PARTITIONING AND TRANSMUTATION PROGRAM "OMEGA" AT JAERI

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ABSTRACT

During the last two decades, JAERI has been carrying out the partitioning and transmutation program in the following areas: (a) development of the four-group partitioning process, (b) design study of the transmutation systems, one with actinide burner reactor, the other with an accelerator-driven subcritical reactor, (c) development of an intense proton accelerator, (d) development of nitride fuel manufacturing and pyrochemical reprocessing, and (e) basic research for supporting the development of transmutation systems. Activities and recent achievements are overviewed.

JAERI is about to launch the Neutron Science Project which aims at bringing scientific and technological innovation for the 21st century in the fields of basic science and nuclear technology using neutrons. The accelerator-driven transmutation system study and the development of an intense proton accelerator are also under way as important parts of this project.

I. INTRODUCTION

Since the mid 1970s, JAERI has been developing a partitioning process of high-level waste (HLW) and a concept of a dedicated transmutation system of minor actinides (MAs) as an effective and efficient measure to alleviate the long-term burden of nuclear energy which may arise from the management of HLW. During these studies, the double strata fuel-cycle concept was developed. JAERI's activities cover the following areas of the partitioning and transmutation: (a) development of the four-group partitioning process, (b) design study of the actinide burner reactor and the accelerator-driven hybrid system, (c) development of an intense proton accelerator, (d) development of nitride fuel cycle technologies, and (e) basic research for supporting the development of transmutation systems. [(1)]

In the course of planning of an intense proton accelerator for transmutation, we recognized that neutron scattering researchers need intense neutron beams for basic science. JAERI is planning to launch the Neutron Science Project which aims at accomplishing the scientific and technological innovation using neutrons for the 21st century. An intense proton accelerator will be constructed for dual duty as a neutron producer for transmutation and for basic science including neutron scattering.

II. DOUBLE STRATA FUEL CYCLE

JAERI has been proposing the concept of the double strata fuel cycle consisting of the power reactor fuel cycle and the Partitioning-Transmutation (P-T) cycle. (2) In this scenario, a reprocessing plant, a partitioning plant and a transmutation plant will be co-located in one site. This configuration of plants composes a high level radioactive waste (HLW) management park.

The concept of the double strata fuel cycle is illustrated in Fig. 1. The final HLW from this fuel cycle contains only short-lived fission products. The separate treatment or the isolation of MAs from the commercial cycle is preferable from two reasons: (a) complete recovering and transmutation of MAs and long-lived fission products are possible, and (b) MAs are strong neutron emitters, and, thus, MA recycling through the conventional fuel cycle may cause problems in the radiation shielding of the fuel cycle facilities. This may cause the cost of electricity generation to increase. [(3)]

III. DEVELOPMENT OF FOUR-GROUP PARTITIONING PROCESS [(4, 5)]

Firstly, a partitioning process was developed for separating elements in HLW into three groups, namely, transuranium elements (TRU), Sr-Cs, and others. The process consists of three steps: (a) the solvent extraction of U and Pu with tributylphosphate (TBP), (b) the solvent extraction of Am and Cm with diisodecylphosphoric acid (DIDPA), and (c) the adsorption of Sr and Cs with inorganic ion exchangers. The process was demonstrated with actual HLW and more than 99.99% of the Am and Cm were extracted with DIDPA.

Later, a four-group partitioning process has been developed in which one step for separating the Tc-PGM group was developed in addition to the three-group separation. Effective methods for separating TRU (especially Np) and Tc have been developed.

(a) Np Separation

Neptunium, which is dominant in HLW, is the most difficult actinide element to be extracted with general organic solvents. A new process was developed in which more than 99.97% of penta-valent Np was extracted when hydrogen peroxide was fed to a level to compensate for its decomposition in the DIDPA extraction process.

(b) Am and Cm Separation from rare earths

Selective stripping of Am and Cm from DIDPA with the complex agent DTPA is being studied for their separation from rare earths (RE). Batch experiments showed that the separation factor between Am and RE (ratio of distribution ratios) is larger than 10. Experiments are now in progress to find the optimum process conditions.

(c) Tc Separation

Two methods have been developed to separate Tc, precipitation by denitrating HLW and adsorption with active carbon. More than 95% of Tc in a simulated HLW was recovered as precipitate by denitration of HLW. An active carbon column was used for the quantitative adsorption of Tc from a 0.5 M nitric acid solution. Desorption of Tc from the column was achieved quantitatively by the use of an alkaline thiocyanate solution as eluant.

The four-group partitioning process was developed as shown in Fig. 2. This process is to be tested with actual HLW at NUCEF (Nuclear Fuel Cycle Safety Engineering Facility). (6) Hot operation with actual HLW is scheduled in 1998.

IV. DESIGN STUDY OF THE TRANSMUTATION SYSTEMS

A transmutation system with a very hard neutron energy spectrum and high neutron flux would be very efficient and effective for MA transmutation. The concepts of MA burner reactors (ABR: Actinide Burner Reactor) and a proton accelerator-driven MA transmutation system have been developed at JAERI. When a dedicated transmutation system becomes available, the scheme of an entire fuel cycle will be a strata structure fuel cycle.

IV.1. MA BURNER REACTORS (ABR: ACTINIDE BURNER REACTOR) (7, 8)

Two types of ABRs are designed at JAERI. Fuel material of these ABRs is MA-U nitride mixture. One type is a lead-cooled pin fuel ABR (L-ABR) and the other is a He-cooled particle fuel ABR (P-ABR). Nitride was selected as the fuel material of these ABRs because of its good thermal property. The other advantage of nitride fuel is that it can be processed with the pyrochemical reprocessing, and, hence, the fuel cycle facilities can be very compact and cost effective. The fuel concept of P-ABR is shown in Fig. 3. The reactor core design parameters of these ABRs are given in Table 1. In these ABRs, neutron energy spectrum is very hard and the core-averaged neutron energy is around 720 keV. These hard neutron spectra are very effective for direct fission of MAs which has fission threshold above 600 keV. The MA burnup in the ABRs of 1 GW thermal output is 190 to 200 kg per year.

IV.2. ACCELERATOR-DRIVEN TRANSMUTATION SYSTEM (9)

Two types of accelerator-driven system concepts are being studied; namely, a solid system a molten-salt system. Either system utilizes the hard neutron spectrum of spallation neutrons to transmute MAs efficiently by fission.

(a) Concept of solid target/core system

An accelerator injects proton beam through a beam window into the solid tungsten target located at the center of the target/blanket. Surrounding the target is the subcritical blanket loaded with actinide alloy fuel. Spallation neutrons emitted from the target induce fission in the actinide blanket region. The schematic diagram of the proposed transmutation system concept is shown in Fig. 4. With a 1.5 GeV, 39 mA proton beam, a sodium-cooled solid target/core having k_{eff} of 0.89 produces 820 MW of thermal power. Assuming a load factor of 80%, MA burnup is approximately 250 kg/y, or 8% of inventory per year.

(b) Concept of molten-salt target/core system

The molten salt acts both as fuel and as target material, and also serves as coolant. Its main advantage is the capability of continuous on-line processing of MAs and reaction products. Chloride salt is chosen based on the consideration about actinide solubility and nuclear characteristics. The molten-salt target/core with k_{eff} of 0.92 produces 800 MW thermal power with a 1.5 GeV, 25 mA proton beam. Assuming a load factor of 80%, the MA burnup is approximately 250 kg/y, or 4.6% of inventory per year.

The support factor, which is defined as the number of power reactor units whose MAs are transmuted by one unit of a transmutation system, is about 10 to 15 for dedicated transmutation systems, while the support factor of FBR proposed so far is between 4 to 6.

V. TRU NITRIDE FUEL AND FUEL CYCLE [10, 11]

In a double-strata concept, MAs from the commercial fuel cycle flow into the second-stratum of transmutation ("dedicated actinide-burner") cycle. MAs are concentrated and confined in the second stratum, exiting only after being converted to fission products. Considering the inherent difficulty in handling MAs, an innovative approach is required in designing a fuel cycle system for the actinide burning.

Concepts of dense fuel cycles for the second stratum (Fig. 5) have been proposed, where high atom densities of the actinides are maintained throughout the whole cycle, and the system volume and envelope are minimized. JAERI is studying the feasibility of employing the nitride fuel and pyrochemical reprocessing. The favorable thermal properties of the nitride fuels that make full utilization of a cold-fuel concept possible are: (a) lower fuel temperatures and hence lower fission gas release, (b) a thinner cladding to achieve a harder neutron spectrum, and (c) a relatively large Doppler effect in the over-power events.

Efforts are directed to technical developments and fundamental property studies of (a) the sol-gel process to obtain nitride microspheres from the actinide nitrate solution, and (b) the electrorefining process of the nitride fuel with a LiCl-KCl-$AnCl_3$ (An: actinides) melt.

(a) Nitride fabrication from actinide salts

Actinide nitrate solution from the partitioning of HLW can be solidified to a ceramic form by an internal gelation method. A droplet of the actinide nitrate solution with a carbon suspension turns into a solid mixture of oxide (hydroxide) and carbon in a form of microspheres. A microwave gelation apparatus has been developed and tested at JAERI. The (oxide+carbon) microspheres thus obtained are converted to the nitride by a carbothermic synthesis.

The nitride fuels can be used in the form of either pellets or TiN-coated particles. In the particle-fuel concept, the TiN coating consists of both high-density and low-density layers.

(b) Electrorefining of nitrides

In the proposed pyrochemical process, the irradiated nitride fuels are electrorefined in a LiCl-KCl eutectic melt. Like metal fuels, the actinide nitrides would be anodically dissolved. The design of the electrorefiner may be very similar to that for the metal fuels. The recovered metals then have to be converted to nitrides.

Laboratory runs of the fused salt electrolysis of UN have been made. The recovery of uranium metal has been demonstrated. During the electrolysis, the system was purged with purified helium. Conversion of the metal to the nitride has been readily made in liquid Cd with nitrogen cover gas. The reaction products were U_2N_3 and (U, Gd)N.

VI. DEVELOPMENT OF A HIGH-INTENSITY PROTON ACCELERATOR [12]

A high-intensity proton linear accelerator with a beam power up to about 10 MW has been proposed for basic science and various engineering tests of the transmutation system.

The R&D work has been carried out for the components of the front-end part of the proton accelerator: ion source, radio-frequency quadrupole (RFQ), drift tube linac (DTL), and RF source. In the beam test, a current of 70 mA with a duty factor of 10% has been accelerated from the RFQ at an energy of 2 MeV. A hot test model of the DTL for high-power and high-duty operation has been fabricated and tested.

For the high-energy portion above 100 MeV, superconducting accelerating cavity is studied as a main option. The superconducting linac is expected to have several favorable characteristics for high-intensity accelerator such as shorter length acceleration, large bore radius resulting in low beam losses, and cost effectiveness for construction and operation. The design work for superconducting cavities is in progress in collaboration with the KEK (National Laboratory for High Energy Physics). A test stand with the equipment of cryogenics system, vacuum system, RF system, and cavity processing and cleaning has been prepared to test the physics issues and fabrication process.

The main accelerator components such as high-current hydrogen-ion source, RFQ, DTL, and RF power source have been constructed and tested. A high brightness (140 mA) hydrogen ion beam has been extracted.

The first 2 MeV beam test with the ion source and RFQ in combination of a single unit of high power RF source was successfully carried out with the peak acceleration current of 70 mA (a duty factor of 10%) . [13]

VII. BASIC RESEARCH SUPPORTING THE DEVELOPMENT OF TRANSMUTATION SYSTEMS

To support the above-mentioned activities on transmutation system development, several basic research activities are in progress.

VII.1. NUCLEAR DATA FILE FOR TRANSMUTATION SYSTEM DESIGN STUDY [14]

Two types of nuclear data files are being compiled. The JENDL Actinide File is compiled for use in the ABR design study. It contains neutron-induced reaction data for about 90 nuclides from ^{208}Tl to ^{255}Fm. This compilation will be completed by 1997. The JENDL High-Energy File contains the data for protons and neutrons up to a few GeV which are used for the design of the accelerator and for studying an accelerator-driven transmutation system.

VII.2. MEASUREMENT AND EVALUATION OF ACTINIDE NUCLEAR DATA

MA nuclear data are measured for fission-neutron yields, delayed-neutron yields, and fission yields in collaboration with the Oak Ridge National Laboratory (ORNL) and Texas A&M University. Actinide nuclear data in the JENDL File are evaluated using the integral experiments at the fast critical facility FCA. [15]

Spallation integral experiments have been carried out to obtain data on nuclide production, to estimate the yields of neutrons and spallation products, and to evaluate the validity of the simulation code system. The 500 MeV booster proton synchrotron facility at the National Laboratory for High Energy Physics (KEK) is used for the activation experiments. In general, the calculation with the codes agree fairly well with the experimental results. There are however some discrepancies in both the nuclide production cross section and the neutron energy spectrum . [9, 16]

VII.3. FUEL MATERIAL THERMODYNAMIC DATABASE [10, 11]

In view of the feasibility study of the above dense fuel cycles, thermodynamic data base needs to be expanded in three major areas: actinide alloys and intermetallics, nitrides, and actinide-containing molten salts. In JAERI, a pure substance and solution data base, related to the actinide burning, is being formed as a minor supplement to the existing thermodynamic data base.

VIII. NEUTRON SCIENCE PROJECT OF JAERI

Neutron scattering has achieved some notable successes in recent years, such as unraveling the crystal structures of high-temperature superconductors, and is now exciting a lot of interest among biologists for probing large organic molecules. A limiting factor for neutron scattering is the intensity of the neutron beams. High intensities of neutron beams allow researchers to carry out experiments that would otherwise be impossible. In Europe, the European Spallation Source ESS is under design, and in the USA, the National Spallation Neutron Source (NSNS) will be constructed at ORNL, both with 5 MW proton beam power.

JAERI is preparing to launch the neutron science project next year. The objective of the project is to construct an intense proton accelerator with proton energy of 1.5 GeV, proton beam power of 6 to 8 MW, and research facilities to be dedicated for the neutron science research center of JAERI Tokai. The spallation neutron source has double duty as a neutron source for basic science and as a neutron source for nuclear-energy-related research such as an accelerator-driven transmutation study.

Basic science in this center covers the fields of structural biology for investigating the structure and function of biological molecules such as DNA, advanced material science (e.g., under extreme conditions), high-energy neutron science (e.g., spallation phenomena), nuclear cross-section measurements for transmutation study, heavy-ion science for creating unstable heavy nuclei through spallation, and decay property measurements of extremely-neutron-rich nuclei.

The layout of the proposed facilities and the proposed schedule for construction of facilities are shown in Figs. 6 and 7.

Transmutation study at this facility will cover the two steps of accelerator-driven transmutation system development. The first step will be the feasibility study of the hybrid system concept at low power level of uranium subcritical system. These experiments will check the stable operation of a hybrid system and MA burnup with use of MA foils or pellets for activation measurements or destructive analysis. The data base for designing a spallation target for high-power experiments will be obtained at the second step. In the first step, most of the transmutation experiments will be performed with pulse-mode operation so that proton beams from the accelerator can be shared in time among the other experiments.

Once the feasibility of the hybrid system concept for the MA transmutation is proven, a test reactor facility with 10-30 MW thermal power will be proposed for the second phase of development. Operation of the hybrid system with uranium subcritical system will be tested at this high power level and transmutation capability will be tested using MA target pins. Technical feasibility of spallation target and beam window will be also tested in the second-step experiments.

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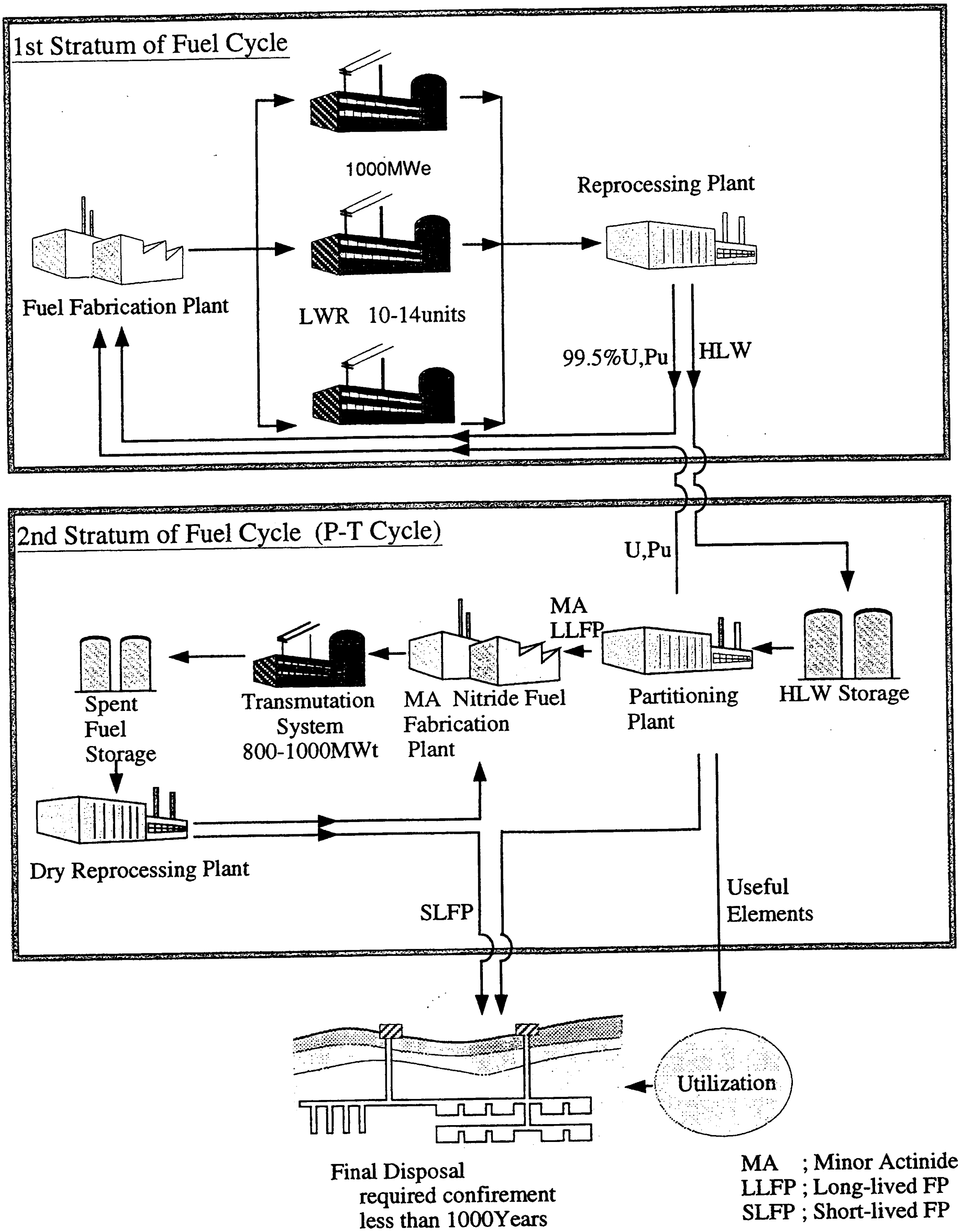

Fig. 1 JAERI's Concept of Double Strata Fuel Cycle for Complete HLW Management

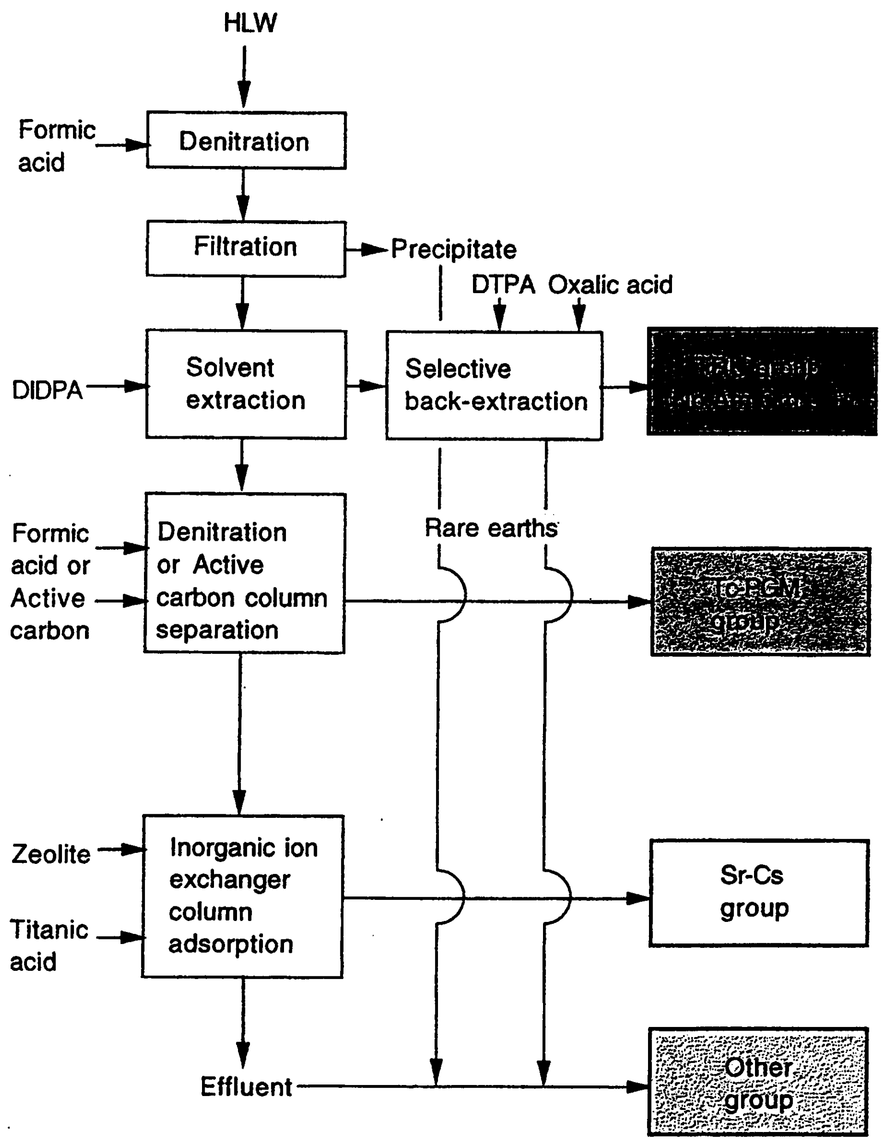

Fig. 2 Flow Sheet of Four Group Partitioning Process

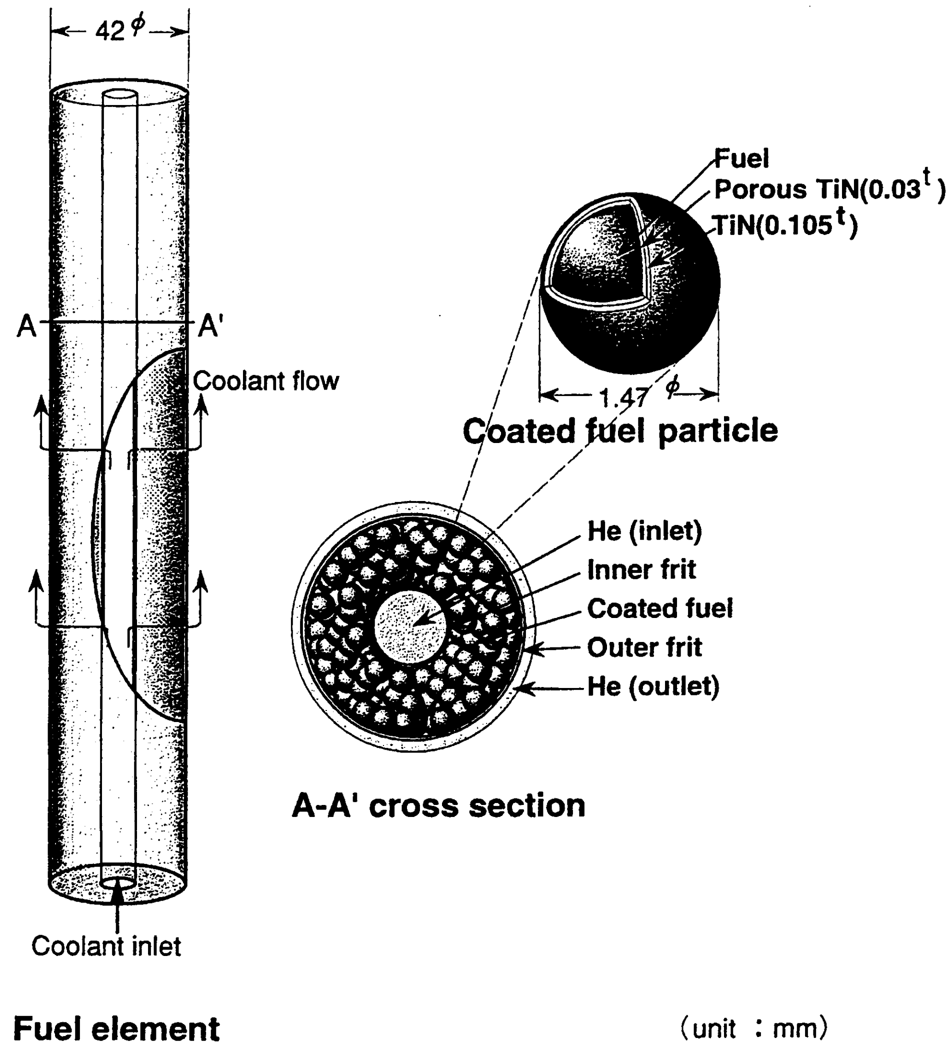

Fig. 3 Fuel element and coated fuel particle for He gas cooled reactor

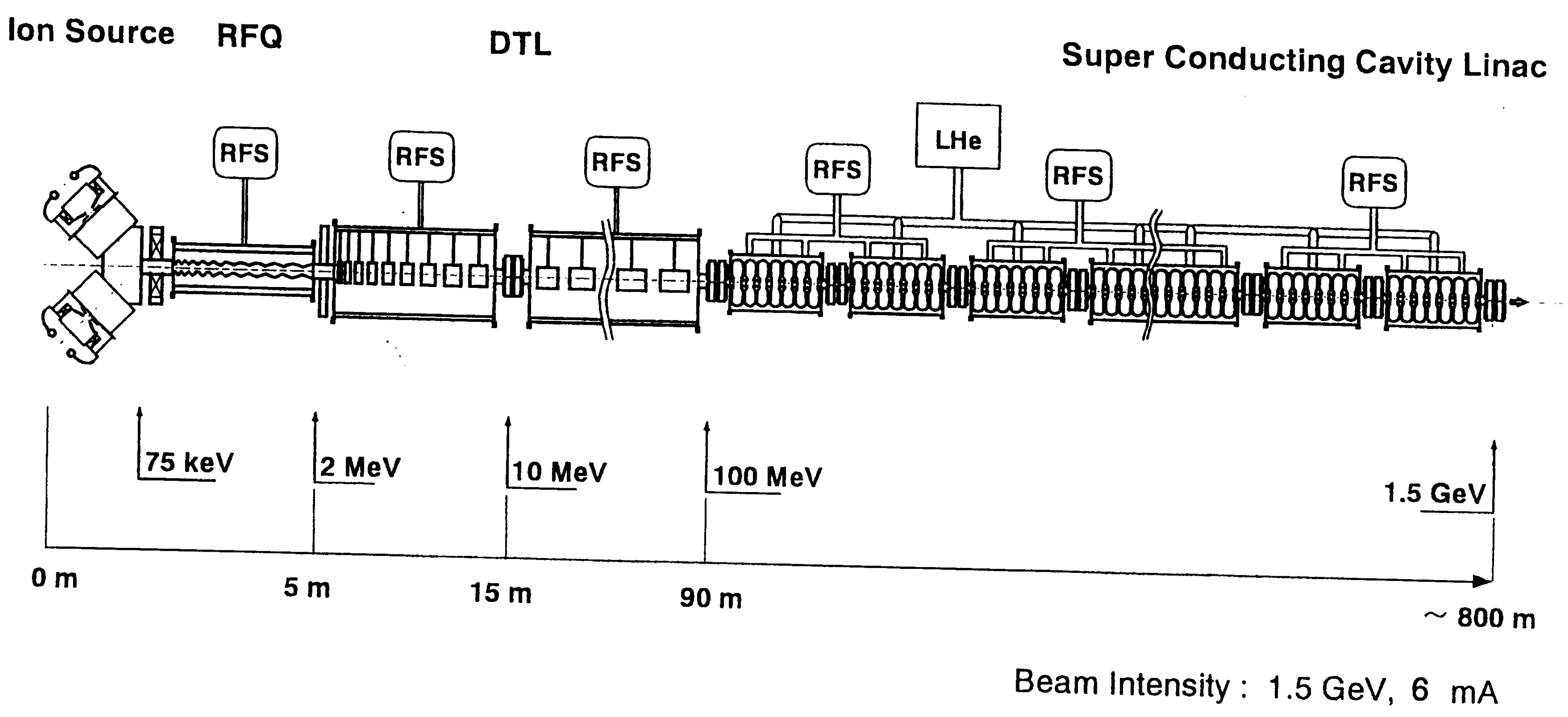

Fig. 4 Conceptual Layout of Intense Proton Accelerator with Superconductive Cavity

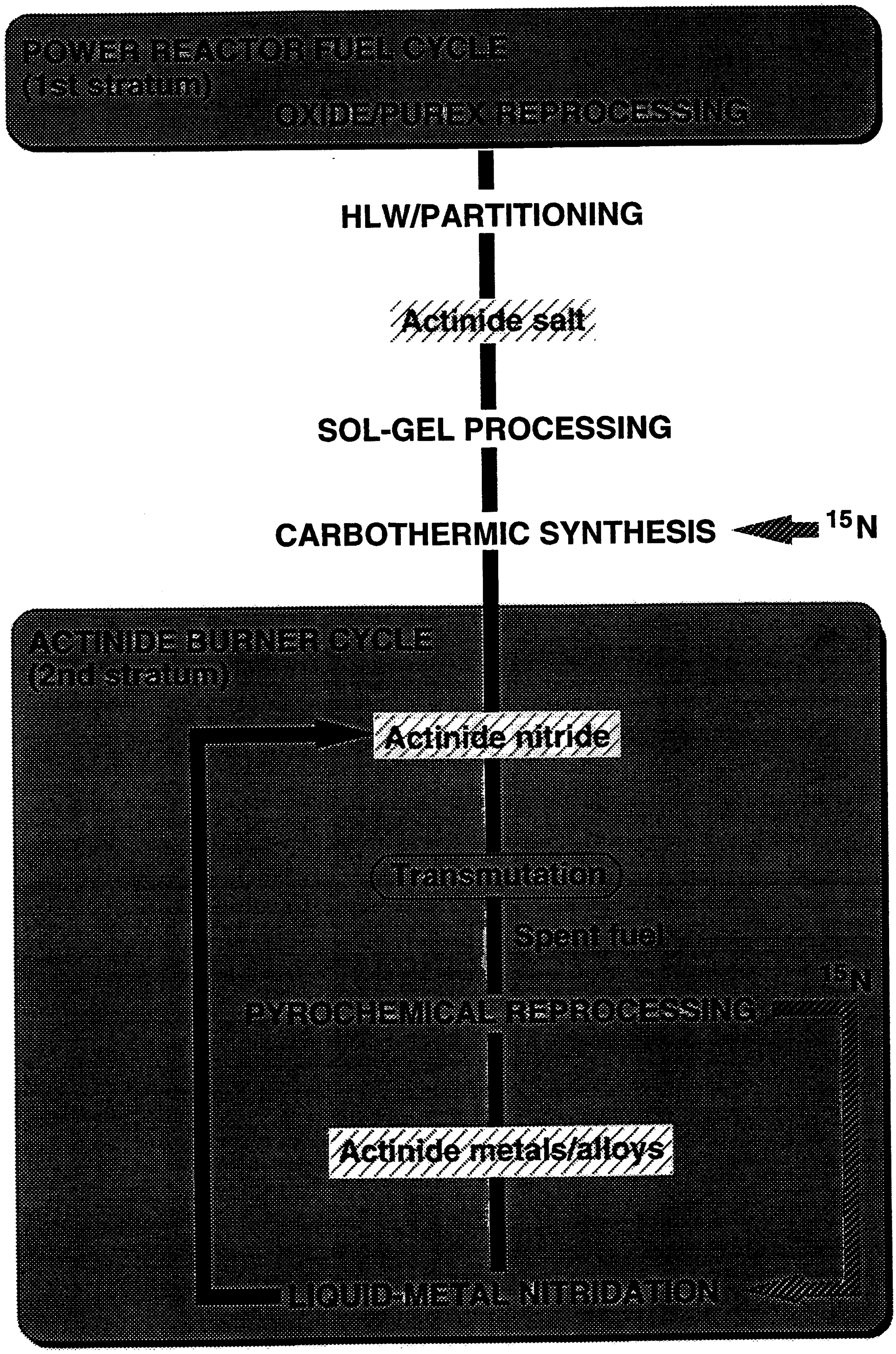

Fig. 5 Double Strata Fuel Cycle with Nitride/Pyrochemical Process

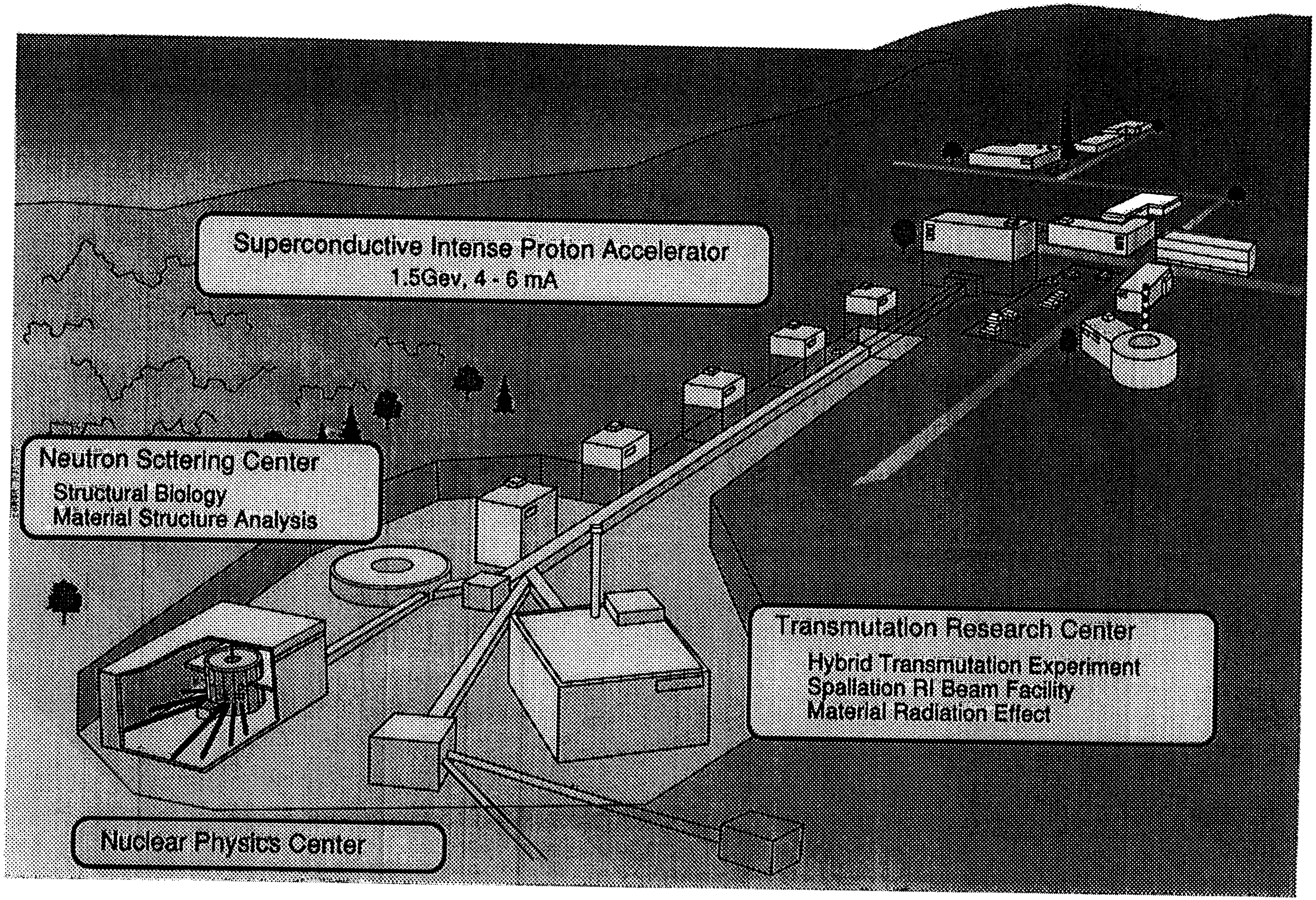

Fig. 6 Layout of proposed Facilities for Neutron Science

Fig. 7 Proposed Schedule for Construction of Neutron Science Research Facilities