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Energy from thorium

Introduction

Current generation commercial nuclear energy harnessing systems predominantly utilise uranium based nuclear fuel cycles[1]. In most of the industralised nuclear power producing countries, the nuclear fuel cycle employed currently is of the once-through[2] type, leading to wastage of precious energy resources. While a beginning has been made to exploit the larger energy potential in uranium through recycle of nuclear fuel by adopting MOX [3] (U-Pu) system, very little attention has been paid worldwide towards commercial utilisation of nuclear fuel cycles based on thorium. However, in the future, increasing energy demands and decreasing fuel resources may strongly dictate the use of thorium, a fertile material, since thorium is more abundant in earth's crust than uranium. The Indian uranium resources amount to only 78,000 te as against 5,18,000 te of thorium resources. Since naturally occurring thorium does not contain any fissile isotope as is the case with uranium, the use of thorium needs to start when there are sufficiently large uranium resources available. In the Indian context, therefore, to achieve long-term energy security, the use of thorium for large-scale commercial electricity generation, should begin much earlier than in most other countries. This paper presents a review of the various avenues of deployment for thorium as a fuel resource during the next several decades for large-scale power generation.

The three stages of Indian nuclear power programme

Out of the many naturally occurring elements, only a single isotope is fissionable. This is the isotope of uranium, uranium-235. The natural uranium, which forms the basis of the present nuclear industry, contains only 0.71% fissile U^{235}. The rest is U^{238}, which is a fertile material. It can absorb neutrons and through a series of nuclear reactions get converted to plutonium, which is a man-made fissionable element. Such materials, which can be made fissionable artificially, are called 'fertile' materials. Similarly, thorium-232, abundantly available in India, constituting almost one third of the world resources, is one such fertile material. On absorbing a neutron, thorium-232 finally yields uranium-233, which is another fissile material. Thus, in the first stage of a self-reliant nuclear power programme, one has no options but to start with natural uranium (or enriched uranium, in which the content of uranium-235 isotope is increased through an enrichment process). If one has an inexhaustible supply of uranium, probably nothing else needs to be done for continuing the programme indefinitely.

However, the available and currently viable uranium resources are not large enough for meeting future energy needs, in a sustainable manner, in this wasteful, 'once-through' fuel cycle. This fact is applicable for the entire world in a long-term time frame and for India in the shorter term itself. It is, therefore, essential to multiply the available fissionable material resources by progressively converting the fertile materials (uranium-238 and thorium-232) to fissionable materials in nuclear reactors, and use these man-made fissionable materials also, for progressively enhancing nuclear energy generation base.

This implies separation of these man-made fissionable materials from spent nuclear fuel, by reprocessing these fuels, and bringing these fissionable materials back as fuel, as a part of, what is called, a 'closed nuclear fuel cycle'. One must note that no other energy producing system is capable of expanding its own generation base, thus making nuclear energy inherently sustainable.

Keeping these facts and the goals, mentioned earlier, in mind, the Indian Nuclear Power Programme (Fig. 1) envisages the following three stages:

1. Natural uranium fuelled pressurised heavy water reactors
2. Fast breeder reactors utilising plutonium based fuel
3. Advanced nuclear power systems for utilisation of thorium

Here it must be mentioned that the three stages have considerable, and necessary, overlaps in the time frames of

1. A nuclear fuel cycle is the complete lifecycle of the fuel, right from its production to its final disposal as waste.
2. A once-through nuclear fuel cycle is one in which nuclear fuel is disposed off as waste upon its exit from the reactor after one time use.
3. Mixed oxide fuel.

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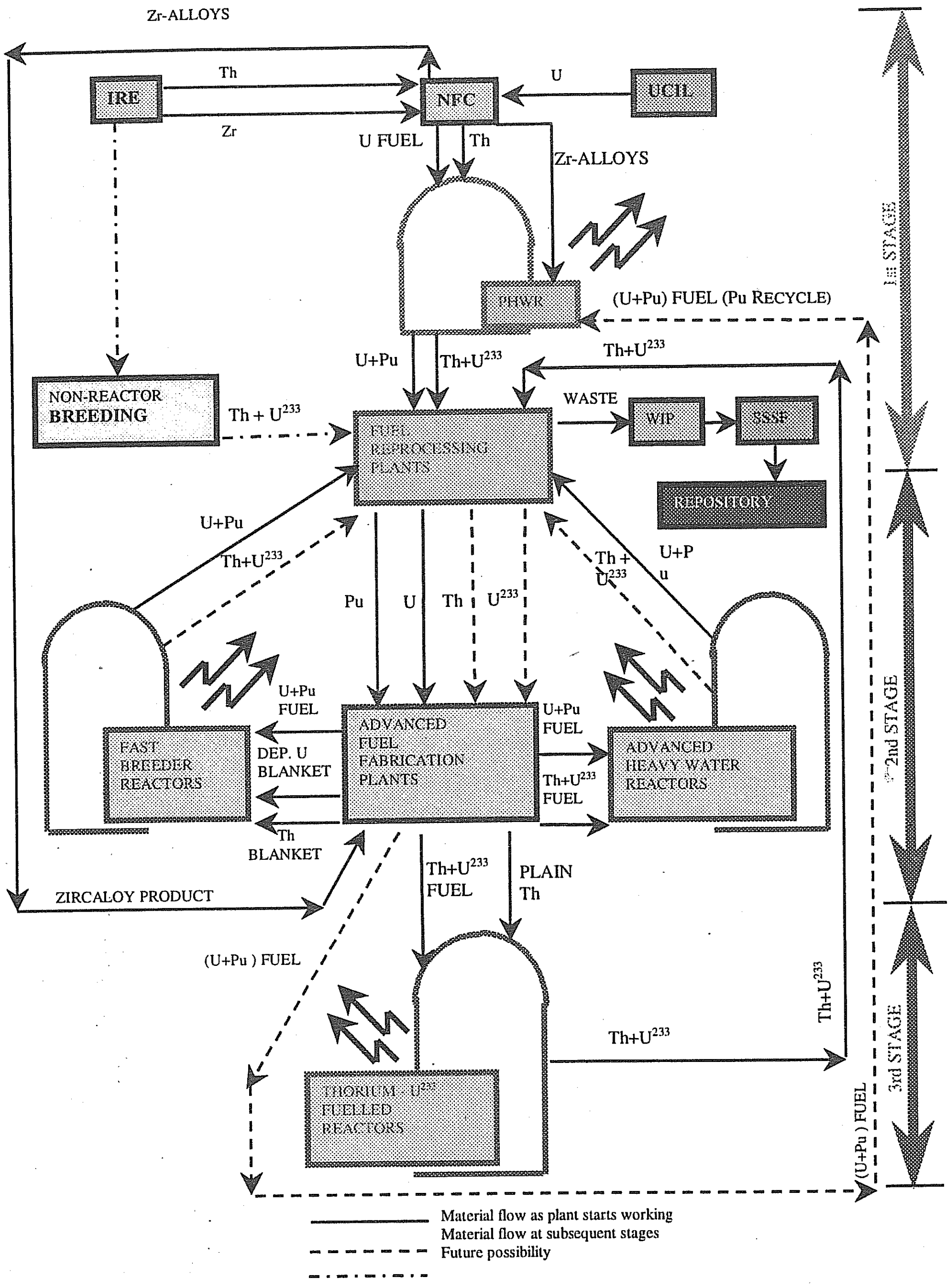


Fig.1 Three stages of the Indian Nuclear Power Programme

their development and deployment, It should be noted that though the three stages comprise the main stream of the Indian nuclear power programme for electricity generation, the programme has enough flexibility to accommodate some variants and additional elements, including light water reactors, for augmenting the nuclear power base as needed. In addition, flexibility also exists for use of nuclear energy for applications other than generation of grid-based electricity.

4. The probability of a nuclear reaction is quantified in terms of "cross section". It is measured in terms of "barns"; one barn is defined as 10^{-24} cm^2 – typical nuclear cross sectional area.

5. Resonance: In this energy range (resonance) the cross section of some nuclear reactions (mostly capture) suddenly raises fairly sharply, implying enhanced probability of capture at these energy levels.

Thorium utilisation

Thorium cycles are feasible in all the existing thermal reactors and fast reactors as well. In the short term, it should be possible to incorporate the thorium fuel cycle in existing reactors without necessitating major modifications in the engineered systems, reactor control and the reactivity devices. These options could run for the next 10-20 years of operation. For some of the fuel cycles described in international literature, only the reactor physics studies have been carried out and a lot of other technological developments are very much needed before these could be implemented.

Properties of Thorium

The interest in thorium is determined by a number of its advantages over its counterpart uranium:

1. Availability of raw materials: Worldwide the deposits of thorium exceed the uranium deposits by a factor of three [1].
2. Improved neutron balance in thorium reactors: Thorium as such does not contain any fissile material. However, if one looks at the absorption cross section[4] for thermal neutrons of Th^{232} and its counterpart U^{238} (7.4 barns vs. 2.7 barns), Th^{232} offers greater competition to capture the extra neutrons and as such a lower proportion of the extra neutrons will be lost to structural materials and other parasitic capture. That is to say that a higher amount of energy could be extracted from U^{235} fissile feed with Th^{232} as host than with U^{238} as host, owing to the higher neutron capture cross section of Th^{232} compared to U^{238}, both in thermal and resonance[5] region (Fig.2). So thorium is a better fertile material. It may be mentioned here that high discharge burnup is a prerequisite for realising this advantage. Such burnups are now well within the current day technologies.

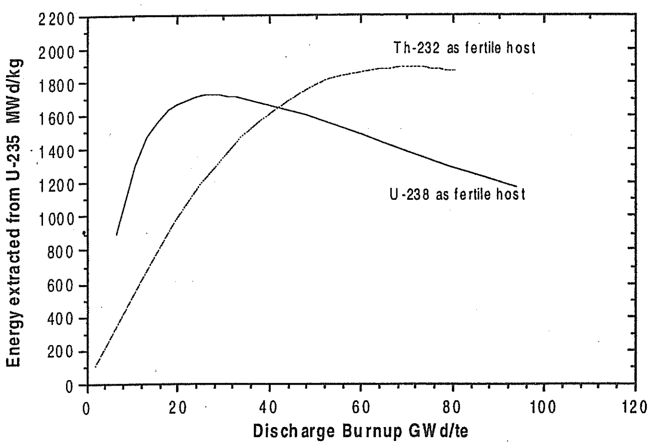


Fig.2 High burnup open cycle: Th-232 Vs U-238 as fertile host

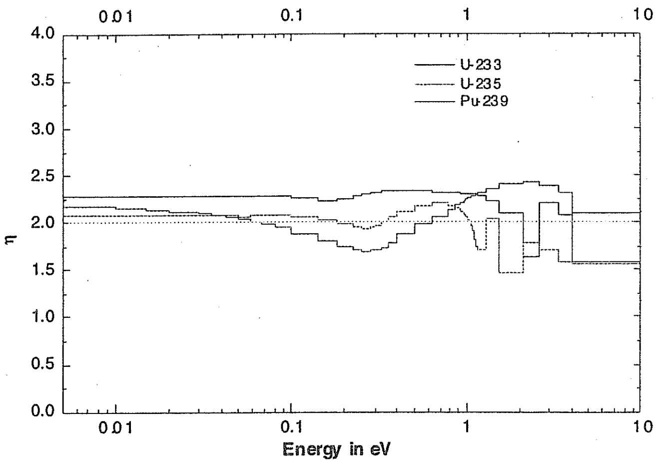


Fig.3 Variation of h for fissile isotopes

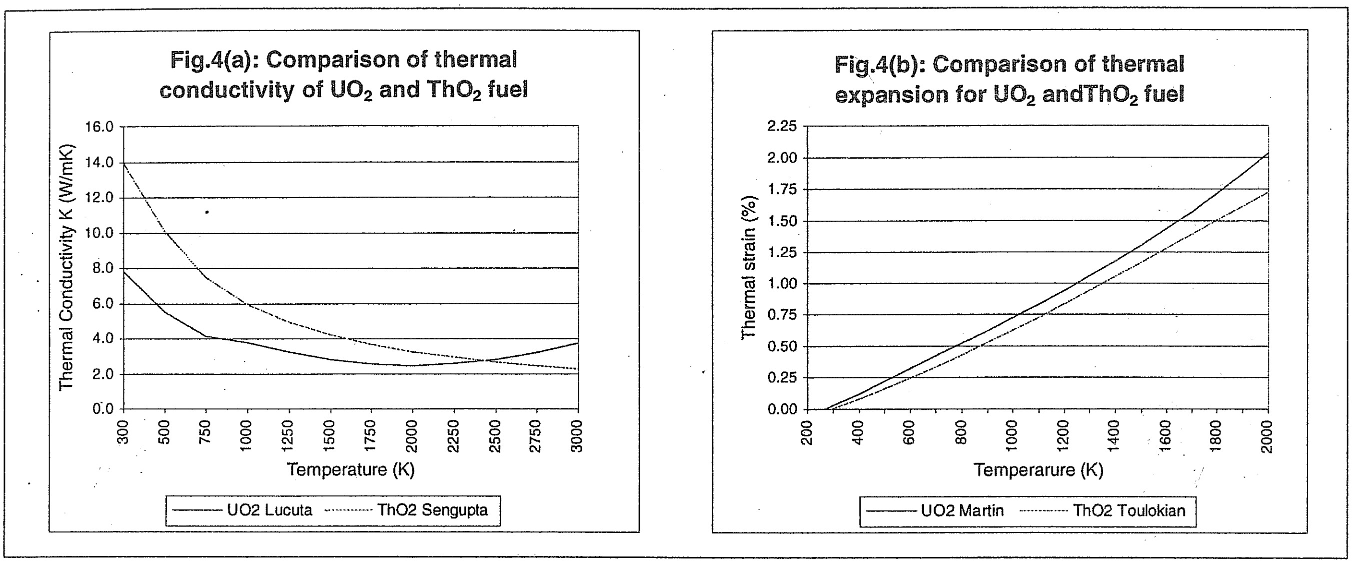


Fig.4 Thermal performance of thorium and uranium based fuel

3. High η^6/η value of bred fissile material U^{233}. U^{233} is the only fissile nuclide with η value greater than 2 per fission caused by thermal neutrons. Moreover the η value of U^{233} does not change much with the neutron energy as with U^{235} and Pu^{239} (Fig.3). This stable nature of the η value of U^{233} makes the thorium fuel cycle less sensitive to the type of reactor.
4. Improved technological properties and high radiation resistance.
5. Reduced quantity of long-lived minor actinides resulting from the fission has advantage in long-term waste management.

A comparison of the properties of thorium di-oxide (ThO_2) and uranium-di-oxide (UO_2) show ThO_2 to be superior from fuel performance point of view [2]. The thermal conductivity of ThO_2 is higher than UO_2 (Fig.4a). As a result fuel temperatures for thorium fuel will be lower than UO_2 fuel resulting in reduced fission gas release. The thermal expansion coefficient of ThO_2 is less as compared to UO_2 inducing less strain on the clad (Fig.4b). ThO_2 retains dimensional stability at high burn-ups. It is a very stable oxide and does not oxidise resulting in less release of fission products in the coolant in the event of a clad breach. The fission product release rates for thoria-based fuels (ThO_2) are one order of magnitude lower than that of UO_2. Fuel deterioration is slow allowing the fuel to reside in the reactor for longer periods of operation before significant deterioration occurs.

6. Eta (η) defines the number of fast neutrons emitted in a fission process per neutron absorbed by the fissile nuclei; one neutron is required to continue the chain reaction.
7. Pressurised heavy water reactor moderated and cooled nuclear reactor.

Utilisation of Thorium in Existing Reactors

The thorium fuel cycles in the near term can be implemented in the existing reactors without any change in the reactor hardware. The design changes are expected to be in the fuel. There are several possible options for utilisation of thorium in reactor systems of existing designs, as described in the following paragraphs.

Pressurised heavy water reactors[7] (PHWR)

Fuel cycle studies [3]

The thorium fuel cycles can be incorporated into PHWR in three different cycles mainly the self-sustaining equilibrium thorium cycle (SSET), the high burn-up open cycles and the once through thorium cycle (OTT).

(a) In SSET cycle, the fuel is thorium-U^{233}. As the name suggests in SSET, the fuel U^{233} is contained in the discharged fuel in such quantities so as to provide the charge for the next fuel cycle. Once initiated, this cycle does not require any external fissile input. However, SSET has to be initiated by adding fissile input either in the form of U^{235} or Pu to the thorium, extracting U^{233} from the spent fuel and then using it for the first charge of SSET. The discharge burn-up with this cycle is low (about 11000 MWd/te). The cycle also depends heavily on reprocessing of thorium to chemically separate U^{233} and re-fabrication of U^{233} containing U^{232}. Once this technology is well developed, either through advances in robotics or through separation of U^{233} and U^{232}, the cycle may become attractive.

(b) In the high burn-up open cycle, the intent is to burn thorium to a high burn-up (about 50,000 MWd/te). In the process, the spent fuel inventory is reduced to a large extent. The fuel utilisation is lower in this cycle. The fuel utilisation could be improved by reprocessing the spent fuel to obtain U^{233}. The fissile enrichment needed could be in the form of either U^{235} or Pu.

(c) The OTT cycle uses plain thorium along with Slightly Enriched Uranium (SEU) or Mixed Oxide (MOX) in segregated channels of PHWR and allowing the thorium to reside in the reactor for a longer period of time. Thorium acts as a load causing the SEU/MOX to be discharged at a lower burn-up. The energy extracted from this cycle is the sum of energy obtained from thorium and SEU/MOX. This cycle is favourable for long residence time of thorium fuel.

Engineering studies

The fuel cycle studies of the past are slowly maturing to studies on thorium utilisation in specific reactors for better understanding and evaluation of the reactor specific problems, changes in fuel design, fuel fabrication, engineering development, safety implications and back end of the fuel cycle.

Presently thoria bundles are used in Indian PHWRs for achieving the initial flux flattening in the core. This represents a unique way of utilising thorium without any loss of burn-up in UO_2 fuel. The ThO_2 bundles have been irradiated in Kakrapar-1 & 2 (70 bundles) and Rajasthan-2, 3 & 4 (88 bundles), Kalpakkam (4 bundles) and Kaiga-1 & 2 (70 bundles) [2]. These bundles will be highly useful in development and demonstration of reprocessing of ThO_2 bundles irradiated in power reactors and will provide U^{233} for the development of fabrication technology and irradiation experiments.

An intermediate in the formation of U^{233} is Protactinium (Pa^{233}), which deteriorates the neutron balance and causes some difficulties in the reactor control. The Pa^{233} absorbs neutrons during reactor operation. It has a half-life of 27 days and undergoes decay to form U^{233}. During a prolonged shutdown, there is a build up of fissile U^{233} due to decay of Pa^{233}, increasing the reactivity of the fuel. The design of control system has to take this factor into account.

Reprocessing and refabrication [4]

Thorium, a fertile material, on absorption of a neutron in the reactor forms U^{233}. The thorium dioxide is very stable. This poses difficulty in dissolution of thorium during reprocessing. Stronger reagents and better material of construction are required to circumvent this problem. Addition of MgO to thorium dioxide during fabrication also facilitates dissolution. Thorex process is used for reprocessing of thorium dioxide, which is similar to Purex process followed for reprocessing of uranium. FUS, a plant for separation of uranium from thorium is coming up at BARC, Trombay. Fig.5 shows operation in glove boxes in FUS.

Another isotope U^{232} is always associated with U^{233}. It is formed by (n,2n) reaction. The half-life of U^{232} is 73.6 years. The daughter products of U^{232} are gamma emitters (>2 MeV) with small half-lives. As a result the dose rate increases with time. Hence, conventional powder metallurgy process of fabrication of fuel cannot be adopted for U^{233} fuel. Alternate processes that are dust free and amenable to automation and remotisation like solgel, pellet-impregnation techniques are being evaluated for fabrication of thoria-U^{233} fuel. Developmental work on various fabrication processes is carried out at Advanced Fuel Fabrication Facility (AFFF), Tarapur. Fig.6 shows some of the products fabricated at AFFF – annular pellet and granules produced by advanced agglomeration technique.

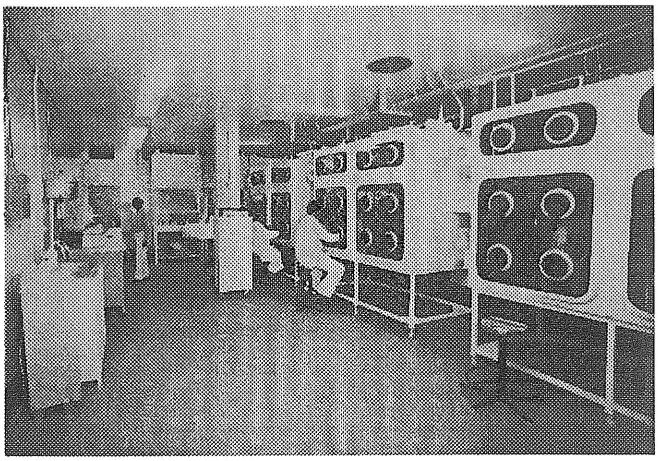
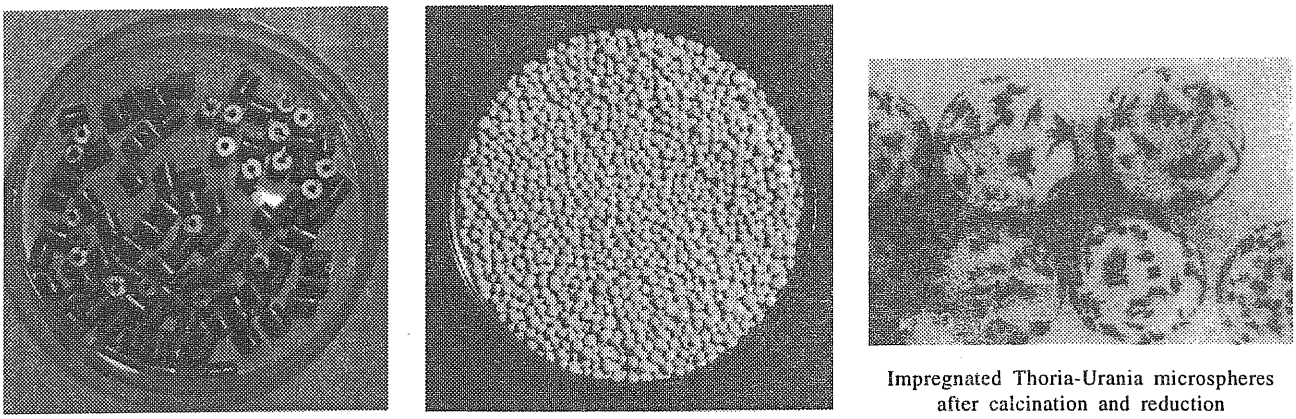


Fig.5 Facility for Uranium-233 separation (FUS)



Annular pellet

Granules by advanced agglomeration technique

Impregnated Thoria-Urania microspheres after calcination and reduction

Fig.6 Developmental work at advanced fuel fabrication facility (AFFF)

Advanced heavy water reactor (AHWR) [5]

Breeding in Th-^{233}U cycle has been actually demonstrated in the seed and blanket type core in light water reactor at Shippingport, USA. Attractive characteristics of thorium were also seen in high temperature gas cooled reactors. Several innovative fuel cycles have been worked out with thorium for light and heavy water reactors. The possibility of considerable energy that can be extracted through ^{233}U-generated in-situ, when thorium fuel is taken to high burn-up, has been a common feature in most of these fuel cycles. The design of advanced heavy water reactor, shown in Fig.7, is based on a requirement to derive maximum benefit from the neutronic characteristics of thorium recognised above, in the context of the Indian nuclear power programme.

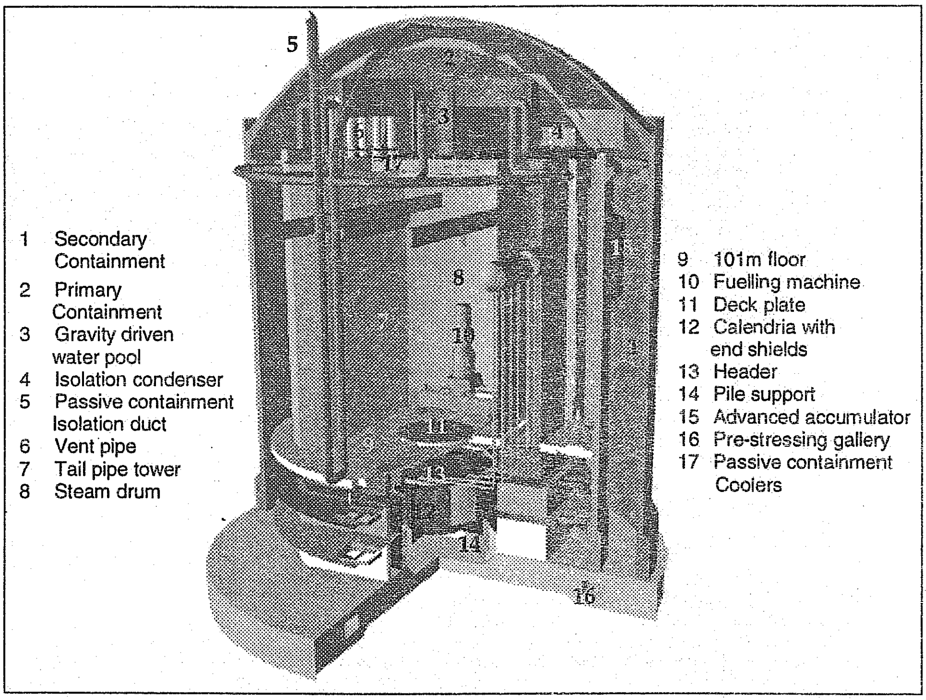


Fig.7 Advanced heavy water reactor

Thorium being a very strong absorber of thermal neutrons, it is clear that in a thorium based system, thermal absorption in coolant, moderator and structural material would be relatively lower. Management of heavy water coolant in PHWRs has always been a problem due to frequent leakages leading to a requirement to manage tritium activity, and associated economic penalty. In a thorium system, it is therefore logical to change the coolant to light water while retaining heavy water as moderator for better neutron economy. With improvement in void reactivity in such pressure tube type of reactor, heat removal in boiling mode, thereby effecting a reduction of about 50% in the overall quantity of coolant and improvement in steam cycle efficiency becomes attractive. This results into a vertical pressure tube type reactor with boiling light water as coolant and heavy water as moderator.

Yet, for an evolutionary design to be acceptable the overall cost of the system must be decreased and at the same time safety should be enhanced. In a low power density core needed to facilitate improved conversion, boiling coolant offers an attractive option for natural circulation. The general arrangement of flow paths in various reactor systems of AHWR is shown in Fig. 8.

It is well known that, in a PHWR, operating with self-

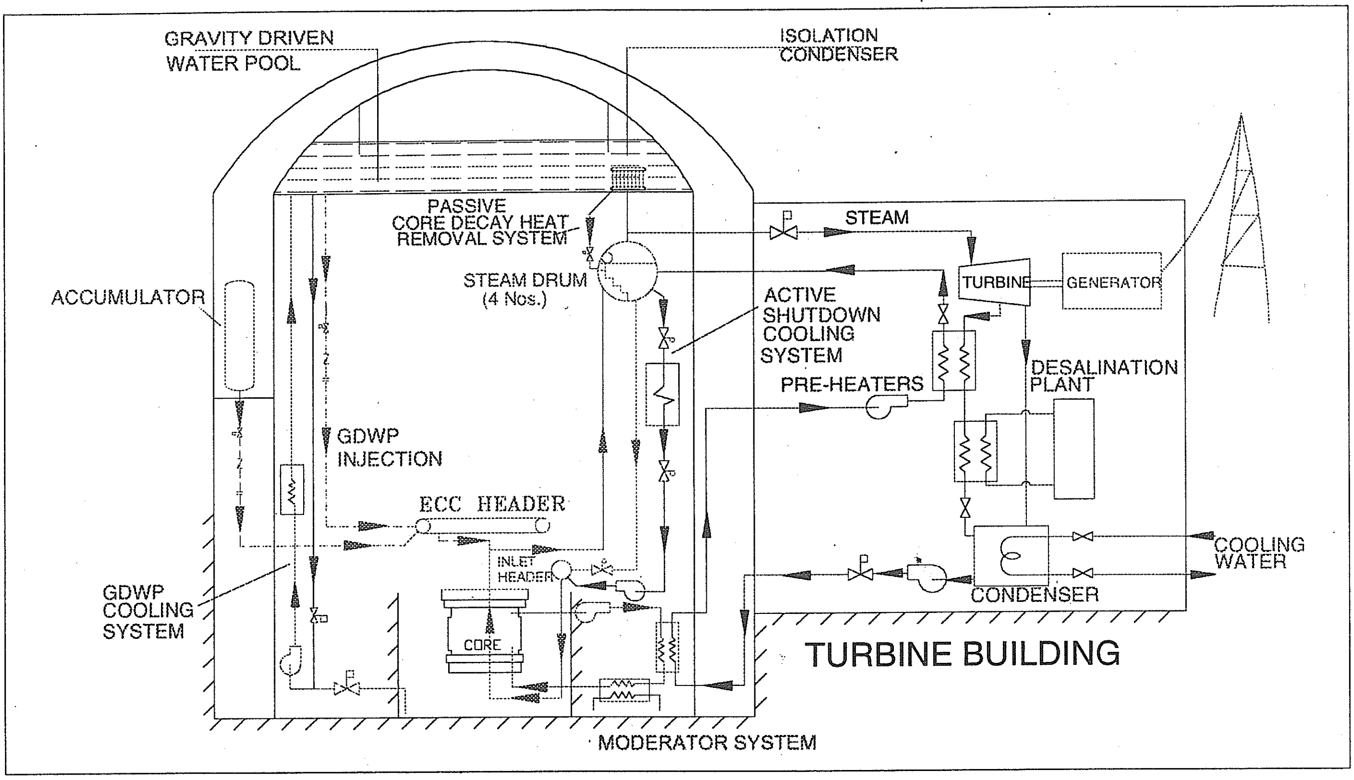


Fig.8 General arrangement of AHWR systems

sustaining equilibrium thorium (SSET) cycle practically any amount of energy can be extracted from thorium after the initial fissile charge of ^{233}U is arranged for. The discharge burn-up is, however, very low (12000 MWd/te) and, additionally, this cycle requires continuous reprocessing for recovering the fissile ^{233}U from spent fuel, which is again mixed with thorium for the next charge. As such, a low burn up with reprocessing is not a sound economic proposition; it is prudent to add some fissile topping to increase the burn-up at the cost of a slight reduction in fuel utilisation. In this way, the energy extracted can be higher and at the same time, the frequency of reprocessing can be decreased considerably. This feature, namely, slight addition of external fissile feed to reduce considerably reprocessing and refabrication load, is used in the design of AHWR. The external fissile feed is plutonium and a composite cluster with (Th, ^{233}U) MOX and (Th, Pu) MOX fuel pins is used throughout the core. Reconstitution of the fuel cluster is envisaged to achieve a differential burn-up between Pu and ^{233}U pins i.e., allowing a high burn-up for (Th, ^{233}U) MOX pins and a relatively lower burn-up for (Th, Pu) MOX pins. Thus, by keeping the plutonium bearing fuel in separate pins, there is an increase in thorium fuel utilisation and plutonium too, is effectively utilised.

While initial power flattening in PHWR cores is already established through irradiation of thorium, a scheme to irradiate thorium regularly in PHWR cores is being worked out. This would enable accumulation of initial inventory needed for AHWR partially. This would also enable considerable scale-up in thorium fuel cycle activities.

While discussing about development of thorium utilisation technologies for the third stage, it is to be kept in mind that large-scale availability of ^{233}U is an important pre-requisite for meaningful exploitation of thorium in the third stage. This can be realised only through fast reactors, which are going to be the mainstay of the second stage of the Indian nuclear power programme. These reactor systems, while they enhance energy potential from uranium, would also help us in large-scale production of ^{233}U from thorium. Several studies on irradiation of thorium in fast reactors have been carried out.

Compact high temperature reactor based power packs

It is well known that for India a large fraction of the import bill goes to purchase crude oil and petroleum-based hydrocarbon fuels. With continuously increasing demands, new technological solutions have to be looked at for generation of alternate fluid fuels. Most of the technologies for this need temperatures in the range of 700° C to 900° C. In particular, generation of hydrogen from water using chemico-thermal processes needs high temperatures exceeding 800° C. Keeping this in mind, a programme to design and develop a high temperature reactor system mainly for process heat, and non-grid based electricity generation applications, has been initiated. A conceptual design of the reactor is shown in Fig.9. Using U^{233} as fuel, a very compact design of core has been

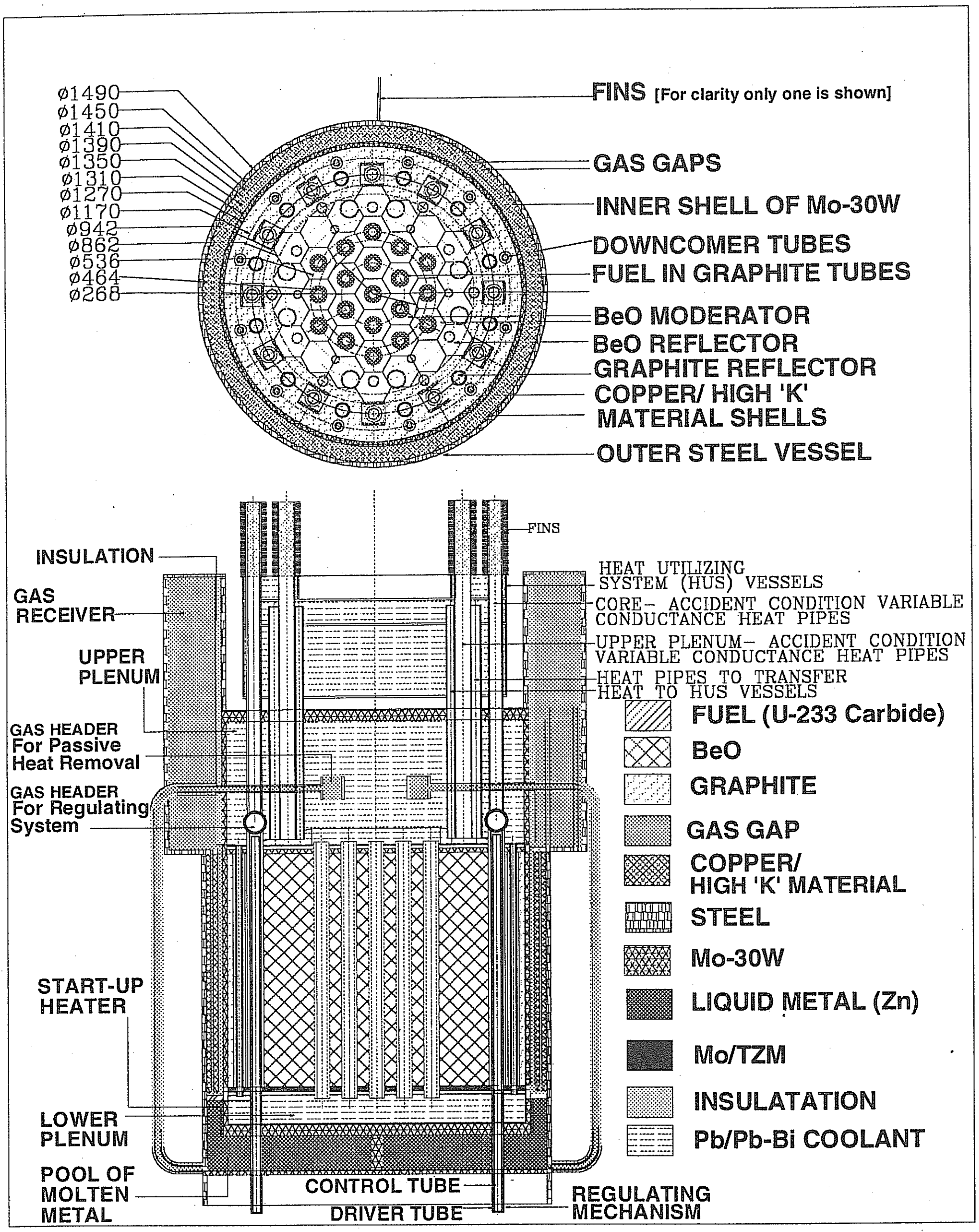


Fig.9 Compact high temperature reactor

worked out. A reactor based on this concept can thus; serve as compact power pack in remote locations. Several new technologies and materials are associated with this reactor system. These include liquid lead based cooling system, carbon-based structural components, coated particle type nuclear fuel, high temperature resistant alloys, and control systems which can reliably and passively operate under high temperature conditions, preferably without human intervention.

Some of the technologies required for such a high temperature reactor will also be useful for possible implementation in a future version of AHWR, where use of a carbon-based moderator, along with heavy water, may be considered to further improve the economic performance of the reactor. Molten lead related and high temperature application specific technologies are important for the accelerator driven systems, discussed in the next paragraph.

Accelerator driven subcritical systems

Accelerator driven systems throw open several attractive possibilities for extending the Indian nuclear power programme. High-energy protons, on colliding with a target of high-density element (such as lead, tungsten, uranium etc.) cause detachment of a large number of neutrons from these nuclides in a process known as 'spallation'. These neutrons can provide the required additional population, which can sustain a chain reaction in an otherwise sub-critical blanket (an arrangement similar to a nuclear reactor with not enough fuel to make it critical). Such systems, called 'accelerator driven systems', can be used to produce several times more electrical energy than that required to run the accelerator. Such a system is, therefore, also termed as an 'energy amplifier'. The system can also be so designed as to convert fertile materials, present in the blanket, to fissionable materials. Accelerator driven systems are also eminently suited for transmuting the highly radioactive waste from conventional nuclear power plants to shorter lived radionuclides, which do not require a very long-term storage under surveillance.

A good beginning has been made in acquiring the necessary expertise to design and build linear accelerators as well as cyclotrons. The challenges involved in the design of very large accelerators, and coupling them to sub-critical cores, are quite substantial and are matters of intense R&D effort in several countries. As a part of the roadmap for the third stage of the Indian nuclear power programme, a set of milestones has been identified, along the way of the development the technologies relevant for an accelerator driven system [6]. ADSS will contribute to improvement in the overall energy scenario in the long-term time frame.

Accelerator driven subcritical systems (ADSS) have attracted international attention in recent years. These systems offer the promise of energy production in sub-critical reactor systems based on thorium with considerable gains in terms of safety as well as growth in energy generation capacity. Normally, accelerators with sufficiently high current would be required for this purpose. Such systems are currently under development. A one-way coupled fast-thermal system has been proposed to get around this problem at least partially. An interesting aspect of this design of ADSS is that due to high decoupling of booster and reactor, the specific power density in the main thermal reactor is very low. Specific power density in AHWR is also low on account of natural circulation. Therefore, a modified AHWR core could be very well adopted for the thermal portion of such ADSS.

Conclusion

In India, uranium reserves alone cannot sustain a large nuclear power programme. The reserves of thorium in India are many times more than the uranium reserves. Hence the Indian need and priority for utilisation of thorium is much more urgent. The ThO_2 bundles used for the initial flux flattening will help in standardisation of reprocessing and re-fabrication technology and provide the material for engineering development. The worldwide interest in the utilisation of thorium in different type of reactors will provide the irradiation experience and specific reactor type related issues. India will be building AHWR and thorium will be used in the blanket of fast reactors also.

In the near term (next 10-20 years), the use of thorium in the existing reactors along with enriched uranium or plutonium will provide the necessary technological inputs for the next stage.

During the next stage (25-50 years), the advancement in technologies for reprocessing, remote fabrication, robotics, isotopic separation of U^{232} from U^{233} and production of U^{233} based on accelerators along with the reactor designs specifically optimised for thorium utilisation, the power obtained from thorium is expected to be significant.

In the long-term, due to constraints on raw material availability and very high level of safety standards in addition to other uses like waste incineration etc, thorium fuelled reactors are expected to provide highly innovative safe reactors.

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