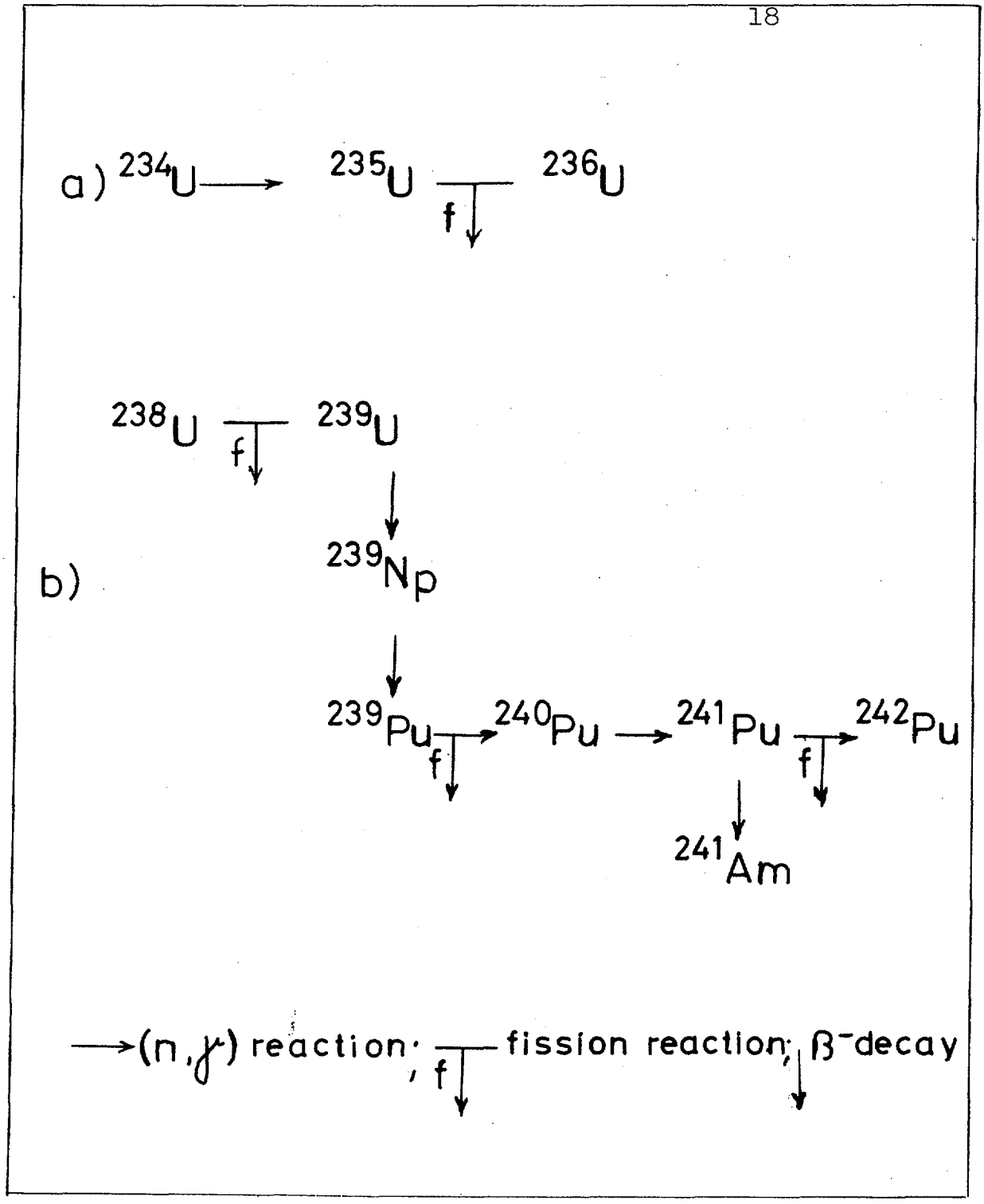

Fig. 2. The most important reactions of the most important heavy atom nuclides in the fuel of the RA reactor.

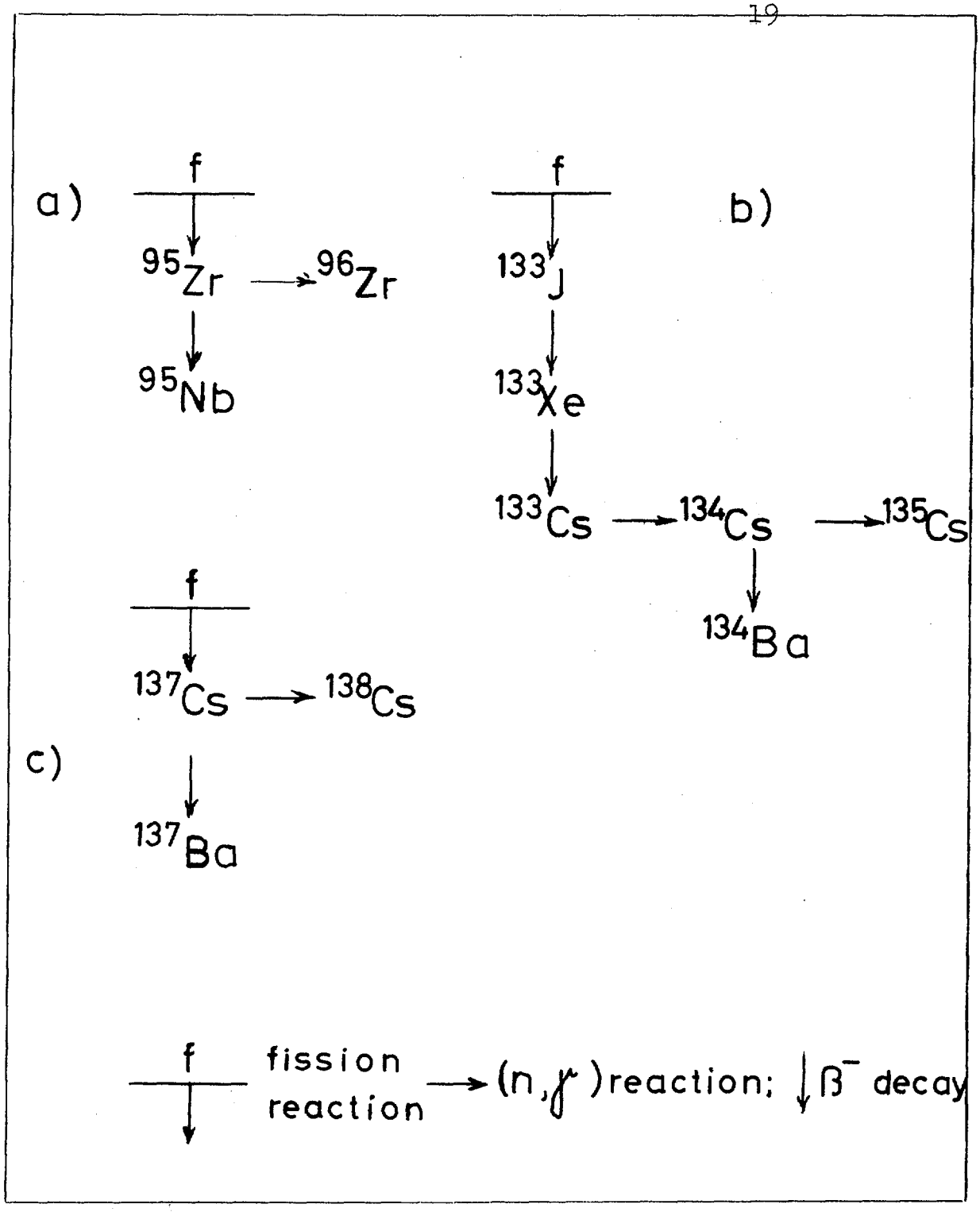

Fig. 3. The most important transformations affecting the contents of ^{95}Zr, ^{134}Cs and ^{137}Cs in the fuel.

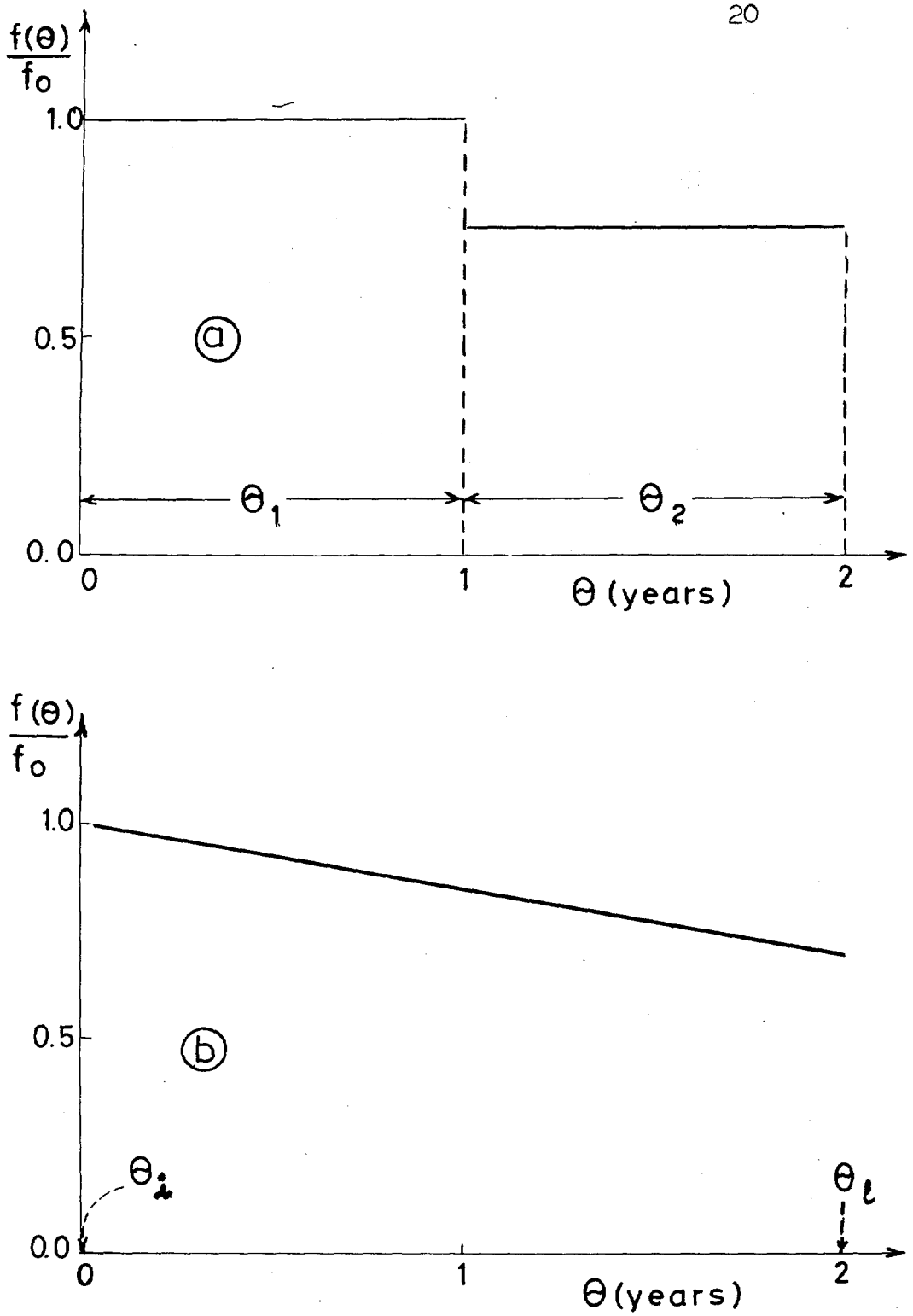

Fig. 4. Two observed manners of change of f factor from eq. 2 versus reactor operation time (Θ).

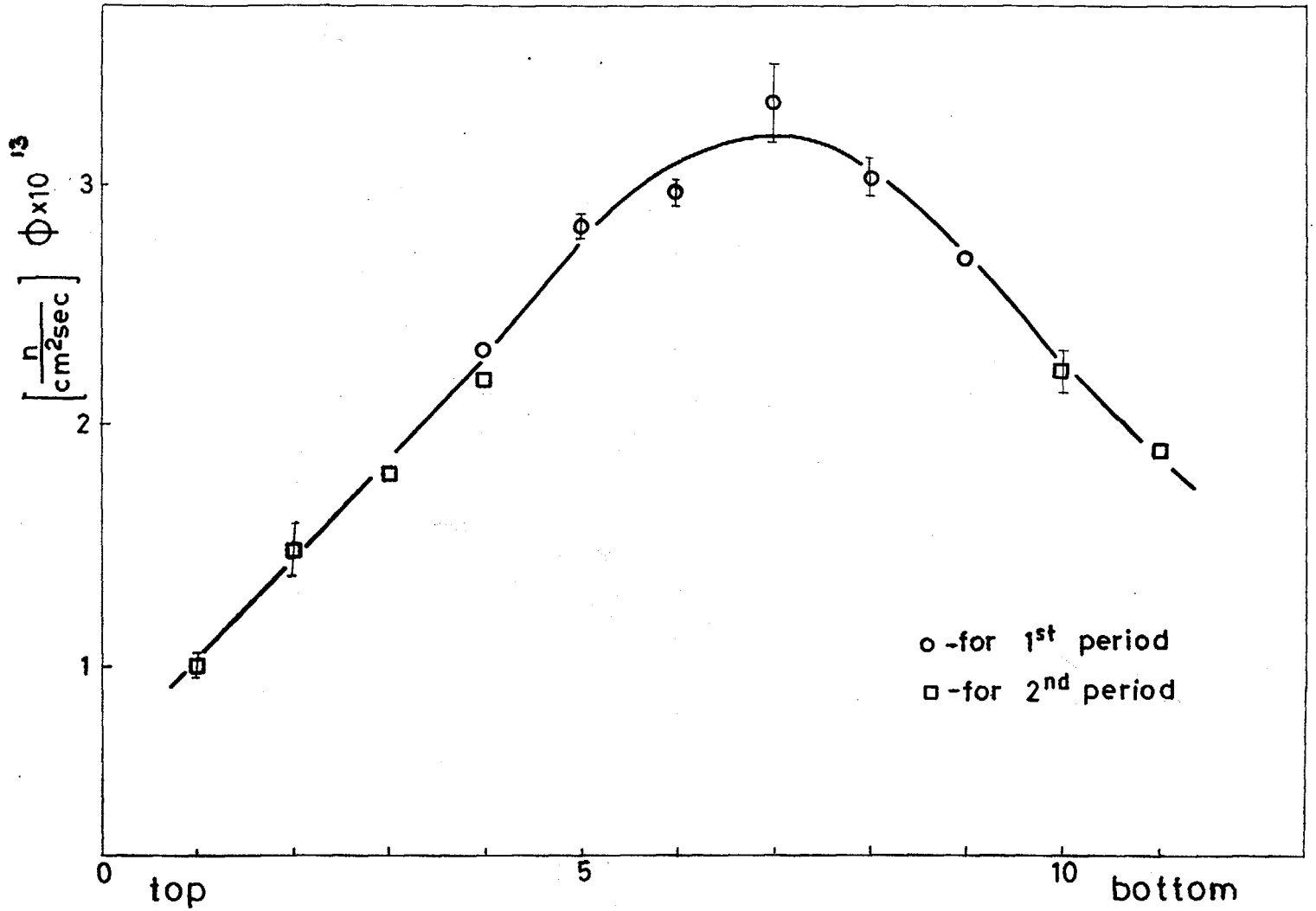

Fig. 5. Neutron flux distribution in the fuel from the channel 0305, at the reactor power of 6.5 MW.

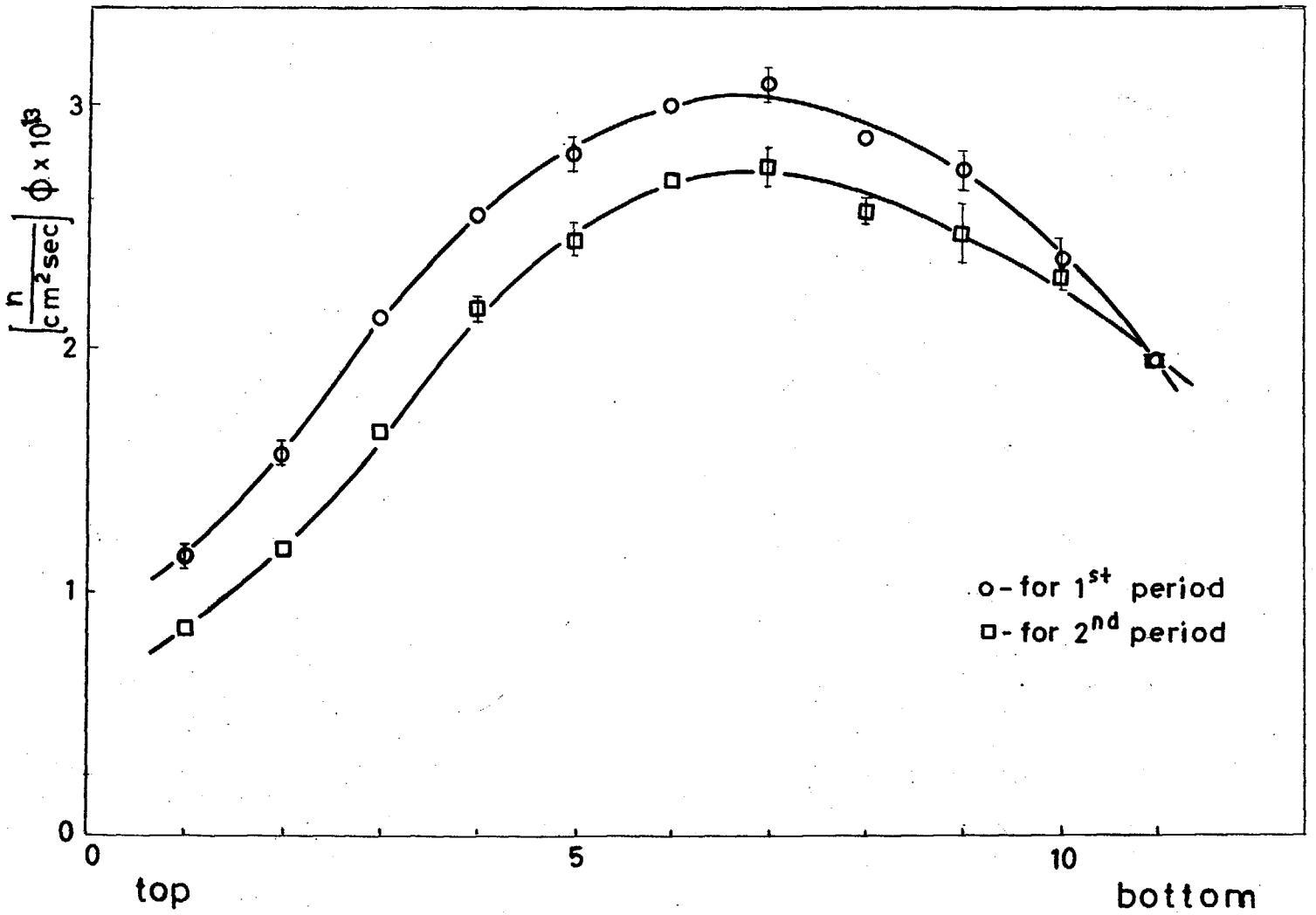

Fig. 6. Neutron flux distributions in the fuel from the channel 0808, at the reactor power of 6.5 MW.

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