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ATOMIC ENERGY
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L'ÉNERGIE ATOMIQUE
DU CANADA LIMITÉE

ONCE-THROUGH THORIUM CYCLES IN CANDU REACTORS

Cycles de thorium à passe unique dans les réacteurs CANDU

M.S. MILGRAM

Chalk River Nuclear Laboratories
Laboratoires nucléaires de Chalk River

Chalk River, Ontario

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Michael S. Milgram

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Résumé

Les cycles de thorium à passe unique sont définis comme des cycles de combustible dans lesquels des grappes de thorium pur peuvent être irradiées conjointement avec des grappes d'uranium dans un réacteur CANDU avec des paramètres judicieusement choisis pour que le coût global du cycle de combustible soit concurrentiel avec les autres possibilités - en particulier l'uranium faiblement enrichi. Cela signifie que U-233 peut être engendré et stocké pour une utilisation éventuelle, sans obligation qu'il soit employé à moins qu'il ne le faille dans l'avenir. On peut donc procéder au stockage d'U-233 indépendamment de l'état d'avancement de la technologie de retraitement. Le but de ce rapport est d'identifier les propriétés générales de ces cycles et de démontrer que ces derniers existent.

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ABSTRACT

Once-through thorium cycles are defined as those fuel cycles in which pure thorium fuel bundles can be irradiated conjointly with uranium fuel bundles in a CANDU reactor with parameters judiciously chosen such that the overall fuel cycle cost is competitive with other possibilities - notably low enriched uranium. This means that U233 can be created and stockpiled for possible future use with no imperative that it be used unless future conditions warrant. Thus production of a U233 stockpile can be begun independently of the state of reprocessing technology. The aim of this report is to identify the general properties of such cycles, and to demonstrate that these cycles exist.

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ONCE-THROUGH THORIUM CYCLES IN CANDU REACTORS

1. INTRODUCTION

The utilization of thorium fuel in CANDU* reactors has been a longstanding goal[1], because of the superior fissile properties of U233 bred from Th232 in a thermal neutron spectrum.

Since naturally occurring thorium does not contain any fissile isotopes, past studies[2] (with a few exceptions[3]) on thorium utilization have concentrated on fuels consisting of homogeneous mixtures of thorium and enriched uranium or plutonium. Two important hindrances to implementation of these cycles are the necessity of

1) reprocessing the fuel to recover the U233; and

2) remotely fabricating new fuel containing radioactive U233.

The one study which did not make use of homogeneous fuel was the "Valubreeder" concept[3], in which fissile and fertile materials were to be irradiated separately; thus fissile and fertile feed rates need not be the same. In this concept, considerable amounts of energy could be obtained from burning U233 formed "in situ". The economics of this cycle, however, postulated a "credit" for U233 recovered, thus presupposing the existence of a marketplace and its concomitant reprocessing infrastructure.

The purpose of this study is to develop a mechanism for identifying fuel cycles which are competitive, but without taking credit for any U233 created. This would eliminate any immediate incentive to reprocess the fuel, yet a potentially valuable commodity can be created and stockpiled for possible future use. The arguments will be based on general principles

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*CANada Deuterium Uranium

of neutron conservation and a representative economic schedule, and it will be shown that "once-through" cycles based on these principles exist.

2. MOTIVATION

An impediment to the utilization of thorium in CANDU reactors is the fact that naturally occurring thorium (100% Th232) contains no fissile isotopes. Thus, to initiate the cycle, thorium must somehow be irradiated in conjunction with fissile material. The simplest way to accomplish this is to design fuel consisting of a homogeneous mixture of thorium and enriched uranium, plutonium or a combination of both. Past studies on thorium utilization in CANDU reactors[2] have concentrated on such designs. But this particular route to inaugurating the thorium cycle has several short-comings.

In the first place, if plutonium enrichment is used, the entire industrial reprocessing infrastructure necessary to produce this plutonium must be in place before the thorium cycle can be implemented. If uranium enrichment is used (presumably by purchase of the enriched material), it might appear that the reprocessing infrastructure is not required beforehand. In this case the uranium may be either highly enriched in U235, which is undesirable[4], or be medium or low enriched (<20% U235) material which degrades the U233 by the presence of U238 in the spent fuel[4]. In either case, such fuel will be extremely expensive, and there can be no economic justification for employing the cycle unless the U233 created is immediately reprocessed and reused. Thus, a demonstrated reprocessing infrastructure would be required with either choice of enrichment.

Secondly, spent fuel containing U233 contains small amounts of U232 (10-50 ppm of fuel), which is the precursor of a family of γ-active nuclides. So refabrication of fuel bearing U233 must be performed by remote control[5], which presupposes that another industrial infrastructure be in place before the economical use of thorium could be countenanced.

Finally, since U233 has superior nuclear characteristics compared to plutonium in a thermal reactor spectrum, its creation should be the direct objective of fuel cycle design. The alternative of creating plutonium, which, in turn, will some day be used to create U233, lacks elegance and efficiency. For example, the plutonium created from natural uranium in a CANDU-PHW is relatively dilute in the spent fuel, whereas U233 created directly in a once-through cycle is about five times more concentrated. Thus by creating U233 directly, ultimate reprocessing costs per gram of fissile material extracted from spent fuel may be reduced if once-through cycles are ever used to produce this material.

The challenge then is to conjure up an economical method of creating U233 now. This means one cannot presuppose the existence of reprocessing, or industries for remote fabrication of fuel; this eliminates the route of plutonium enrichment. The cycle chosen must be competitive with present fuel cycles because of the possibility that the U233 being created will never be used; any additional expense or complexity associated with the cycle can only be justified by the same arguments as are used when insurance is purchased - the premiums must be small if the policy is unlikely to be paid out, or large premiums must be accompanied by the virtual certainty of an eventual return on the investment. In view of the uncertainties associated with the need for advanced fuel cycles [6,7] and the possibilities of future development in other areas (e.g. solar, fusion), any premium paid for the creation of U233 must be adequately small, or non-existent.

Thus we are inexorably led to an evaluation of fuel cycles consisting of thorium enriched with U235, with the added proviso that fuelling costs in such a cycle must be competitive with natural uranium fuel cycle costs, and furthermore, in the economic analysis no credit can be given to the U233 created. This latter point is central to the entire thesis, for, if such a condition is met, it will be possible to economically justify creating U233 now without demanding that it be eventually utilized. If future conditions warrant that advanced fuel cycles be implemented, the U233 will exist in spent fuel from previous irradiation. If advanced fuel cycles are never justified, there will be no impetus to implement such cycles simply to recoup an investment, since no investment will have been made.

It is the purpose of this report to outline the principles of a once-through thorium fuel cycle which meets the criterion outlined above. In addition to this outline, an analysis will be given which illustrates that in at least one economic scenario it is possible to find solutions which satisfy the principles, and hence conclude that such cycles exist. In general we subdivide the cycles into three categories:

- throwaway cycles, where the economics are such that the present use of thorium is <u>beneficial</u> relative to other cycles and so implementation is justified on the grounds of economy without regard to the U233 created;

- stowaway cycles, where the use of thorium has the <u>same cost</u> as other available cycles but the creation of U233 justifies implementation of such a cycle. The U233 is stored for eventual reuse if necessary, but there is no economic imperative that this be done;

- layaway cycles where a <u>slight premium</u> is paid relative to other cycles, with the justification that the premium be sufficiently small that, if no future benefit derives from its payment, little is lost, but, if the material being created is ever utilized, the benefits will greatly exceed the premium.

It is also necessary to distinguish between the terms "economic cost" as a monetary measure, and "resource cost" as a unit of resource utilization. The former term defines the economic burden needed to produce energy, and is measured in mills/(kWe.h). The latter term is a gauge of relative resource husbandry, and may be measured as the quantity of natural uranium (mined) required to produce a certain amount of energy. (In effect, "resource cost" = 1/Burnup where Burnup is measured in MW.d/MgNU*.)

Although it may be possible to minimize "economic cost" by increasing "resource cost", the implementation of an advanced fuel cycle premised on such a combination violates the very objective of advanced fuel cycles - minimization of "resource costs". Conversely, implementing advanced fuel cycles which decrease "resource cost" but increase "economic cost" requires careful consideration and a conscious decision by society to pay the economic premium for resource husbandry. In contrast, an ideal solution would involve no increase in either "resource cost" or "economic cost" during the implementation of an advanced fuel cycle. The purpose of this report is to ascertain if such combinations can be found, within the limits of CANDU technology and the existing industrial infrastructure, focussing on once-through thorium cycles for reasons previously discussed.

Throughout the text, the word "cost" will be used to mean both "economic cost" and "resource cost" if the modifier is omitted. The word "profit", unless modified by the same adjectives, means "decreased cost", and the word "expense" refers to both "economic" and "resource cost" when unmodified.

3. PHYSICAL PRINCIPLES

As discussed previously, the intent is to justify irradiating thorium at either a profit, no cost or slight cost relative to other options according to the category of fuel cycle under consideration. The physical principle relies on the observation that when U233 is formed by the

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*NU = Natural Uranium

irradiation of Th232, the U233 eventually builds up to a dynamic equilibrium level of about 1.3% weight fraction relative to Th232 (fig. 1). The existence of an equilibrium level means that as much U233 is being created by neutron capture in Th232 (fig. 2) as is being destroyed by neutron absorption (absorption equals capture plus fission) in U233. Because of the excellent properties of U233 in a CANDU neutron spectrum, most of the "destruction" of U233 is through the process of fission rather than capture. And, of course, this fission is accompanied by the release of neutrons and heat energy, both of which are partial repayment for the neutrons invested in creating the U233 at an earlier time in the irradiation.

However, repayment of neutrons can never be complete. To understand why this is so, define a quantity $k_T^I(w_T)$, the irradiation integrated multiplication constant for thorium fuel, by

$$k_T^I(w_T) = \int_0^{w_T} Y_T(w')dw' \ / \int_0^{w_T} A_T(w')dw'$$

where $Y_T(w)$ is the w (irradiation) dependent yield of neutrons from a U233-thorium fuelled cell, and $A_T(w)$ is the (irradiation dependent) total absorption of neutrons in the same material. This ratio is a measure of the lifetime excess (if greater than 1.0) or deficiency (if less than 1.0) of neutrons produced by the irradiation of Th232 (in ThO_2) as a function of the thorium irradiation w_T. Because of the presence of Pa233 in the transmutation chain of Th232→U233, this quantity will also be dependent on the magnitude of the flux ϕ in which the sample is irradiated. All calculations are for a 37-element bundle in a Gentilly-II type lattice.

In fig. 3, the dependence of $k_T^I(w_T)$ and its instantaneous value is illustrated as a function of thorium irradiation (w_T) at two levels of flux which are representative of values at which thorium fuel would likely be irradiated. Also shown in this figure for comparison is the dependence of the similar quantity $k_u^I(w_u)$ for a fuel bundle enriched to 1.8% U235 in U238.

Two important properties must be noted:

1) the value of $k_T^I(w_T)$ never exceeds 1.0, so there is no possibility of repaying, in one complete irradiation, the neutrons that have been absorbed by the Th232. This is because fission products build up and act as parasitic absorbers even as the U233 is created and fissions;

2) for burnups greater than 20,000 MW.d/Mg, the value of k_T^I is greater than .8 indicating that U233 has built up in the thorium fuel close to its equilibrium value and the fuel is relatively reactive though subcritical. In contrast, near the end of its irradiation lifetime, the enriched uranium bundle is so depleted of fissile material that its reactivity is exceeded by that of the thorium bundle.

The general principle upon which once-through thorium fuel cycles are based is founded upon these two observations.

Specifically, the intent is to reduce the burnup of the enriched uranium slightly, so that the lifetime value of $k_u^I(w_u)$ exceeds 1.05 by a small margin. (In practice and for the remainder of this paper, we shall use the value $k^I(w) = 1.05$ to represent lifetime "breakeven neutron excess" which allows for variables such as leakage, control and instrumentation, and, for the case of 37-element natural uranium CANDU fuel, gives a burnup prediction of 7630 MW.d/Mg in reasonable agreement with observation.) This lifetime excess of neutrons for the enriched uranium is then invested in irradiating a (different) quantity of thorium to such an extent that U233 is created, and burned "in situ". The energy emitted by fissioning the U233 in situ will be a repayment for the neutrons invested in its creation. In general, we anticipate that the thorium will need to be irradiated to greater than 20,000 MW.d/Mg to retrieve the neutrons invested in it during the early part of its irradiation, because at such large burnups a small increment of reactivity will permit the extraction of considerable amounts of energy according to fig. 3.

Performing this trade-off of burnup between U235 in enriched uranium, and U233 created and burned in situ, requires the introduction of a new design variable - the thorium to uranium feed ratio y. That is, the fuel cycle must have the flexibility of varying the rate at which thorium is fed to the reactor relative to the feed rate of uranium, permitting the thorium to be subjected to an irradiation different from that of uranium. This means that the thorium and uranium must be physically separated and irradiated in different bundles, in contrast to the homogeneous mixture of uranium and thorium considered in other studies. This heterogeneous loading will be referred to by the name "dual fuel".

The introduction of dual fuel leads to a number of consequences.

First is the predication that under some economic conditions, optimal values of y exist that justify the introduction of such dual fuel cycles. This, of course, is the motive for this work.

Second is the flexibility in choosing the ratio y, defined as (number of thorium bundles loaded per day) / (number of uranium bundles loaded per day). Alternatively, the instantaneous loading ratio N_T/N_U, (number of thorium bundles) / (number of uranium bundles, being simultaneously irradiated), is dictated by the choice of y. And the choice of y itself can be made according to "cost" considerations; in the following sections we shall see how y may be chosen to optimize one of the three fuel cycle categories defined previously. Because of this flexibility of dual fuel, the reactor operator may have more freedom to respond to changes in economic conditions. For example, if the price of enriched uranium varies such that a new optimum value for y is indicated, it may prove feasible to vary only the fuelling rate ratio without encountering problems of fuel redesign which would occur in fuel cycles using homogeneous fuel.

Third, the introduction of dual fuel launches a new technical variable in reactor fuel management. The utilization of two kinds of fuel opens the door to possible technical advances in the utilization of low enriched fuel (LEU) in CANDU reactors[8]. Technically, the use of LEU has been impeded because of heat transfer and critical power ratio problems. The introduction of a new variable in fuel management may allow such difficulties to be circumvented. On the other hand, it may make them worse. This will be touched on in a later section.

From the physics point of view, uranium created via a dual fuel route should be superior to that created by means of irradiation of homogeneous bundles, because the U233 will not be degraded by the presence of U238 as would be the case if enriched uranium were used as the fissile component of the homogeneous fuel. This increases the value of the U233 if it is ever to be used in an advanced fuel cycle, because U238 acts as a powerful parasitic absorber[4].

4. CHOICE OF "RESOURCE COST"

The purpose of this section is to demonstrate a methodology for delineating the parameters of "dual fuel" cycles such that "resource costs" never exceed those of competing possibilities: the natural uranium CANDU-PHW in all cases, and LEU in CANDU-PHW for the case of stowaway-throwaway cycles. Given that all choices of "dual fuel" cycles satisfy this criterion, in a later section it will be shown how to further hone the selection of possible cycles according to a particular economic schedule to further guarantee that "economic costs" do not exceed the same competing possibilities. When such a culling has been achieved, the objective of this report will have been met.

Define the combined burnup B_c in MW.d/Mg of uranium in terms of the burnup of each of the component fuels B_u(MW.d/Mg of uranium) and B_T(MW.d/Mg of thorium) by

$$B_c(w_u, w_T, y) = B_u(w_u) + .92y\, B_T(w_T) \tag{4.1}$$

As indicated, B_c is an explicit function of w_u and w_T, the irradiation of the uranium (thorium) fuel as well as the relative feed rate y defined earlier. The factor .92 is included to account for the lower density of thorium relative to uranium. Similarly, define an integrated combined lifetime reactivity[9] for the fuel cycle by

$$k_c^I(w_u, w_T, y) = \frac{\int_o^{w_u} Y_u(w)dw + y\int_o^{w_T} Y_T(w)dw}{\int_o^{w_u} A_u(w)dw + y\int_o^{w_T} A_T(w)dw} \tag{4.2}$$

where subscripts u and T, respectively, refer to uranium and thorium fuels, Y is the yield and A is the absorption of neutrons. In addition to the explicit dependence on the variables indicated, both k_c and B_c

have an implicit dependence on ϕ, the flux level in which the thorium fuel is irradiated, and ε the enrichment level of the uranium fuel. The purpose of the exercise is to obtain optimal values of w_u, w_T, y and ε consistent with one of the three dual fuel cycles.

To begin, note that separate calculations give rise to the estimate that ϕ, the average flux level in which the thorium will be irradiated, will probably lie between 3 and $5\text{x}10^{13}$ $\text{n.cm}^{-2}.\text{s}^{-1}$; these two values will then serve as lower and upper bounds for the calculations. The accurate determination of the flux level will depend on detailed fuel management calculations of an actual embodiment of the cycle.

Proceed by choosing an enrichment ε, a (bounding) flux ϕ and fix y at some convenient value. This leaves w_T and w_u, the irradiation of the two fuel types, free. Temporarily fix w_T at some value in the range of likely acceptable thorium irradiations (in effect $0 \leq w_T \leq 12$ n/kb) and calculate B_c and k_c^I according to Eqs. (4.1) and (4.2) as a function of w_u. For example, fig. 4 demonstrates that if $y=.15$, $\phi=5\text{x}10^{13}$ $\text{n.cm}^{-2}.\text{s}^{-1}$, $w_T = 9.9$ n/kb and $\varepsilon = 1.81\%$, the lifetime combined excess reactivity k_c^I reaches a maximum at $w_u \approx 2.0$ n/kb and decreases thereafter. Significantly, $k_c^I = 1.05$ at $w_u = 4.05$ n/kb, which is illustrated in the figure and is indicated by the point X. This means that for the chosen values of ε, ϕ, y and w_T, any values of w_u greater than 4.05 n/kb will lead to a lifetime neutron deficiency and are therefore not viable. Conversely, values of w_u less than 4.05 n/kb will result in a lifetime neutron excess and are not efficient. However, the value $w_u = 4.05$ n/kb will result in an overall lifetime breakeven in neutron population, and thereby represents a viable dual fuel cycle. From tables of $B_T(w_T)$ and $B_u(w_u)$ we subsequently discover that this particular combination of variables corresponds to

$$B_u(w_u = 4.05) = 25{,}000 \text{ MW.d/Mg U}$$

$$B_T(w_T = 9.9) = 63{,}700 \text{ MW.d/Mg Th}$$

or a combined burnup according to Eq. (4.1) of

$$B_c = 34{,}000 \text{ MW.d/Mg U}$$

In order to compare burnups independently of enrichment levels, it is necessary to relate B_c back to the quantity of natural uranium originally mined to produce one Mg of 1.81% enriched uranium. Assuming a tails enrichment of .2% at the separation plant we find

$$B_c = 10{,}900 \text{ MW.d/Mg NU}$$

demonstrating that for this particular combination of w_u, w_T, y, ϕ and ε it is possible to decrease the "resource cost" by extracting 43% more energy from a given amount of natural uranium relative to employing that uranium in a CANDU natural uranium once-through fuel cycle (10,900/7630 = 1.43) as is presently done. It is now necessary to identify which of the multitude of possible values of w_T, y and are optimal.

Referring back to fig. 4, the point X defines the predicted uranium burnup of a dual fuel cycle based solely on the principle of conservation of neutrons. If the thorium burnup w_T were to be varied, a new, unique value of w_u would be found, keeping y, ϕ and ε constant all the while. And for each explicit combination of y, w_T and w_u (and implicit value of ϕ and ε), it is possible to predict a unique value for the combined burnup B_c (w_u, w_T, y).

In fig. 5, such a computation has been performed; here we find $B_c(w_u, w_T, y)$ plotted as a function of w_T over its likely allowable range of values - each point on this curve represents the predicted burnup of a dual fuel cycle when ϕ,ε and y are fixed and the thorium irradiation is allowed to vary. The point $w_T=0$ represents a low enriched uranium (LEU) fuel cycle; thorium is not used. As w_T increases, the combined burnup decreases as expected, because the thorium has not had sufficient irradiation to create sufficient U233 to generate "payback" energy. At about 2 n/kb, however, the thorium fuel will be sufficiently irradiated that significant amounts of U233 will start to fission in situ, and, as w_T increases from that point, the contribution from the U233 leads to an increase in the combined burnup B_C. Finally, at $w_T = 6.3$ n/kb the combined burnup equals the burnup at $w_T = 0$; this represents the breakeven point of dual fuel cycles. That is, by decreasing the burnup of uranium (w_u) and increasing the burnup of thorium (w_T), the same amount of energy can be extracted from dual fuel cycles as from LEU cycles, with the provisional benefit that some U233 will have been created in the dual fuel cycle, rather than Pu239 in the LEU cycle.

If w_T increases past 6.3 n/kb, a broad maximum is obtained in the combined burnup near $w_T \approx 11.8$ n/kb. This indicates that when 6.3 n/kb$\lesssim w_T \lesssim$11.8 n/kb (for fixed y, ϕ and ε), it may turn out to be beneficial to utilize dual fuel cycles vis-à-vis an LEU cycle at the same enrichment, in order to minimize "resource cost". The energy extractable from a given amount of natural uranium (mined) may be greater for a dual fuel than for a similar LEU cycle, and it may turn out that the "economic cost" too will be smaller.

Alluding to the three categories of fuel cycles defined earlier, the point at $w_T = 6.3$ n/kb denoted as point Q represents a demarcation. In terms of energy output, all values of w_T to the right of point Q are potential throwaway cycles - there is an energy advantage in employing any one of these. Conversely, points to the left of point Q represent possible layaway cycles - the combined burnup B_C exceeds that of a natural uranium cycle at 7630 MW.d/Mg NU - but are not as energy efficient as the LEU cycle indicated by the case $w_T=0$. The minimum of the curve to the left of point Q (denoted as point R) represents the lower bound of layaway cycles, since there is no purpose in considering points to the left of "R" - there is a point with equal burnup to the right which will always have more U233 created. From the figure, we see that in such cycles, U233 will have been beneficially created with a net energy output that exceeds (or perhaps in some cases equals) that of a natural uranium fuel cycle; the "resource cost" (or premium) is that the better burnup potentially available by employing the LEU cycle will have been foregone.

Finally, point O itself represents the energy breakeven or "stowaway" cycle. There is no energy advantage or disadvantage in employing dual fuel cycles operating at point Q relative to an LEU cycle with the same enrichment operating at the point $w_T=0$. So in this case, U233 will have been created at the same energy cost relative to LEU and so there is no compulsion to reprocess; this is the stowaway definition.

Referring to fig. 5, points P, Q and R serve to delimit the dual fuel cycles of interest at a particular value of y, but an optimal value of y has yet to be chosen. To do this, it is necessary to recalculate points P, Q and R for various values of y; the loci of such points for a range of y values is illustrated in fig. 6. Prior to a detailed discussion of this figure, which introduces "economic costs", it is necessary to define cost estimates. This is done in the next section.

5. COSTING

A simple model for the cost of fuel supply (cfs) is calculated[3] according to

$$\text{cfs} = \frac{1000\ C}{24e\ B_c}\ (\text{mills}/(\text{KWe.h})$$

where the electrical/thermal conversion efficiency $e=.3$ and C is the total fuel cost per kg U. To further calculate C we have

$$C = C_u + .92y\ C_T$$

where $C_{u(T)}$ is the specific cost of uranium (thorium) according to the subscript, and

$$C_u = F_u + WC_{fu} + SC_s + \theta(W-1)[WC_{of} + F_{u\varepsilon}]$$

$$C_T = F_T + C_{fT}$$

where

$F_{u(T)}$ = fabrication cost/kg of uranium (thorium)

W = $(X_\varepsilon - X_t)/(.00711 - X_t)$

= weight of uranium mined/weight of enriched uranium

S = $V(X_\varepsilon) + (W-1)V(X_t) - WV(X_f)$

= No. of SWU/kg of uranium

C_s = cost of a SWU (separative work unit)

C_{of} = cost of conversion from oxide → hexafluoride/kgNU

$C_{fu(T)}$ = cost of uranium (thorium) feed / kgNU(Th)

$F_{u\varepsilon}$ = incremental fabrication cost of enriched U/kgU

X_t = tails enrichment = .002

X_ε = fuel enrichment

X_f = feed enrichment = .00711

$\theta(W-1)$ = 1 if $W>1$
= 0 if $W\leq 1$
= allowance for discontinuous incremental charges associated with enriched fuel

$V(x) = (1-2x)\ell n((1-x)/x)$

In addition to this estimate, interest charges should be assigned to each cost component according to its duration and location in an irradiation sequence and initial inventory charges should be included. This will not be done in this note, in order to preserve simplicity and emphasize principles.

Dollar values have been assigned to each of these variables according to schedule 1 below.

SCHEDULE 1

F_u = \$ 50/kg $F_T = F_u$

C_{fu} = \$100/kg $C_{fT} = 1/3\ C_{fu}$

C_s = \$100/kg

C_{of} = \$ 10/kg

$F_{u\varepsilon}$ = \$ 10/kg

The value of C_{fT} was chosen because the relative abundance of thorium to uranium ores is about 3:1.

6. WINNOWING

(a) Economics

Subject to the methods, constraints and costs presented in the previous two sections, the intent is to now demonstrate that dual fuel cycles exist meeting the objective of minimizing both economic and resource costs relative to LEU.

Refer to fig. 6a, where the combined burnup B_c is plotted as a function of y for each of the three cycles; the points P, Q, and R from fig. 5 are shown, as is their locus as y is varied. Since these loci represent boundary points of the cycles, the interior represents a multitude of possibilities, each of which satisfies the constraints defining one of the three cycles, although the associated costs may not be minimal. This is indicated by the crosshatching.

In fig. 6b, the same cycles are depicted, but here it is the fuel cycle cost (cfs) that is plotted against y according to Schedule 1. And in fig 6c for the same three sets of cycles, the thorium burnup B_T is plotted; this is an important engineering parameter.

From these diagrams, the following attributes of the three cycles can be inferred:

- For throwaway cycles, the maximum burnup B_c (minimum resource cost) and minimum economic cost both occur at about the same value of $y \sim 0.1$, representing the best of the possible throwaway cycles at this level of flux and enrichment. This is indicated by the point P*. However, to achieve this cycle, the thorium burnup B_T is excessive, lying in the range $B_T \sim 10^5$ MW.d/Mg, and is probably not achievable. From fig. 6c we see though that throwaway cycles do exist with 30,000 MW.d/Mg $\leq B_T \leq$ 50,000 MW.d/Mg, which probably represents the upper range of achievable technology near-term[†], and from figs. 6a and 6b it can be seen that this can be achieved at very little economic or resource cost, simply by moving slightly into the hatched area.

- For stowaway cycles, B_c is constant as a function of y and cfs is nearly so. The point Q* represents one end of the acceptable range of stowaway cycles, and the thorium burnup B_T probably lies near the upper limit of acceptability. Since the objective is to produce as much U233 as possible at no extra cost, this suggests as large a value of y as possible, so the point Q* designates the best choice for this cycle, although it too can be shifted to the left at little resource or economic cost if $B_T \gtrsim 50{,}000$ MW.d/Mg proves too extreme.

- For layaway cycles, the locus of point R in fig 6a is monotone decreasing, but still exceeds the burnup of a natural uranium cycle up to large values of y. However, the cfs increases rapidly with y, and exceeds that of the natural uranium cycle when $y > .2$, as indicated in fig. 6b. This represents a limit for layaway cycles, and the point at which this occurs is denoted by the symbol R*. In these cases it is seen from fig. 6c, that the thorium burnup B_T is inordinately low, and can be improved (for constant values of y) by moving into the hatched area from the point R*. When this is done the corresponding economic and resource costs are reduced.

Overall, the impression is that at this level of flux and enrichment, the extreme points defining the best choice for each of the three cycles are not optimal due to the associated values of B_T, but optimal cycles do exist in a broad band roughly paralleling the locus of point Q (stowaway cycles).

[†] Indications are that ThO_2 may be successfully irradiated to such large burnups; c.f. reference 10.

Finally, it is necessary to study the sensitivity of these results as a function of enrichment level and flux. In fig. 7, the locus of points P*, Q* and R* is shown as a function of the uranium enrichment ε at two levels of flux ϕ.

First consider the resource and economic costs represented by cfs in fig. 7a and combined burnup B_c in fig. 7b. The cfs for throwaway cycles is less than that for stowaway cycles, which in turn is less than that for the layaway cycles, as expected. For stowaway cycles, the cfs increases rapidly as the enrichment increases, and an enrichment of 2.1% is about an upper limit for these cycles, while for throwaway cycles, ε=1.2% is about a lower limit. Similarly, the combined burnup for throwaway exceeds that for stowaway which in turn exceeds that for layaway, with about the same range of acceptable enrichments. In both cases, the dependence on the thorium flux level ϕ_T is not significant.

In fig. 7c, the thorium burnup B_T is given as a function of enrichment level, emphasizing that for optimal throwaway cycles, B_T is unacceptably high at all enrichments. However, for stowaway cycles, B_T decreases to a reasonable limit with increasing enrichment[10], whereas this parameter is relatively flat and small for layaway cycles over the range of enrichments.

(b) Fissile Material Production

Since the objective of employing such cycles is to create maximal amounts of U233 at low cost, some estimates of the production of this isotope must be examined. In fig. 7d the feed rate ratio y corresponding to each of the optimal cycles is shown over a range of enrichments. For throwaway cycles y is fairly small except when $\varepsilon \gtrsim 1.8\%$. For layaway cycles, this parameter peaks and is of meaningful magnitude and fairly constant over the range $1.2\% \leq \varepsilon \leq 1.8\%$, and for stowaway cycles, rises to significant levels when ε approaches the upper end of its range. In all cases, there is some dependence on ϕ_T. In general, U233 production should increase with increasing values of y.

The further possibility exists of reckoning the thorium to uranium loading ratio N_T/N_u, and PR^3, the U233 production ratio (g U233/day)/(g U235/day) for these cycles. For given levels of uranium and thorium average flux (ϕ_u and ϕ_T) we have

$$N_T/N_u = y \frac{w_T}{w_u} \frac{\phi_u}{\phi_T}$$

where, due to uranium fuel properties being insensitive to flux levels, the value $\phi_u = 7\text{x}10^{13}$ $\text{n.cm}^{-2}.\text{s}^{-1}$ has been used throughout. This ratio is presented for the various cycles in fig. 7e.

Making the further assumption that (U233+Pa233) has built up to a flux independent saturation level of about 1.5% of the Th232 content for all values of w_T under consideration (this is probably invalid only for layaway cycles when $w_T \leq 20{,}000$ MW.d/Mg), we make the further estimate that

$$PR^3 \approx .92\text{x}1.5\ y/\varepsilon \quad \text{(g U233/day)/(g U235/day)}$$

which is an upper limit when the saturation assumption isn't valid. From separate computations, we obtain PR^9, the fissile production rate measured as (g Pu239/day)/(g U235/day) in enriched uranium cycles, shown as the dashed curve in fig. 8 (Pu239 includes Np239). For dual fuel cycles the quantity of interest is the total fissile production ratio PR^{fiss} given by

$$PR^{fiss} = PR^9 + PR^3$$

An examination of fig. 7e now indicates that for layaway cycles very little thorium (<20%) is ever present at one time, whereas for stowaway or layaway cycles near the upper end of the enrichment range, as much as 50% of the tolerable fuel load could be ThO_2 bundles. However, this parameter is very sensitive to flux levels, so careful calculation of fuel management strategies will be required to establish a viable loading that will result in thorium irradiations consistent with expected values of y and B_c.

Turning to fig. 8, the top dashed curve illustrates one inherent feature of LEU cycles - the value of PR^9 decreases as uranium enrichment increases and consequently, relatively less fissile plutonium is created. (It should be noted parenthetically that the plutonium produced in LEU cycles is of different isotopic composition than that of natural uranium cycles. If plutonium produced from LEU were to be used to create U233 in a homogeneous ternary blend of PuO_2-UO_2-ThO_2, the overall burnup of such a tandem cycle differs little from that obtained from a cycle where the PuO_2 emanates from natural uranium. However, the production ratio of U233 from the original U235 mined decreases as LEU enrichment increases, but the concentration of plutonium in spent fuel entering a reprocessing plant increases with increasing LEU enrichment.) The qualitative explanation for this decrease is that the Pu239 concentration saturates at its equilibrium level for reasonable burnups, and this level depends only on the U238 progenitor of Pu239 whose capture cross section is roughly constant. Thus the Pu239/U235 production ratio decreases because of an increase in U235 concentration with enrichment. In contrast, in a thorium cycle the U233 saturates at roughly 1.5% of the Th232 concentration, in almost all conditions.

In dual fuel cycles, the fuels are physically separated, and both Pu239 and U233 are free to saturate independently of one another, which results in a significant increase in the overall fissile production ratio. This is demonstrated in the curves of fig. 8, revealing the U233 production ratio PR^3 for each of the three cycles at both levels of flux. Consistent with the feed rate ratio curves of fig. 7d, the throwaway cycles have very little U233 production, whereas near the high end of the enrichment range both stowaway and layaway cycles give rise to U233 production comparable to that of Pu239 in LEU cycles, although the estimate for layaway cycles will likely be high because of the low thorium burnup in this case (fig. 7c).

Finally, the top solid curve of this figure discloses the fissile production ratio $PR^9 + PR^3$ for the stowaway cycle, at two levels of flux. This ratio reveals some sensitivity to flux level, but, more significantly, it is in a range comparable to that of PR^9 for a natural uranium cycle. Thus with dual fuel cycles in this regime, it appears possible to make use of LEU fuel cycles without losing the fissile production efficiency of natural uranium cycles, and gaining the advantage that U233 in thorium is produced in a more concentrated form (∿5 times) than Pu239 in uranium.

7. CONCLUSIONS

In the previous sections, a winnowing process has been described designed to isolate dual fuel cycles of economic interest. From the cost and resource curves of figs. 6 and 7, the enrichment range of interest is seen to lie in the range $1.5\% \leq \varepsilon \leq 2.1\%$, and from the other curves, it is seen that a choice of fuel management such that ϕ_T lies nearer to $3\mathrm{x}10^{13}$ $\mathrm{n.cm^{-2}.s^{-1}}$ would be beneficial. Although it has been possible to detect a large family of cycles satisfying the initial criterion for throwaway, stowaway and layaway, in practice the throwaway cycles impart only a marginal cost advantage and require large enough values of B_T as to suggest their impracticability. Conversely, the layaway cycles have only a marginal economic cost penalty, but the associated burnup B_T is relatively small so that the implementation of such a cycle may not be worth the effort. In contrast, the stowaway cycles, and a large family of cycles with parameters in the nearby range of values show considerable promise (e.g. a band roughly parallel to the locus of point Q in figure 5). Thorium burnups can be chosen in an acceptable range[10] ($30\mathrm{x}10^3$ MW.d/Mg $\leq B_T \leq 50\mathrm{x}10^3$ MW.d/Mg) and the costs - both economic and resource - are competitive; within the probable accuracy of the estimates given here all costs* are about the same. The loading ratios are in an acceptable range and the feed rate ratios are not onerous. A reactor run on such a cycle would probably have an enrichment of ε=1.8%±.3%, y=.2±.05, B_T=40±10 GW.d/Mg and a fissile production ratio PR^{fiss}=.25±.1. The instantaneous loading ratio would be N_T/N_u=.7±.3.

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*It is worth noting that for a natural uranium CANDU reactor, fuel supply costs (cfs)typically represent 14-18% of the total unit energy cost.

Inside this envelope of parameters all costs would be competitive with both natural uranium and LEU cycles, with the advantage that U233 would be created at little or no extra cost.

It remains to invent a practical fuel management strategy that embodies such cycles consistent with sound engineering principles. As an example, a reactor employing an 8-bundle shift where three of the eight bundles are thorium would have $N_T/N_u=.71$, and $y=.15$ if the irradiated thorium emerging from each channel were re-irradiated[11] three times, accompanied by fresh uranium bundles. An illustrative diagram is given in fig. 9. This strategy has the advantage of simplicity and allows the dispersion of the thorium and uranium bundles so as to hopefully smooth local power peaks or dips. In addition, only 15% of the thorium (and therefore non-reactive) bundles are ever simultaneously fresh, with the intent of reducing power perturbations due to on-power refuelling. An alternative having a similar set of parameters would consist of an array of 9 channels consisting of 4 channels always loaded with thorium and 5 containing LEU fuel. The uranium fuelled channels would be refuelled about 4-5 times more frequently than the thorium channels. If the thorium channel loadings were staggered, this would reduce the refuelling surge, but the simultaneous introduction of 12 pure thorium bundles (one channel) could still be difficult. Future work should focus on investigating such problems.

To summarize, the objective of this report was to demonstrate the existence of competitive once-through thorium fuel cycles on general principles of neutron conservation and simple economics. This has been done. If the parameters of the once-through cycle are judiciously chosen to lie inside an envelope of reasonable values, it is possible to create U233 at no additional cost. The main innovation[3] is the necessity of evoking dual fuel cycles whereby thorium and uranium fuels are physically segregated, and the main conclusion is that if LEU cycles are justified in CANDU economically or on the grounds of resource conservation, so is the use of once-through thorium.

Future research effort is needed. Specifically, experimental irradiations of ThO_2 need to be performed under reactor operating conditions to establish practicality limits[10] on thorium burnup (B_T). Obviously fuel management remains a stumbling block and must be studied in more detail. The economics and physics used in this survey were simplistic and should be repeated with more realistic models to obtain a better grasp of the optimal values of the operating parameters and their sensitivities to different economic costs. The impact of once-through thorium cycles on the growth of nuclear power systems must be studied and compared to alternatives. And finally, the physics of high burnup fuel and highly heterogeneous reactor cores will require investigation.

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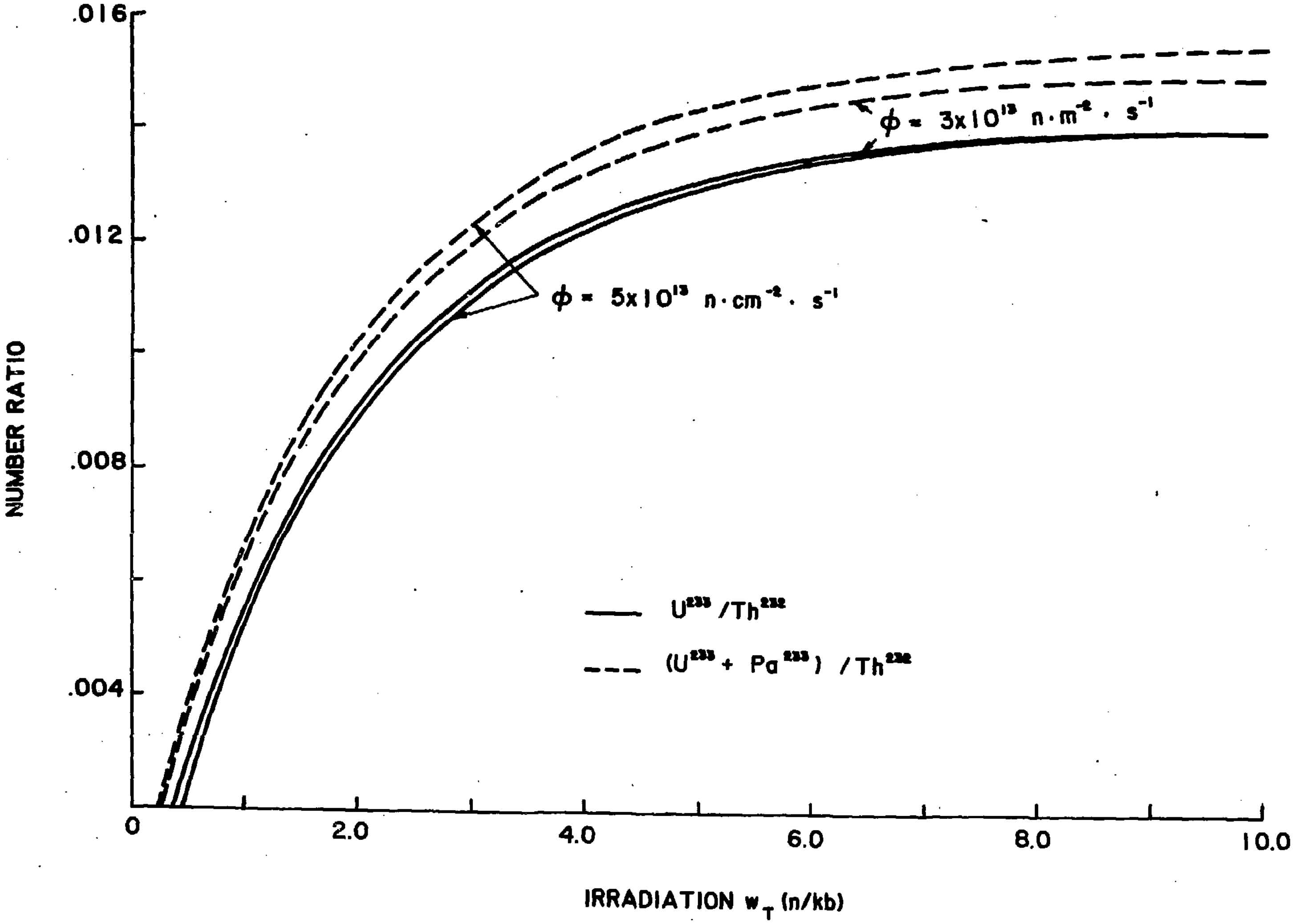
FIGURE 1: BUILDUP OF U^{233} VS. THORIUM IRRADIATION (w_T)



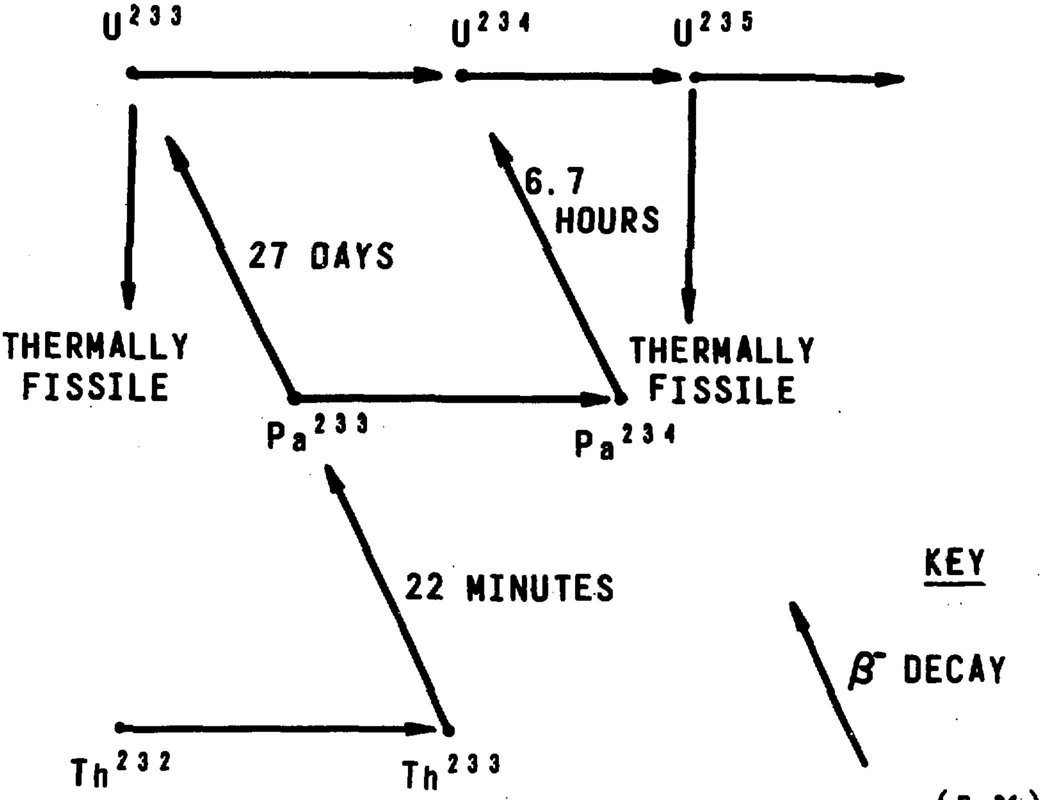


Figure 2 - Transmutation chain leading to the creation of U233 from Th232.

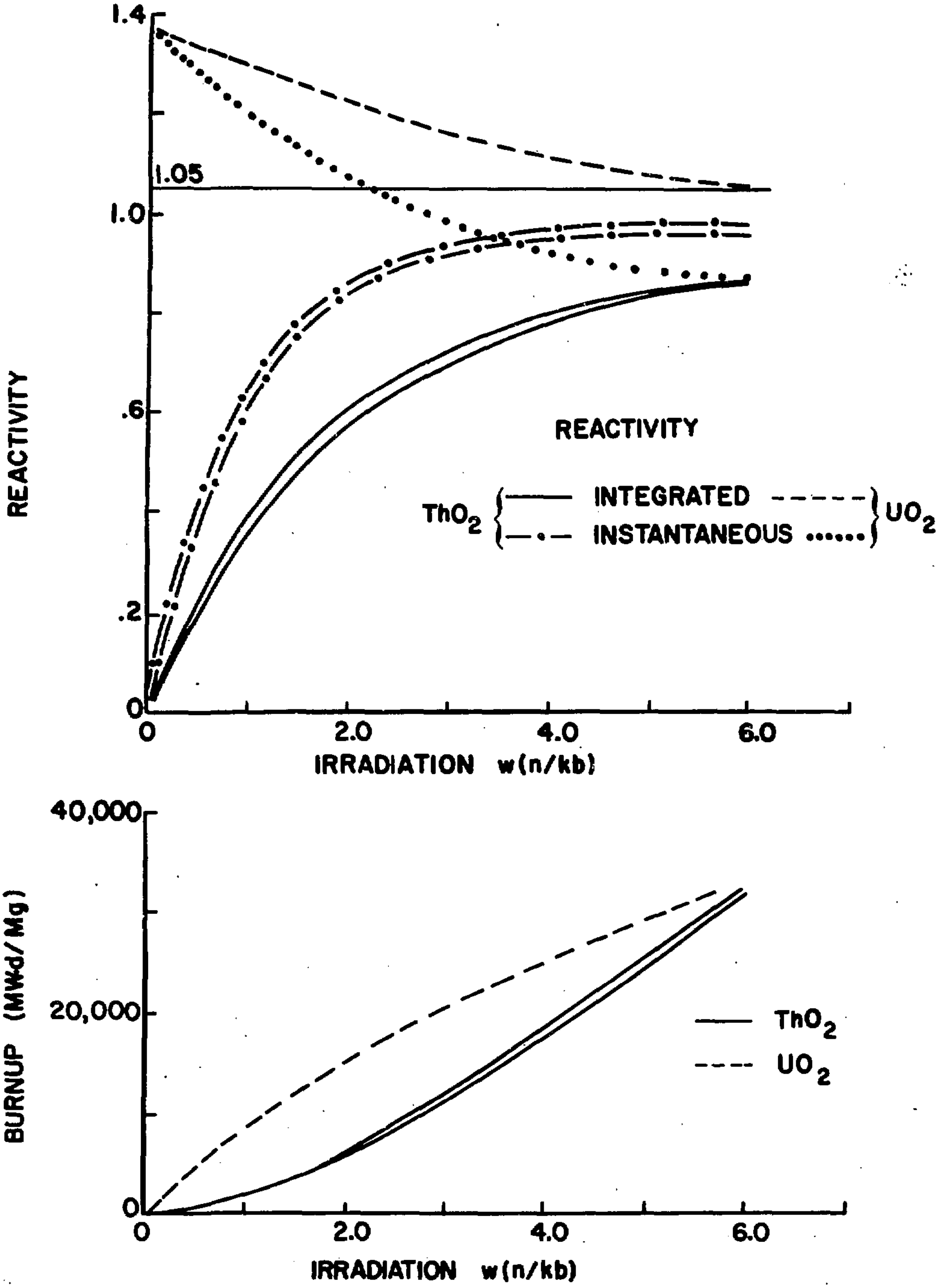


Figure 3 - Burnup, Integrated and Instantaneous Reactivity versus irradiation for a ThO_2 and 1.8% enriched UO_2 fuel bundle. In the case of the thorium bundle, the upper and lower curves refer to a flux level of 3 or 5×10^{13} $n.cm^{-2}.s^{-1}$ respectively.

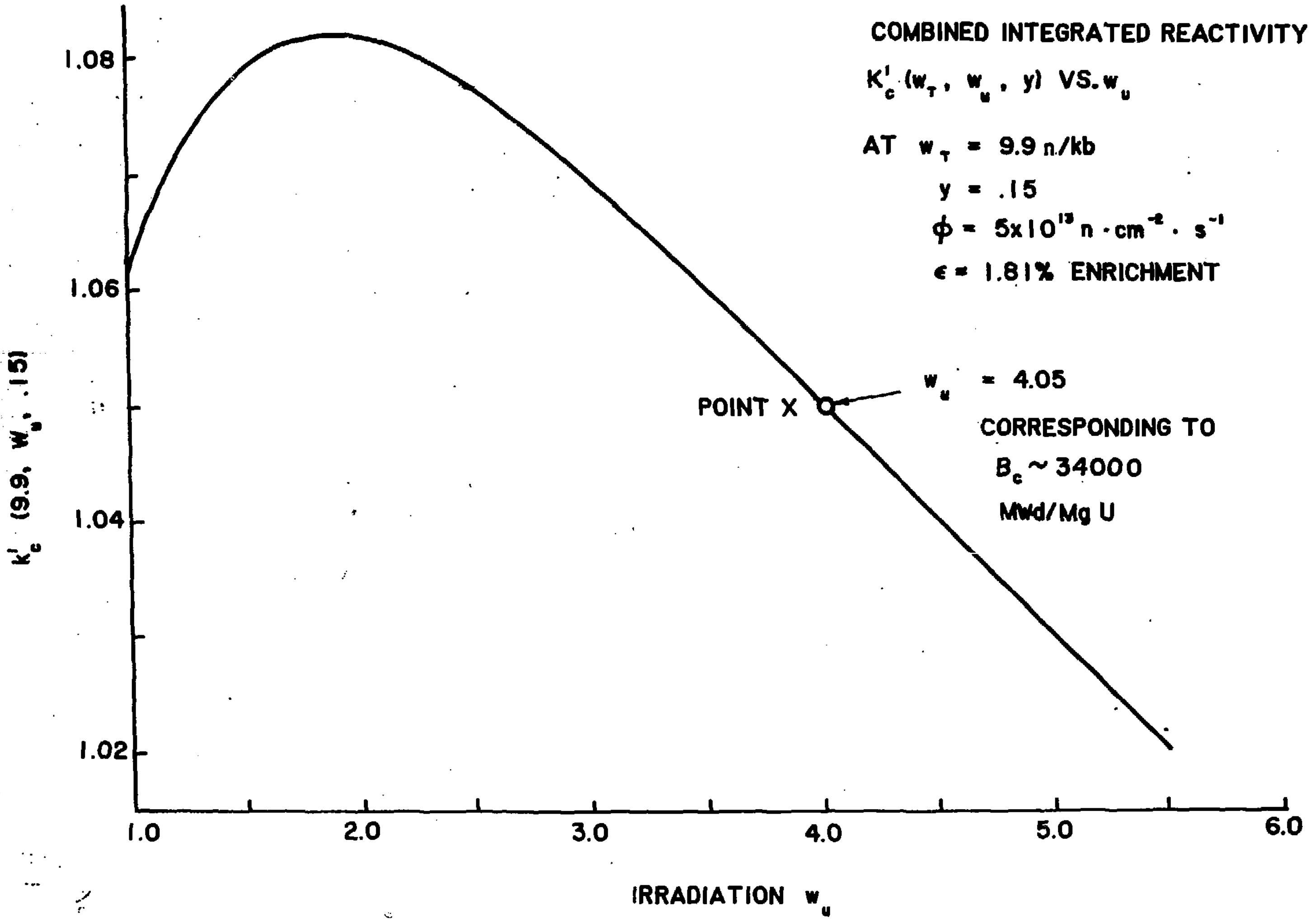


Figure 4 - Combined integrated reactivity for one possible dual fuel cycle.

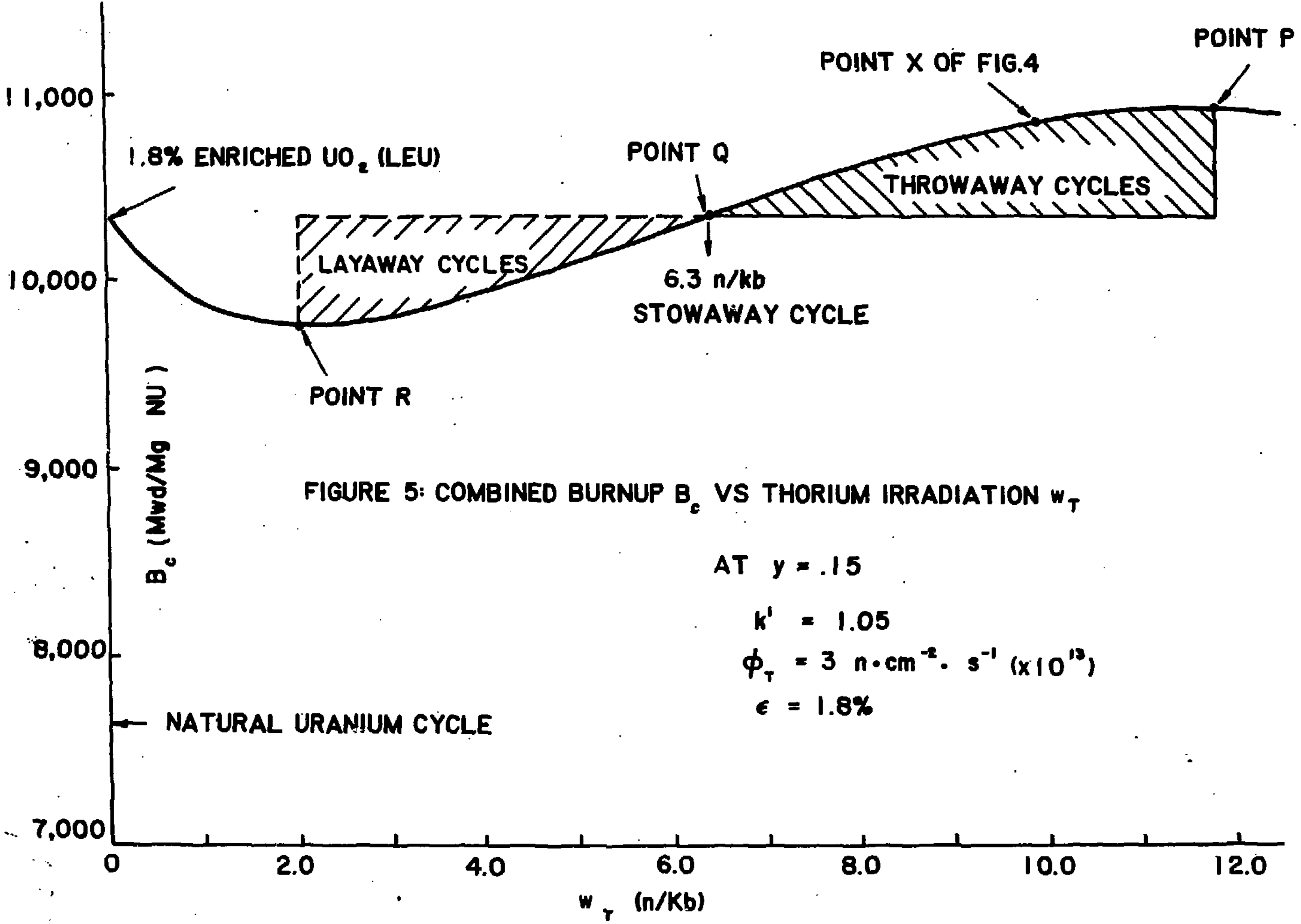


Figure 5 - Combined burnup as a function of thorium irradiation for several dual fuel cycles, illustrating the different kinds of cycles.

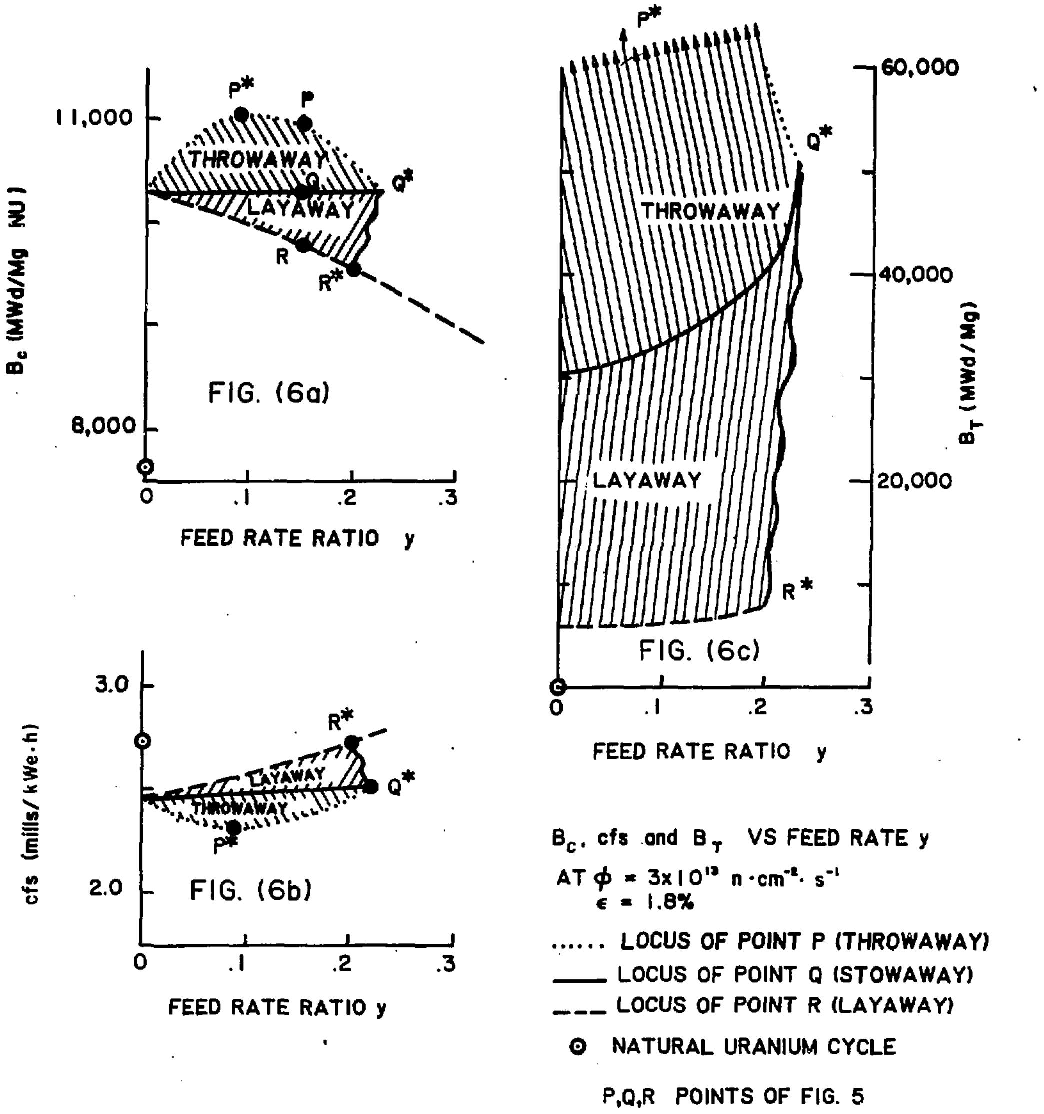


Figure 6 - Cost of fuel supply, thorium (B_T) and combined (B_c) burnup for a once-through cycle showing optimal choices of feed rate ratio y at one enrichment.

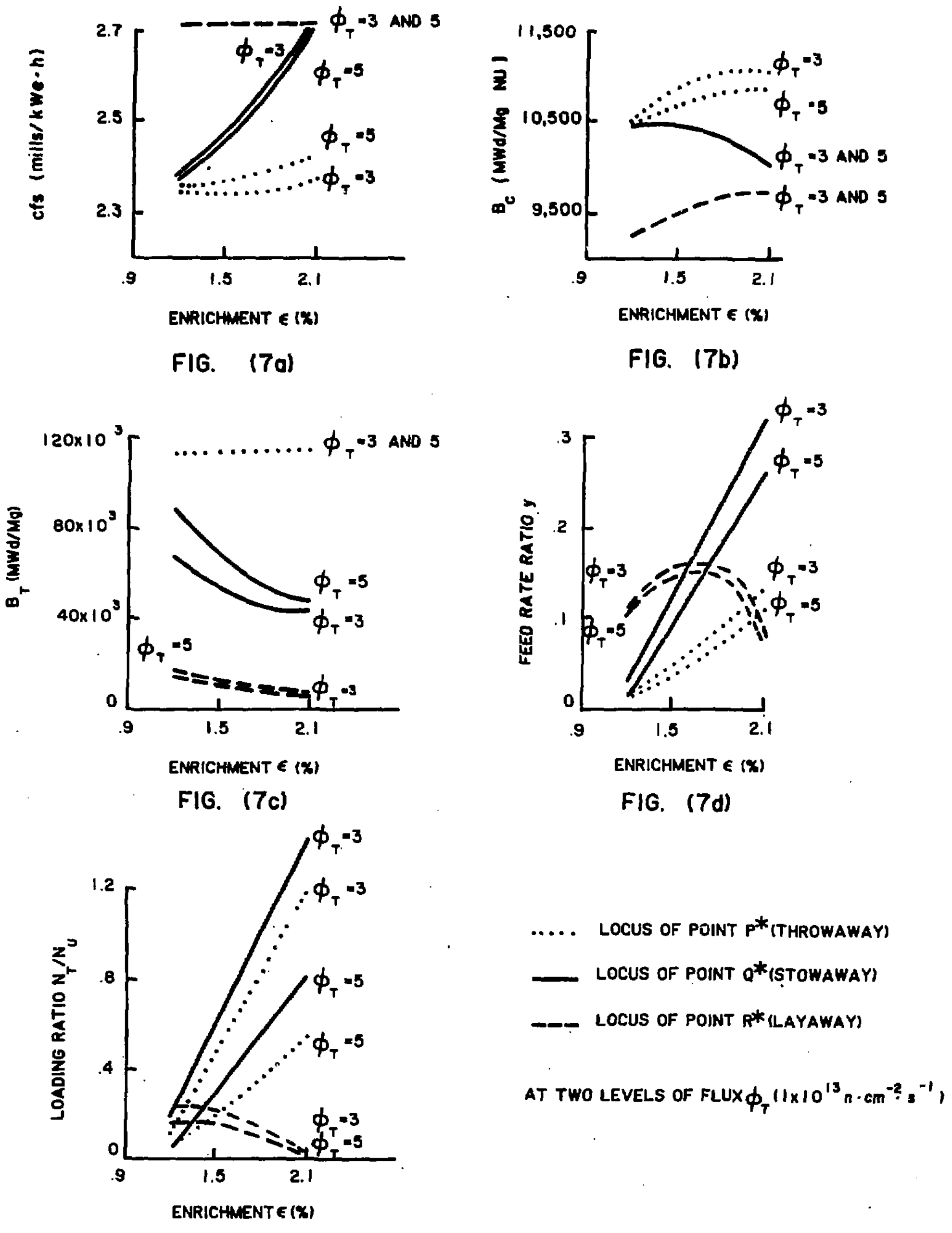


FIG. (7a)

FIG. (7b)

FIG. (7c)

FIG. (7d)

FIG. (7e)

Figure 7 - Important parameters of optimal once-through thorium cycles at different levels of enrichment and flux.

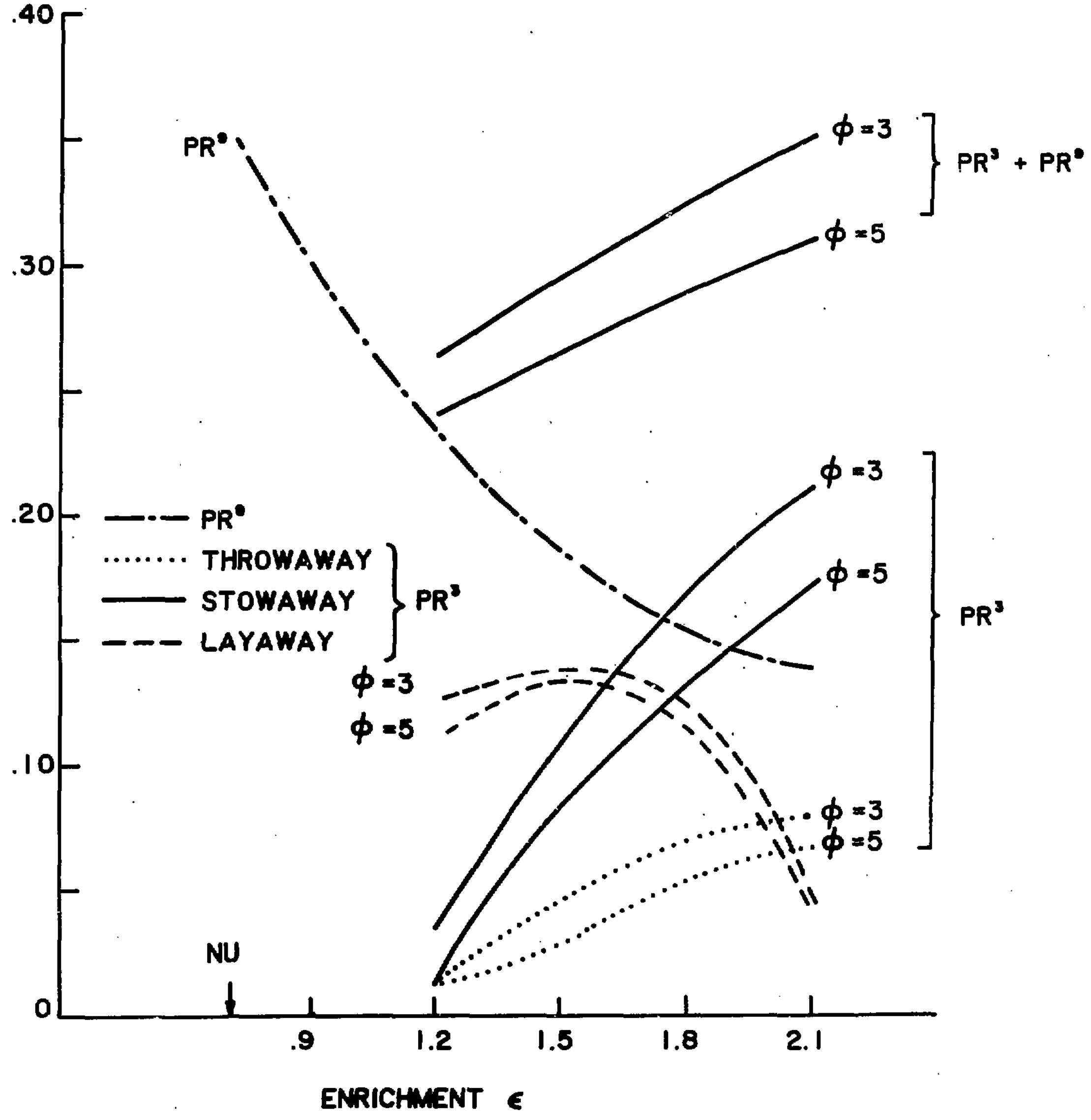


Figure 8 - Production ratio versus enrichment for optimal once-through thorium cycles.

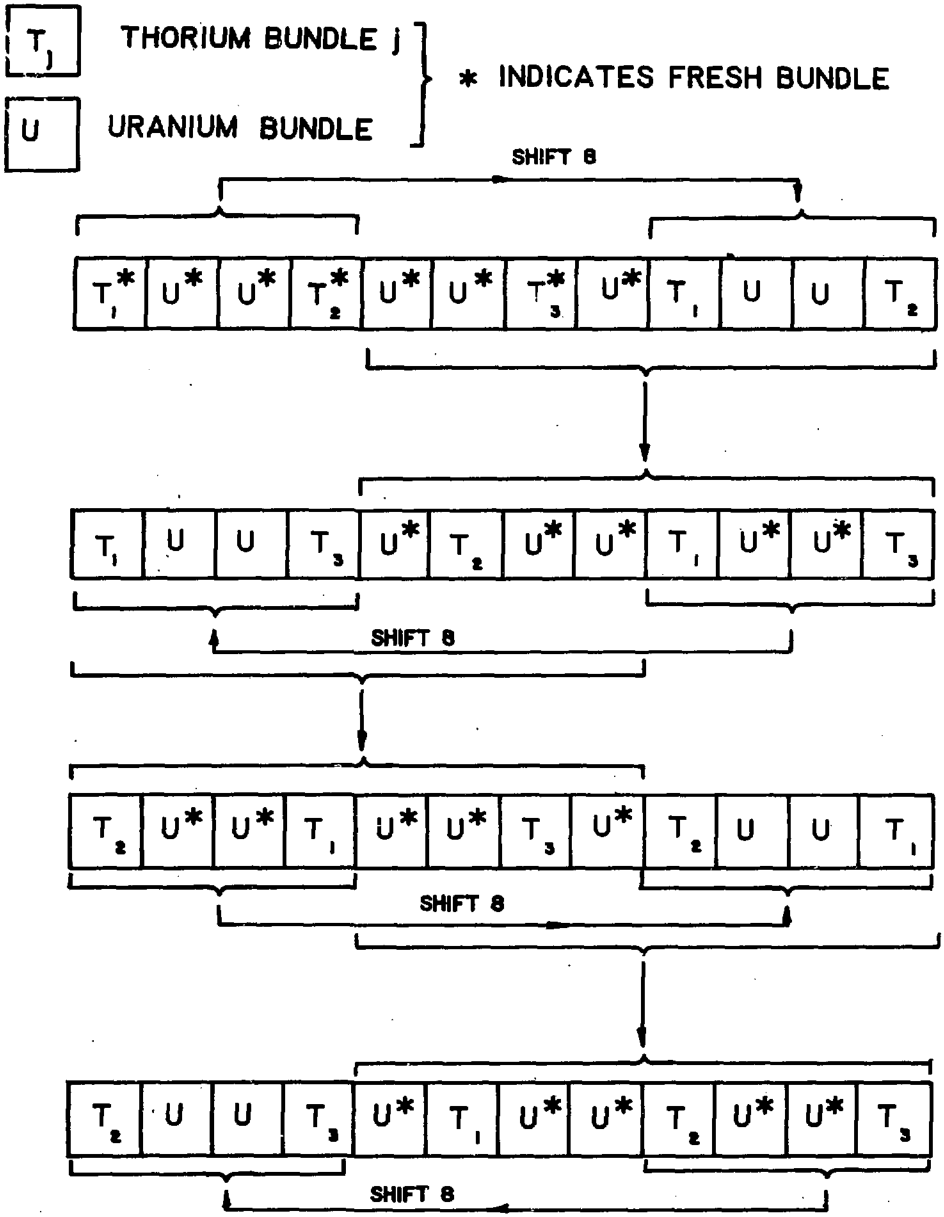


FIGURE 9: 8-BUNDLE SHIFT AND IRRADIATION HISTORY OF THREE FRESH THORIUM BUNDLES IN 4 CHANNELS

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