Plutonium and thorium in the Indian nuclear programme*

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The major applications of plutonium in India have so far been the development and production of plutonium-rich, hitherto untried, mixed uranium–plutonium monocarbide fuel for FBTR, mixed uranium–plutonium oxide (MOX) substitute fuel for the BWRs at Tarapur and the peaceful nuclear explosion experiment at Pokhran in 1974. India is pursuing a three-stage nuclear power programme linking the fuel cycles of PHWR and LMFBR. Presently, all activities of PHWR fuel cycle, including fuel fabrication, spent fuel reprocessing and waste treatment, are being carried out on an industrial scale. India has one of the largest reserves of thorium which could be converted by neutron capture reaction to fissile ^{233}U and utilized most efficiently in thermal reactors as a self-sustaining (Th, ^{233}U) O_2 fuel. A programme of 20,000 MWe nuclear power in the course of the coming 25 years with a mix of PHWRs, LMFBRs and LWRs may constitute an effective base, from which we can expand our programme into the later part of the next century.

THORIUM, uranium and plutonium are the basic elements for utilization of 'nuclear fission' energy. The earth's crust contains nearly 12 ppm thorium, about 4 ppm uranium and practically no plutonium. Natural uranium has two main isotopes, $^{238}_{92}U$(99.3%) and $^{235}_{92}U$ (0.7%) but thorium occurs in nature only as $^{232}_{90}Th$. ^{235}U is the only naturally occurring 'fissile' material. Neutrons of all energy can induce fission of ^{235}U nuclei, leading to the release of fission fragments (consisting usually of two lighter elements), two or three or more neutrons and about 200 MeV energy. Some of the released neutrons can go on to cause fission of other ^{235}U nuclei, thus sustaining the 'fission chain reaction'. The extra neutrons available after maintaining the chain reaction could be utilized for transmuting naturally occurring $^{238}_{92}U$ and $^{232}_{90}Th$ isotopes to produce man-made 'fissile' isotopes $^{239}_{94}Pu$ and $^{233}_{92}U$ respectively. ^{238}U and ^{232}Th are called 'fertile' isotopes.

$$^{235}_{92}U + ^{1}_{0}n \rightarrow ^{236}_{92}U \rightarrow \text{fission fragments} + (2-3)^{1}_{0}n + 200\ \text{MeV}$$
(lighter elements)

$$^{238}_{92}U + ^{1}_{0}n \rightarrow ^{239}_{92}U \xrightarrow[23.5\ m]{\beta^-} ^{239}_{93}Np \xrightarrow[2.3\ d]{\beta^-} ^{239}_{94}Pu$$

$$^{232}_{90}Th + ^{1}_{0}n \rightarrow ^{233}_{90}Th \xrightarrow[23\ m]{\beta^-} ^{233}_{91}Pa \xrightarrow[27\ d]{\beta^-} ^{233}_{92}U\ .$$

The fission energy is utilized in a nuclear power reactor for generation of electricity or in a nuclear explosive device for blast and thermal radiation damage. The minimum mass of 'fissile' material, necessary to sustain a nuclear chain reaction is called its 'critical mass'. This critical mass varies depending on the configuration of the fissile and other material in the system and is the smallest when the configuration is spherical. The critical mass of bare spherical fissile material could be further reduced by surrounding the same with a neutron reflector or by compressing the fissile material and increasing its density. The latter technique is used in an explosive device using the method of 'implosion'. Fissile and fertile materials and fission energy have the following unique features:

High energy density

The fission process emits about 200 MeV per fission while a chemical reaction occurring in fossil fuels releases about 2–4 eV only per atom of carbon.

Singular application

The fissile and fertile materials are used only for generating fission energy and additional fissile materials respectively, while fossil fuels like coal or oil have several applications other than generating energy.

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Absence of greenhouse or acid gases

The most obvious environmental advantage of nuclear power stations over coal-fired thermal plants is the absence of acid gases SO_2, NO_x and of the 'greenhouse' gas CO_2 which is considered responsible for potential global warming. If the electric energy now generated from nuclear power each year in the world were produced by coal-fired power plants, there would be additional annual emission of about 1600 million tonnes of CO_2 (ref. 1).

Requirement of high levels of safety and security

'Th', 'U', 'Pu' and fission products are radioactive and health hazardous to varying degrees and require proper containment, and sometimes remote and automated handling. The radioactive waste has to be properly treated and physical protection of nuclear material has to be provided to avoid proliferation risks.

Relatively new

The science and technology of nuclear fission energy are not even 60 years old and are, therefore, very recent in the time scale of human history. The major scientific discoveries associated with nuclear fission energy, namely (a) 'fission' (January, 1939), (b) 'plutonium' (February, 1941), (c) neutron-induced fission of ^{239}Pu (March, 1941), and (d) 'self-sustaining fission chain reaction' (December, 1942) took place within a short span of time during the Second World War and paved the way, to start with, for military, rather than civilian, application of nuclear energy.

The Manhattan project for nuclear weapons was secretly initiated in the Los Alamos Laboratories, USA during 1943–45, which soon led to the development of 'nuclear explosive devices' based on weapon grade ^{239}Pu ($< 10\%$ ^{240}Pu) and high enriched uranium (HEU: $>90\%$ ^{235}U). The first explosive device had a 'Pu' core weighing about 6.1 kg and was tested on 16 July 1945 at Almogordo, New Mexico. This was followed by the actual and tragic use of uranium and plutonium weapons by USA in the Japanese soil in Hiroshima and Nagasaki almost exactly fifty years back – on 6 August and 9 August 1945 respectively – which led to unprecedented loss of human life and property damage. The nuclear weapons activities continued in USA after the second world war. The former Soviet Union, UK and France started their own nuclear weapons-development programme. Soon, the nuclear fusion device was also developed. Thereafter, in the 1960s China joined the nuclear weapons club by developing and testing both fission and fusion devices. The nuclear weapons race continued, in these five countries, particularly between USA and Russia, till 1990 when the estimated warheads were in the range of several tens of thousands. These warheads are estimated to contain several hundred tons of HEU and of weapon grade 'Pu'.

India's first and only peaceful nuclear explosion (PNE) experiment was carried out in the Rajasthan desert on 18 May 1974 at a place near Pokhran. A plutonium device of yield 12 kt equivalent of TNT was successfully tested. It is interesting to note that, among the six countries which have manufactured and exploded nuclear devices, India is the only one which carried out its first test underground. And it has observed the longest moratorium of more than 20 years on testing!

Civil nuclear fission technology had its birth in the early weapons programme but later followed a separate evolutionary path. The first nuclear power plant, a 5 MWe unit, was commissioned in Obninsk, Russia, in 1955. Thereafter, in the last 4 decades, the design, development, construction, commissioning, operation and safety features of nuclear power plants have progressed in leaps and bounds. So far, nearly 8000 reactor years of operating experience have been acquired and several operating nuclear reactors today have capacities as high as 1400 MWe. Currently, 431 nuclear power reactors are operating in 30 countries, generating approximately 340 GWe, which accounts for nearly 17% of global electricity[1]. In France, Belgium, Sweden and Lithuania, more than half of the electricity comes from nuclear energy and in 14 other countries including USA, UK, Germany, Hungary, Finland, Japan, South Korea and Taiwan more than 20% electricity is obtained from nuclear power plants. In India, the current contribution of nuclear power (~1940 MWe) to electricity generation is about 2%, but this is likely to increase progressively in the coming decades.

Light Water cooled Reactors (LWRs) consisting of Pressurized Water Reactors (PWRs) and Boiling Water Reactors (BWRs) account for more than 80% of the operating nuclear power reactors followed by the Pressurized Heavy Water Reactors (PHWRs). Liquid Metal cooled Fast Breeder Reactors (LMFBRs) are few in number, at present, but are likely to be further commercialized during the first quarter of the 21st century. LWRs are popular in USA, France, Germany, Russia, East European countries, Sweden and Japan. The PHWR system originated in Canada and is the backbone of the nuclear power programmes in Canada and India. Apart from the 'power reactors', nearly 325 research reactors are utilized all over the world as 'neutron sources' for isotope production, irradiation-testing of materials, neutron activation analysis, radiography and for basic and applied research.

Nuclear properties of ^{233}U, ^{235}U and ^{239}Pu

Table 1 summarizes some important nuclear properties of the three fissile isotopes ^{233}U, ^{235}U and ^{239}Pu. It may

Table 1. Nuclear properties of ^{233}U, ^{235}U and ^{239}Pu

Property	$^{233}_{92}U$	$^{235}_{92}U$	$^{239}_{94}Pu$
Occurrence in nature	No	Yes	No
Main mode of radioactive decay (half-life)	α (1.6×10^5 y)	α (7.1×10^8 y)	α (2.4×10^4 y)
Isotopic impurities, their half-life and characteristics	^{232}U, α (72 y) Has strong γ emitting daughter products	^{238}U in HEU α (4.5×10^9 y)	^{240}Pu, α (6.5×10^3 y) Emits spontaneous fission neutrons ^{241}Pu, β^- (14.4 y) is fissile; decays to ^{241}Am (γ-emitter) ^{242}Pu, α (3.8×10^5 y) ^{238}Pu, α (87.7 y)
Fission cross-section, σ_f			
Thermal (barns)	531 ± 2	580 ± 2	742 ± 3
Fast (barns)	1.9	1.3	2.0
Neutron capture cross-section σ_c (barns)	47 ± 1	98 ± 1	271 ± 3
Ratio of captures to fissions	0.089 ± 0.002	0.169 ± 0.002	0.366 ± 0.004
Number of neutrons produced per fission, ν	2.487 ± 0.007	2.423 ± 0.007	2.880 ± 0.009
Number of neutrons created per neutron absorbed by the nuclide, η			
Thermal neutrons	2.284 ± 0.006	2.072 ± 0.006	2.109 ± 0.007
Fast neutrons	2.31	1.93	2.49
Critical mass (bare)	15 kg	43 kg	10 kg

Table 2. Weapons grade v/s reactor grade Pu

	Weapons grade	Reactor grade
Typical ^{239}Pu isotopic content	~ 94%	~ 75%
Critical mass (bare) (kg)	11	13
Specific activity (Cu/g)	3	10
Spontaneous fission rate ($ns^{-1}\,kg^{-1}$)	70,000	300,000

be noted that the critical masses for bare spheres[2] of ^{239}Pu (10 kg) and ^{233}U (15 kg) are lower than that of ^{235}U (43 kg). This is because they have higher fission cross sections (σ_{fiss}) and produce more neutrons per fission than ^{235}U. However, the isotope ^{240}Pu, a by-product of the plutonium produced in a reactor, is undesirable because of its higher radioactivity, higher heat production and higher spontaneous fission rate as shown in Table 2. The latter factor may lead to premature initiation of a nuclear device when high ^{240}Pu content plutonium is used and this makes the yield low and uncertain. Therefore, for these purposes, the build up of ^{240}Pu is restricted to a few per cent in so-called weapon-grade plutonium.

The number of neutrons produced per neutron absorbed by the fissile nucleus, known as η, is an important physics parameter. Figure 1 shows the variation of η with the energy of the neutron. ^{233}U has a higher value compared to ^{239}Pu and ^{235}U in the thermal (neutron energy $E < 0.025$ eV) and epithermal energy ranges of neutrons, whereas ^{239}Pu has the highest η value amongst the three fissile nuclei in the fast neutron range ($E > 0.1$ MeV). ^{233}U and ^{239}Pu are, therefore, the best fissile materials for use with thermal and fast neutrons respectively. Unfortunately, neither of them is available in nature. ^{233}U always contains a small amount of ^{232}U which has strong gamma-emitting daughter products as shown in Figure 2. The presence of ^{232}U renders ^{233}U somewhat more difficult to handle.

Plutonium metallurgy and peaceful nuclear explosion

Spent fuel reprocessing technology

The need for a strong plutonium base for the nuclear programme in India was recognized in the late 1950s. Accordingly, a decision was taken to build a small demonstration plant at Trombay for reprocessing research reactor fuel that would be a forerunner for the much bigger plants to be built later for reprocessing the spent fuel discharged from our power reactors. The basic process chosen for spent fuel reprocessing in India was combined solvent extraction and ion exchange separation technique. The plutonium plant was commissioned at Trombay in 1964 and was utilized for reprocessing

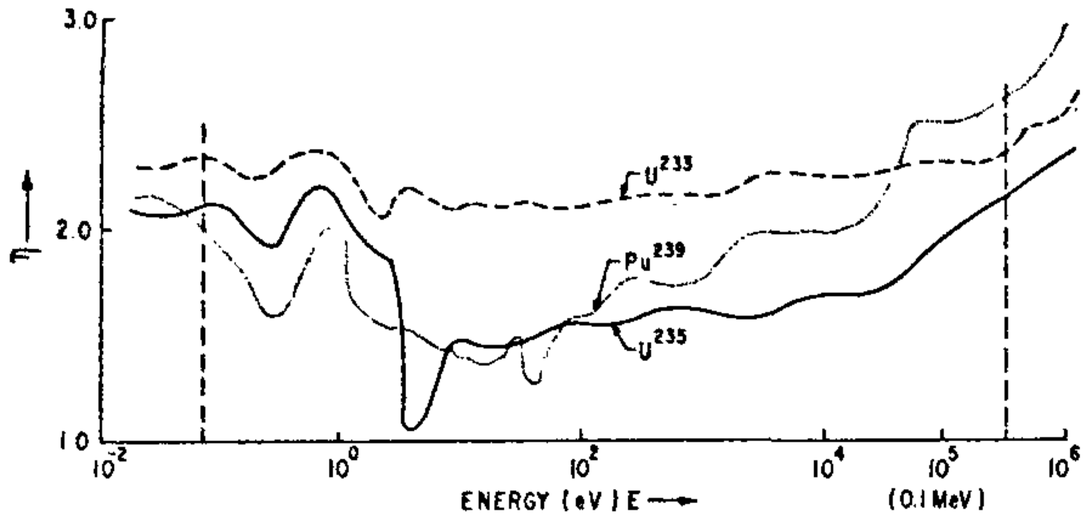

Figure 1. Variation of η with energy.

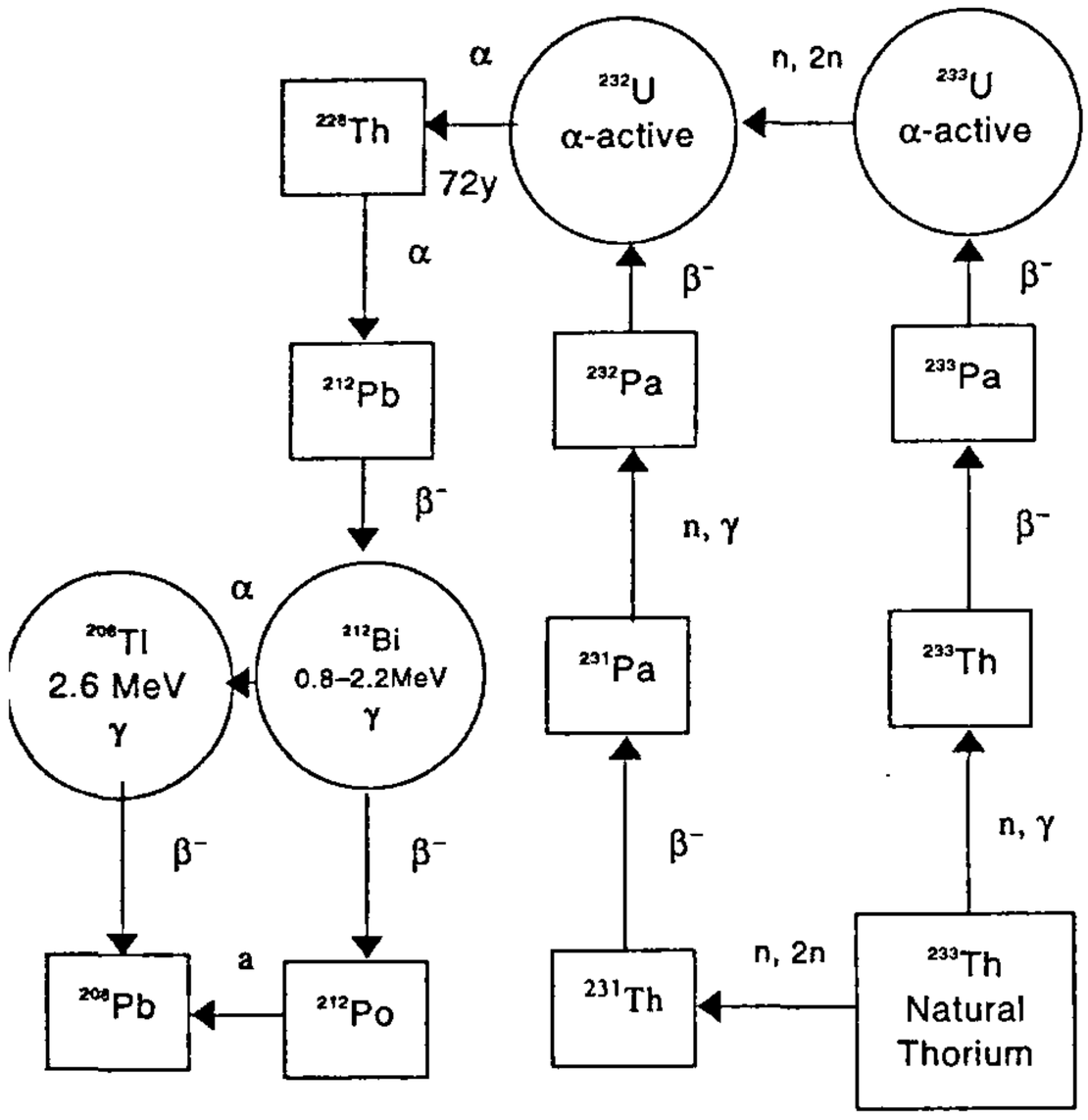

Figure 2. γ-activity of ^{232}U present in ^{233}U.

Al clad natural uranium metal spent fuel rods from the CIRUS research reactor at Trombay[3]. The high radioactivity associated with the irradiated fuel requires all operations to be carried out remotely inside thick concrete shields with reliable instrumentation for process control. Because of the high radiotoxicity and the criticality hazard associated with plutonium, safety measures are required to be built into the design and a close surveillance has to be kept continuously throughout the operation.

The plutonium obtained from the Trombay plant was utilized for the following three major fabrication activities at BARC:

- stainless steel 316 clad PuO_2 fuel pins for the first fast critical assembly PURNIMA-I at Trombay in 1972.
- plutonium metal for the peaceful nuclear explosion (PNE) experiment at Pokhran in 1974, and
- stainless steel 316 clad mixed uranium–plutonium monocarbide fuel in 1984 for the first core of the Fast Breeder Test Reactor (FBTR) at Kalpakkam.

The Trombay plant has been subsequently augmented and is being used for reprocessing spent uranium metal fuels from CIRUS and DHRUVA research reactors at BARC.

A second fuel reprocessing unit of higher capacity known as the Power Reactor Fuel Reprocessing (PREFRE) plant was commissioned at Tarapur in the 1970s on the basis of the satisfactory experience and feedback from the Trombay plant[4]. The PREFRE plant has been extensively utilized for reprocessing zircaloy clad UO_2 spent fuel from our operating water cooled power reactors. A third reprocessing plant is nearing completion at Kalpakkam[5]. The Kalpakkam Reprocessing Plant (KARP) would also have a provision for reprocessing spent fuel elements from FBTR and the proposed Prototype Fast Breeder Reactor (PFBR).

The irradiated or spent uranium fuel elements from nuclear reactors are a mixture of depleted uranium (< 0.7% ^{235}U), fission products and plutonium. These are first adequately cooled in spent fuel storage pool adjacent to the reactor for the short-lived fission products to die down. Next, the spent fuel elements are transported in a special vehicle to the reprocessing plant, where they are first mechanically or chemically dejacketed from the cladding material. After this, the fuel is dissolved in nitric acid and the solution is subjected to solvent extraction for removal of fission products in the first stage and uranium in the second stage. The separated plutonium is further purified by ion-exchange process and obtained as plutonium nitrate solution, which is converted to the oxide powder by the oxalate precipitation route followed by air-calcination. PuO_2 powder is usually the end product of spent fuel reprocessing plant because it is most convenient to store plutonium in the form of oxide from the point of view of chemical stability and transportation.

Plutonium metallurgy at Trombay

The metal plutonium is man-made and is no doubt the most complex, interesting and strategic of all metals. It undergoes six phase changes before it melts. It has very low critical mass and is a storehouse of energy which could be utilized in a peaceful way. At Trombay, plutonium metallurgy was started on a laboratory scale in the early 1970s. The plutonium produced was used as starting material for remelting and casting of Pu metal, Al–Pu, Pu–Be and other plutonium alloys[6].

Phenomenology of Pokhran PNE experiment

The objective of India's first and only nuclear explosion experiment was to study the explosion phenomenology, fracturing effects in rocks, ground motion, containment of radioactivity, etc. in the context of the possible applications of peaceful nuclear explosions. In this experiment, the 12 kt device was emplaced in a shale medium at a depth of 107 m. Upon detonation, the ground surface above the emplacement point rose with a velocity of 25–30 m/s to form a dome 170 m in diameter and 34 m in height. There was no venting of radioactivity in the experiment. The resultant apparent crater, measured with respect to the preshot ground surface, had an average radius of 47 m and a depth of 10 m. In fact this is the only case of an underground nuclear explosion which produced a crater (though shallow), and yet was completely contained from the radioactivity point of view[7].

The phenomenology of this experiment was studied using a one-dimensional spherical symmetric rock mechanics computer code which simulates the various physico-chemical processes set up in the rock medium on sudden release of energy[8]. The constitutive relations of rocks were inputs to these calculations. It was shown among other things, that the smaller cratering efficiency of the shale–sandstone medium (compared to the hard rock in which US experiments have been carried out) is related to the lower kinetic energy imparted to the mound because of lower density, high rock porosity and lower modulus (i.e. a softer equation of state).

The calculations made show that this resulted in 640 tons of rock, extending up to 4.1 m from shot point, being vapourized. About 2000 tons of rock, extending up to a radial distance 6.2 m, was shock-melted. At the vapour–liquid interface, the pressure is expected to be about 160 GPa. The final cavity radius is calculated to be about 28–29 m compared to the post-shot measured value of 30 m (Figure 3). The calculated spall velocity of 30 m/s and the extent of rock fracturing, 114 m, are in good agreement with the measured values of 25–30 m/s and 80–100 m respectively. The spall was found to be the principal mechanism of cratering at Pokhran.

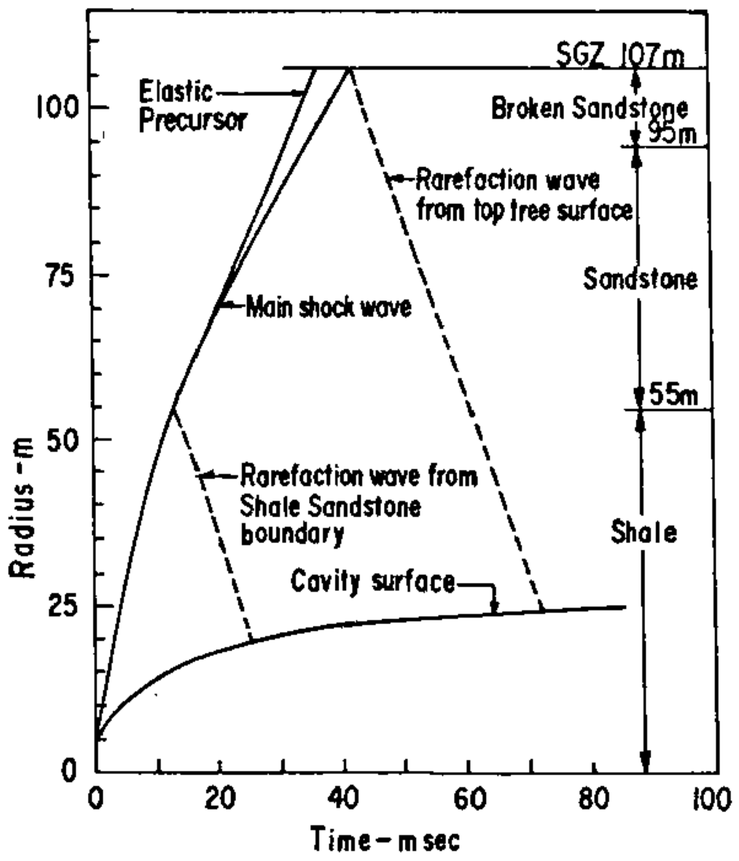

Figure 3. Computed wave propagation and cavity growth i direction for the POKHRAN experiment.

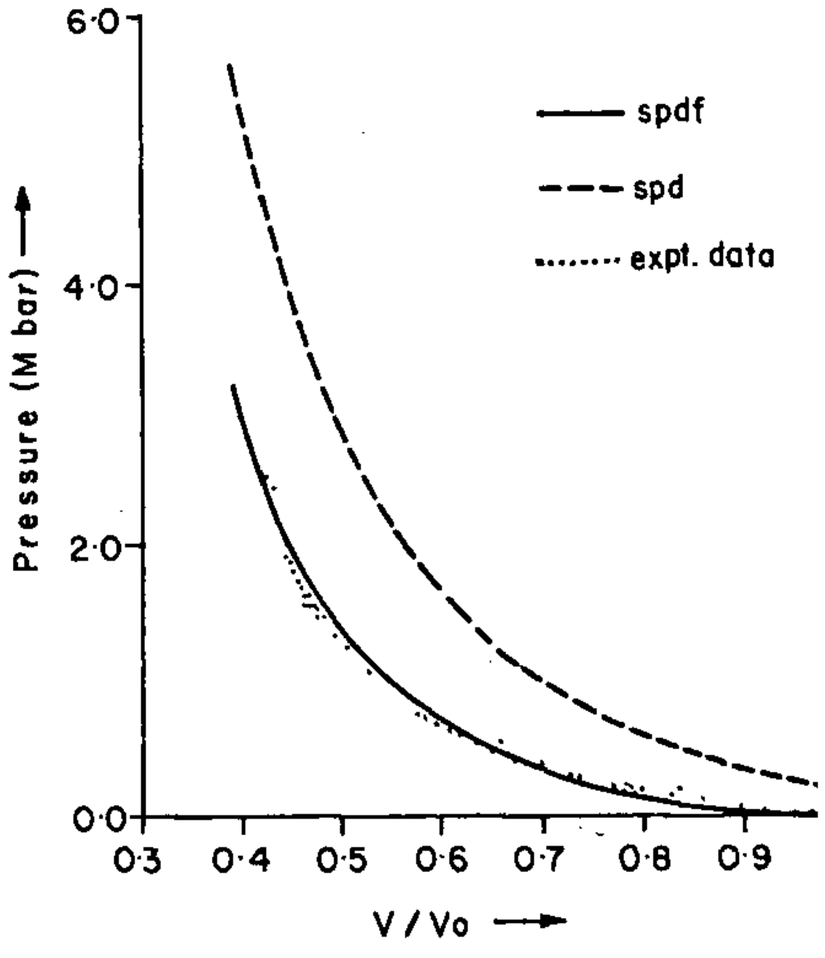

Figure 4. Theoretical 300 K isotherm of thorium

Equations of state of nuclear materials

The equation of state (EOS) of a system is a relationship between thermodynamic variables like pressure and energy with volume and temperature. It is a vital input for a wide spectrum of practical applications in an atomic energy programme, e.g. in design of fission and fusion explosive devices, for understanding the rock mechanical effects of shock propagation in earth due to underground nuclear explosions and simulations of reactor accidents. For example, as noted above, in the case of

fission explosions, the critical mass can be lo
the fissile material is compressed to increase its
In 1968, when the design of the device use
Pokhran explosion experiment was initiated, ve
was known in open literature about the compres
haviour of materials at ultra high pressures and
formation was not at all available for nuclear m
especially for Pu (it is still classified). So our f
was to calculate the EOS of Pu. Using empiric
of EOS with constants evaluated carefully f
analysis of scarce data on related elements in th
ture, we demonstrated that desired compression
achieved by shock waves. Of course, EOS stu
important also from a basic research point of v

we now have more sophisticated first principle solid state theories to compute equations of state[9,10]. The kind of confidence one has in these calculations may be illustrated from the recent work of Rao *et al.*[11] on Th (Figure 4).

Nuclear fuel cycles and nuclear power programme in India

The nuclear fuel cycle activities encompass mining of uranium and thorium ores, their chemical processing and concentration, enrichment of ^{235}U content in uranium in some cases, fabrication of natural or low enriched uranium (LEU: < 20% ^{235}U) fuel elements, irradiation of these fuels in reactors, reprocessing of spent fuels, refabrication of Pu- or ^{233}U-bearing fuel elements, recycling of these fuels in reactors and treatment and disposal of nuclear wastes as shown in Figure 5. The evolution of nuclear fuels and nuclear fuel cycles in different countries essentially depends on the availability of uranium and thorium resources, the energy demands, the industrial know-how and infrastructure and various economic and political pressures. In addition, recent dismantling of large quantities of weapon grade plutonium from the nuclear warheads from the former Soviet Union and USA has led to the development of a wide variety of plutonium-based ceramic, metallic and dispersion type nuclear fuels with the objective of burning and not breeding plutonium in thermal and fast reactors. The nuclear fuel cycles fall under the following three main categories:

Open 'once-through' cycle

In this cycle, natural (0.7% ^{235}U) or low enriched uranium (LEU: <20% ^{235}U) fuels are utilized in the reactor on a 'once-through' basis. The spent fuel is not reprocessed but cooled, conditioned and sent for permanent disposal in repositories. The total uranium utilization in this scheme is very low (≤ 3%). Countries like USA, Canada and Sweden are following the 'once-through' cycle. These countries have relatively small population, very low population growth, a high living standard and other sources of energy, apart from liberal access to world resources of uranium. Hence, reprocessing of spent fuel for recovery and utilization of plutonium has not much of an incentive for them at this stage.

Closed ^{238}U–^{239}Pu cycle

In the closed ^{238}U–^{239}Pu fuel cycle, the spent natural or LEU fuel is reprocessed for recovering plutonium by-product and unused ^{235}U and ^{238}U. The fission products are separated and sent for disposal as waste. The plutonium and depleted or reprocessed uranium are fabricated into fuel elements and recycled either in thermal or fast reactors. Since the breeding efficiency of plutonium in ^{238}U–^{239}Pu cycle is best in fast neutron spectrum, the plutonium should be recycled in fast reactors. On recycling the plutonium bred in each cycle, along with depleted uranium repeatedly, it is possible to utilize realistically almost 60–70% of the total uranium. Countries like France, Japan, UK, Russia and Germany have opted for the closed ^{238}U–^{239}Pu fuel cycle utilizing plutonium in fast reactors. Since the LMFBR programme has been delayed all over the world, plutonium is being recycled in the form of mixed uranium–plutonium oxide (MOX) in LWRs for interim utilization. MOX fuel is regularly being used in commercial power plants in France, Germany and Japan.

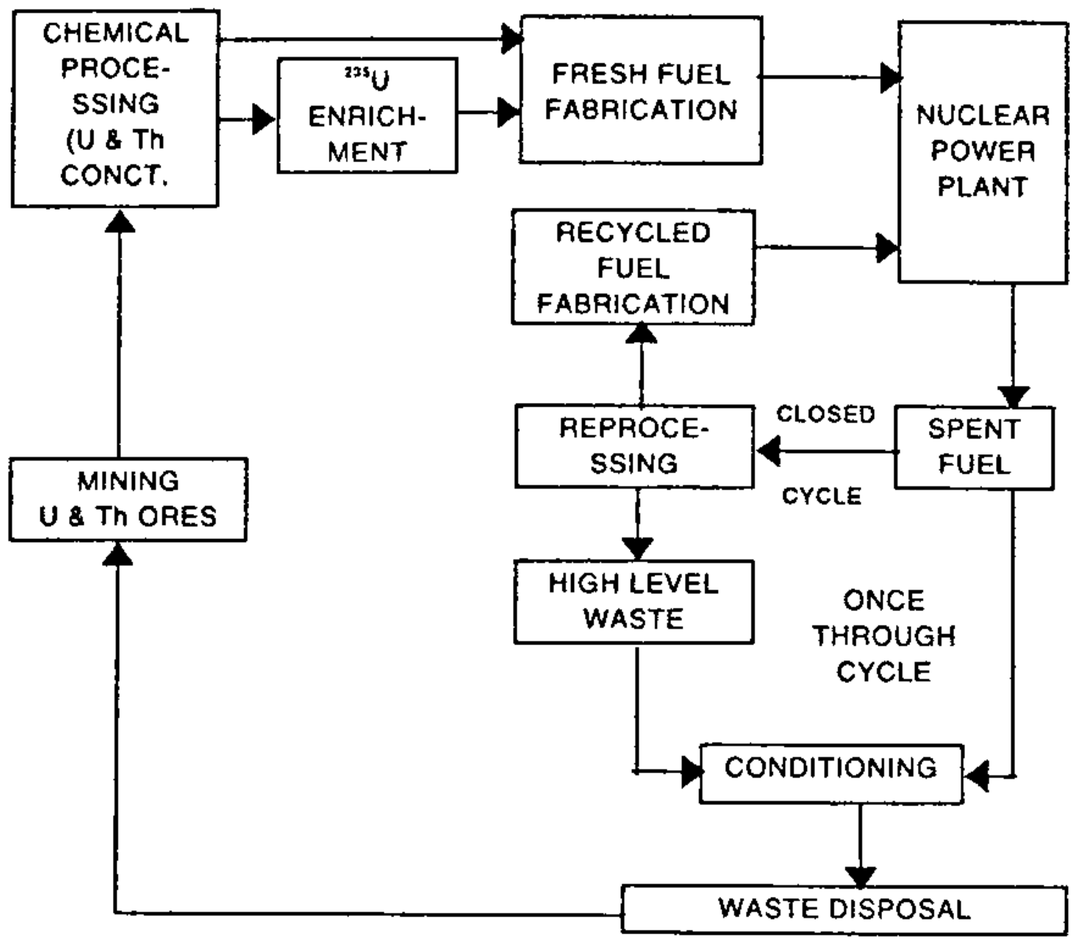

Figure 5. Schematic of 'open' and 'closed' nuclear fuel cycles.

Closed and combined ^{238}U–^{239}Pu and ^{232}Th–^{233}U fuel cycles

In this scheme, the spent uranium fuel from water-cooled reactors is reprocessed for recovering the plutonium which is recycled in a fast reactor in combination with depleted, natural or recycled uranium in order to breed plutonium most efficiently. In these fast reactors thorium is used in the radial and axial blankets in order to breed ^{233}U. When sufficient quantity of ^{233}U is stockpiled, it is to be recycled in thermal reactors in combination with thorium. It is possible to have a self-sustaining ^{232}Th–^{233}U fuel cycle in thermal reactors, including PHWRs and High Temperature Gas cooled Reactor (HTGR). With such combined cycles, it is theoretically possible to utilize all the uranium and thorium resources

in nature for generation of nuclear power. The combined cycle is most attractive for countries like India and Brazil which have large reserves of thorium.

Nuclear power in India

From the very inception of nuclear power programme in India, in the mid 1950s, great emphasis was given to self-reliance and indigenization. Accordingly, a three-stage nuclear power programme (Figure 6), linking the fuel cycles of PHWR and LMFBR was planned for judicious utilization of limited and low grade ($\leq 0.1\%$ U_3O_8) uranium ores (78,000 tonnes) but vast thorium resources (>360,000 tonnes) in India. Thorium utilization has been one of the most important long term objectives of the Indian nuclear power programme.

Apart from the two Boiling Water Reactors (2×160 MWe) at Tarapur, there are eight operating PHWRs in India of total capacity 1520 MWe. Four 220 MWe PHWRs are in different stages of construction, two each at Kaiga and Rawathbhata. An indigenous design for 500 MWe PHWRs is also ready and construction activities of two such units would start soon at Tarapur. The known uranium resources in India can support a PHWR-only programme of around 10,000 MWe. All aspects of PHWR fuel cycle, namely mining and concentration of uranium, fabrication of uranium fuels, use of these fuels in PHWRs, reprocessing of spent fuel, fabrication of mixed uranium plutonium oxide fuel and treatment and fixation of nuclear wastes are being carried on an industrial scale in India. In addition to PHWRs and LMFBRs, it is proposed to include LWRs and Advanced Heavy Water Reactors (AHWRs) in the Indian nuclear power programme in the coming decades. The nuclear fuel cycle programme in India, implemented in different units of the Department of Atomic Energy is shown in Figure 7.

As a first step to LMFBR programme, India could leapfrog and use an advanced LMFBR fuel of high breeding ratio and thermal conductivity, namely, the mixed uranium plutonium monocarbide, for the first time in the world, in the Fast Breeder Test Reactor (FBTR) at Kalpakkam. FBTR is operating with a unique and hitherto untried plutonium-rich mixed carbide fuel namely $(Pu_{0.7}U_{0.3})C$ since October 1985 (ref. 12). A Prototype Fast Breeder Reactor (PFBR) of 500 MWe is at the design and development stage. Though the reference fuel for PFBR is mixed uranium plutonium oxide, mixed uranium plutonium mononitride and U–Pu–Zr are being developed as alternate stand-by advanced fuels. It is proposed to construct several LMFBRs of capacity 500 MWe by the first quarter of the 21st century. 'Th' blankets would be mostly used in these LMFBRs to breed and stock pile ^{233}U for the third stage of the nuclear power programme, where ^{232}Th–^{233}U fuel cycle would be used in some advanced thermal reactors, to be decided later.

Plutonium and thorium fuels

The present generation of nuclear power reactors all over the world derive energy from the fission of ^{235}U by slow or thermal neutrons mainly in LWRs and to some extent in PHWRs. In LWRs and PHWRs, 2–4% enriched ^{235}U and natural uranium respectively are used as fuel in the form of high density ($\geq 94\%$ TD) UO_2 pellets clad in zircaloy-2 or zircaloy-4. In these reactors, the coolant water temperature and pressure are kept in the range of 280–330°C and 70–150 MPa respectively. Zircaloy is chosen as cladding material because of its low thermal neutron absorption cross section, excellent corrosion resistance and good mechanical properties at elevated temperature. The water cooled reactors, apart from generating electricity, produce ^{239}Pu and depleted uranium as by-products. The plutonium is best utilized as primary fuels in LMFBRs because of high breeding ratio (>1.0). Since there has been a delay in the progress of LMFBRs technology several countries in the world, including France, Germany, Belgium, Japan and India are considering interim recycling of plutonium in the form of mixed uranium–plutonium oxide (MOX) in LWRs and PHWRs. The MOX fuel for water cooled reactor would contain plutonium in the range of 1–8%. It is also possible to recycle plutonium in combination with thorium in PHWRs in the form of $(Th, Pu)O_2$ with the plutonium content not exceeding 4%. The ^{233}U produced from thorium could be recycled and substituted for plutonium in subsequent cycles till the PHWRs are completely converted to a self-sustaining $(Th, {}^{235}U)O_2$ fuel[13].

The fuel cycle cost can be reduced to a great extent by improved burn-up of the fuel and by minimizing the fuel fabrication cost. Burn-up is defined to be the amount of energy extracted from the fuel per unit weight before it is withdrawn from the reactor either because of its premature failure or inadequate fissile material content and poison built up.

UO_2, PuO_2 and ThO_2 are isostructural (FCC, CaF_2), completely solid soluble and have very similar thermodynamic and thermophysical properties. The oxide or mixed oxide fuels of the actinides are fabricated by the classical 'powder-pellet' route consisting of cold-pelletization and high temperature (1600–1700°C) sintering in hydrogen atmosphere, starting from UO_2 ThO_2 and PuO_2 powders. High density ($\geq 94\%$ TD) pellets of UO_2, $(U, Pu)O_2$ and $(Th, Pu)O_2$ containing up to 4% PuO_2 with controlled porosity ('closed', uniformly distributed and diameter greater than 10 micron) have been reproducibly fabricated at BARC by the 'powder-pellet' route and tested in the pressurized water loop (PWL) of

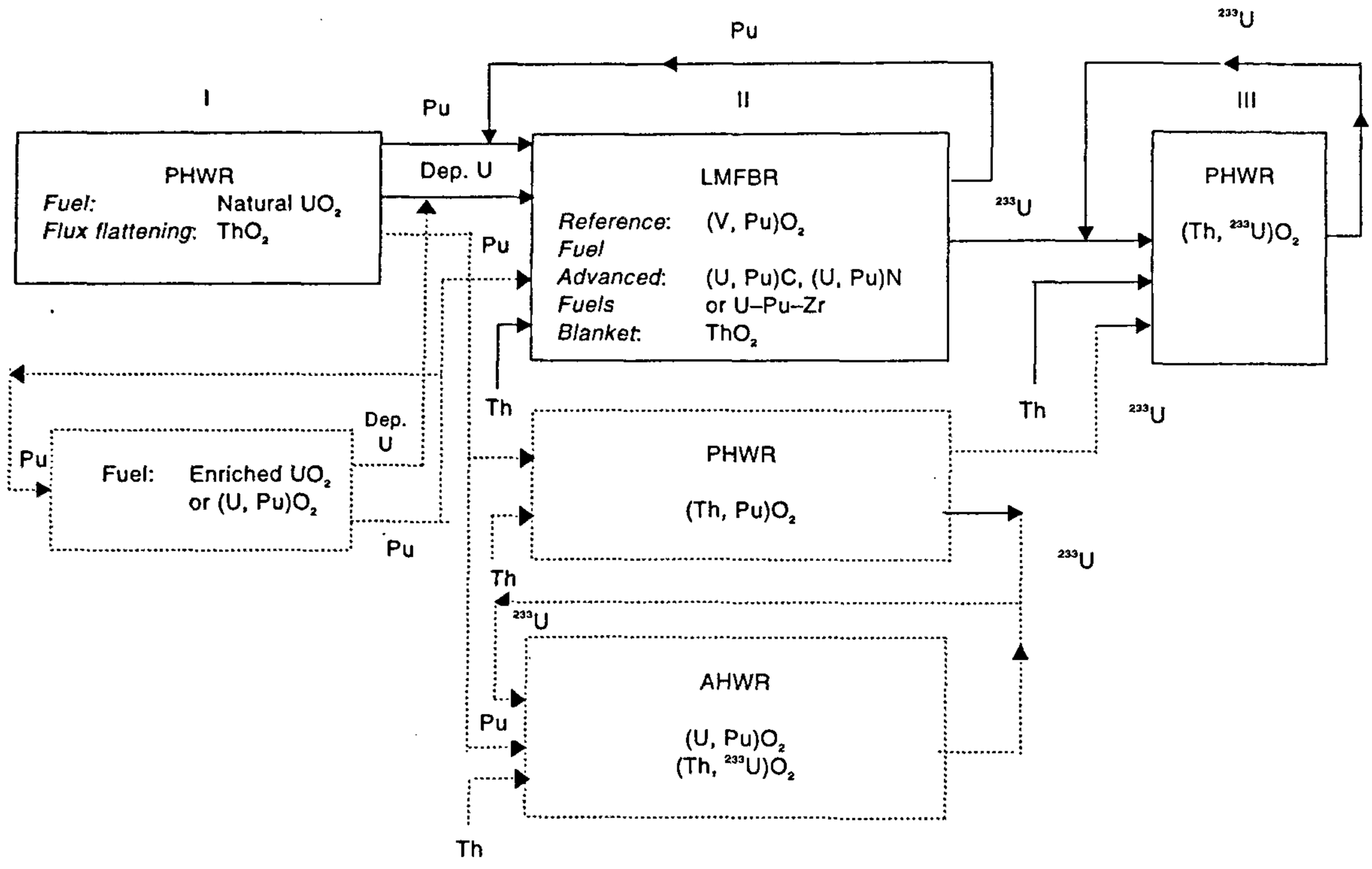

Figure 6. Nuclear fuel cycles and power programme in India.

CIRUS reactor at Trombay. Post-irradiation examinations of these zircaloy-2 clad fuel pins in the hot cells at BARC have revealed their satisfactory performance up to burn up of ~20,000 MWD/t. Additional irradiation testing of zircaloy-2 clad ThO_2 and ThO_2–6.7% PuO_2 fuel pins have been in progress in CIRUS, PWL for more than $2\frac{1}{2}$ years without any fuel pin failure. On the basis of these satisfactory results, two 6×6 zircaloy-2 clad mixed uranium plutonium oxide fuel assemblies have been introduced in Unit I of the Tarapur Atomic Power Station (TAPS) in July 1994 and five more MOX assemblies are to be introduced in Unit II of TAPS by the end of 1995 (Figure 8). The plutonium fuel fabrication facilities at Trombay and at Tarapur would be able to meet the requirement of MOX fuels for the two BWRs at Tarapur and the two PHWRs at Rawatbhata (RAPS I and II).

The advanced fuels for water cooled reactors and LMFBRs aim at judicious utilization of uranium and thorium resources and achieving high fuel burn-up without failure. For LMFBR, an additional requirement is high breeding ratio or in other words, short doubling time. The mixed uranium–plutonium monocarbide (MC) and mononitride (MN) fuels[14] belong to the same family and are considered to be advanced LMFBR fuels all over the world, compared to mixed oxide, because of their higher heavy atom density, higher breeding ratio, shorter doubling time, higher thermal conductivity and excellent chemical compatibility with sodium coolant. The carbide fuel fabrication facility at Trombay (which could also be used for MOX and MN) is unique in the world and has the capacity to meet the annual requirement of fuel for FBTR. India is one of the very few countries in the world to have experience with mixed carbide and nitride advanced fuels[15]. The mixed carbide fuel has so far reached a burn-up of around 16,000 MWD/t in FBTR and is expected to reach a target burn-up of 25,000 MWD/t and beyond without failure.

The advanced methods of fabrication of ceramic nuclear fuel pellets for thermal and fast reactors mainly aim at reduction in the personnel exposure to radiation and improved microstructure. The man-rem in fuel fabrication plant is minimized by automation, remotization and by developing fabrication flowsheets that do not involve generation and handling of fine powders, so that radiotoxic dust hazards are minimized. In addition, the advanced methods for fabrication of fuel pellets aim at minimizing fuel fabrication cost by reducing fuel synthesis and sintering temperatures, as shown in Table 3.

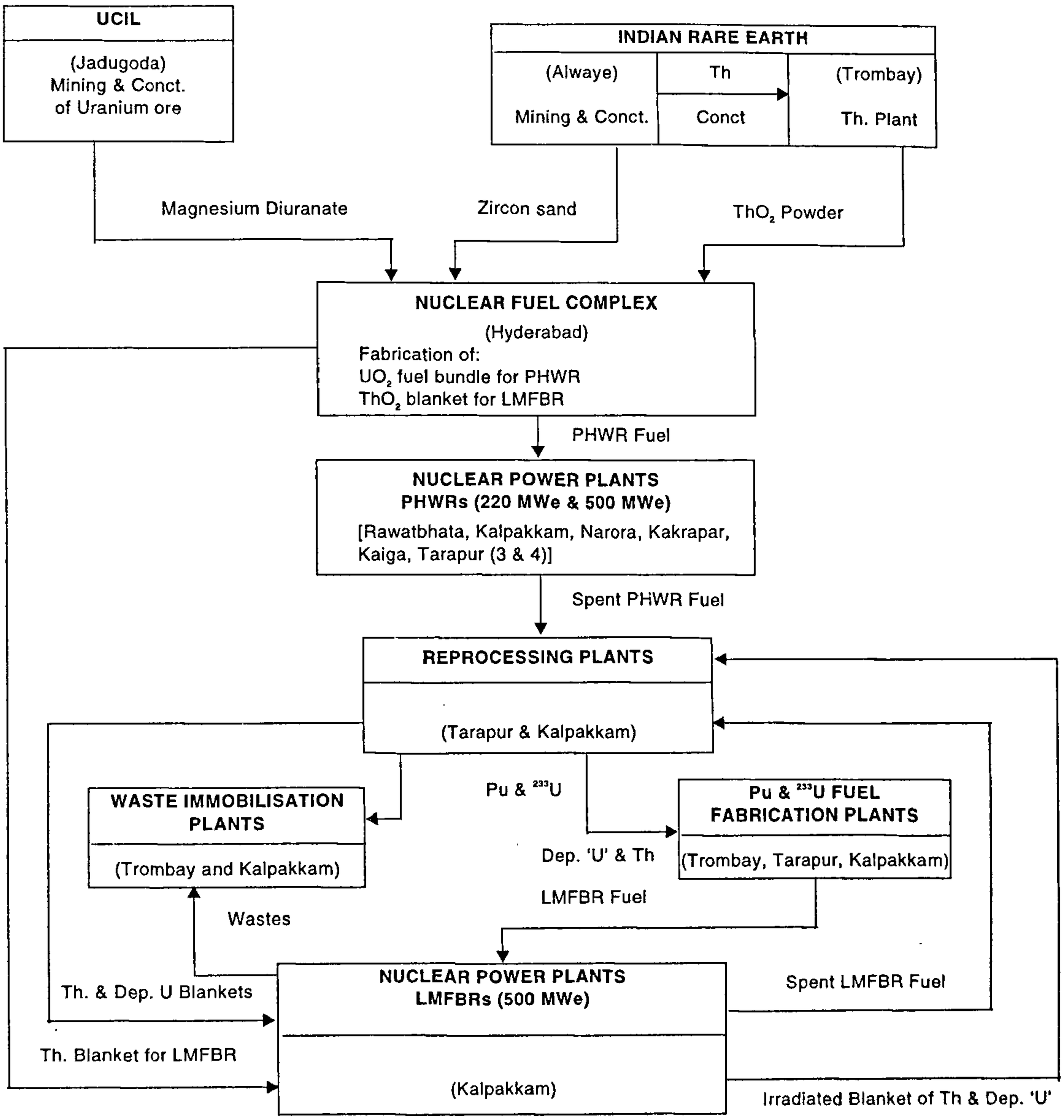

Figure 7. ^{238}U–^{239}Pu and ^{232}Th–^{233}U fuel cycles in India involving PHWRs and LMFBRs.

The sol–gel microsphere pelletization (SGMP) process, developed in BARC, is a dust-free method of fabrication of oxide, carbide and nitride fuel pellets of controlled density and microstructure starting from nitrate solution of uranium, thorium and plutonium. The nitrate solutions are subjected to 'ammonia external or internal gelation' processes to obtain free-flowing hydrated gel-microspheres of the fuel oxide (400–1200 micron diameter), which after controlled calcination leads to oxide microspheres (200–600 micron diameter) suit for direct pelletization and sintering[16–17]. For prep tion of 'porous' oxide microspheres or microsphere carbide or nitride fuels, carbon black is mixed with sol or solution prior to gelation. The hydrated oxide crospheres containing black carbon particles are calcined to obtain 'porous' oxide microspheres or s jected to carbothermic synthesis in vacuum or flow N_2 to obtain carbide and nitride microspheres resp

Table 3. Objectives of advanced methods of fabrication of ceramic nuclear fuel pellets

Safety	Economics	Performance
• Avoid generation or handling of fine powders of fuels	• Minimize process steps	• Tailor-make microstructure of fuel pellets for higher burn up
– for minimizing 'radiotoxic dust hazard'	• Reduce fuel synthesis and Sintering temperatures	– high density (≥94% TD) 'closed' pore and large (≥40 μm) grain size (LWR and PHWR)
– for minimizing 'fire hazard' (for carbide and nitride)	• Reduce gas cost during synthesis and sintering	– low density (≤85% TD) 'open' pore and small grain size (< 5 μm) (LMFBR)
• Fabrication flow sheet should be easy for automation and remotization	– use recirculation and purification	– excellent microhomogeneity of fissile material
– for minimizing personnel exposure to radiation	– use less expensive gas	– avoid fine pores (< 1 μm)
	• Reduce process losses and rejects	

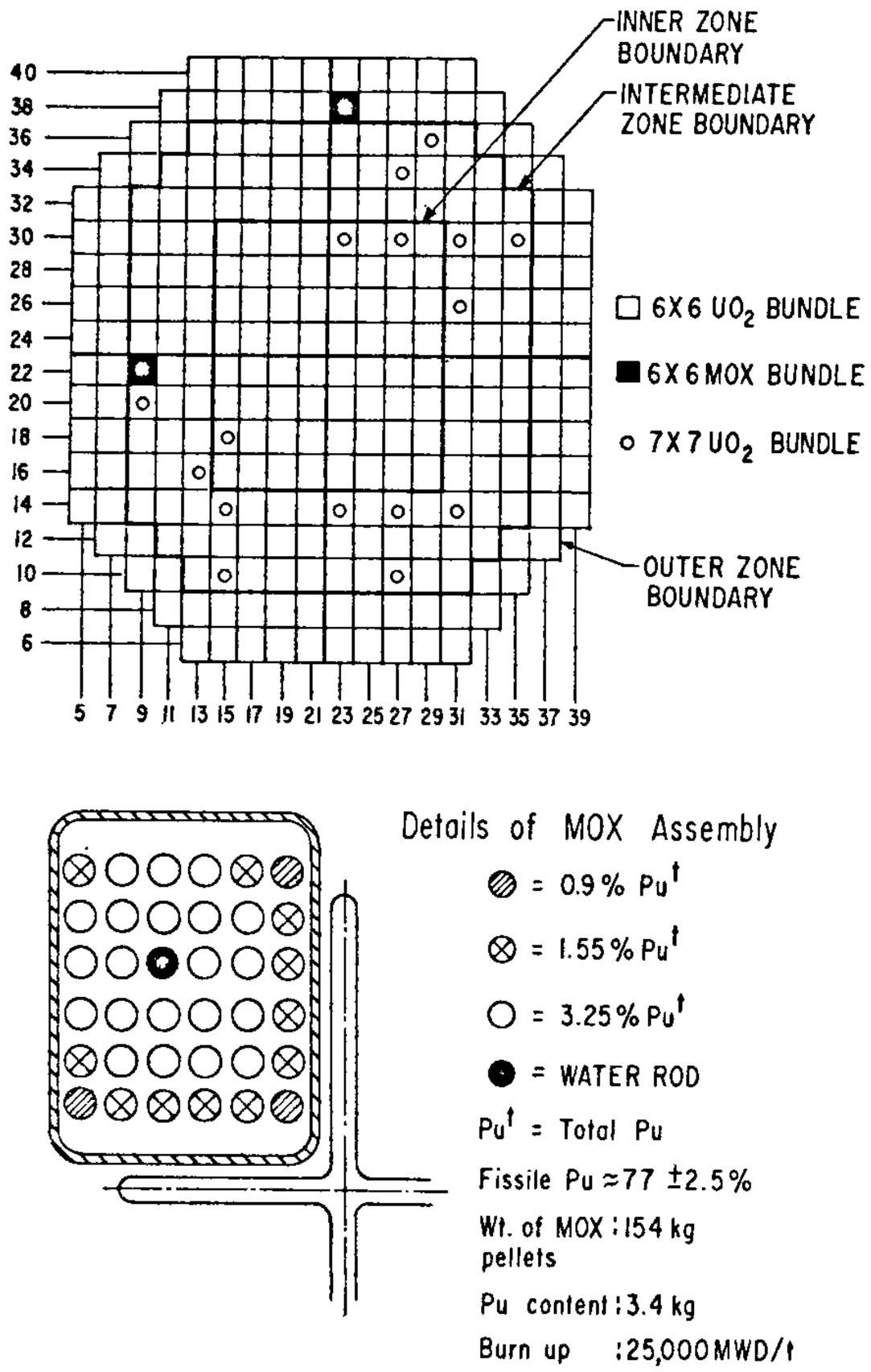

Figure 8. Present core configuration in TAPS I with two mixed uranium plutonium oxide (MOX) assemblies.

tively. For fabrication of UO_2 and (U, Pu)O_2 pellets for PHWR and LWR, BARC has also developed[18] the combined and unique sol–gel microsphere pelletization–low temperature oxidative sintering (SGMP–LTS) process, which facilitates automation and remotization, avoids radiotoxic dust and pyrophoricity hazards, minimizes fuel fabrication cost and produces fuel pellets of improved microstructure suitable for achieving high burn-up. Figure 9 summarizes the important process steps in 'powder-pellet' and SGMP processes for fabrication of oxide, carbide and nitride fuel pellets.

^{232}Th–^{233}U fuel cycle – Prospects, problems and advances

Irradiated thorium bundles from CIRUS research reactor have been reprocessed for recovery of ^{233}U. This ^{233}U has been utilized for fabrication of aluminium clad, Al-20% ^{233}U plate fuel element for the PURNIMA III critical facility at Trombay. The same fuel would be utilized in the 30 kWt research reactor, KAMINI at Kalpakkam. India had reached a major milestone in thorium utilization, in the beginning of 1994, with the use of zircaloy clad ThO_2 bundles, in place of depleted UO_2, in the PHWR-220 MWe of Kakrapar Unit I for neutron flux flattening[19]. In Kakrapar Unit II also, ThO_2 has been introduced for the same purpose. In fact, all subsequent PHWRs in India would use ThO_2 for flux flattening. In addition, several tons of stainless steel 316 clad ThO_2 pins have been fabricated for use as axial and radial blanket materials in the second core of FBTR.

An alternative scheme for interim utilization of thorium would be in an Advanced Heavy Water Reactor (AHWR) which has several passive safety features[20]. AHWR would derive between 75 and 80% of its energy from thorium in a self-sustained mode of ^{233}U-Th. This reactor would, however, need small input of Pu which would be in the form of mixed uranium plutonium oxide and will contribute to 20–25% of the power output of the reactor. In one of the versions, MOX driver fuel is

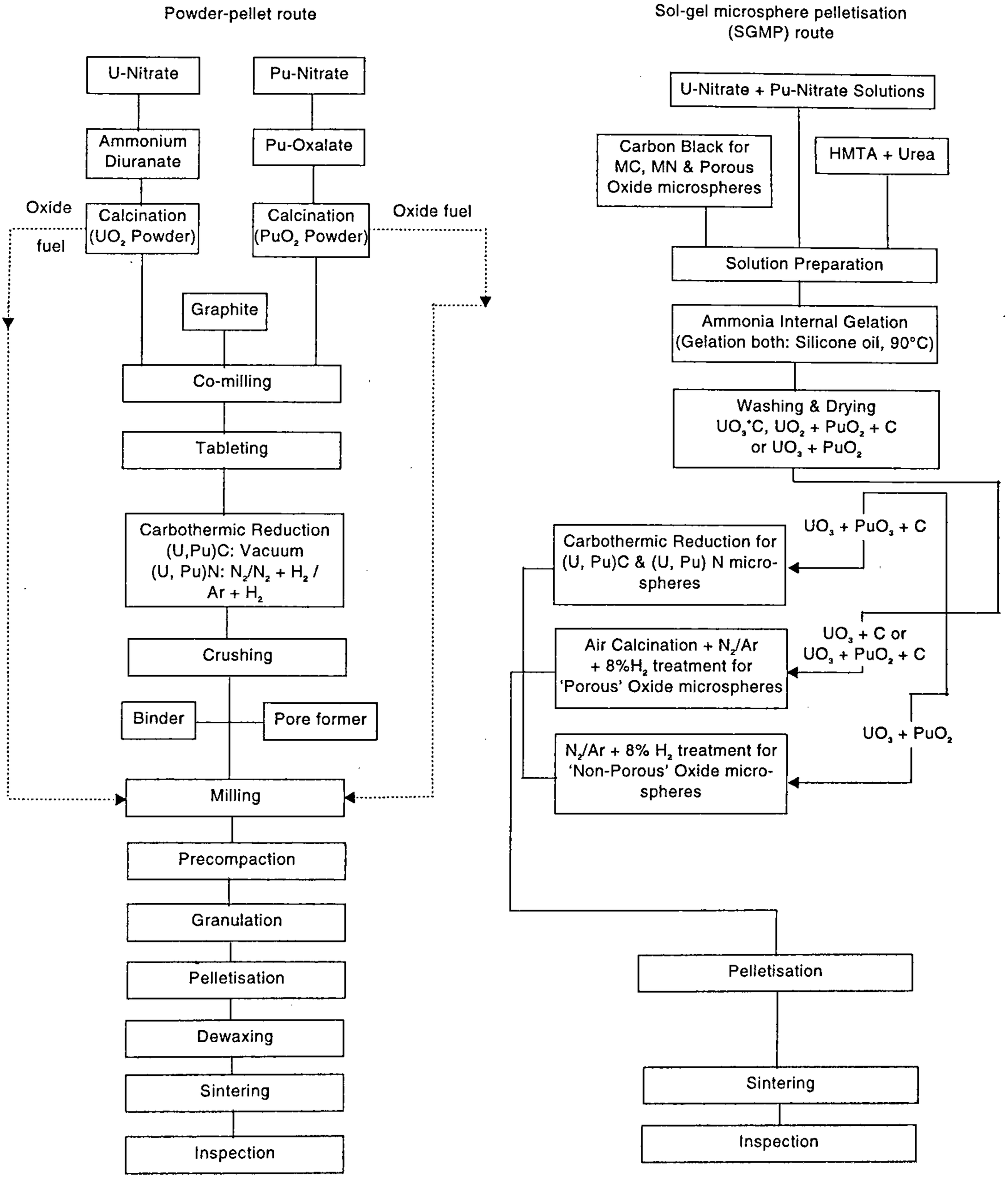

Figure 9. Flowsheets for fabrication of UO_2, (U, Pu)O_2,(U, Pu)C and (U, Pu)N fuel pellets by 'powder-pellet' and 'SGMP' routes.

Table 4. Comparison of processes for isotopic purification

Process	Separation factor for $^{232}U/^{233}U$	Process characteristics for ^{233}U clean up
Gas centrifuge	10.7	Large area of radioactive contamination
Gas diffusion	1.0012	Large area and high inventory
Separation nozzle	1.005	Large area
Chemex process	1.002	High inventory, not suitable for fissile material
Laser separation	>100	Compact, low inventory

located in discrete islands at a limited number of positions within the reactor core. This core configuration is ideally suited for incorporating neutron source targets for driving the balance sub-critical reactor core through the use of accelerator produced neutrons.

Thorium fuel cycle is becoming increasingly important for burning large quantities of weapon grade plutonium which are getting released on account of dismantlement of nuclear warheads in Russia and USA. Of all the options available for burning this plutonium, thorium cycle offers the promise of extracting maximum energy out of plutonium as well as ensuring that the resultant spent fuel would be proliferation-resistant and at the same time contribute to enhancing global energy resources. Another reason for thorium's gaining importance is on account of the fact that production of long-lived actinides waste products is several orders of magnitude smaller with the thorium–uranium cycle as compared with the uranium–plutonium cycle.

Laser purification of ^{233}U

The conventional uranium isotope separation processes namely, gas centrifuge, gas diffusion and chemical exchange have separation factors close to 1.0 for ^{233}U–^{232}U separation and are therefore not attractive (Table 4). In addition, these processes require large number of units and stages spread over large areas, which have to be shielded against gamma activity. The alternative atomic vapour laser isotope separation (AVLIS) technique is a compact and single step process with high selectivity and separation factor greater than 100. In this technique, a small isotope shift (0.01 to 0.03 Å), in the resonance excitation spectra of uranium can possibly be exploited by using tunable lasers, which have bandwidths much narrower than the isotope shift[21]. Fortunately, ^{232}U absorption spectrum does not have hyperfine splitting, which makes the entire population more accessible to the laser. ^{232}U atom (which has an ionization potential of ~6.2 eV) can be selectively photoionized by 1-photon (> 6 eV), 2-photon (~3 eV) or 3-photon (~2 eV) processes. The 3-photon processes use visible spectrum optics and lasers with economic advantage.

For a 200 MWe thermal reactor (like our ongoing PHWRs or the proposed AHWR), the amount of ^{233}U which may have to be processed and put back into the reactor at equilibrium is around 150 kg/y. To support 5 to 6 such reactors, ~1000 kg/y of ^{233}U containing 1 kg of ^{232}U has to be processed producing, say, 990 kg of clean ^{233}U containing a few ppm of ^{232}U, leaving 10 kg of waste ^{233}U containing a large quantity of ^{232}U.

Nuclear waste – Programme and strategies in India

As in other industrial activities, wastes are generated during various steps of the nuclear fuel cycle, namely, mining, fuel fabrication, reactor operation, fuel reprocessing and decommissioning. The major areas of nuclear waste generation are nuclear power plants and spent fuel reprocessing plants. Since the inception of the nuclear energy programme, R&D work on indigenous development of matrices and processes was started at BARC, Trombay which has paved the way for safe management of various types of wastes from nuclear facilities in India.

Low and intermediate level wastes

The liquid waste from nuclear power plants basically contains fission products (^{131}I and ^{137}Cs) and activation products (^{60}Co). Chemical precipitation method is used for removal of trace quantity of fission products from liquid effluents and the resultant sludges are immobilized in vermiculite cement composite matrix[22]. In order to further improve the decontamination factor, an ion-specific exchanger has been developed at BARC which has given encouraging results on a pilot scale. Use of these materials will further enhance the effectiveness of waste management plants for low and intermediate level waste. Similarly, use of ion specific exchanger is helpful in separation of useful isotopes like ^{90}Sr, ^{137}Cs, etc. from the waste. This helps in very significant reduction of final waste volume and resultant savings in the ultimate disposal space.

High level liquid waste

The category of nuclear waste which is getting worldwide attention is high level liquid radioactive waste. This stream is generated during reprocessing of spent nuclear fuel and contains about 88% of all radioactivity generated in the entire nuclear fuel cycle. However, the volumes generated are comparatively small. The major components of this waste are fission products and acti-

nides. The decay heat due to the presence of fission products makes it necessary to continuously cool the waste; at the same time long-lived isotopes in the waste make it imperative that it is isolated from the human environment for extended period of time.

High level waste (HLW) management – Indian efforts

HLW immobilization. Various matrices have been extensively studied for immobilization of high level waste. Among these, borosilicate glass and ceramic-based systems have been found acceptable from the point of view of desired product characteristics. At BARC, sufficient expertise has been developed in this frontier technology and this culminated in setting up of the first Indian vitrification facility as a part of Waste Immobilization Plant (WIP) at Tarapur[23]. Due to prevailing high radiation field, all operations and maintenance jobs are carried out remotely using various viewing aids (oil-filled glass shielding windows and CCTV) and remotization gadgets (like power manipulator, master slave manipulator and specially designed grapplers).

Vitrified waste product – storage and surveillance. The vitrified waste product canisters are stored in an air-cooled engineered facility for surveillance and decay heat removal. The surveillance programme consists of long-term evaluation of thermal stability, chemical durability and homogeneity of the waste products under repository conditions. To meet this objective, various hot cells and experimental facilities have been established at the waste-treatment facilities at Tarapur, which consist of ultrasonic core drilling for specimen preparation, ICP-AES for leachant analysis and XRD/SEM/EDX systems for homogeneity studies[24]. This R&D work is done as a part of international collaborative work co-ordinated by IAEA.

HLW – final disposal. After interim storage for a period of 20–30 years, high level waste products will be placed in a geological disposal facility at an optimal depth adopting a multi-barrier approach. The selection of suitable rock formation is a multidisciplinary activity involving physicochemical, thermal, thermomechanical, hydrological and radiological studies for detailed evaluation and assessment. Our repository programme includes performance evaluation of multibarrier system components, individually as well as jointly followed by theoretical modelling, laboratory and field investigation and *in situ* tests.

Actinides in HLW. There is a growing awareness amongst scientific community working on nuclear waste management with respect to separation of long-lived isotopes from the waste. Various laboratories are working on development of process and flow sheet for separation of actinides from HLW. These studies show that crown ether and carbamoyl methyl phosphine oxide (CMPO) are two promising extractants. These efforts have two dimensions. One, separated actinides like ^{237}Np, ^{241}Am and ^{241}Pu are suitable for recycling as a nuclear fuel. Secondly, long-lived isotopes which cannot be recycled could either be subjected to transmutation or immobilized in thermodynamically stabler matrices like ceramics. One such promising material is titanium-based assemblage of crystals, commonly referred to as synroc[25]. It consists of zirconolite, hollandite, pervoskite and rutile together with minor alloy phase. A distinct advantage of using such formulations is the improved isolation for extended periods of time.

Accelerators in nuclear fuel cycle

The task of disposal of the radioactive waste produced in the nuclear reactors in a cost-effective manner is complex and is being attacked, primarily, on two fronts. Firstly, to find ways to minimize the waste production and, secondly, to develop techniques to transmute the existing long-lived waste into short-lived waste. Attention has been focused on developing reactors which either recycle and transmute their own actinides (e.g. in the integral fast reactors) or specifically burn the actinides (actinide-fuelled fast reactors). Such reactors can operate in the hard neutron spectrum regime in order to exploit the high fission to capture ratio.

An alternative method with increased safety features is to operate such systems in subcritical state by feeding them with neutrons, externally. These are spallation neutrons which are produced using accelerators delivering very high power proton beams in the energy range of 1 to 2 GeV. Because of operation in sub-critical mode, a sudden excursion in power is not possible. In the event of an undesired power increase, the accelerator is quickly shut-down starving the system of the 'external' neutrons. This results in quick decrease in the power level, thereby ensuring safety of the system.

The other advantage of sub-critical assemblies lies in the fact that the 'externally' fed spallation neutrons are multiplied by fission as the system is reactor-like. This can make the accelerator-based transmutation cost-effective because the power produced in fission is not only sufficient to run the accelerator complex but is also available for feeding into the grid at, presumably, competitive cost. The construction of accelerators to produce such powerful beams approaches the cutting edge of the technology. While achieving the beam energy of 1 to 2 GeV is not difficult with the existing accelerator technology, accelerating beam currents of the order of 10 to 250 mA is rather tough, more so at low energy end of the accelerator complex because of space charge blow-up. Such beam currents are one to two orders of

magnitude higher than the existing machines. For the lower beam currents (several mA) cyclotrons are less expensive to build, but to obtain several tens or hundreds of mA, linacs seem to be the only solution. Linacs are very expensive machines to build (also import component is much higher in the Indian context). However, enough of confidence exists in the accelerator technologists to build both kinds of accelerators. The desired beam current depends on the type of application.

The so-called energy amplifier concept of Carlo Rubbia[26] employs a 1 GeV cyclotron complex delivering 10 mA proton beam[27]. The beam is directed on a Pb target inside the assembly producing spallation neutrons. The target is enclosed by a thorium–uranium-233 blanket; thorium is converted into ^{233}U while ^{233}U burns to release energy and the two can operate in a self-sustaining mode. Plutonium production is virtually non-existent. Fast neutron operation and lead moderator are preferred. Lead leads to much more homogeneous distribution of neutrons. 'Poisons' like ^{135}Xe are not produced (they are present in the thermal neutron case). In the transmutation mode, some selected long-lived fission products, e.g. ^{135}Cs, ^{129}I, are recycled in the rods depending on neutron availability. They are transformed into short-lived or non-radioactive elements. The energy amplification of 60 to 120 can be expected. In one such scenario, the system will feed the required 20 MW for the accelerator complex and in addition can feed minimum 180 MW to the grid.

For transmutation of the existing nuclear waste, several different concepts have been developed which differ on the grounds of the particular actinides to be burnt and/or the particular fission product to be transmuted. Depending on the application, fast neutron or thermal neutron regime is exploited. Complexity of the system varies accordingly and depends also on the number of operations to be performed simultaneously, namely, fission, transmutation, breeding, etc. The required beam current (energy of 1 to 2 GeV) depends on the amount of 'load' in the sub-critical assembly as well as on the operating multiplication factor. For example, calculations from Los Alamos show that a 1.6 GeV, 13 mA proton beam will convert waste of one 3 GWt LWR reactor and generate 1060 MWT power[28].

Concluding remarks

In spite of impressive progress made in the power sector in the country, power shortages continue to persist. The annual per capita electricity consumption for India is around 300 kWh, which is far below the world average and is a small fraction of that in developed countries like USA. The Central Electricity Authority has projected a demand of 142,000 MWe of installed capacity of electricity during the 15 years from 1992 to 2007. Currently the installed capacity is 81,000 MW with coal-based power stations providing the major part of the demand followed by the hydel ones. The energy demand of the country necessitates utilization of all forms of energy without exception and as efficiently as possible. All forms of energy have their own merits and their own constraints.

While coal-based power stations will continue to play a major part for many more years to come, they are likely to pose serious problems in the future arising out of transport of large quantities of coal across the country, apart from environmental problems related to disposal of ash and emission of greenhouse and acid gases. Clean technologies for thermal power are also more expensive. A 1000 MWe station needs 4–5 million tonnes of coal a year. Coastal shipping and high voltage DC transmission which are being proposed may alleviate the severity of the problem, but cannot be a solution if coal-based power stations have to bear the major burden of power production in the country. Further, reserves of coal are finite. It may not be possible to meet the future requirements without import of coal, which will pressurize India's foreign exchange resources.

Hydel power is cheap and has no emissions affecting the environment. But a hydel station occupies a large area and suffers from social constraints arising out of evacuation and rehabilitation of a large population and ecological considerations. No doubt, small hydro projects are making good progress, but capacity based on them will be limited. While every attempt must be made to utilize solar energy to the extent feasible, this form of energy appears good only as a decentralized source. Its economic viability is yet to be established. A 1 MW solar station needs nearly 7500 m^2 of mirror area which has to be sustained dust-free to maintain efficiency of the solar cells. Wind power is also now being availed for small power requirements as a local source. However, large power stations of this type will suffer from infrastructural problems, particularly as they will have a large number of wind mills with many moving parts. It is also reported to be interfering in TV transmissions.

As mentioned earlier, it is evident that to meet the power demand in India, all forms of energy with an appropriate harmonious mix is essential. In this energy mix, nuclear power has an important role to play in the coming years. France which has nuclear power constituting nearly 80% of its total electricity, Japan which has issued a White Paper stressing the importance of nuclear power for its national development, and China which has embarked on a large nuclear power programme, in spite of large reserves of coal, have chosen nuclear power for energy security and independence. This is also true for India. A programme of 20,000 MWe installed capacity with a mix of reactors including Fast Breeder Reactors in the course of the coming 25 years may constitute an effective base for nuclear power, from

which we can expand our programme into the later part of the next century.

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