

DR. H.L. ROY MEMORIAL LECTURE

Chemical Engineering Perspectives in Atomic Energy Programme

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DISTINGUISHED members of the Indian Institute of Chemical Engineers, Ladies and Gentlemen,

I am indeed delighted and consider it to be a matter of great honour for me to be asked by the Indian Institute of Chemical Engineers to deliver the Dr. H.L. Roy Memorial Lecture in the year 2000. Dr. H.L. Roy has been one of the most respected Chemical Engineers of our country. It was Dr. Hira Lal Roy, the elected overseas Vice President of Institute of Chemical Engineers, UK, who realised the importance of the professional body of Indian chemical engineers and alongwith two other stalwarts of chemical engineering profession, viz. Dr. G.P. Kane from Bombay, and Dr. M.A. Govinda Rao from Madras, founded the Indian Institute of Chemical Engineers in the year 1947 and dedicated his entire life for the growth of chemical engineering science in India. One feels extremely happy to observe that this versatile branch of engineering has always been playing an enviable role in meeting the basic needs and desires of the society. The task of providing sufficient food, adequate shelter, reasonable clothing and certain minimal medical health to our one billion people is stupendous, but to those involved, it is professionally satisfying. The perspectives of chemical industry in India are to be governed by Indian conditions, viz., high density of population, tropical climate, poor quality of water and others and these are entirely different from those prevailing in the Western world. In today's talk, which is dedicated to Dr. H.L. Roy, I propose to discuss the perspectives of chemical engineering in a major programme in India, viz. Atomic Energy.

1. Nuclear Power Option is Inevitable for India

The nuclear energy programme in India is directed towards our main two mandates, viz. production of electricity which is reliable, safe, economic and yet eco-friendly and production of radioisotopes for their applications in the fields of medicine, agriculture, industry and research (in the frontier areas of science and technology). We also continue to develop advanced technologies and do basic

research to support the two main mandates.

For sustained growth of nuclear energy in any country, there are three basic conditions that need to be satisfied — first of all, there should be a need for electricity; secondly, the various energy options available should favour nuclear energy; and finally, there should exist an appropriate technological base to exploit the nuclear energy. India meets these conditions unquestionably. With our per capita electricity generation of only about 480 kWhr (which is 20 to 30 times less than those prevailing in economically advanced countries), we do not have to debate further — definitely not with this audience — to arrive at the message that nuclear energy is not just essential, but it is an inevitable option for India if we are to improve the quality of our vast population. We must ensure rapid induction of additional electrical power from all available resources (Table-1) — from our thermal resources (coal), from hydro-resources (which has the associated problems of submergence and rehabilitation) and finally more and more from our nuclear resources.

Table-1-Potential of all Resources for Power Generation in India

Resource		Amount	Potential (Gwe - YR)
A.	Conventional		
i)	Coal	206 Billion Tonne	41,000
ii)	Oil	0.75 Billion Tonne	300
iii)	Natural Gas	692 Billion Cu.M	250
iv)	Hydro	84 GW At 60% PLF	84 GW at 60% PLF
B.	Nuclear		
i)	Uranium	78,000 Tonne U-Metal	In PHW Rs - 420 In FB Rs - 54,000
ii)	Thorium	518,000 Tonne Th-Meta	In Breeders 358,000
			Total Nuclear : 4,12,420

- Does not include Non-Conventional Energy Resources
- Assumptions in Calculations of Potential
- For Coal, Oil and Gas: Complete source is used for electricity generation with $\square = 30\%$ and calorific values for coal = 5000 kcal/kg. Oil = 10,200 kcal/kg & Gas = 9150 kcal/m³
- For Nuclear : Fuel burn-up in PHWRs = 6700 MWD per tonne of U & $\square = 29\%$
- FBRs can use 60% uranium with $\square = 42\%$
- Breeders can use 60% thorium and $\square = 42\%$

* Dr. Kakodkar, Chairman, Atomic Energy Commission, presented this lecture at CHEMCON-2000, in Calcutta

2. Three Stage Nuclear Energy Programme in India

Our uranium resources are limited and can sustain a modest programme of electricity generation based on Pressurised Heavy Water (PHWR) presently being set up under our nuclear power programme. However, our thorium reserves are vast (third largest in the world) and the real impact of our nuclear power programme will depend on our success to tap nuclear power from this source. For ensuring a good growth potential of the nuclear power, a three stage programme was accordingly formulated for development of nuclear power in the country based on our modest uranium and vast thorium resources. In the first stage, our uranium resources would be utilized in PHWR systems for electricity generation by burning its U-235 fissile isotopes along with part of the Pu-239 fissile material (generated by conversion of the fertile U-238 isotopes content of natural uranium).

Based on the fissile Pu-239 and the depleted uranium from the thermal reactors of the first stage, second stage of the programme would involve installation of a series of Fast Breeder Reactors (FBRs) which would breed more fissile material Pu-239 in the U-238 blanket material than what would be consumed in the core of the reactor.

Third stage of our nuclear power programme would be based on yet another type of reactors which use Th-U-233 at the core to produce excess U-233 material at their thorium blankets. Following the three stage programme, it would be possible to generate about 3,58,000 GWe-Y of nuclear power from our thorium reserves in the country.

3. Present Status of Nuclear Energy Programme of India

Setting up of nuclear power plants calls for development of a host of technologies that are highly complex and involved. Our R&D efforts over the last four and half decades have already started paying rich dividends in our entire programme of nuclear science and technology. The first phase of our nuclear energy programme is firmly established with 12 units of our indigenous PHWR nuclear power stations with 2400 MW(e) installed capacity (excluding 2 units of BWRs) are performing as per world standard, both in terms of capacity factor (>80% average) as well as safety standards. Two additional PHWR units of 500 MW(e) each are under construction.

For our Fast Reactor development programme of the second stage, a Fast Breeder Test Reactor (FBTR) has been constructed and commissioned at IGCAR, Kalpakkam, Chennai along with a string of supporting R&D facilities needed for indigenous development of all the necessary technologies involved in FBR fuel cycle. Design work for

a 500MW(e) Prototype Fast Breeder Reactor (PFBR) based on mixed oxide fuel (Pu-U)₂O₇ has since been the prime activity at IGCAR. Parellel to the R&D activities for indigenous development of various sub-systems of PFBR, a detailed project report of 500 MW(e) PFBR is nearing completion. Subject to availability of financial support, the construction of the first unit of 500 MW(e) FBR is expected to start in the year 2002.

Similarly, for implementation of our third stage, we have established utilisation of thorium for initial power flattening in existing PHWR core. Instead of using 384 bundles of depleted uranium with ~0.6% w/w U-235 for initial flux/power flattening in a PHWR, we have already started introducing thorium bundles (35 bundles/reactor).

4.0 Chemical Engineering Science and the Nuclear Energy Programme in India

With the Reactor system located at the center of the entire spectrum of activities involved in what we call "Nuclear Fuel Cycle" (Fig.1), the dominance of chemical engineering activities are clearly visible not only in the technologies needed to provide all the necessary inputs (also called "Front End Activities") to the reactor system but also in those involved to handle the outputs (also called "Back End Activities") from the reactor system. In fact, DAE chemical engineers are equally involved in the entire spectrum of "Nuclear Fuel Cycle", right from production of nuclear fuel from the ore deposits to the final disposal of the high level waste deep into a geological repository.

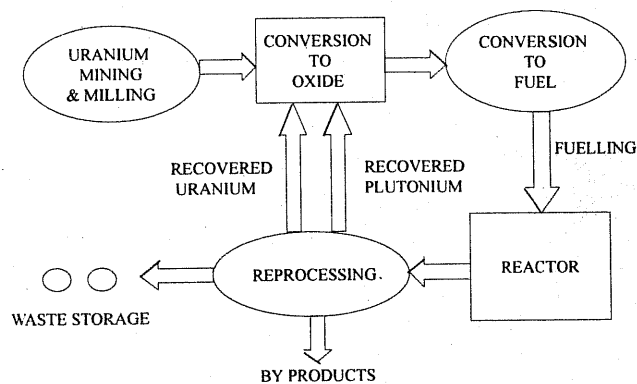


Fig 1 : The Nuclear Fuel Cycle

For proper appreciation of the perspectives of chemical engineering in our totally indigenous nuclear energy programme based on the present generation of Pressurised Heavy Water Reactor (PHWR) systems, I would like to draw your attention first to two simplified Tables (Table-2A & Table-2B) that show the various major inputs and outputs of our totally indigenous 220 MW(e) and 500

MW(e) PHWR systems. This task of chemical engineers gets immensely magnified because of some added complexities, viz. technologies to be developed for the "front end" as well as the "back end" activities must ensure not only the production of all the materials involved with stringent quality standard (of what we call "nuclear grade") but also guarantee release, if any, of various radioactive wastes strictly as per International Commission for Radiation Protection (ICRP) standards. Coming to their involvement in the nuclear reactor itself, the role of chemical engineers is equally significant found in the design and analyses of "reactor thermal hydraulics" as well as "reactor safety".

Table 2A - Major Input to PHWR Systems

Front End Activities	220 MW(e)	500 MW(e)
1. Fuel Elements	Tonnes	Tonnes
1.1 Initial Core Loading – UO ₂	60	125
Annual Requirement at 80% capacity factor - UO ₂	42	84
1.2 Initial requirement for		
- Pressure Tubes (Zr, 2.5 Nb)	12.3	30
- Calendria Tube (Zircalloy-4)	5.2	9.7
- Cladding of Fuels (Zircalloy-4)	5.5	12.4
Periodic requirement for		
- Cladding of Fuels (80% CF) - Annual	3.8	9.8
- Pressure Tubes replacement (80% CF) once in 10 years upto KAPP-1 and once in 20 years KAPP-2 onwards	-	-
2. Moderator/Coolant - D ₂ O		
Initial Loading	275	500
Annual Make-up	12	8

Table 2B - Major Outputs from PHWR System

Back End Activities	220 MW(e)	500 MW(e)
1. Electricity MW(e)	220	500
2. Spent Fuel (Reprocessed & separated Pu/U recycled as MOX fuel) - % of annual U-fuel	99.9%	99.9%
3. High level wastes (HLW) (separated during reprocessing and subjected to calcination & vitrification) - % of annual U-fuel	0.1%	0.1%
3.1 Amount of vitrified HLW - Cubic meter/year (max.)	33	66

4.1 "Front End Activities" of Present Nuclear Fuel Cycle

In the context of our first phase programme on nuclear

power based on our totally indigenous PHWR technology, the front end activities of chemical engineering science covers essentially the production of nuclear grade uranium, thorium, zirconium and heavy water. Success stories of DAE chemical engineers in producing the required quantities of nuclear grade uranium (from very low grade deposits of uranium at Jaduguda, Bihar), thorium (from monazite mineral of Kerala beach sand), zirconium (from zirconium material present in beach sands of Kerala) and heavy water (from both water source and H₂ source of syn-gas from fertilizer plants) are well known to all chemical engineering professionals in the country. Here, I would like to spend some time on some of the other major challenges to the chemical engineering professionals of DAE in the front-end activities, viz. production of uranium from phosphoric acid as well as from sea water (to augment uranium resources in the country), development of NH₃-H₂O Front End technology for Heavy Water industry, upgradation of the fluorine technology (that has been developed for production of fluorine for in-house consumption in some strategic applications) for making it available to Indian industries, production of Rare Earths and finally, the desalination technology coupled to nuclear power plants.

4.1.1 Uranium from Phosphate Rock

Phosphate rocks are recognised to be an important sustainable secondary source for uranium. Depending on the uranium content of the Indian phosphate rocks used, the phosphoric acid produced can contain 0.005 – 0.05 gms U₃O₈ per litre. Separation of uranium on pilot scale (~1 m³/hr) by solvent extraction employing synergistic combination of DEPHA with TOPO has already been successfully completed. Trials with most attractive alternative solvent mixtures like (DNPPA + DBBP) and (DNPP + TOPO) are also in progress. Recovery of uranium from phosphate rock of Mussorie has been taken up using high capacity (between 50–300 m³/hr capacity) solvent extraction equipment.

4.1.2 Uranium from Sea Water

Sea water contains around 3.3 µg of uranium per litre (3.3 ppb) and constitutes a very significant source of uranium from our sea water resources along the long coastline of our country. Table-3 shows the possible uranium inventory in different sea water outfalls in various nuclear power units in the country. Sorption is considered a suitable technique for selective extraction of uranium from sea water for which both inorganic as well as organic adsorbents are being tried out. Among the inorganic sorbents, hydrous titanium oxide has proved to be promising. On the other hand, amongst

the organic sorbents, amidoxime based sorbents has proved to have highest potential for recovery of uranium from seawater. R&D work has been initiated at BARC to synthesize and characterize the organic adsorbent based on Amidoxime by radiation induced grafting as well as by conventional chemical route. These organic sorbents could be prepared in sheet, fibre as well as in particulate form and our in-house expertise on development of membrane

economical aspects for coupling this $\text{NH}_3\text{-H}_2\text{O}$ exchange process at the front end of all heavy water plants based on $\text{NH}_3\text{-H}_2$ exchange process.

4.1.4 Upgradation of Fluorine Technology

Pursuing the goal of self-reliance in critical technologies, BARC had embarked on development of fluorine

Table 3 - Recovery of Uranium from Seawater

Site	Gross quantity of Uranium per year	Recovery planned per year (from experimental facility)	Adsorbent required *	Quantity of 40 μ fibre PP required in 1.5 mm non woven sheet form as stem material	
				Weight	Area
Trombay Estuary	Abundant	100 g	50 Kgs	100 Kgs	200 m ²
NDDP	80 Kg @	1 Kg	500 Kgs	1000 Kgs	2000 m ²
MAPS	4000 Kg @	10 Kgs	5000 Kgs	10000 Kgs	20000 m ²

* Based on 10 days submergence and a life of 20 cycles and adsorption rate of 0.1 mg/g/10 days.

@ Based on flow rates at outfall from respective plant.

technology will be effectively utilised for designing a suitable contactor for uranium sorption with minimal pressure drop under sea water flow conditions (without any external energy input). Initial laboratory scale experiments on the adsorbents, developed in BARC, has given promising results with respect to selective uranium recovery from sea water. However, we have a long way to cover before an acceptable module with optimum parameters is arrived at.

4.1.3 $\text{NH}_3\text{-H}_2\text{O}$ Exchange Technology

Dependency of Heavy Water Plants based on $\text{NH}_3\text{-H}_2\text{O}$ Exchange process on the adjoining Fertilizer plant for its hydrogen source (synthesis gas) has been known from the beginning. Accordingly, R&D efforts have been initiated in time to find a suitable technical solution for delinking the heavy water plants from adjacent fertilizers plants. Of late, with the advent of special catalysts being developed, fertilizer plants are favouring more and more of low pressure technologies for production of NH_3 . If heavy water plants are continued to be linked with such low pressure syn-gas source from fertilizer plants, economic production of heavy water cannot be sustained. This has necessitated accelerated R&D work by our chemical engineers for establishing a front end $\text{NH}_3\text{-H}_2\text{O}$ exchange technology at a pilot scale which can be coupled to the $\text{NH}_3\text{-H}_2$ exchange plants (for changing over of the feed source of deuterium from syn-gas of fertilizer plants to abundant natural water). Studies on this $\text{NH}_3\text{-H}_2\text{O}$ exchange at pilot scale has already been completed at Baroda based on which a demonstration plant is under construction for working out details of techno-

technology essentially for catering to the in-house need of one of our yet another major technology of strategic importance where chemical engineers of BARC have played a commendable role. The performance data of the fluorine cells developed by BARC compare well with those of industrial scale fluorine cells in the USA/UK. However, the only parameter which needs to be improved is Space – Time Yield ($\text{Kg/M}^3\text{-hr}$), which we have deliberately kept conservative by use of low current density in order to avoid certain engineering uncertainties and complexities (like polarisation which has a bearing on availability and efficiency of cell; and explosive recombination of fluorine and hydrogen which has bearing on safety service life, etc.).

In order to effectively utilize the knowledge base and expertise gained over the years of operating experience, priority has now been assigned for commercial exploitation of this technology. Employing cheaper material of construction with innovative design features to minimize corrosion, a large scale fluorine cell is presently being fabricated as a demonstration cell for commercial exploitation. It is hoped that this will facilitate establishment of a host of fluorine based “Spin-off-technologies” of International Standard in the country.

4.1.5 Separation of Rare Earths

Purified rare earth elements with high neutron absorption cross sections, e.g. gadolinium and dysprosium, are useful as reactor control materials during start-up/and or operation. While Gd_2O_3 is already in use in existing PHWR reactors, Dy_2O_3 (dispersed in yttrium stabilised ZrO_2) is a candidate

material for our proposed Advanced Heavy Water Reactor (AHWR) as a burnable poison.

Extremely high purity (>99.99%) Rare Earth phosphors (like $Gd_2O_3:S:Tb^{+3}/GdVO_4:Bi^{+3}$) are excellent materials for detection of radiations in the nuclear energy applications, particularly in the field of microdosimetry. As x-ray phosphors, Rare Earth materials have revolutionised the medical applications of x-rays due to their high sensitivity (arising out of very high efficiency in conversion of x-rays to photons compared to that of conventional phosphors - $CaWO_4$) resulting in reduction of the exposure dose by a factor of four to five.

Strategic applications of rare earths include high powered pulsed laser for defence weapon systems as well as super conductivity materials (based on Y-Ba-Cu-O system with Tc 90). Other promising applications of rare earths include high sensitivity magnetic field detection (SQUID) and high powered permanent magnets.

Source of Rare Earths in India is the significant deposits of monazite (a source of the Rare Earths present alongwith thorium). Due to the close separation factors, Rare Earths are known for their difficult separation but the chemical engineers of BARC has established the processes based on solvent extraction using mixer-settlers for separation of high purity individual Rare Earths (starting from the mixed crude Rare Earths concentrate produced by Indian Rare Earths Ltd. at Alwaye, Kerala).

4.2 Desalination

BARC has presently undertaken establishment of a demonstration scale hybrid MSF-RO desalination plant coupled to our 170 MW(e) PHWR station at Kalpakkam, Chennai. This desalination project will have a 4500 m³/d (1 MGD) MSF plant along with a 1800 m³/d (0.4 MGD) SWRO plant to provide 6500 m³/d of desalted water which would be good enough to meet the dual needs of about 1000 m³/d process water for NPP and the drinking water for the neighbouring people. In this connection, it may be observed that the relatively lower thermal efficiency and generation of saturated steam at low pressure and temperature in our present generation PHWR system leads to higher steam consumption per MW(e) generated compared to that in the thermal power plants in the country. Availability of large quantity of steam at low pressure has certain techno-economic advantages for its utilisation in thermal desalination process.

This NDDP plant at Kalpakkam is in an advanced stage of construction and is scheduled for commissioning by mid 2002. At present this is the largest capacity seawater desalination plant in the world being set up which is coupled to a nuclear reactor. A larger capacity plant was earlier operated in former Soviet Union at Aktau (coupled to their

Fast Breeder Reactor) which is now closed down. There are some plants operating in Japan that are coupled to NPP but these are of small capacity catering to the needs of the process water requirements of the station.

The purpose of our NDDP at Kalpakkam is to demonstrate the feasibility, economic viability and safety of coupling the desalination plant with the nuclear power reactor. Based on couple of years of experience of this project, standardization of 10 MGD plants both on MSF process and RO process would be completed by the year 2005 that can be utilized for commercial use in the country.

5.0 Back End Activities of Present Nuclear Fuel Cycle

Reprocessing, refabrication and management of radioactive wastes are the major three areas of activities that constitute the back-end of nuclear fuel cycle. Out of these, reprocessing and waste management are of major challenges to chemical engineers. Reprocessing of spent fuel establish the main link between various stages of our nuclear power generation programme by way of providing the needed inputs to the second as well as third stages. The success of the overall nuclear energy programme, therefore, hinges on a safe, cost effective and efficient reprocessing programme.

Reprocessing of spent fuel is required to recover the valuable fissile material (e.g. Pu in case of uranium fuel) that has been produced in the reactor during irradiation of the fertile component (e.g. U-238 in PHWR) of the fuel alongwith unutilized portion of the original fuel (e.g. U-238 with residual U-235 in case of uranium fuel) from the unwanted fission products. The fissile as well as the fertile materials thus recovered are utilized to refabricate the nuclear fuel for further generation of electricity. The other major reason why countries like India, France and UK have developed the reprocessing technology is the added incentive of significant reduction in the volume of the high level radioactive waste (which accounts for 3.0% of the original fuel quantity). The radioactive wastes that result from separation of the fissile/fertile components of the fuel from the fission products are called High Level Waste (HLW).

5.1 Reprocessing of PHWR Fuel

Reprocessing is a chemical or electro-metallurgical process for separation of fissile material (e.g. plutonium in case of uranium fuel) from spent fuel. The main method followed world over is a technology based on a chemical separation process called "PUREX" (Plutonium Uranium Extraction) process which is quite complex and it was developed over fifty years ago to produce plutonium. India's first

reprocessing plant was set up at Trombay in 1964. Decades of expertise in design and operational aspects of reprocessing technology has already resulted in allround improvement in the areas of materials, process technology, instrumentation and automation techniques. Some areas of further R&D in reprocessing of PHWR Fuel are as follows:

i) State-of-the-Art Technology for Large Sized Reprocessing Plants

The need for reprocessing plants with large throughputs still pose challenge that calls for change in design philosophy, fabrication-erection technologies and O&M concepts to bring the reprocessing technology to the state-of-the-art level. Greater reliability needs to be built into the systems to ensure capacity utilization and to cut down plant outages. Greater automation to assist the operator; a new approach towards an integrated remote maintenance of the active equipment through increased application of robotics; reduction in occupational exposures and dealing with actinides are some of the areas that now call for major thrusts.

ii) Non-Aqueous Process of Reprocessing

For reprocessing of high burn-up fuels (e.g. spent fuel from Fast Reactors), the time tested PUREX process appears to be adequate if the problem of solvent degradation due to increased radioanalysis resulting from such high burn-up are taken care of by employing fast contactors (e.g. centrifugal contactors) to reduce the residence time alongwith more efficient clean-up procedures of the solvent. However, keeping in mind that nuclear fuels will reach progressively high burn-ups in future generation reactors and it would be desirable to recycle the reprocessed fuels from such reactors at much shorter cooling period (to be economically as well as ecologically attractive), various alternative concepts of pyrometallurgical reprocessing techniques, also called dry reprocessing techniques), are being studied throughout the world.

In addition to the main objective of attaining utmost simplification and shortening of number of steps in reprocessing process in pyrometallurgical techniques (by avoiding dissolution and handling of large amount of highly radioactive aqueous liquids) these dry processes have a number of added attractions, viz.:

- * Extremely low waste volume (because the actinides are recycled along with fissile materials leaving only the FPs).
- * Possibility of segregating the transplutonium actinides (present in the spent fuel) along with the plutonium stream (which not only opens up the possibility of recycling the separated transplutonium elements (as fissile material) along with plutonium but also avoids

the safety related problems associated with long term storage of vitrified waste product.

- * High radioactive resistance of inorganic reagents leads to shorter cooling time and hence low out-of-pile inventory.
- * Absence of criticality safety considerations, normally associated with aqueous techniques.
- * Compact plant size which allows such reprocessing plant to be integrated with nuclear power plant (thus avoiding the need for transportation of spent fuel to a centralized large capacity reprocessing plant).

5.2 Management of Radioactive Waste

The main objective of radioactive waste management is to deal with radioactive waste in a manner that protects human health and the environment — now and also in the future — without imposing undue burdens on future generations.

High Level Waste (HLW) constitutes about 99% of the total non-volatile radioactivity generated during the reactor operation and HLW from PHWR fuel contains i) FPs like Cs-137, Sr-90, Ru-160, Ce-144 etc., ii) corrosion products like CO, Ni, Mn, etc., iii) added chemicals like HNO_3 , iv) unextracted quantities of uranium and plutonium, and v) small amount of transuranic elements TRUS like trivalent actinides, e.g. Neptunium, Americium and Curium, which have very long half-lives.

High level waste is largely in liquid form which generates a considerable amount of heat due to radiation decay of the FPs and hence it needs cooling. Because of the highly radioactive FPs alongwith the presence of transuranic (actinides) elements with long life, HLW pose a hazard for extended periods of time. Disposal in deep underground geological formation is one option which has received wide attention world-wide. The basic requirements for geological formation to be suitable for the location of the radioactive waste disposal facility are i) remoteness from environment, ii) absence of circulating ground water and iii) ability to contain radionuclides for geological periods of time. In India the choice is restricted to igneous rock formations (granitic) and some selected sedimentary deposits. Indian programme of site selection and host rock characterisation for geological repository is in evaluation stage.

Current technologies that are adopted for treatment of HLW for its safe storage and transportation are vitrification and concrete solidification. Areas of further development in handling of HLW from PHWR are the following:

i) Development of Ceramic Melter

Challenges in vitrification encompass material performance at elevated temperature (1000-1200 deg.C) in contact with molten glass environment. Ceramic melters using Joule heating as an alternative to metallic melter for vitrification is being considered to mitigate high temperature corrosion of processing pot made out of Inconel-690. The Joule heated ceramic melter has an added advantage over the metallic melter as it offers continuous mode of operation, a homogeneous quality of product glass with the added attraction of operational flexibility.

ii) Development of Cold Crucible Process

The cold crucible process is an innovative solution to the day-to-day challenges facing radioactive waste management. Compared to electric-arc and induction furnaces with ceramic crucibles, the main merits of the cold crucible process are its compactness and long life-time of crucibles (~10 years). The technology employs inductively heated "cold crucible" melters which are small-sized units capable of operating at very high temperatures (~3000°C) without corrosion. This results into more efficient melting and opens up feasibility of a variety of viable glass and crystalline matrices for the final waste forms. A layer of solidified waste material between the crucible walls and the high-temperature melt provide the refractory function (by protecting the walls from corrosion). An industrial scale plant for vitrification of the intermediate level wastes is under successful operation in Russia since October 1997. At present no other technology matches the performance of cold crucible vitrification.

iii) Development of Synroc Technology

Although glass is a satisfactory immobilization medium, a newer technology — Synroc (synthetic rock) is a more sophisticated way to immobilize such waste since it offers significant improvements in long term performance. Synroc is an advanced ceramic form principally comprising three natural titanate minerals (hollandite, zirconolite and perovskite) which are geochemically stable and which together have the capacity to incorporate into their crystal structures nearly all of the elements present in high level radioactive waste (HLW).

iv) Need for Partitioning and Transmutation

As we have seen earlier, HLW from reprocessing plant contain small amount of TRUs (like Np, Am and Pu) which have extremely long half-lives along with long-lived fission products (LLFPs) like Cs-135, Tc-99, T-129 and shortlived ($t_{1/2}$ ~300 years) FPs. Since the short lived fission products (SLFPs) constitute ~90% of the initial radiotoxicity of HLWs, if we can separate these SLFPs and keep them under surveillance for ~400 years, the associated activity of this

part will go down to that of natural uranium levels which can be easily buried. Similarly, the separated actinides are no longer being considered as "wastes" but as valuable fissionable fuel materials which can be transmuted (converted) into stable or shortlived nucleides in low flux FBRs in a way as uranium/plutonium are recycled. In fact, LLFPs are also being considered to be ideally suited for transmutation in a high flux Thermal Reactor system (like Accelerator Driven Subcritical System (ADSS)). Broadly speaking, there appear several possibilities to transmute the actinides and ILFPs through nuclear reaction which would reduce the net radioactivity per KWHr of electricity generated by about 20,000 times.

While on the subject, it is to be kept in mind that as the radioactive nucleides of actinides/ILFPs can be transmuted by neutron capture into stable ones, a stable nucleus could also simultaneously get transmuted into radioactive one (P.S. Te-99 and I-129 do not require such separation). It is clear that such transmutation process calls for separation of the isotopes to be transmuted. At BARC, reprocessing schemes have been successfully developed using a powerful bifunctional extractant octl (phenyl), N, N-diisobutyl carbamoylmethyl phosphine oxide (CMPO). This extractant is mixed with a PUREX solvent to selectively extract the minor actinides (MAs) together with the lanthanides (rare earths) from strong acid solutions. This process known as TRUX (Trans-Uranium Extraction) is fully compatible with the currently followed aqueous PUREX process.

6. Advanced Heavy Water Reactor (AHWR)

Slow progress of FBR technology based on thorium worldover and realization of the fact that there is no specific advantage in use of U-233 in fast reactors (over its induction in thermal reactors — in fact there is definite disadvantage of Th/U-233 cycle in FBR because of more generation of U-232 with its highly radioactive daughter products) — engineers and scientists of BARC have taken the lead in the world to introduce large scale thorium in an entire new thermal reactor system called Advanced Heavy Water Reactor (AHWR) system. Table-4 shows some of the salient features of PHWR and AHWR system to highlight the basic differences/similarities between these two systems.

The detailed physics design of AWHR has been made to ensure six basic objectives of our 750 MW(th) AHWR viz. a) About 75% of the power produced by AWHR should be extracted from the thorium fuel, b) The system should have negative void coefficient of reactivity under all conditions, c) The discharge burn-up of the fuel should be above 20,000 MWD/Te, d) The consumption of plutonium is to be kept to the minimum, e) Initial plutonium inventory should be as low as possible, f) The system should be self

sustaining in U-233. One of the important characteristic in Th-U-233 fuel is that if we can provide sufficient U-233 to adjust the initial concentration of U-233 to a certain minimum level (about 1.5% U-233 which is called Self Sustaining Equilibrium of Thorium - SSET), thorium is capable to produce large amount of energy through burning of in-situ bred U-233 with spent fuel remaining practically same as the initial fuel. The burn-up achievable in such SSET fuel is, however, limited to ~12,000 MWD/Te without significant loss in the reactivity. If, however, we can arrange from external source some net supply of fissile isotope (e.g. Pu-239 which is a logical choice in our case) we can easily augment the burn-up level to ~20,000 MWD/Te.

Some areas related to Chemical Engineering Science in AHWR technology development are the following:

Table 4 : Comparison of PHWR (220 MWe) and AHWR

Feature	PHWR Actuals	AHWR Estimates
Core Configuration	Horizontal	Vertical
D2O Inventory	252 Tonnes	190 Tonnes
PHT Pressure	102 Kg/cm ²	70 Kg/cm ²
Steam Pressure	40 Kg/cm ²	70 Kg/cm ²
Station Loads	25 MW	15 MW
Fuel Burn up	7000 MWD/T of HM	20000 MWD/T of HM
Net Efficiency	26.5%	29.5%
Steam Generators	4 Nos.	Nil
Outlet Headers	2 Nos.	Nil
Inlet Headers	2 Nos.	1 No.
PHT Pumps	4 Nos.	Nil
Net Station Output	210.81 MW	235 MW

6.1 Thermal Hydraulic Studies Related to AHWR

Thermal Hydraulics plays a key role in the design of AHWR mainly because of the incorporation of a number of passive heat removal systems. Some important areas relevant to chemical engineering in which extensive thermal hydraulic studies are being carried out are as follows :

- i) Natural circulation and stability
- ii) Carryover in steam drum
- iii) Effect of noncondensable on steam condensation
- iv) Thermohydraulic relationship
 - a) Critical heat flux
 - b) Void fraction
 - c) Single and two phase pressure drop
 - v) Thermal stratification in large water pool

6.2 Fuel Cycle for AHWR

For successful implementation of AHWR, a number of new technologies need to be developed to have a closed Th-U²³³ fuel cycle. Since AHWR fuel contains Th, Pu and U, a reprocessing technology needs to be developed for separating these three fuels from the fission products.

Owing to more inert chemical properties and higher melting points of the relevant thorium compounds and to the inevitable production of U-232 and Th-228 during irradiation, the production and reprocessing of thorium fuel is more complicated and expensive. Some of the special aspects of reprocessing of AHWR spent fuel are:

- i) Thorium dioxide being a stable material is very difficult to dissolve (unlike sintered UO₂ which can be dissolved in boiling HNO₃). ThO₂ needs little amount of HF or NaF in HNO₃ for its dissolution. The addition of fluorides increases the corrosion rates of stainless steel vessels.
- ii) The radioactive contamination of U-232 in the separated U-233 product and of Th-228 and Th-229 in the separated thorium product will have to be taken into while handling these products for refabrication of the fuels.
- iii) The Pu-233 formed by (h₁) reaction from Th-232 decays to fissile U-233 with a half life of 27 days. This necessitates longer cooling period for complete recovery of U-233 in one step.

6.3 Technology for U-233 Clean-up in Thorium Fuel Cycle

One technology that would greatly simplify the major hurdle in induction of Thorium Fuel Cycle is the technology for separation of U-232 impurity from U-233 fissile material based on Laser Isotope Separation Process. The difference between the isotopes of U-232 and U-233 being less than the same existing between U-235 and U-238, we need to develop advanced Laser Isotope Separation (LIS) process. The major tasks involved in the LIS process are i) Evaporation of feed material to generate the vapour stream, ii) Selective multistep photoionization of the targeted isotope present in the vapour by tunable lasers, iii) Extraction of the photoions formed using the electrostatic fields, and iv) Collection of the depleted stream.

7.0 Accelerator Driven Subcritical System (ADSS)

As the name implies, ADSS is a Subcritical System (reactor) whose chain reaction is sustained by extra source neutrons generated in the core with the help of an external beam of protons driven by an accelerator. Unlike present generation of reactors which operates in "critical mode" with reserve of fissile material more than what is just necessary in the core, ADSS works in "subcritical mode". Essentially, therefore, ADSS consists of three components viz. an accelerator, a target and a blanket in addition to systems for heat removal and electricity generation. Accelerator produces an intense high energy proton beam (typically

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1 GeV with 10 mA \times 10 MW(th) which impinges on a heavy metal target (e.g. lead, the other preferred ones being W, Ta, Bi, Th, U_{nat} , U_{enrich} or Pu-239) to produce copious neutrons by spallation reaction (also called electro-nuclear reaction). These neutrons enter the subcritical blanket of the reactor (which has a multiplication factor of typically K_{eff} 0.95 – 0.98) to produce more neutrons by way of fissions of the fissile material along with generation of heat.

Liquid heavy metal lead (melting point 327°C) or Lead-Bismuth Eutectic (LBE) alloy (melting point = 127°C) are the typical coolants (circulated under neutral convection) for ADSS which also serves as spallation targets, reflectors as well as shields. Both Pb and LBE have low absorption of neutron. But the apparent edge of LBE over Pb is offset by much larger generation of radioactive Polonium.

A road map for ADSS development is being worked out at BARC mainly for utilization of thorium to breed U-233 material. Since the accelerator to generate 10-20 mA, 1 GeV proton beam needed for various ADSS in the world is about one order of magnitude above the best achieved so far, BARC proposes to circumvent this problem by using a 2 mA, 1 GeV accelerator on an entirely different type of reactor concept called "One Way Coupled" Booster Reactor system. For the development of the highly multi-disciplinary ADSS, a host of technological challenges are to be tackled. Some of those that are of special concern to chemical engineering community are :

- i) The ADSS fuel cycle does not need separation of fertile (Th) and fissile (U-233) material from the irradiated fuel. Unlike reprocessing of spent fuel from conventional reactors, one does not require separation of all the transuranic elements from the fissile material and only the FPs are to be removed by reprocessing. In order to take advantage of this nuclear characteristic of ADSS, a new simplified reprocessing technology (based on pyrochemical reprocessing) is to be established.
- ii) Spallation target design requires very efficient removal of concentrated heat deposited by the

interaction of energetic proton beam with target material.

- iii) Highly stabilized thermal conditions (at ambient or cryogenic temperature) are to be developed to meet the needs of many sub-systems of accelerators, e.g. D.M. process water system is required to maintain structural systems (with internal heat generation of hundreds of KW) within $\pm 0.1^\circ\text{C}$.
- iv) Circulation of heavy liquid metal coolant (HLMC) Pb/LBE under spallation condition (P.S. These have earlier been used as reactor coolant in some Russian submarine reactors) with debris of spallation is new and the spallation products may create problem of material compatibility (for which duplex ferritic martensite steel are being considered for the whole range of plant equipment and structural material). Fresh development work is required for control of process chemistry of purification of the coolant from contamination of corrosion and erosion of wetted surfaces.

8. Concluding Remarks

From the highlights of the various technological development activities already in hand and those which are yet to be pursued in Nuclear Fuel Cycle, it goes without saying that our programme on Nuclear Science and Technology not only provides the widest and toughest technological challenges to the chemical engineering community but the very success of our programme also depends on their success in finding out cost effective solutions to these challenges. Before I conclude, I am happy to acknowledge that chemical engineers of DAE have played a very crucial role to shape our nuclear energy programme to a stage, which has earned the status of an "advanced country" from IAEA as far as our programme on Nuclear Science and Technology is concerned. In fact, we are one of the few countries in the world who have established the entire spectrum of industries involved in "Nuclear Fuel Cycle".