

STUDIES ON ACCELERATOR-DRIVEN TRANSMUTATION AT JAERI

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Abstract

Research and development on intense proton accelerator-driven transmutation of long-lived radioactive nuclides are being carried out at the Japan Atomic Energy Research Institute (JAERI) under the Japanese long-term program for research and development on nuclide partitioning and transmutation technology, called the OMEGA Program. In the conceptual design study of transmutation system, two options of solid system and molten-salt system have been pursued as dedicated transmutation systems with hard neutron spectrum and high neutron flux. Spallation integral experiments are being made on a lead assembly with 500 MeV proton beam to validate and improve the cascade code NMTC/JAERI. Front-end components for the prototype intense proton accelerator such as ion source, RFQ, DTL, and RF source were developed and tested successfully. JAERI is proposing the development of an intense proton linear accelerator of 1.5 GeV class with a maximum beam power of around 10 MW for tests in accelerator-driven transmutation technology and various experiments in basic sciences. The project of the accelerator-based research complex is expected to be launched in 1997 with conceptual designs of the accelerator and research facilities.

1. Introduction

Under the framework of the Japanese long-term research and development program on nuclide partitioning and transmutation (P-T) technology, called the OMEGA Program, the Japan Atomic Energy Research Institute (JAERI) is carrying out R&Ds on P-T [1,2]. The OMEGA Program is aiming at establishing P-T technologies to provide an attractive option for high-level waste (HLW) management in the future. The technologies under study include an advanced partitioning to separate HLW into four element groups [3], transmutation with actinide burner reactor [4], and transmutation with accelerator-driven system. The proposed transmuters are dedicated systems specifically designed for nuclear waste transmutation purpose. With dedicated transmuters, the fuel cycle for P-T can be separated completely from the conventional commercial fuel cycle for power reactors, forming a double strata structure of nuclear fuel cycle [5].

The project on accelerator-driven transmutation technology at JAERI includes the conceptual design study of transmutation plants, the development of spallation simulation code system [6], the spallation integral experiment [7], and the development of a high-intensity proton linear accelerator [8].

In the conceptual design study of accelerator-based transmutation plant, two types of dedicated system concepts are being investigated; solid system and molten-salt system. The design of the solid system is based on the current technology and design practice for sodium-cooled fast breeder reactors. Another option is the molten-salt system with flowing actinide chloride that plays three principal roles of as fuel, target material, and coolant. Its main advantage is the capability of continuous on-line processing of actinides and reaction products.

JAERI is proposing the development of an intense proton linear accelerator for tests in accelerator-driven transmutation technology together with various experiments in basic sciences. The project is called the Neutron Science Project. The proposed accelerator will be of 1.5 GeV class with a final beam power of around 10 MW. It will share the beam among research facilities, which altogether form an accelerator-based research complex.

This paper describes the conceptual designs of accelerator-driven solid system and molten-salt system for nuclear waste transmutation. Outline of the program plan of tests in accelerator-driven transmutation is presented.

2. Accelerator-Driven Transmutation System

JAERI is aiming to develop technologies of an advanced partitioning process of transuranic elements (TRUs) and long-lived fission products from HLW and dedicated systems for their subsequent transmutation. The dedicated transmutation system is specially designed to burn minor actinides (MAs) efficiently in a very hard neutron energy spectrum and high neutron flux.

In this context, JAERI has been pursuing feasible concepts of accelerator-driven system as dedicated transmuters to be introduced in the second stratum (P-T fuel cycle) of the double stratum fuel cycle. The concept of the double stratum fuel cycle is schematically depicted in Fig. 1.

In the conceptual design study of accelerator-driven transmutation plant, two types of system concepts are being investigated. One is the solid system consisting of tungsten target and solid fuel core cooled by liquid sodium. Mononitride fuel is selected mainly on the basis of its good thermal performance and adaptability of dry reprocessing. The design of the solid system is based on the current technology and design practice for sodium-cooled fast breeder reactors.

Another advanced option is the molten-salt system with flowing chloride fuel. The molten-salt fuel also serves as target material and as coolant. One of the main advantages of fluid fuel is the capability of continuous on-line processing of actinides and reaction products. Chloride salt is selected mainly on the basis of actinide solubility.

2.1 Solid System

The design of the solid target/core system is based on the state of the art of technology for a sodium-cooled fast breeder reactor. An accelerator injects proton beam through a beam window into the solid tungsten target located at the center of the target/core. Surrounding the target is the subcritical core loaded with actinide nitride fuel. Spallation neutrons emitted from the target induce further fission in the actinide core region.

The target consists of multiple layers of tungsten disk with through holes for coolant passage. The target is designed to maximize the number of emitted neutrons and to flatten the shape of axial distribution of

neutron flux. The coolant holes are arranged to stagger from layer to layer to avoid the possibility of direct penetration of high energy particles through the target.

The target/core is surrounded by reflectors made of stainless steel. The target and fuel subassemblies are cooled by forced upward flow of primary sodium. The whole target/core including reflectors is contained within a target/core vessel made of steel as shown in Fig. 2. The target/core is of loop type configuration, having two sodium primary loops. A vertical duct for beam path is inserted into the target/core vessel down to just above the target region. The bottom end of the duct is the hemispherical beam window made of oxide dispersion strengthened (ODS) steel. The beam window is cooled by impinging coolant flow from the target exit.

Mononitride of minor actinides is used as fuel in the design of the solid system. Nitride fuel has advantages of a high thermal conductivity around 15 W/m/K and a high melting point around 3100 K. This excellent thermal performance permits a high linear rating and high power density, leading to a high transmutation rate. It also results in a low fission gas release.

High atom density of actinides is beneficial to achieve a hard neutron energy spectrum. This is also preferable to realize a dense fuel cycle for the second fuel cycle stratum, where actinides are maintained in a high concentration throughout the whole cycle [9]. The mutual miscibility of actinide mononitrides also leads to the fuel design of high concentration of actinide elements and simplifies the process of fuel fabrication. For reprocessing, an innovative fuel cycle combined with molten-salt electrorefining will be available in the case of nitride as well as metallic fuel.

With a 1.0 GeV-22 mA incident proton beam, the target/core having an effective neutron multiplication factor of 0.927 produces 360-MW thermal power. The net MA transmutation rate is approximately 110 kg per year, or 9.7 %/y, at a load factor of 80%.

Heat transport and power conversion systems in the plant design are similar to those for a sodium-cooled fast breeder reactor plant. The heat generated in the target/core is transported through the primary and the secondary sodium loops to the power conversion system. In the energy conversion system, thermal energy is converted into electricity through conventional steam turbine. A part of electric power is supplied to its own accelerator.

Major parameters of the nitride fuel solid system are summarized in Table 1. Further study is needed to detail and optimize the system design, taking full advantage of the excellent thermal performance of nitride fuel.

2.2 Molten-Salt System

A conceptual design study is being performed on a molten-salt target/core system as an advanced option for an accelerator-based transmutation system. Figure 3 schematically shows the proposed molten-salt system concept. Chloride salt with a composition of $64\text{NaCl}-5\text{PuCl}_3-31\text{MAnCl}_3$ is chosen as the fuel for its sufficiently high actinide solubility. The molten-salt fuel serves also as target material and as coolant. This eliminates the physical and functional separation of target and core, and thus significantly simplifies the target/core configuration.

One of the attractive features of the molten-salt fuel is the possibility of continuous fuel feed and on-line processing of reaction product removal.

The target/core is designed for 800-MW thermal power. Proton beam at 1.5 GeV is injected vertically into the central target/core region through the beam window. The target/core region is surrounded by an internal reflector. Intermediate heat exchangers and salt pumps are installed in the annular region around the internal reflector to reduce radiation damages. The intermediate heat exchangers are of compact type with plate-and-fin configuration, where heat in the primary molten salt is transferred to the secondary fluid of molten fluoride salt.

In the fluid fuel system, the heat removal problem usually appears in the intermediate heat exchangers than in the core itself. External shell-and-tube type heat exchangers of conventional design would require a large volume, leading to an unacceptably high actinide inventory and low transmutation rate for a dedicated transmutation system. In the present design, highly efficient compact type heat exchangers are incorporated within a reactor vessel. This in-vessel heat exchanger arrangement minimizes the total volume of the primary system and thus the total actinide inventory.

Another leading candidate fuel for a fast spectrum molten-salt system is $\text{PbCl}_2\text{-AnCl}_3$ (An: actinide). A preliminary nuclear calculation was made for the lead-based molten-salt system with the same geometry and dimensions as the sodium-based molten-salt system. The energy of incident proton is 1.5 GeV. Table 2 lists the calculated results in comparison with the sodium-based salt system.

A comparison is also made for two candidate molten salts from the point of thermal-hydraulics design view.

Primary flow cross section area S in a heat exchanger can be estimated from the Blasius equation $f = 0.3164 Re^{-0.25}$ for friction and the Dittus-Boelter equation $Nu = 0.023 Re^{0.8} Pr^{0.4}$ for heat transfer, where f = friction factor, Nu = Nusselt number, Re = Reynolds number, Pr = Prandtl number. In case when the maximum fluid velocity is a limiting factor in the design, S is expressed as

$$S = c (d Q / \lambda) (1 / \Delta\theta) Re^{-1} Pr^{-1},$$

where d = hydraulic diameter of the flow channel, Q = thermal power exchanged, λ = thermal conductivity $\Delta\theta$ = temperature drop through heat exchanger. It is noted that S is independent on the temperature difference between primary and secondary coolants. Assumptions were made that the maximum fluid velocity is limited by the pressure drop and the dynamic pressure in the primary system and the viscosity, thermal conductivity, and thermal capacity of the fluid are constant. Under these assumptions, S becomes proportional to the square root of the fluid density ρ . For other primary flow cross sections, the same relation holds if the pressure drop and dynamic pressure are to be unchanged. This results in the relation that the total volume of the primary system varies as $\rho^{1/2}$.

Table 3 compares system parameters of the sodium-based molten-salt system and those of the corresponding lead-based molten-salt system. The total inventory of the lead-based salt in the system is about 65% larger due to its higher fluid density than that of sodium-based salt, but the actinide inventory is about 25% smaller. The comparison is preliminary since reliable data of thermodynamic and transport properties are lacking for lead-based salt.

3. Spallation Integral Experiment

In the design study of accelerator-based transmutation system, a nucleon meson transport code, NMTC/JAERI is used to simulate nuclear reactions and the particle transport in the energy range above 15 MeV. To estimate and validate the accuracy of the code system, a spallation integral experiment using a large scale lead assembly is in progress. The lead assembly is bombarded with 500 MeV protons at the booster synchrotron facility of the National Laboratory for High Energy Physics (KEK).

The lead assembly was 60 cm in diameter and 100 cm in length. The protons were injected through the beam entrance hole into the target of 16 cm in diameter and 30 cm in length installed in the assembly. The target is made of lead or tungsten alloy. High purity metal activation samples of Al, Fe, Ni, Cu and Au were inserted into irradiation holes drilled parallel to the beam axis in the assembly at various radial distances. The gamma-ray spectrometry was used to obtain yields of the nuclides produced in the samples.

Neutron energy spectrum from thick lead target and nuclide yield distributions of various threshold reactions in a lead assembly were measured in the experiment. In general, the calculation with the codes NMTC/JAERI and MCNP4.2 was agreed fairly well with the experimental results.

There are however some discrepancies in both the nuclide production cross section and the neutron energy spectrum, requiring further improvements in the calculation model. Comparison of the yield of proton induced spallation products is planned in the next step in the integral spallation experiment to validate the accuracy of the NMTC/JAERI prediction.

4. Intense Proton Accelerator and Transmutation Test Facility

JAERI is proposing the development of an intense proton accelerator for tests in accelerator-driven transmutation technology together with various experiments in basic sciences. The plan called the Neutron Science Project is aiming at the scientific and technological innovation for the 21st century using neutrons. The study on accelerator-driven transmutation and the development of an intense proton accelerator are under way as the important part of the Project. The proposed accelerator will be of 1.5 GeV class with a beam power around 10 MW and employ superconducting linac in the high-energy portion. It will share the beam among research facilities, comprising an accelerator-based multi-purpose research complex.

To develop intense accelerator technology, R&D work has been carried out for the accelerator components for the front-end part; ion source, radiofrequency quadrupole (RFQ), a drift tube linac (DTL), and RF source. Prototype components of the ion source, RFQ, and RF source have been developed and tested successfully. The design study for an intense proton accelerator is also underway and efforts are focused on a superconducting

accelerating cavity as a main option for the high-energy portion above 100 MeV. The superconducting linac has attractive features for a high-intensity accelerator such as shorter length acceleration, large bore radius resulting in low beam losses and possible cost reduction in the construction and operation. A test stand equipped with cryogenics system, vacuum system, RF system and cavity processing and cleaning is being prepared to test the physics issues and fabrication process.

The preconceptual study for the transmutation experimental facility started in the late 1995. The experimental program is to be proceeded in two steps. The first step will be the feasibility study of the hybrid system concept with a spallation target and a uranium subcritical system at a low power level with a pulsed beam. The experiments will demonstrate the stable operation of a hybrid system and MA burnup with use of MA foils or pellets. It will provide and verify the database for the development and design of a test reactor facility in the second step.

In the second phase, a test reactor facility will be constructed to perform tests of integrated target/core system at a moderate thermal power level of 10-30MW with a CW beam. Transmutation capability of the hybrid system will be tested using MA target pins. These system level tests will demonstrate the technical viability of the accelerator-based transmutation system. Technical feasibility of spallation target and beam window will be also tested at a high beam power in the second step experiments. These experiments are expected to open up possibility for the development of a future nuclear option of accelerator-driven system for not only waste transmutation but also energy production and fuel breeding.

The JAERI Neutron Science Project of the accelerator-based research complex is planned to start in 1997 with conceptual design of the intense proton accelerator, transmutation test facility, and other research facilities. Construction will be started around 1999. Operation with 1.5-MW beam power is expected to begin as early as 2003, and upgrading up to the full beam power around 10 MW will be completed in 2006.

5. Summary

The conceptual designs of accelerator-driven solid system and molten-salt system for nuclear waste transmutation were presented in this paper together with brief outline of the program plan of tests in accelerator-driven transmutation technology.

JAERI accelerator-driven transmutation project is being proceeded under the Japanese long-term R&D program on P-T technology, called OMEGA. In the conceptual design study, both solid system and molten-salt system are being pursued as dedicated transmuters to be introduced in the second stratum of the proposed double stratum fuel cycle system.

In either system, an MA-loaded subcritical core is driven by a high-intensity proton accelerator and makes use of hard neutron energy spectrum to transmute MAs efficiently by fission. The design of the solid system is based on the status of liquid-metal fast reactor technology, whereas the design of the molten-salt system relies on the next generation technology.

The solid system employs actinide mononitride fuel that has several favorable features as fuel for dedicated transmutation systems in the proposed double stratum fuel cycle. Its main advantages are excellent thermal performance and high atom density of actinides. It is expected that further improvement in the transmutation performance can be achieved by taking full advantage of the excellent thermal performance of nitride fuel.

The molten-salt system employs flowing chloride salt fuel which also acts at the same time both as target material and as coolant. The fuel salt selected for the reference design is NaCl-AnCl_3 . Another leading candidate salt is $\text{PbCl}_2\text{-AnCl}_3$. A preliminary comparative nuclear and thermal-hydraulics design study was made for these two compositions of fuel salts. Results obtained seems to be not conclusive mainly due to the lack of reliable property data for $\text{PbCl}_2\text{-AnCl}_3$.

JAERI is proposing the development of an intense proton linear accelerator with a beam power of around 10 MW for tests in accelerator-driven transmutation technology together with various experiments in basic sciences. The preconceptual study for the transmutation test facility was initiated recently. The project is planned to start in 1997 with conceptual design of the accelerator and research facilities. Operation with 1.5-MW beam power is expected to begin as early as 2003, followed by gradual upgrading to the full beam power around 10 MW.

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Table 1 Major Parameters of Nitride Fuel Solid System

Fuel	Mononitride (90MA-10Pu)N MA: Np, Am, Cm
Target	Tungsten Multi-Layer Disk Type
Actinide Core Height	800 mm
Target/Core Volume	0.06/0.35 m ³
Actinide Inventory	1150 kg
Neutron Multiplication Factor	0.927
Proton Beam, Energy - Current	1.0 GeV - 22 mA
Average Neutron Flux	5.2×10^{15} n/cm ² /s
Power Density Average/Maximum	1020/1600 MW/m ³
Thermal Power	360 MW
Burnup	110 kg/y (9.7 %/y)
Coolant Temperature Inlet/Outlet	663/793K
Clad Temperature, Maximum	988 K
Fuel Temperature, Peak	1440 K

Table 2 Neutronics of Na-based Salt and Pb-based Salt Systems

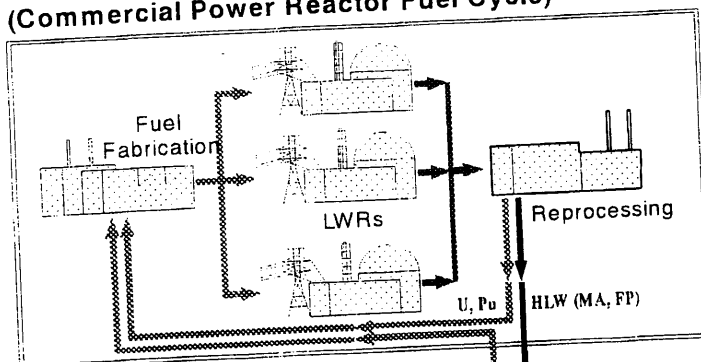
	64NaCl-36AnCl ₃	70PbCl ₂ -30AnCl ₃
Neutron Multiplication Factor	0.93	0.88
Spallation Neutron Yield (n/p)	37	40
Average Neutron Energy * (keV)	800	768
Power Density (keV/cm ³ /p)		
Average*	27	16
Peak	66	54
Peaking Factor	2.5	3.5

* averaged over target/core region, excluding IHX region.

Table 3 Property and System Parameters for Na-based Salt and Pb-based Salt

	64NaCl-36AnCl ₃	70PbCl ₂ -30AnCl ₃
Fluid Density (kg/m ³ at 923K)	3800	5300
Melting Point (K)	~726	~783
Primary System Volume (m ³)	2.7	3.2
Molten-Salt Inventory (kg)	10,000	17,000
Actinide Inventory (kg)	5,400	4,100

**First Stratum of Fuel Cycle
(Commercial Power Reactor Fuel Cycle)**



**Second Stratum of Fuel Cycle
(P-T Cycle)**

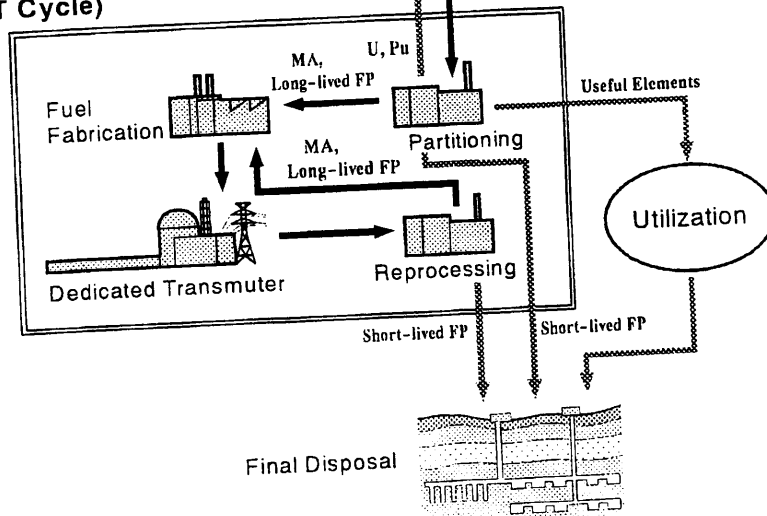


Fig. 1 Double-stratum fuel cycle

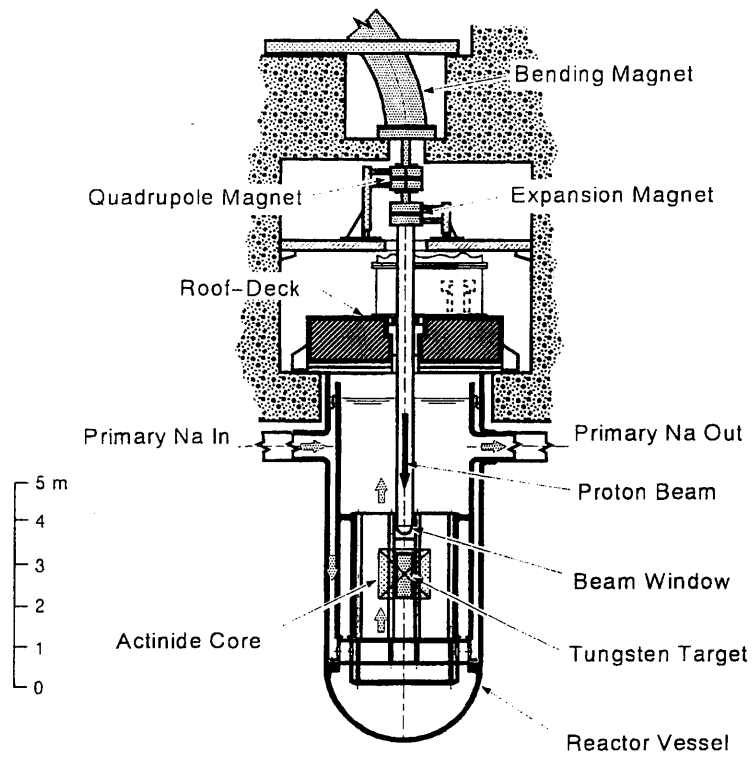


Fig. 2 Accelerator-driven nitride fuel solid target/core system

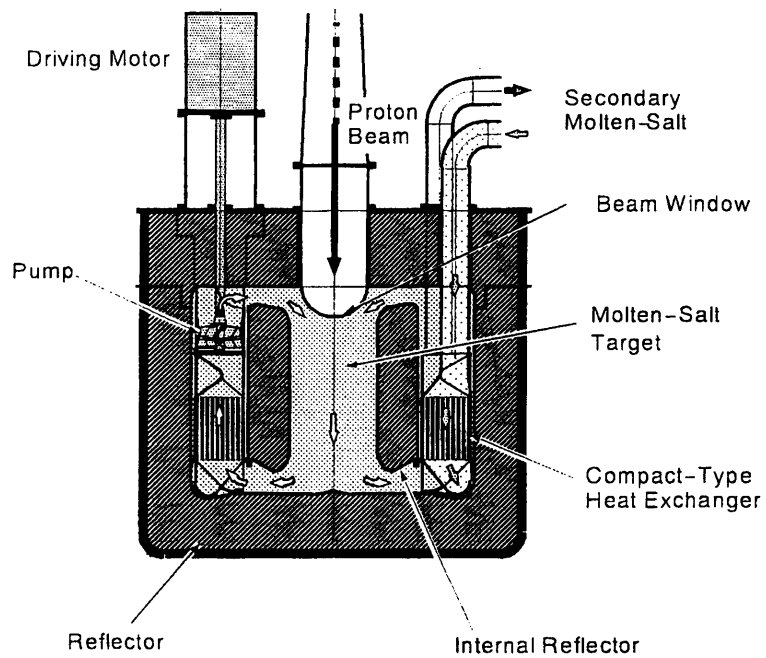


Fig. 3 Conceptual drawing of molten-salt target/core system