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CLOSING THE FUEL CYCLE — A SUPERIOR OPTION FOR INDIA

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Abstract

The closed fuel cycle option with reprocessing and recycle of uranium and plutonium (U & Pu)for power generation allows better utilization of the uranium resources. On its part, plutonium is a unique energy source. During the initial years of nuclear fuel cycle activities, reprocessing and recycle of uranium and plutonium for power generation was perceived by many countries to be among the best of long term strategies for the management of spent fuel. But, over the years, some of the countries have taken a position that once-through fuel cycle is both economical and proliferation-resistant. However, such perceptions do vary as a function of economic growth and energy security of a given country.

This paper deals with techno-economic perspectives of reprocessing and recycling in the Indian nuclear power programme. Experience of developing Mixed Oxide UO_2 -PuO_2 (MOX) fuel and its actual use in a power reactor (BWR) is presented. The paper further deals with the use of MOX in PHWRs in the future and current thinking, in the Indian context, in respect of advanced fuel cycles for the future. From environmental safety considerations, the separation of long-lived isotopes and minor actinides from high level waste (HLW) would enhance the acceptability of reprocessing and recycle option. The separated actinides are suitable for recycling with MOX fuel. However, the advanced fuel cycles with such recycling of Uranium and transuranium elements call for additional sophisticated fuel cycle activities which are yet to be mastered.

India is interested in both uranium and thorium fuel cycles. This paper describes the current status of the Indian nuclear power scenario with reference to the program on reactors, reprocessing and radioactive waste management, plutonium recycle options, thorium-U233 fuel cycle studies and investigations on partitioning of actinides from Purex HLW as relevant to PHWR spent fuels.

1. NUCLEAR ENERGY

In the coming decades, the global energy demand will be growing by leaps and bounds mainly due to the developing countries. These nations have only limited options at their disposal to meet the steep increase in energy requirement and cannot ignore the role of nuclear energy as an alternate energy source with the potential to sustain the energy demand at the projected rates. Further, from the environmental point of view, there is a global need to deploy non-fossil sources to limit the carbon dioxide liberation to the atmosphere. As of today, the proven resources of low-priced uranium are insufficient to support a long-term and meaningful contribution to global energy demand by way of the nuclear energy.

2. FUEL CYCLE OPTIONS

So far as nuclear power production is concerned, there are two fuel cycle options of relevance and under consideration at the present juncture, Viz. the once -through cycle with permanent disposal of spent fuel and the closed fuel cycle with reprocessing and recycle of U and Pu. Both the options require efficient and safe waste management strategies and whichever is the option, the need for a long term geological repository cannot be eliminated, at the present stage.

In comparison to the waste from reprocessing and recycle, the disposal of spent fuel on once -through basis does not eliminate plutonium inventory and the hazards from long term perspective can only be reduced by its sustained irradiation in reactors. The reprocessing option provides a solution to radioactive waste arisings now rather than burdening future generations with this problem..

The key factor in the success of the closed fuel cycle lies in the efficient utilization of plutonium for power generation as it can increase the quantum of energy that can be derived from a given amount of uranium which varies depending on the reactor systems used. Thus closing the nuclear fuel cycle by reprocessing the spent fuel and recycle of U & Pu helps in achieving the goal of exploiting the full potential of nuclear power.

Plutonium is an unique energy source and "by allowing us to burn virtually all of the available uranium rather than just 1% as we do at present, the use of plutonium makes nuclear energy by far the largest energy resource available, indeed one that is virtually inexhaustible"[1]. Disposal of a fossil fuel after such a low level utilization of its potential as we do now with nuclear fuel would be unheard of.

Further, with the depletion of the natural uranium and fossil resources, the recycle of reprocessed uranium with an altered isotopic content of U235 may become economically viable. The redeployment of uranium as fuel from one type of reactor to another based on its depleted U235 content is an attractive proposition. The Dupic Process based on dry route and the Purex process followed by a U/Pu co-processing and precipitation route are again recycle options with minimum processing and are considered to be proliferation resistant. Over the years, the research reactor systems have spawned HTGRs and fast reactors for better utilization of uranium and plutonium. The challenges posed by fast reactors can be met by appropriate technology development in the coming years. The interim use of plutonium in LWR and PHWR as MOX fuel has been gaining acceptance.

From the safety point of view, reprocessing technology has made vast improvements by complying with national and international regulatory requirements and its annual radioactivity releases through various forms of effluents have steadily decreased over the years. As of today, these releases are very small in comparison to the present environmental burden of Pu and other radioactive elements released through atmospheric testing of nuclear weapons.

3. EVOLVING PRECEPTIONS AND INDIAN OPTIONS

Over the past few decades, the operation of the uranium fueled power reactors and the various research reactor systems have led to increased fissile inventories of Pu in the spent fuel. During the first generation nuclear fuel cycle activities, reprocessing and recycle of U & Pu for power generation was perceived by many countries to be among the best of long term strategies for the management of spent fuel. For several reasons, this perception has changed gradually over the years in some of the energy surplus countries which now consider that once-through fuel option as the economical and proliferation resistant approach that should be accepted globally. However these two criteria may differ from country to country and their perceptions would also differ accordingly.

Several nuclear energy countries like France, The United Kingdom, Russia, India and Japan had used reprocessing as part of their strategy for spent fuel management whereas some countries like Canada, Sweden and USA have opted to use the uranium once-through cycle option. USA has abundant reserves of coal, natural gas and oil. Canada has abundant natural uranium reserves and has no need to reprocess and recycle Pu. Among European countries, France has pursued a very active program on reprocessing and recycle of plutonium as MOX fuel. Thus for any country, the choice of its fuel cycle options with its minor variations should rightly be governed by its own assessment of its energy requirements on a long-term perspective, the alternate energy resources available at its command with their cost and its technological infrastructure capabilities to support and sustain modern sophisticated technologies such as nuclear power and subsequent spent fuel management.

The availability of uranium resources in India is limited. Other than the constraints to be overcome in meeting the energy security due to uncertainties in fuel supplies, purely from economic considerations, uranium procurement would add substantially to the foreign exchange component of the energy bill. The Indian reprocessing and storage costs in terms of installation and operation are substantially lower in comparison with the figures reported for western countries. Scaling up the facilities would result in cost reductions.

Further in our view from a long-term standpoint, reprocessing and recycling is more eco-friendly and provides better safeguards against plutonium getting into wrong hands as compared to once-through option, because recycling consumes plutonium while once-through leaves behind huge stocks of spent fuel which contain recoverable plutonium that may prove to be a rich and easy source for Pu after several hundred years of cooling.

Thus from the Indian stand point, the reprocessing and plutonium recycle option is not only considered to be superior option but also inevitable . This perception had emerged some four decades ago and has since remained unaltered.

4. THE INDIAN NUCLEAR ENERGY PROGRAMME

The Indian nuclear resources have been estimated [2] to be around 60,000 tons of U and around 360000 tons of Th. In terms of fossil fuel, this is equivalent to around 1.2 billion tons of coal equivalent through pressurized heavy water reactors (PHWR) and around 800 billion tons of coal equivalent through fast breeder reactors (FBR) and other reactor systems using thorium. Clearly, this constitutes a resource several times larger than any other resource that we have in our country for bulk electricity production. Table I. gives the profile of the energy resources in India in terms of coal equivalent.

The nuclear energy programme in India envisages three stages of implementation involving installation of uranium fueled thermal reactors in the first phase followed by utilization of plutonium in fast breeder and other types of reactors and in the third phase, utilization of reactor systems based on U233- Th cycle, which we consider to be the ideal fuel cycle of the future, from Indian context.

The first phase of our Programme is essentially based on the utilization of PHWRs for power generation with fuel reprocessing, plutonium recycle and efficient waste management as the strategies for the back end of the Fuel Cycle.

The choice of the Reprocessing and Plutonium Recycle option has endowed the program with a variety of mid-course options in both U and Th fuel cycle with Pu forming the vital link between the two.

TABLE I
ENERGY RESOURCES IN INDIA

RESOURCES	QUANTITY*
COAL	196 billion T
OIL	0.6 billion T (coal eqvt.)
GAS	540 billion m^3
HYDROELECTRIC (per year)	84 Gwe at 60% CF
URANIUM (in heavy water reactors)	380 Gwe-yr
URANIUM (in breeder reactors)	50, 000 Gwe-yr
THORIUM (in breeder reactors)	360, 000 T (> 200,000 Gwe-yr)

4.1. NUCLEAR REACTORS AND POWER GENERATION

Besides the two BWRs at Tarapur, there are several operating PHWRs with a design capacity of 220 MWe each. Some more reactors of similar type including two each of 500MWe are under different stages of planning, construction and commissioning. Under Fast Breeder Reactor (FBR) technology development programme, a 40 MW_{th} Fast Breeder Test Reactor (FBTR) is operational at Kalpakkam and the design of a 500 MWe Prototype Fast Breeder Reactor (PFBR) is in progress. In addition to PHWRs and FBRs, it is proposed to include LWRs and Advanced Heavy Water Reactors (AHWR) in the power programme.

These activities call for extensive recycling of Pu generated from PHWR's in FBR's or in the existing PHWRs or in newly conceived reactors of the AHWR type. These concepts are evolved to maximize the use of available resources and are heavily dependent on successful reprocessing and recycle of Pu.

4.2. FUEL REPROCESSING

Over the years, in tandem with the increase in spent fuel arisings from the growth of nuclear power, the reprocessing and nuclear waste management capabilities have been augmented to keep pace with the plutonium demands. There are now two reprocessing facilities to treat spent fuels from PHWRs.

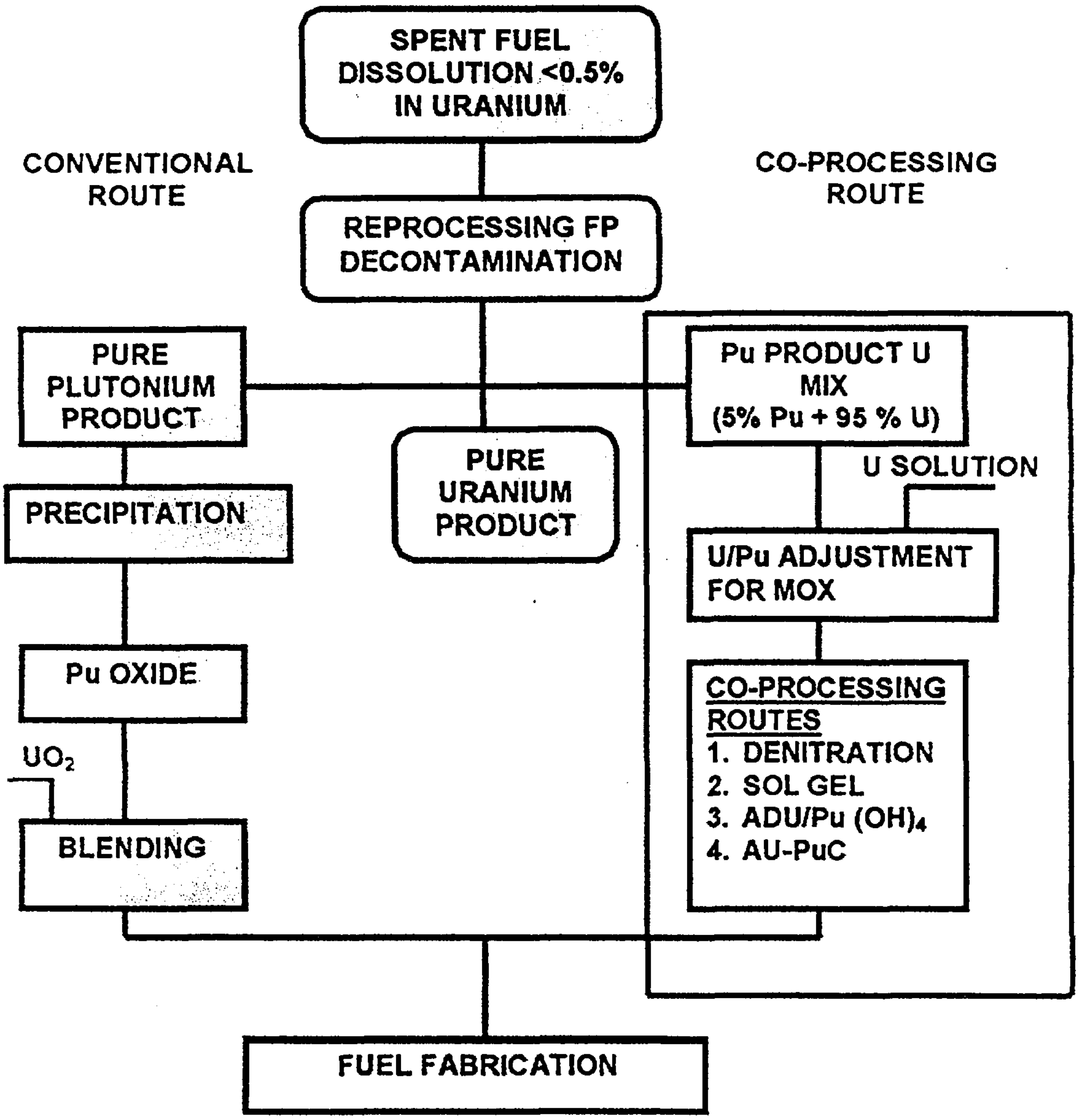

Fig. 1 Schematic Diagram of the Fuel Fabrication Route

4.2.1. Recovery of U/Pu by co-processing for MOX fuel

Presently reprocessing and recycle of uranium and plutonium together without their individual separation is considered as one of the alternate approaches for closing the nuclear fuel cycle. This approach becomes attractive for reprocessing the BWR spent fuel so that the fissile values available in it can be recovered and utilized for fabrication of MOX fuels for PHWRs. From the reprocessing and MOX fuel fabrication point of view, this approach comprises the following three major tasks : 1) Co-Processing and recovery of U and Pu together in Purex process, 2) Co-conversion of U and Pu product mix into their oxides and 3) Fabrication of PHWR - MOX fuel from the mixed oxide product.

The Purex flow-sheet can be modified for reprocessing spent fuel from LWR by co-processing as shown in Fig.1. The flow sheet conditions at the partitioning stage of the process can be adjusted so that the U/Pu composition in the stripped product will be about 95%U and 5% Pu. This would form the master feed that can be diluted further with uranium to the required Pu content and processed further for conversion to MOX using any of the several possible routes..

In the wet process route, certain amount of fission product decontamination is envisaged and the process does not eliminate enriched ADU generation during LWR fuel reprocessing. A major fraction of decontaminated LEU follows the ADU route as a separate product. About 10-15% of LEU (of the total input U) with enhanced Pu content (Pu enrichment 5%) forms the second product. Both the streams can be used for PHWR fuel fabrication.

Over the years BARC have been working on the development of several alternate routes for the conversion of U, Pu and Th products in their nitrate form to their corresponding oxides. Expertise and technological back-up for the various co-conversion process routes like Sol-gel, mixed oxalate precipitation, hydroxide precipitation, ammonium uranyl-plutonyl carbonate precipitation and direct denitration are available with us.

The Purex process, with a co-processing route for the production of Pu/U MOX in an integrated facility, may not pose major technical problems. The economic aspects and safety from radiological point of view are being examined.

4.3 THORIUM/URANIUM 233 FUEL CYCLE

To meet the challenges of thorium based fuel cycle, R&D efforts are directed towards extractive metallurgy of thorium, fuel fabrication and its utilization in reactors, reprocessing of irradiated thorium for U233 recovery and studies on U233 based reactor systems. Demonstration facilities have been operated in all these domains. With U233 as fuel, India has operated three low power research reactors, PURNIMA-II & III and KAMINI, a 30KW reactor with U233-Al alloy as fuel.

Thorium in the form of oxide fuel bundles was used in each of the two units of the Kakrapar atomic power station (KAPS I & II) for the purpose of initial flux flattening. It is proposed to follow this scheme for all future PHWRs as well. Additionally, there has been a continuous programme for irradiation of thorium rods in the research reactors located at Trombay. The irradiation of thorium in the blanket region of FBTR will commence shortly and it will also help in U233 generation.

4.4 FUEL FABRICATION AND EMERGING CONCEPTS IN Pu RECYCLE

Based on the plutonium-based fuel fabrication experience on pilot plant scale at Trombay, an industrial scale Advanced Fuel Fabrication Facility (AFFF) has been setup at Tarapur to meet the MOX fuel fabrication requirements. This facility will meet the fuel requirements of MOX for thermal reactors, and the initial startup requirements of Prototype Fast Breeder Reactor (PFBR). A facility to cater to the regular requirements for PFBR is being planned to be set up at Kalpakkam.

Though the FBRs are the best long term options for Pu recycle and burning, utilization of plutonium in PHWRs also offers considerable flexibility in terms of fuel cycle variations. Using U-Pu MOX in PHWR, it is possible to get improved fuel utilization and extended burnup resulting in significant increases in the overall installed capacities .

The conceptual once through thorium fuel cycle studies reveal that Th can be used in combination with Pu in reactors to high discharge burnups. It can burn Pu to a very significant extent. Studies on various reactor concepts indicate that heavy water reactors are second only to molten salt reactor systems as a choice for thorium utilization. An attractive option would be the use of Pu as a key to initiate the thorium cycle. As part of this programme, India is working on the design of an AHWR. This reactor requires an initial inventory of uranium-233 as well as plutonium. It derives 75-80% of its power from thorium in a self-sustaining mode of U233-Th cycle. The reactor needs an initial input of Pu in the form of mixed U-Pu oxide which contributes to 20-25% of the power and the recurring need for plutonium is relatively small. U233 in the thorium is adjusted to be at the self sustaining level and a discharge burnup of 20,000MWD/T is attained using plutonium as additional make up in the form of U,Pu oxide pins. The Pu pins are placed where neutron spectrum is most advantageous to Pu and the thorium fuel remains uncontaminated by the long lived plutonium and transplutonium actinides.

Coprocessing Thorium-Uranium 233 for the AHWR, which maintains Uranium proportion unaltered, is yet another attractive option that we are examining.

4.5. Pu Recycle - Technology Developments

The Indian policy has been “Reprocess and Recycle” from the very beginning. The experimental work on Pu fuel development has been initiated in the late seventies which has lead to the fabrication of mixed carbide fuel for FBTR and to the loading of MOX lead test assemblies in TAPS.

4.5.1. *Irradiation Experiments*

The irradiation experiments started in the late seventies with the irradiation of UO_2 - PuO_2 pins in Pressurized Water Loop (PWL) in Cirus Reactor. These irradiation experiments were mainly aimed at proving MOX fuel pins of BWR design. In addition, pins with PHWR design were also irradiated in PWL. A list of important irradiation experiments are given in Table-II. The experience obtained during fabrication lead to the establishment of a flowsheet and formed the basis for the design of our Advanced Fuel Fabrication Facility (AFFF).

TABLE II

EXPERIMENTAL IRRADIATIONS OF MOX FUEL (BWR) TYPE CLUSTERS IN CIRUS

Fuel Composition : UO_2 - 4% PuO_2

Sr. No.	Designation	Max.linear Rating W/cm	Burn-up MWD/Te	Remarks
1.	AC-2	414	16,265	Standard
2.	AC-3	490	16,000	Standard
3.	AC-4	490	2,000	Limited burn-up planned. Design variables studied include fuel clad gap, annular pellets, LTS, grain size and Pu cluster size.

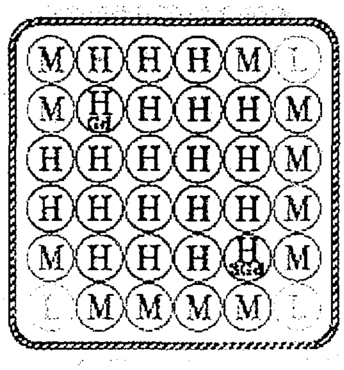

Fig. 2 (a) Standard LEU Design
$L = 1.6\ \%\ U^{235}$
$M = 2.1\ \%\ U^{235}$
$H = 2.6\ \%\ U^{235}$

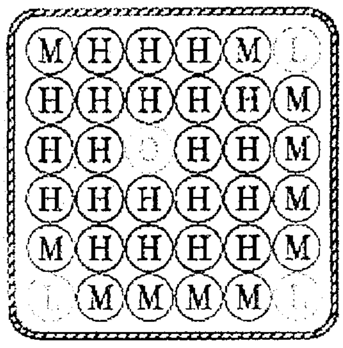

Fig. 2 (b) Replacement All Pu Design
$L = 0.9\ \%\ Pu^{235}$
$M = 1.55\ \%\ Pu^{235}$
$H = 3.25\ \%\ Pu^{235}$

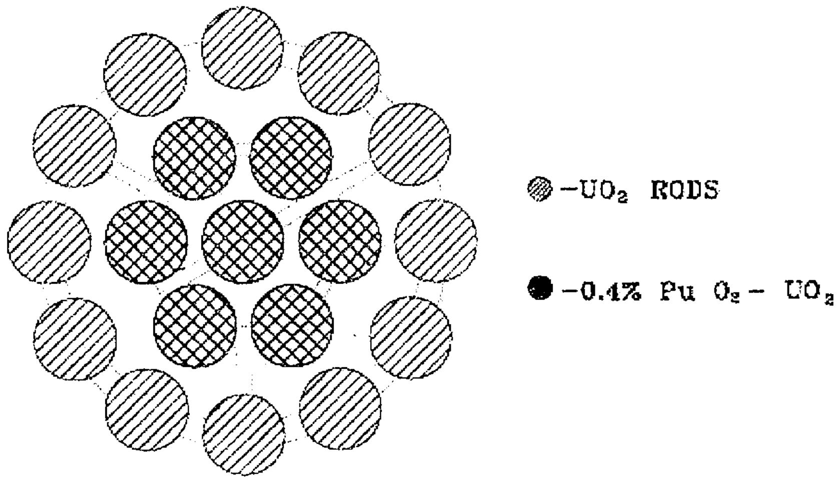

Fig. 3 MOX Fuel Bundle for PHWR

4.5.2. Fast Breeder Test Reactor (FBTR) Fuel

For the sodium-cooled FBTR, built at Kalpakkam, we have used an indigenously developed mixed carbide (MC) fuel, which has a composition of $(U_{0.3}Pu_{0.7})C$. Initially designed for a burnup of 50,000 MWD/Te, the performance of the fuel, on recent evaluation, has been found to be quite satisfactory and an extension of burnup to 70,000 MWD/Te is now under consideration. This programme on the fast reactor is very important to us for enhancing the potential for nuclear power generation using the limited uranium resources available in our country. The confidence generated in building and operating this reactor has given us the impetus for designing a prototype 500 MWe fast breeder reactor, proposed to be taken up for construction shortly.

TABLE III

CORE OPTIMISATION FOR TARGET MOX REACTOR

Fuelling Scheme No.	Inner zone Fuel/Burn-up MWD/Te	Outer zone Fuel/Burn-up MWD/Te	Average Burn-up MWD/Te	Max. Bundle Power (KW)	Max. channel power (MW)
1[*]	N.U./10700	N.U./6000	6600	395	2.89
2	MOX-7/14500	MOX-7/10200	10840	429.6 in MOX-7 bundle	3.14
3	N.U./11000	MOX-7/10200	10300[#]	422.5 in N.U. bundle	3.11
4	MOX-7/12/17350	MOX-7/12/13700	14300	470 in MOX-7/12 bundle	3.45

Note : Powers are normalised to total power of 6.551 MW to coolant

[*] Reference Natural UO_2 fuel bundles in all the channels

[#] Observed burn-up will be about 10,700 MWD/Te

4.5.3 TAPS Fuel

A programme to partially substitute the LEU (Low Enriched Uranium) assemblies by MOX assemblies in the BWRs at Tarapur is in progress. These assemblies have been fabricated at Advanced Fuel Fabrication Facility. Figs. 2 (a) and 2 (b) show the designs of the fuel assembly; the standard LEU fuel and the substitute all-Pu assembly. At present ten assemblies have been fabricated and six are undergoing irradiation in reactors. Some of these assemblies have so far crossed a burnup of 10,000 MWD/Te and their irradiation is continuing. They have been loaded adjacent to TIP (Travelling Incore Probe) locations to facilitate continuous monitoring. Their performance has been good based on off-gas monitoring and sipping. The fraction of MOX in the core is planned to be progressively raised as experience is gained.

4.5.4. PHWR Fuel

A study has been made in the use of MOX fuel in PHWRs with minimum deviation in the basic design of the fuel bundle, the reactor hardware, the control system etc. For the introduction of MOX in PHWRs, the limits on bundle power and channel power are already fixed by the present Nat.UO_2 design. Likewise the locations and the number of control and safety devices and fuel handling system are fixed. After analysing different cases (Table III), a MOX-7 design (Fig.3) with central seven rods containing 0.4% $Pu0_2$ in nat.UO_2 was selected for further implementation. The selected fuelling scheme will utilise nat.UO_2 for the central 44 channels. The studies indicate that the core average discharge burnup will improve to 10,700 MWD/Te instead of 6,700 MWD/Te at present.

4.6. RADIOACTIVE WASTE MANAGEMENT AND PARTITIONING AND TRANSMUTATION OPTIONS

The Indian program on safe management of radioactive wastes envisages two distinct modes of final disposal in respect of radioactive wastes; near-surface engineered, extended storage for low and intermediate-level active wastes and deep geological disposal for high-level and alpha bearing wastes.

A waste immobilization plant for the treatment of HLW is operational at Tarapur. It is a semicontinuous pot glass process involving calcination followed by melting in the process vessels. Two more waste immobilization plants are being set up at Trombay and Kalpakkam. Use of joule heated ceramic melters is under development. A solid storage and surveillance facility (SSSF) has also been set up for interim storage of vitrified HLW.

As regards ultimate disposal, the Indian choice is focused on igneous rock formations and some selected sedimentary deposits. Investigations are in progress for evaluation of candidate sites for a repository.

From environmental safety considerations, the separation of long-lived isotopes and minor actinides (MA) from reprocessing high level waste would enhance the acceptability of reprocessing and recycle option. The separated MA are suitable for recycling with MOX fuel. However, the advanced fuel cycles with such recycling of uranium and transuranium elements call for additional sophisticated fuel cycle activities which are yet to be mastered. Fig. 4 shows the conceptual schematic diagram of the advanced closed fuel cycle.

The main objective of partitioning high level waste is that it shall lead to a safer waste, more acceptable to the public. The removal of long-lived alpha emitting actinides from these wastes under P&T option would greatly reduce their long term radiological hazards. Removal of shorter lived fission products like Sr90 can reduce the heat generation from these wastes. Further, recovery of useful nuclides from this waste will make the waste management with P&T more economical and viable.

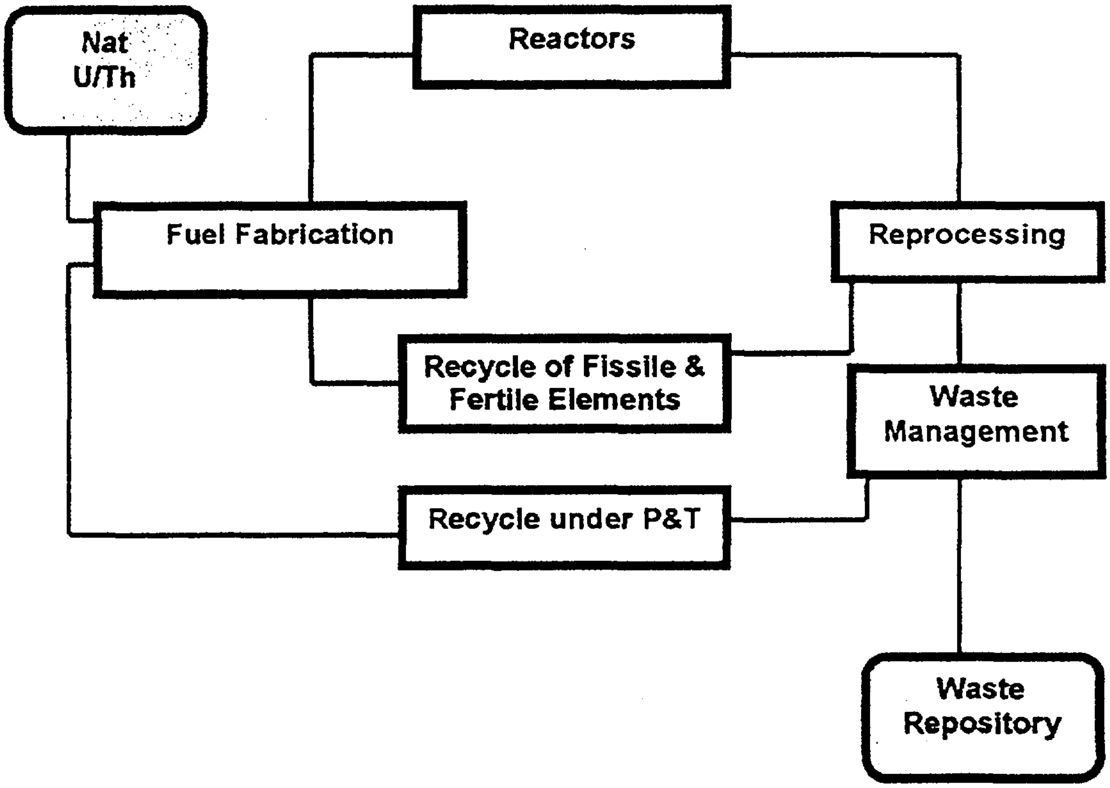

Fig.4 Schematic Diagram of Advanced Closed Fuel Cycle

From the Indian context, the present efforts are limited to the partitioning of the long lived actinides from the HLW as any reduction in the alpha burden of these wastes would render them safer with respect to long term disposal. At an appropriate time, a long term policy on the final utilization/ transmutation of the recovered actinides would be evolved, based on the available state of the art technology at that point of time.

Studies are in progress for the quantification of the PHWR spent fuel arisings, the radiological source terms of the relevant actinides and fission products in Purex HLW after reprocessing and evaluation of their hazard ranking. CMPO based solvent extraction and extraction chromatographic studies with HLW are in progress to propose suitable flow sheets for partitioning of the relevant actinides from these wastes and to reduce the alpha burden to very low levels. Other extractants are also being explored in this context.

The man-rem expenditures associated with P&T tasks should be evaluated and compared with the dose the future generation would be expected to receive in the distant future, in the event of leachates from geological repository reaching the biosphere without P&T. Such a comparison would be of help in reaching a decision regarding P&T option.

In the Th/U233 fuel cycle, the amount of transuranium nuclides generated is smaller by several orders of magnitude as compared to that arising from U235/U238 fuel cycle. In the case of LWR, the major hazard is from Am, Np, Cm isotopes and the left out Pu and U, whereas in the ^{232}Th-^{233}U fueled reactor, the hazard is mainly from ^{231}Pa. In the very long term perspective, the presence of U233 and U234 would also need consideration as Ra226 in the case of U fuel cycle.

5. CONCLUSION

Opting for a closed nuclear fuel cycle, a significant fraction of the energy output could come from the materials recycled from reprocessing. From Indian stand-point, under given limited resources of nat. U, this option is not only superior but also an inevitable one. Closed fuel cycle with Pu recovery on a `reprocess to recycle mode' can lead to a viable, safe and eco-friendly reprocessing and waste management strategy. Development of flow-sheets for co-processing of Pu together with U, without their individual separation by Purex process for conversion to MOX in integrated facilities, appears attractive . Advancements in reactor research have spawned several new alternatives for the better utilization of Pu and U. These systems are yet to be perfected prior to commercial exploitation and public acceptability. Meanwhile, the available fissile inventory can be redeployed on an interim basis in the existing reactor systems which can lead to an enhanced energy profile. The emerging reactor concepts such as the AHWR, which integrates both U/Pu and Th/U233 fuel cycles can yield valuable information and can lead India closer to its ultimate goal, ie. the Th-U233 fuel cycle.

From environmental safety considerations, the separation of long-lived isotopes and minor actinides (MA) from reprocessing high level waste would enhance the acceptability of reprocessing and recycle option.. However, the advanced fuel cycles with such recycling of uranium and transuranium elements call for additional sophisticated fuel cycle activities which are yet to be mastered. Any strategy for sustained nuclear power generation that involves recycling of reprocessed U and Pu will have to face these challenges.

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