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GOVERNMENT OF INDIA BHABHA ATOMIC RESEARCH CENTRE

HUCLEAR INDUSTRY - CHALLENGES IN CHEMICAL ENGINEERING

By

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ARSTRACT

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Chemical engineering processes and operations are closely involved in every step of the nuclear fuel cycle. Starting from mining and milling of the ore through the production of fuel and other materials and their use in nuclear reactors, fuel reprocessing, fissile material recycle and treatment and disposal of fission product wastes. each step presents a challenge to the chemical engineer to evolve and innovate processes and techniques for more efficient utilization of the energy in the atom. The requirement of high recovery of the desired components at high purity levels is in itself a challenge. "Nuclear Grade" specifications for materials puts a requirement which very few industries can satisfy. Recovery of uranium and thorium from low grade ores, of heavy water from ray water, etc. are examples. Economical and large scale separation of isotopes particularly those of heavy elements is a task for which processes are under various stages of development. Further design of chemical plants such as fuel reprocessing plants and high level waste treatment plants, which are to be operated and maintained remotely due to the high levels of fadio-activity call for engineering skills which are boing continually evolved. In the reactor, analysis of the fluid machanics and optimum design of heat removal system are other examples where a chanical engineer can play a useful role. In addition to the above. the activities in the Nuclear Industry cover a very wide range of chemical engineering applications, such asdesalination and other energy intensive processes, radioisotope and radiation applications in industry, medicine and agriculture.

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1. INTRODUCTION

It is only about five decades since teaching of chemical engineering began in this country; it was during this period that a tremendous transformation took place in the science and technology scenario in the country. Processing of minerals, crude oil and other ray materials, preparation of basic chemicals and production of fertilizers have to a large extent been the mainstay of chemical engineering profession in the past. With the advent of nuclear technology among others, a whole new spectrum of processes and techniques has emerged, thus expanding the scope of chemical engineering. Right from the stages of ore-beneficiation, milling and refining of fuels and other reactor materials to nuclear grade purity, nuclear reactor operation, reprocessing of irradiated fuel and safe treatment of radioactive wastes - each stage presents a challenge to the chemical engineer.

In this presentation, we shall attempt to highlight the areas in the nuclear industry where the chemical engineer plays a vital role. In view of the strategic importance of the various materials, processes and products involved in the nuclear industry, it is essential not to depend on any external assistance but to develop entirely on indigenous capabilities. The training of chemical engineers to enable them to shoulder the responsibilities effectively in this newly emerging industry, would be a valueable input to the efforts of self-sufficiency in nuclear power development.

2. ORE RENEFICIATION

2.1. Separation of minerals from beach sand

The separation of useful minerals from the beach sand may be described as one of the important industrial operations and historically the first one to be carried out.

Important minerals present in the beach sand of Tamil Nadu and Kerala on the West Coast, and Orissa on the East Coast are monazite, ilmenite, rutile, zircon and silliamanite.

Taking advantage of the difference in physical properties such as specific gravity, magnetic susceptibility, electrical conductivity and surface property, schemes were worked out at BARC, and plants have been set up by Indian Rare Earths at two locations in the West Coast and another plant is to be set up in Orissa.

2.2 Recovery of Uranium from Copper Tailings

The copper tailings in the Bihar region after processing for copper production have been found to contain uranium in the range of 0.01%. Although this grade is very low, the tailings are available in a ground form at no extra cost. Taking advantage of the fact that uranium minerals are quite heavy, a process based on gravity separation using the tables was developed at BARC and has now been adopted by the Uranium Corporation of India Ltd. at Surda. The possibility of recovering uranium from copper tailings of other plants in the area is also being explored.

3. FUEL PREPARATION

3.1. Uranium

Comparatively uranium ores in India are of lower grades containing less than 0.1% uranium. The economic recovery of uranium from such low grade eres was a challenge

for developing highly efficient chemical processes and separation techniques. The process developed at BARC consists of size reduction, acid leaching, solid-liquid separation, partial purification and upgrading of solution by ion-exchange technique and fractional precipitation to obtain uranium as a concentrate containing about $70\%~U_308$. A uranium mill for processing 1000 tonnes of ore per day based on this process has been in operation at Jaduguda in Mihar for over ten years.

The significant steps in refining the uranium concentrate to nuclear purity intermediate and in metal production, are solvent extraction using tri-butyl phosphate diluted with kerosene as solvent, precipitation of the purified uranyl nitrate solution to ammonium diuranate and calcination, reduction of the calcined UO; to UO2, conversion of UO2 to UF, by anhydrous, HF and finally calcio-thermic reduction of UF, to metallic uranium. A plant for producing nuclear grade uranium metal has been in operation at Trombay since 1959 for meeting the requirements of fuel for research reactors based on natural uranium. Power reactors use UO2 as fuel rather than metallic uranium, However, to obtain UO2 suitable for compacting and sintering to high densities, necessary optimisation studies of process parameters for ammonium diuranate precipitation, calcination and reduction were carried out. Half of the initial loading of uranium oxide pellets for reactor No.1 of the Rajasthan Atomic Power Station was produced at BARC. Plants for making oxide fuel for the power reactors have now been set up at the Nuclear Fuel Complex, Hyderahad.

The R & D work done in this field at BARC has enabled India to become one of the very few countries with the capability of producing nuclear fuel for its power reactors.

3.2. Thorium

India's long term nuclear programme is based on the utilisation of the wast deposits of thorium, with the use of plutonium produced in the first generation power reactors. Thorium is the fertile material to be used in the next generation of reactors, i.e. the fast breeder reactors which will breed the fissile material uranium-233. Monagite is being processed by alkali digestion for the production of rare earths and a "Concentrate" containing most of the thorium and uranium. This concentrate is further processed by chemical precipitation to get "mantle-grade" thorium nitrate. In order to prepare thorium nitrate of muclear purity, a process based on solvent extraction using tributyl phosphate diluted with kerosene has been developed. A plant based on this process starting from an intermediating feed is in operation. Another process has been developed which involves the direct dissolution of the crude hydroxide concentrate in nitric acid, extraction of uranium and thorium nitrates successively with 10% and 40% of tri-butyl phosphate and conversion of wure nitrate solution to solid thorium nitrate. This process simplifies the recovery of thorium and uranium from the concentrate. A process for converting thorium nitrate into sinterable grade thorium oxide has also been developed.

4. NUCLEAR MATERIALS

4.1. Heavy Water

Except for the Tarapur Atomic Power Station, heavy water moderated natural uranium fuelled reactor has been selected for the first phase of our nuclear power programme. The availability of heavy water is, therefore, of pivotal importance in implementing this programme. A small heavy water plant based on electrolysis for pre-enrichment and

hydrogen distillation for final enrichment has been in operation since 1951 at Nangal. Of the various processes available for the enrichment of heavy water the one based on H2S-H2O exchange has been developed at BARC and a plant for producing 100 tonnes of heavy water per annum is under construction at Kota. Natural water contains approximately 150 ppm of heavy water. In view of the low natural abundance of deuterium, low separation factor, low rate of reaction, etc. the number of stages, inter-stage flows, heat transfer and recovery systems were optimised and highly efficient contacting devices for heat and mass transfer were developed. Process dynamics studies were carried out for the control of large number of inter-related process variables arising out of the large number of stages and allied equipment. In view of the urgency of the requirement, a few more plants are being constructed based on ammonia hydrogen exchange process developed elsewhere.

For upgrading the heavy water diluted during reactor operation, processes based on distillation and electrolysis have been studied at WARC. Based on these studies, upgrading plants are being set up at reactor sites. Studies are also in progress on the amine-hydrogen and hydrogen-water exchange processes.

4.2. Zirconium

Processing of sircon sand to obtain zirconium metal (an excellent material for fuel canning and other structural components in the nuclear reactor) involves preparation of hafnium-free zirconium oxide, chlorination of the oxide and reduction of the chloride so obtained with magnesium. The separation of hafnium which is chemically similar to zirconium and which is present to the extent of about 2.5%, is an important step in this process since hafnium has a high

absorption cross-section for thermal neutrons. Solvent extraction using tri-butyl phosphate has again been adopted for this separation. Processes for the extraction of zirconium from the ores and its separation from hafnium, and for the production of zirconium metal were developed at BARC and are now being used in the plants at Nuclear Fuel Complex, Hyderabad for the commercial production of zirconium metal sponge starting from zircon sand.

5. NUCLEAR REACTORS

Most R & D activities relating to reactor engineering are carried out at BARC. These include thermal and hydraulic design of nuclear reactor, evaluation of dynamic performance of its components and systems, critical heat flux, burn out and safety analysis of nuclear reactor, compatability of materials in reactor environment, etc. The aim is to develop methods and to collect data required for designing sophisticated reactor components and systems. An estimation of heat transfer from the fuel to the coclant cannot always be made accurately on the basis of theoretical analysis, as many of the problems related to flow distribution in the complex geometries, single and two phase flow in rod bundles, mechanism of heat, mass and momentum inter-change between various channels within the fuel bundles, etc. remain unsolved. Fuel testing and heat transfer loops have, therefore, been set up to carry cut investigations for understanding the heat removal mechanism. As part of the R & D programme the Centre has set up special facilities for testing of power reactor components and evaluating the performance of various reactor components, equipment and systems. In the field of safety of nuclear reactors and systems, blowdown, critical heat flux and emergency core cooling are major topics on which studies are in progress. A corrosion loop has been set up

to study the effect of sea-water under different flow conditions on various materials of construction used in the heat transfer equipment.

6. FUEL REPROCESSING

The reprocessing of the irradiated fuel is one of the major steps in the nuclear fuel cycle wherein the fuel after use in the nuclear reactors is reprocessed to separate plutonium (formed in the reactor by the absorption of neutrons in U-238), uranium and the highly radioactive fission products. If thorium is used as the fertile material, the fissile material uranium-233 produced from it in the reactor has to be separated out. A number of special problems arise in fuel reprocessing because of (1) the necessity of carrying out operation remotely behind thick shielding to guard against high radioactivity, (2) limitation of equipment size and geometry from consideration of nuclear criticality involving hazard of uncontrolled fission chain reaction and (3) very high degree of decontamination, viz. 106 to 108 from fission product are required to be achieved. Aqueous reprocessing with modifications and refinements and employing solvent extraction techniques in pulsed perforated plate columns has become the universal method for thermal reactor fuels on account of its proved performance. For Fast Reactor fuels the choice lies between aqueous and non-aqueous methods.

A plant was set up in Trombay to process the irradiated fuel from the research reactor. The experience in the operation of the plant has provided not only the know-how for the design of future bigger plants but has indicated the fruitful areas of research and development. A bigger plant for processing irradiated fuel from the power reactors has recently been set up at Tarapur. The Indian programme visualizes development work in short residence solvent extraction contactors as well as on the various non-aqueous processes.

7. RADIOACTIVE WASTE MANAGEMENT

Effluents from the nuclear industry differ from the waste from conventional industries in the sense that they contain radioactive materials which emit biologically harmful radiation and which cannot be destroyed. The two concepts, viz. "dilute and disperse" and "concentrate and contain" are adopted for the management of radioactive waste depending upon the level of radioactivity. For radioactive liquid waste of low to moderate concentration, chemical treatment and ion exchange are two of the most commonly used techniques. Due to very low concentrations of some of the radionuclides that can be permitted in the effluents to be discharged. it is required to aim at and achieve high decontarination factors. Design and operation of the various units get complicated due to the fact that stoichiometrically the concentration of the radioactive ions in the waste is on a microscale when compared to the inactive species which are present in macro-quantities.

The highly radioactive wastes arising from the fuel reprocessing plant require particular attention since they have to be stored for a very long period for the radioactivity to decay. The most accepted practice for such high level wastes calls for vitrification of the streams at temperatures ranging from 900 to 1100°C along with suitable additives to result in a stable inert glass matrix which could be stored almost indefinitely. The corrosive and high temperature process conditions require the use of corrosion resistant materials like titanium. As in the case of fuel reprocessing plant, all operations are to be carried out remotely in highly shielded cells. Most of the equipment such as evaporator, fractionator, condenser, multi-some control industion furnace, inline pump, etc. have been custom designed and developed at BARC for specific uses.

8. ENERGY UTILIZATION STUDIES

The impact that nuclear power could make on the countryside in starting agro-industrial complex is so striking that development work on utilization of nuclear energy in industrial processes as well as agriculture through the production of fresh water by desalination has been initiated at BARC. A number of large electricity consuming processes such as the production of hydrogen by electrolysis, electrothermal production of elemental phosphorous and production of aluminium and caustic-chlorine have been studied. The electric furnace technology for the production of elemental phosphorous has been developed at BARC and a pilot plant with a 600 K.W. electric furnace has been set up.

In the field of desalination, studies have been carried out on multi-stage flash distillation, vertical tube evaporators, reverse osmosis and electro-dialysis processes. A pilot plant based on the vertical tube evaporators is in operation at Trombay for collecting design data and evaluating heat transfer performance of advanced heat transfer tubes like single and double fluted tubes, axially and spirally corrugated tubes, etc. developed in the Centre. An experimental facility based on multi-stage flash distillation process is being installed. Work on ship-borne desalination plant has also been taken up. Work on the development and testing of various types of semi-permeable membranes suitable for reverse osmosis application is also in progress.

9. APPLICATION OF RADIO-ISOTOPES AND RADIATION

BARC has been active in developing multifarious application of radioisotopes and radiation to serve industry, agriculture and medicine. BARG makes over 300 radioisotope products including radio-phatmaceuticals which are supplied

to many institutions in India and abroad. Separation of stable isotopes of argon, nitrogen, boron, etc. and production of nitrogen-15 labelled compounds are also carried out. The Centre has developed self-luminous compounds incorporating promethium-147, and tritium.

Cobalt-60 occupies a pride of place as the most important radiation source. Radiographic cameras housing cobalt-60 have been developed and supplied to industrial organizations in the country for quality control of castings and inspection of welding. The Centre has also developed various equipment incorporating radioisotope sources for control of level in closed containers, automatic filling of packages on the production line, measuring thickness of pipes in the chemical industry, detection of smoke, etc. A number of these devices have already been installed in industrial plants. A radio-tracer technique has also been developed at BARC for the detection of leaks in buried pipelines, trunk telephone cables, intricate process equipment and for the determination of process parameters in industry such as blending of viscose in the manufacture of reyon.

Chemical effects of radiation are also being investigated at BARC with a view to producing new and modified materials. Using gamma radiation techniques, BARC has developed wood polymer composites from low grade timber. Two other examples of successful application of radiation are polymerization of trioxane to produce industrially important plastics and modification of cotton and synthetic fibre to achieve desired properties. These processes are ready for commercial exploitation.

Food preservation by radiation should gain wide acceptance in the near future in view of its distinct benefits. Radiation disinfestation of stored wheat, inhibition

of sprouting in onions and potatoes, delayed ripening of mangoes and bananas and extending shelf-life of certain sea-foods using gamma radiation techniques have been developed at BARC. Recently, a plant has been set up at Trombay with UNDP aid for the radiation sterilization of various medical products. Studies are also in progress to improve the heat and mass transfer processes involved in food processing and preservation.

10. CONCLUSION

Extraction of energy from the atom and realization of other spin-off benefits from atomic energy involves a wide variety of scientific and technical disciplines. The role of the chemical engineer is indeed pivotal in many of the challenging areas of nuclear industry. About 250 chemical engineers are at present employed at various levels in the nuclear industry. Our programme calls for the induction of a large number of chemical engineers in the next decade. An industry could contribute only if it is manned by efficient manpower. In this endeavour, our universities and institutes, specially the IITs, can play a vital role.

We have reciprocal arrangements whereby professors from the universities lecture to our Training School and our specialists give guest lectures and practical training to their students. We are setting up a system of retraining our engineers particularly in IITs in specialised fields to make them more useful for our expanding programmes. We have also finalised methods by which we pass on specific research and development projects in various disciplines to the IITs and universities.

The purpose of this lecture would be served if we have conveyed to the faculty members of the universities,

the role of the chemical engineer in the nuclear industry so that they can appreciate our requirements and take appropriate steps to prepare the chemical engineers for shouldering the responsibilities in this newly emerging industry and collaborate in expanding our R & D activities by carrying out applied as well as basic research in the universities.

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