THE INDIAN PERSPECTIVE ON THORIUM FUEL CYCLES*

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Abstract. India is a country with limited deposits of uranium and large deposits of thorium. This fact ensures that India's nuclear power program cannot be totally uranium based the way it today is in most countries that have gone for nuclear power. Therefore, almost from the beginning of India nuclear power program, some effort has always been expended towards developing the technology of the thorium cycle. This has included fuel cycle studies, technology development, inpile irradiations and health physics aspects. A complete study of thorium cycles in various reactor types led to the conclusion that heavy water reactors were second only to molten salt reactors in this respect. HWR then, was the natural option for India. Currently India is working on the design of an advanced heavy water reactor (AHWR), specially designed with thorium in mind. In this paper, the results of a few interesting studies involving the recycling of plutonium with thorium in PHWR are presented.

1. INTRODUCTION

Of all countries that have ongoing nuclear power programmes, India is unique in being a country with limited deposits of uranium, but vast deposits of thorium. Unlike other countries therefore, India could not set thorium aside and settle for the uranium cycle. India followed a steady, even though low key, programme on thorium fuel cycle studies, technology development, and inpile irradiation. A complete study of thorium cycles in various reactor types led to the conclusion that heavy water reactors were second only to molten salt reactors in this respect. HWR then, was the natural option for India. Consequent to the decision to go for thorium cycles in heavy water reactors, the following things were done:

(1) 500 kg of thorium in the form of fuel bundles has been loaded in each of the two units of the Kakrapar Atomic Power Station (KAPS-1 & 2) for the purpose of initial power flattening. This scheme will be followed for all future PHWRs as well.
(2) There has been a fairly continuous programme of (irradiating thorium rods in the reflector of the research reactor CIRUS.
(3) A ^{233}U fuelled experimental reactor PURNIMA-II was commissioned at Trombay in 1984. Another system, called PURNIMA-III was made critical in 1990. This made use of the same fuel as was used in KAMINI at a later date.
(4) A research reactor KAMINI fuelled by ^{233}U -A1 alloy was commissioned in 1996. This was designed at BARC and built at Kalpakkam. It will be primarily used for neutron radiography on active components.
(5) Capability for reprocessing thorium to extract ^{233}U has been developed here and the ^{233}U used in PURNIMA and KAMINI was extracted here.
(6) On the fuel fabrication side work has been done on automatisation and remotisation which is needed for the highly active ^{233}U fuel. Already completed work includes the fabrication of thorium bundles for PHWRs, and the fabrication of thorium-plutonium mixed oxide fuel clusters.
(7) (Th, Pu) oxide fuel has been irradiated in the pressurised water loop of the CIRUS reactor to a burnup of 18000 MWD/T, and performed well without failure.
(8) Currently India is working on the design of an advanced heavy water reactor (AHWR), specially designed with thorium in mind.

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2. FUEL CYCLE STUDIES

Thorium cycles were analysed in all the extant thermal reactor types. These include:
a. Light water reactor;
b. High temperature gas cooled graphite reactor;
c. Molten salt breeder reactor;
d. Aqueous suspension heavy water reactor;
e. Open lattice pressure vessel heavy water reactor;
f. Pressurised heavy water reactor with pressure tubes;
g. Advanced heavy water reactor;
h. Source driven reactor.

All reactor types have their own special advantages, but the accent in our studies was on fuel utilization. From this perspective, it turns out that the best system (barring the source driven system) is the molten salt breeder reactor, with the heavy water reactors coming in second best. The molten salt technology being very different from what India is accustomed to, and the heavy water reactor being Indians chosen reactor type, the logical sequel was to continue the examination of thorium cycles in heavy water reactors. We shall return to the fuel cycles later on in this paper.

3. WORK DONE IN INDIA TOWARDS THORIUM UTILISATION

(i) Thorium has been used for initial power flattening in the PHWR for the first time in the Kakrapar Atomic Power Station (KAPS). This has been done in both units KAPS-1 & 2. About 500 kg of thorium in the form of 35 fuel bundles was used for this purpose in each unit. This also involved solving a somewhat intricate reactor physics problem [1] whereby these 35 thorium bundles could be distributed in the core in such a fashion as to achieve power flattening without adversely affecting the reactivity worth of the two shutdown systems in the reactor. These bundles have performed satisfactorily in the reactor. In unit-1, all thorium bundles have been discharged. It has been decided to follow this scheme in all future PHWRs.

(ii) A program of irradiating thorium rods in the reflector of the CIRUS reactor was started almost along with the commissioning of the reactor. This is being continued in a fairly sustained manner.

(iii) In 1984, an experimental reactor using ^{233}U as fuel commissioned in Trombay. This reactor was named PURNIMA-II [2]. It was a solution reactor in which ^{233}U in the form of uranyl nitrate was dissolved in water, and this solution was surrounded by a reflector of beryllium oxide. In 1990, a zero energy reactor using ^{233}U -A1 alloy plates as fuel was commissioned. It had nine fuel subassemblies and was named PURNIMA-III [3].

(iv) The 30 kW research reactor KAMINI [4] was built at Indira Gandhi Centre for Atomic Research (IGCAR), for neutron radiography, activation analysis, and radiation physics research. This used ^{233}U -A1 alloy plates as fuel. It had nine fuel subassemblies having a total of 72 U-A1 alloy fuel plates. This reactor was made critical for the first time on October 29, 1996.

(v) Two pilot scale facilities have been operated for the recovery of ^{233}U from thorium rods irradiated at CIRUS using a modified version of thorex process employing a 5% TBP extraction flowsheet.

The ^{233}U recovered from the above operations have been used to meet the fuel inventory requirements of Purnima- II, Purnime-III and Kamini. Efficient recovery and recycling of ^{233}U during the reactor experiments and from alloy scraps have played a crucial role in economic utilization of the ^{233}U resource. Periodic purification of ^{233}U to remove the daughter products of ^{233}U that emit high energy gamma radiations also forms a part of this effort. The expertise gained in all these domains will go a long way in the implementation of scaled up operations.

For the future, reprocessing of zircaloy clad thorium rods from PHWRs, which require chop-leach dissolution treatment, and the processing of thorium, uranium and Plutonium bearing experimental fuels are some of the tasks that need attention. In the long range, better dissolution techniques for thorium oxide, and an assessment of the impact of the presence of ^{231}Pa in thorex HLW and ^{228}Th in the separated thorium product would greatly enhance the viability of the back end processes.

(vi) Fabrication of high density sintered ThO_2 pellets for the ThO_2 bundles used for flux flattening of the initial core of PHWR is carried out by the conventional powder metallurgy technique of cold-compaction and high temperature sintering either under reducing or oxidizing atmosphere. ThO_2, being a perfectly stoichiometric compound, with a high melting point (-3400 °C) needs a sintering temperature of > 1800 °C for obtaining high density (>96%T.D.). Addition of 500-600 ppm of MgO can lower this temperature to 1650-1680 °C.

The following techniques have been tried for thorium based fuels: (a) Cold pressing of powder mixture of $(Th, Pu)O_2$ or $(Th/^{233}U)O_2$ followed by high temperature sintering, (b) vacuum impregnation of partially sintered low density (~ 70 - 80 % T.D.) ThO_2 pellets with uranyl nitrate or plutonium nitrate solution followed by drying and final sinterning, (c) Sol-gel derived microsphere pelletisation followed by sintering. The sol-gel microsphere pelletization process (SGMP), utilizes sol-gel derived dust-free and free flowing soft microspheres of $(Th, U)O_2$ in the size range of 100-600 microns, which are cold compacted and sintered to high density pellets just the way powder pellets are fabricated.

(vii) Fuel irradiation program is carried out in Pressurised Water Loop (PWL) of the 40 MW(th) research reactor, CIRUS. To study the behaviour of (Pu. Th) oxide fuel under high power and high burnup conditions, a six-pin assembly of (Pu, Th) oxide pins was installed in the PWL. The pressure and temperature conditions in the PWL were similar to those in the PHWR power reactor. The cluster was irradiated to a burnup of 18,500 MW·d/t, and its performance was satisfactory [5].

Subsequently, another cluster, which is a mixed cluster of (Th, Pu) oxide pins and (U, Pu) MOX pins has been loaded into the PWL. This has already seen about 10,000 MW·d/t burnup. At the time of writing, it is still inside the loop.

(viii) Currently, India is working on the design of an Advanced Heavy Water Reactor (AHWR). This reactor is a pressure tube kind of reactor [6], which is specially designed with the thorium cycle in mind. ^{233}U enrichment in thorium has been adjusted to be at the self-sustaining level. A discharge burnup of 20,000 MW·d/t is attained by using a certain amount of plutonium makeup. The plutonium is not mixed with the thorium, but is used in the form of (U, Pu) oxide pins. This has three advantages: (a) the plutonium pins can be placed wherever the spectrum is most advantageous to plutonium, (b) the discharge burnup of thorium pins can be adjusted independent of reactivity considerations, and (c) the thorium fuel remains uncontaminated by the long-lived actinides produced by the uranium cycle.

Since the thermal absorption of thorium is high as compared to uranium, the deleterious effects of parasitic absorption are less prominent in thorium systems. Thus it is possible to consider light water as coolant. The AHWR is cooled by boiling light water. The pressure tubes are vertical, and it is possible to have 100% heat removal by natural circulation, thus ensuring passive safety. The reactor has been designed to have negative void coefficient of reactivity.

4. FUEL CYCLE STUDIES IN THE PHWR

The most important thorium cycle is the self-sustaining equilibrium cycle (SSET). The salient features of our findings on this cycle were reported in the previous AGM and have been included in the report which is being presented to this AGM as working material [7]. As such, we do not repeat it here.

Next in importance is the thorium cycle without reprocessing. Figure 1 shows the findings from some studies carried out for a cycle in which thorium in combination with plutonium was burnt in a reactor to very high discharge irradiations. The plutonium considered was of an isotopic purity corresponding to the fuel discharged from the PHWR, and was about 75% fissile. Shown in this figure as a function of the discharge burnup of the fuel are two parameters. One is the fissile inventory ratio, which is the ratio between the fissile contents of the discharged fuel and the initial fuel. The other one we have called the fissile plutonium ratio, and it is the ratio of the fissile plutonium contained in the discharged fuel to that of the initial fuel. What is noteworthy here is the practically complete burning of the plutonium for even moderately high discharge burnups.

Figure 2 depicts a similar cycle with higher grade plutonium. This is almost pure ^{239}Pu, 96% fissile. Though we are not in a position to adopt this cycle, it has been analysed because of the current international interest in disposing of the plutonium that has become available from dismantled nuclear weapons. The independent variable is once again the discharge burnup of the thorium-plutonium fuel. Three quantities are plotted in this figure. One is the fissile plutonium in the initial fuel. Second is the fissile plutonium in the spent fuel, and the third is the fissile uranium, mainly ^{233}U, in the spent fuel. Once again the excellent efficacy of the cycle in burning plutonium is clear. A modest amount of ^{233}U gets built up.

Mixing plutonium with thorium contaminates the thorium with the long lived actinides of the uranium cycle, and thus damages one of the attractive features of the thorium cycle. In this context, Milgram's [8] once through thorium (OTT) cycle is of interest, although Milgram originally proposed it as an incentive for reluctant thorium users. This cycle keeps the uranium and thorium fuels apart, offers a measure of flexibility in that one can decide to start or discontinue the loading of thorium fuels at almost any time, permits the discharged thorium to be stored pending decisions about reprocessing or disposal, and does all this without any penalty in terms of either cost or fuel utilization in the event a decision is taken not to reprocess the thorium to recover ^{233}U. If extracted, the ^{233}U is thus obtained as a bonus.

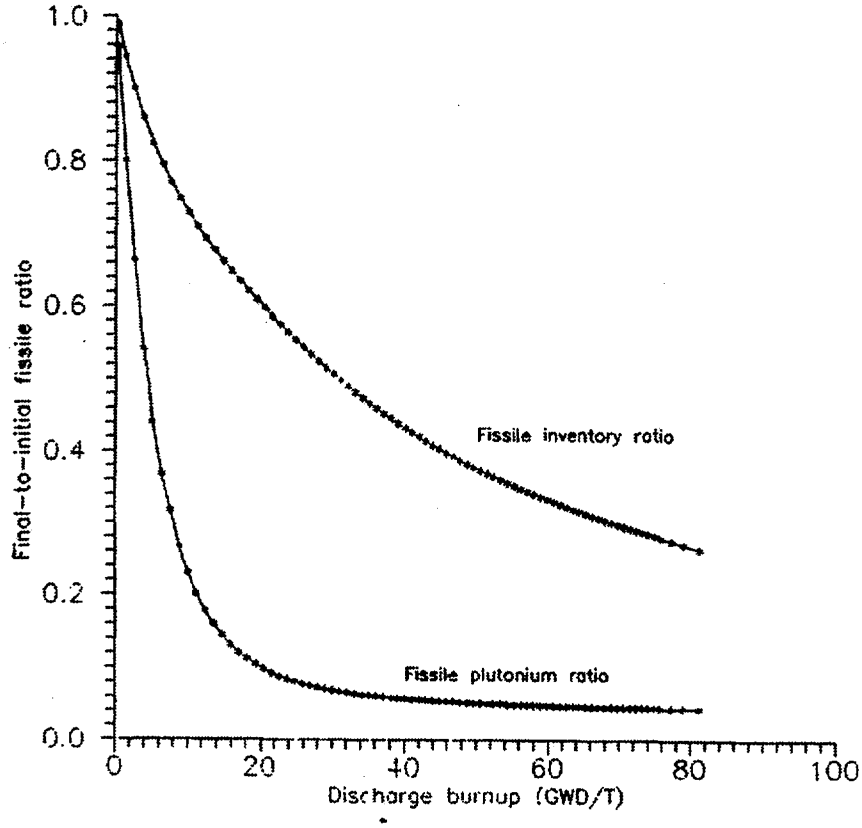

Figure 1. Thorium-plutonium in PHWR without reprocessing.

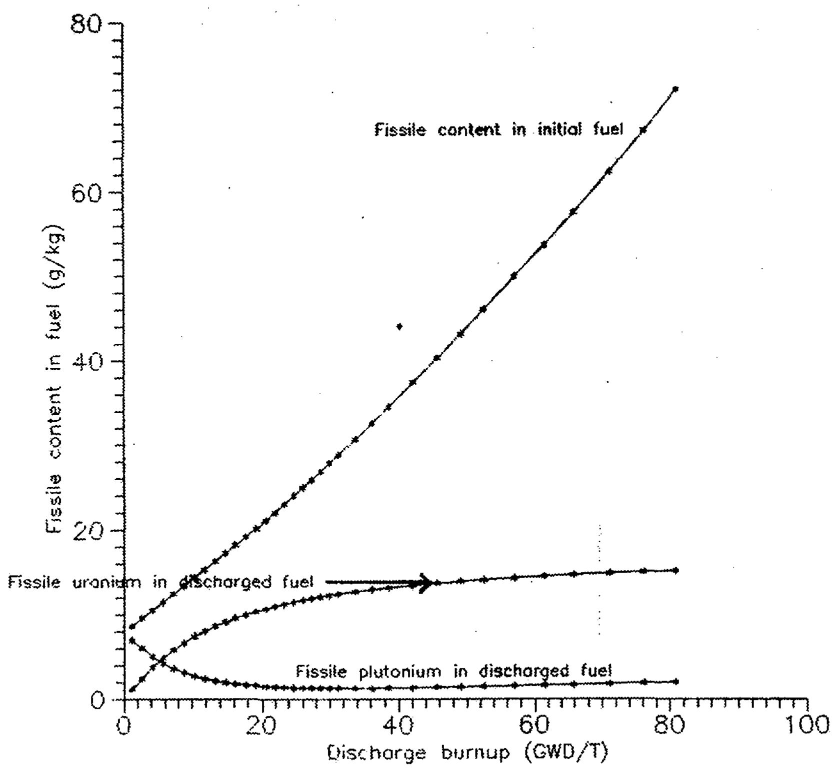

Figure 2. Fissile inventories in the thorium-plutonium cycle without reprocessing.

Figure 3 shows the essential feature of the OTT cycle. The central idea is to fuel the core with a combined loading of uranium fuel and plain thorium fuel. The thorium will act as a load on the uranium and decrease its discharge burnup. But as the thorium resides in the neutron flux, it will build up ^{233}U and begin to produce energy by ^{233}U fission. As the residence time of thorium increases, so will the penalty suffered by the burnup of uranium. However, if we express the energy obtained from a unit of uranium mined by taking credit for the power produced in thorium as well, we get the curve of Figure 3. As the residence time of the thorium increases, the energy from uranium mined decreases at first, but with growing production of energy from ^{233}U fission, the curve turns upward and climbs until it overtakes the value corresponding to the uranium fuel without thorium. It continues to climb, but finally turns downward again due to the accumulated fission products acting as a load. The figure also shows the discharge burnup of the uranium fuel, which decreases all the way. The gain in this cycle also depends upon the level of enrichment of the uranium fuel, the feed ratio of thorium to uranium fuel, and the flux level in the system.

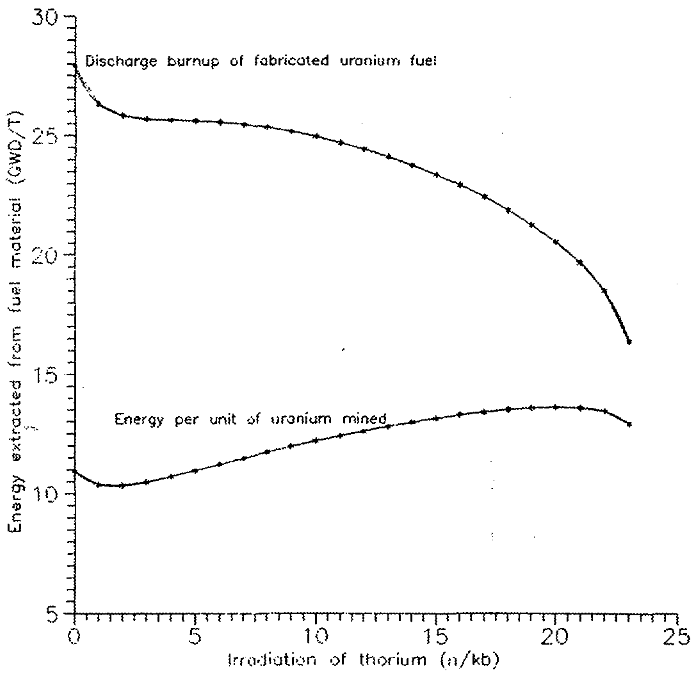

Figure 3. Energy from uranium mined in the OTT cycle.

Figure 4 shows a plot of the energy from mined uranium as a function of the enrichment of the uranium fuel. The cycle works best for low enrichments. Figure 5 shows the same quantity as a function of feed ratio of thorium fuel to uranium fuel. It would appear that a feed ratio in the vicinity of about 10% is most suitable. Figure 6 shows the sensitivity of the energy from uranium mined to the flux level at which the thorium fuel is operated.

An interesting extension of the OTT would be to follow the same kind of segregated loading as the OTT does, but at the same time, reprocess the discharged thorium to recover ^{233}U and feed the thorium fuel in the form of thorium-^{233}U fuel, keeping an enrichment level that would be self-sustaining in ^{233}U. Figure 7 shows the energy extracted as a function of the thorium-to-uranium feed ratio. This quantity increases as the feed ratio increases. For high feed ratios, the major part of the energy is actually coming from the thorium fuel.

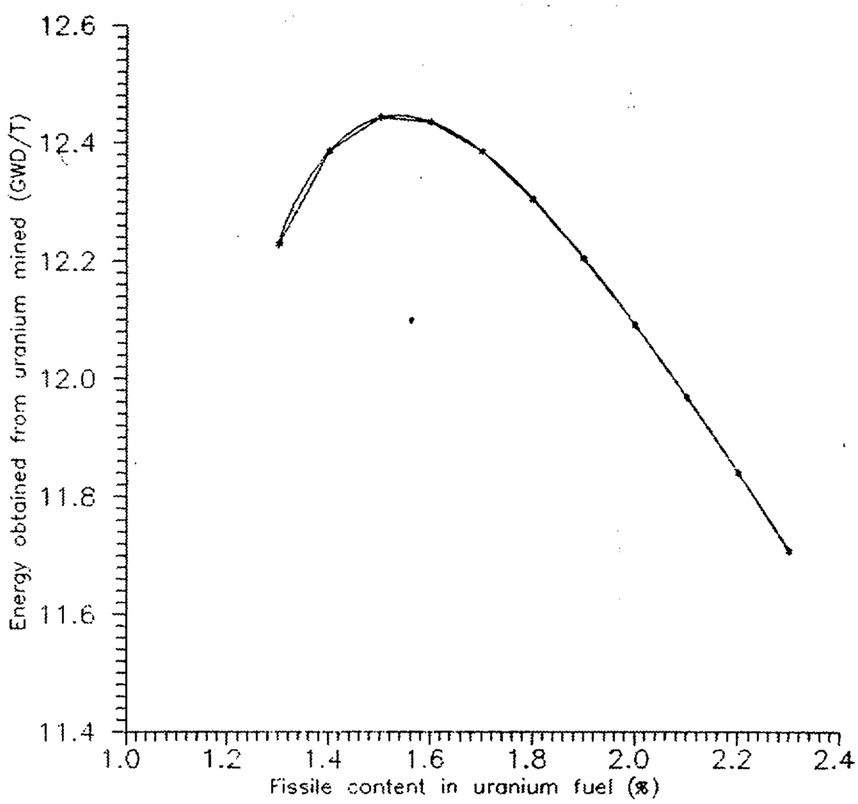

Figure 4. Effect of enrichment on the efficiency of the OTT cycle.

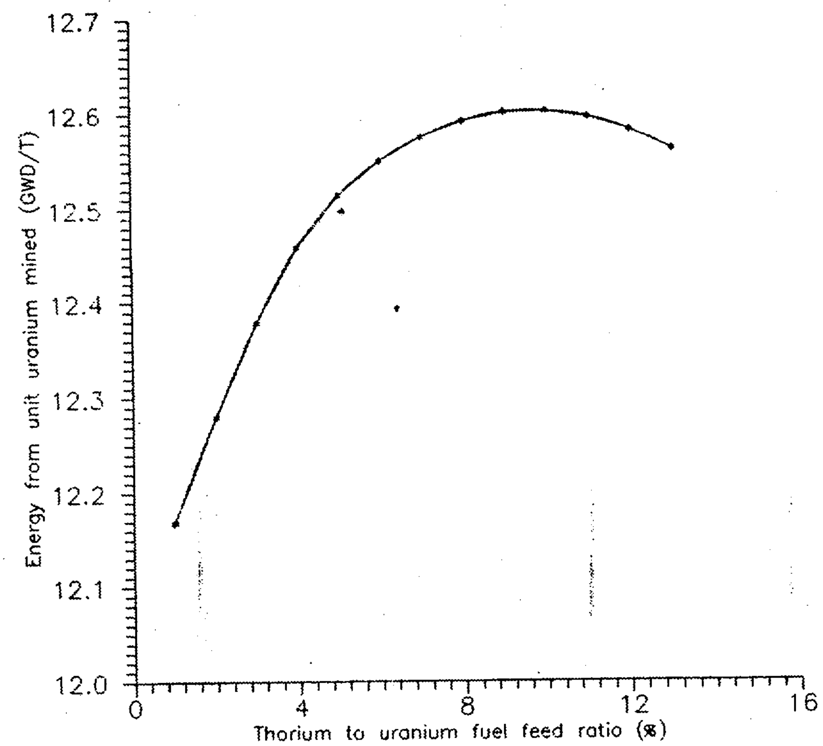

Figure 5. Dependence of fuel utilization on thorium-to-uranium feed ratio in the OTT cycle.

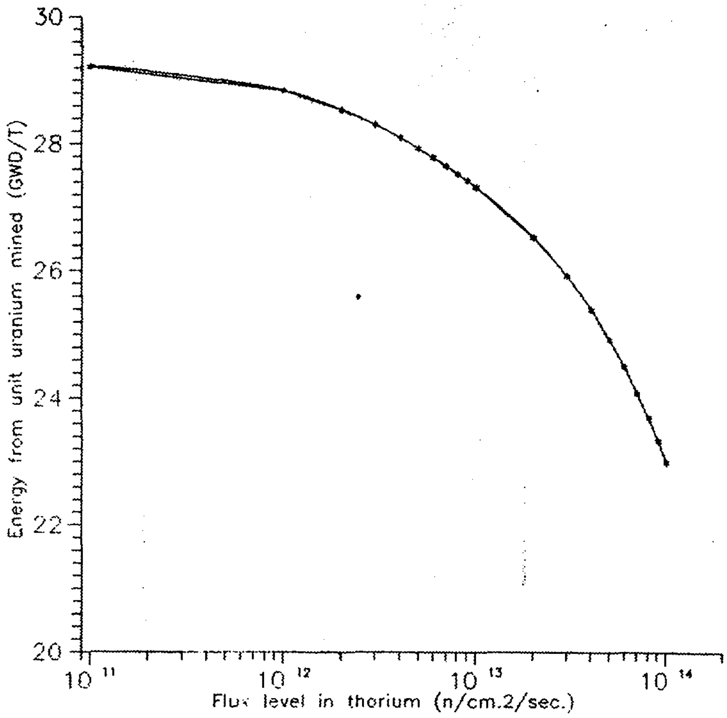

Figure 6. Influence of flux level in thorium on the fuel utilization in the OTT cycle.

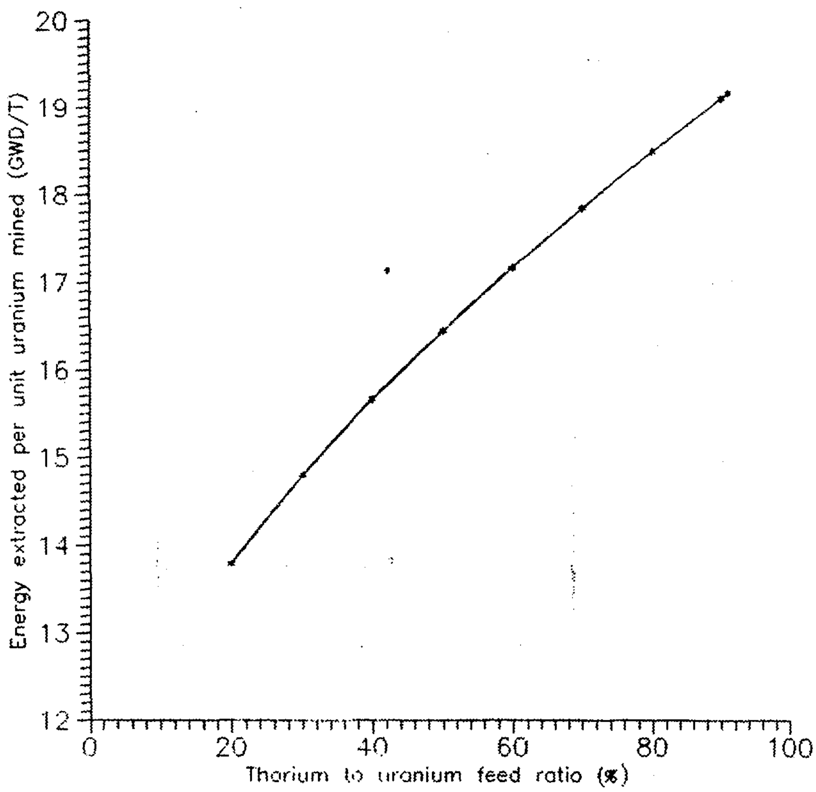

Figure 7. Segregated loading of uranium and thorium fuels with reprocessing of thorium and recycling of ^{233}U.

5. CONCLUSIONS

The versatile nature of thorium makes the thorium cycle very suitable for retrofitting into existing reactor designs. The most attractive feature of thorium lies in the nuclear properties of ^{233}U. Unlike ^{235}U and ^{239}Pu, the reactor physics performance characteristic of ^{233}U is almost spectrum independent. This property of being able to perform well in any spectrum gives thorium fuels great flexibility in the context of reactor systems designed for the uranium cycle.

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