Code Development and Design Study on Accelerator-driven Transmutation Resaerch at JAERI

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Abstract

The calculation models in the cascade code NMTC/JAERI have been improved on bases of the new nuclear theory and new measured data. The intranuclear nucleon-nucleon scattering process calculation has been modified taking into accounts the effects of reflection and refraction of nucleon in nucleus and nucleon-nucleus scattering cross section. The preequilibrium process model was adopted to improve the prediction about nucleon back scattering. Adjustment was made for parameters in the high energy fission model and the adoption of total reaction cross section estimated systematically. Some benchmark calculations were carried out to verify the peidiction capability of the modified cascade code. The ACCEL code system for designing an accelerator-driven system has recently been upgraded as the ATRAS system. The Sn transport code TWOTRAN2 and Monte Carlo transport code MORSE have been replaced by new codes TWODANT with faster computation time and MCNP4A with the continuous Monte Carlo method repectively. The 73 groupwised cross sections upto 20 MeV have newly been produced to be used with the TWODANT code. The burnup code BURNER linked with TWODANT was also prepared well.

The conceputal design study on proton accelerator-driven transmutation systems with nitride fuel and new molten salt of minor actinides were carried out as another promising options. The solid fuel system consists of a sodium-cooled subcritical core with pin-bundle type assemblies of actinide nitride fuel and a multi-layer type spallation target of tungsten. It is expected to have the better thermal charachertistics than metal fuel system. When the system is driven by several tens mA proton beam with an energy of 1 GeV, amounts of minor actinides transmuted per year and the power generation were given respectively. The burnup calculations upto 300 days and the preliminary dynamics analysis were performed to examine the transmutation capability and tne nuclear safety in this system. The nuclear characteristics of the system with MA-$PbCl_3$ type chrolide molten salt fuel was examined and compared with MA-NaCl type one researched already.

1. Introduction

According to the national research and development program OMEGA on partitioning and transmutation [1], the research activities on the accelerator-based transmutation technologies are being promoted at JAERI in the following items,

(1) Development of simulation code system including the spallation cascade code,

(2) Design study on accelerator-based transmutation system,

(3) Spallation experiment with high energy proton beams [2)-3)],

(4) Development of an intense proton linac (1.5 GeV, 10 mA).[4]

In the present paper the recent works on the conceptual design study of the transmutation system and the development of the design code system are reported. Descriptions on the items (3) to (4) are given at other sessions in this meeting.

As well known, the calculation for designing the accelerator-driven transmutation system must treat the nuclear proocess in the wide range from ~eV to a few GeV. Unfortunitely there is not yet the single code that covers all reaction and transport in the energy range because of few available nuclear data in the high energy range. For the sake the hadron cascade code simulates the nuclear reaction and particle transpoet in the energy range higher than 20 MeV. Its prediction accuracy affects heavily on estimation of ability of accelerator-based transmutation system. So the cascade code NMTC/JAERI[5),6)] has been recently upgraded, taking into accounts new experiment data and calculational model, and the new version code named NMTC/JAERI94. In order to calculate the nuclear process in the accelerator-based system, the code system ACCEL[7] had been originally developed at JAERI before~ 1985 and includes the NMTC/JAERI and the neutron transport codes corresponding to ENDF-B4. To calculate more accurately and efficiently the nuclear reaction and particle transport process in the accelerator transmutation system over the energy range, the "ATRAS"(code system) has been recently developed at JAERI by adopting more powerful codes and the nuclear data library urgraded for higher actinides.

The radioactive nuclides to be transmuted in the JAERI transmutation research program are MAs such as ^{237}Np, ^{241}Am, ^{243}Am, ^{243}Cm, ^{244}Cm, ^{245}Cm and Long-Lived Fission Product (LLFP) such as ^{99}Tc, ^{129}I wtih half lives between a few hundred years and several million years. MA should be transmuted through fission reactions because the transmutation of MA by neutron capture has the possibility of increasing higher actinides. For designing the accelerator transmutation system at JAERI, we adopted the design guide lines as

(I) application of spallation neutrons to transmutation system,

(II) exclusive system for MA and long-lived FP burning,

(III) self-supporting system in energy ,

(IV) transmutation rate of MA (260 kg/y) from ten units of 1 GWe LWRs.

Fissioning of MA, which is a non-fissile material for thermal neutrons, occures dominantly to neutron capture above the threshold energy of about 500 keV. So JAERI selected the transmutation method of MA by fast fission reactions controlled by spallation neutron source although it requires the large initial MA inventory. LLFP can be transmuted only through thermal neutron capture to stable nuclides because of small capture cross sections for fast neutron. From the reason the thermalized neutron region may be set up around fast subcritical target/core to transmute MA and LLFP simultaneously in one system.

The design studies for two different types of accelerator-driven transmutation system, that is, solid fuel type and liquid fuel type, are being carried out at JAERI. The solid MA (alloy, nitride) fuelled system is composed of tungsten target and solid fuel subassemblies like fast reactor for MA burning. Its R&D works are considered to be the relatively short term project, except an intense proton accelerator, since most of technologies and know-hows already obtained in the fast reactor development can be effectively apllied to perform the design study of this type system. The system with the liquid fuels will be developed in the long term project. The molten salt system is expected to be the more promising transmutation system with possibility of on-line fuel reprocessing but of containing more problems to be resolved than the solid fuel one. The liquid alloy fuel system has a graphite moderator blanket to transmute LLFP by using thermalized neutrons. Both liqud fuel systems have the active, subcritical core region with the hard neutron energy spectra.

For the proton beam-controlled core operated in the subcritical mode, some advantages are considered in the following,

(a) No control rod, no safety rod, simple core configuration,

(b) Large flexibility in the core design due to less severe requirement to reactivity coefficient ,

(c) Operation stop by shutting down the proton baem,

(d) Less limitation to the burnup time of fuel and

(e) Mild requirement to the fine adjustment of fuel composition.

2. Development of code system for desiging the accelerator-driven tranamtor [8)-10)]

In the intranuclear cascade calculation in the NMTC/JAERI code, the nuclear medium effect for nucleon (N) scattering in a nucleus was sutdied and the accuracy of perdictions of reactions induced by lower energy protons was improved. For the the OECD/NEA code intercomparison task force, the benchmark calculations on the proton-induced nuclide production cross sections in some targets at the energy of 20 MeV to 5 GeV were carried out using the HETC/3STEP code. The calculated cross sections for heavier resultants had good agreements with experimental one except the accuracy of producing lighter nuclides. Neutronic calculations below 20 MeV in the transmutation system are carried out using the well-prepared transport code connected with the cascade code. Here informations about spallation neutrons with energies below 20 MeV is transfered from the cascade code to the neutron tanasport code as source neutron terms. For the calculation in the entire energy region, more excellent new code sytem ATRAS, including the new version cascade code and the 73 groupwised cross sections below 20 MeV extending the JAERI-fast set corresponding to the JENDL3.2, has been newly developed.

2.1 Improvement of cascade codes

For the energy region above 20 MeV, the spallation reaction with many reaction channels occurs dominantly as shown in Fig.1. However there is few nuclear reaction data in this range for various nuclides. At present the best way to analyze these high energy processes is to use the cascade simulation codes. JAERI is using the NMTC/JAERI (High Energy Nuclear Rreaction and Nucleon-Meson Transport Code)and NUCLEUS (only intranuclear cascade). The Bertini model in the cascade code[11)] successfully had treated the intranuclear cascade(INC) above 200 MeVof incident particles, using " two-bodies collision approximation between nucleons in a nucleus. Since the nucleons with the energy of 20 MeV to 200 MeV in a nucleus have gradually more wave-like nature, the disagreement between experimental data and predictions calculated by the INC model becomes larger. Some of calculation models and collision cross sections in the simulation code were installed before ~ 1975.

They have been improved and upgraded by the newly developed nuclear theory and the data measured on the recent high enegry experiments in the following items,

a) adoption of the in-medium nucleon-nucleon scattering cross section ,
b) the effects of reflection and refraction of nucleon in nucleus,
c) addition of preequilibrium process based on the exiton model,
d) modification of high energy fission model (Nakahara model),
e) replacement of original total cross section by Parlstein's systematics one,
f) adoption of level density parameter a_0 by Baba and new mass formulas.

In the original INC calculation the mean free path (mfp) for nucleon collision in a nucleus was approximated by the nucleon(N)-nucleon(N) scattering cross section in free space. The influence of nuclear matter mean field (nucleon-nucleus collision) becomes larger for an incident nucleon with less than ~200 MeV. So the parameterized in-medium N-N cross sections dependent on nucleon energy[12) - 15)] have been installed through the subcode ISOBAR intoNUCLEUS to compare more accurate calculation data with recent experimental ones. Figure 2 shows the in-medium total neutron-proton cross sections in the energy range of 50 to 300 MeVcalculated by taking into accounts the nucleon-nucleon pairing effect. The use of upgraded cross sections result in prolongation of mfp of nucleon in a nucleus. In Fig.3 the calculated angular distributions of emitted protons with 60 MeV are compared with experiment one in the Au thin foil bombarded by 100 MeV protons[16)]. The lines except solid line represent the contribution from each step in multiple scattering. As seen in these figures the distribution calculated by ISOBAR code with free space N-N cross section gives about a half experimental one, whereas the calculated distribution with in-medium N-N cross section agrees well with experimental one, in particular, in the backward direction. Figure 4 compared the calculated neutron production differential cross sections (DDX) with the experimental data by Amian et al.[17)] for the reaction Al(p,xn) induced by 800 MeV proton irradiations. There are some differences between both data in the energy range of a few MeV and a few tens MeV when the free space NN cross sections are used, while the result calculated with the in-medium cross section shows good agreement in each direction.

For the cross section of (p,n) and (n,p) reactions the threshold value has been adopted to treat the quasi-elastic colloision properly as many researchers suggested. The refraction and reflection effect[18)] has been also taken into acounts at boundaries assumed in a nucleus according nuclear matter density. Then a moving nucleon with a lower energy changes its direction even when crossing the boundaries. The exiton model[19) - 21)] was inserted in the Bertini INC model to consider the nucleon emission at the preequilibrium state. The calculated data using this improved version code were compared with ones measured in thin and thick (stopping lengh) targets. Figure 5 shows e xperimental and calculated production yields of 56Co in thenatNi reaction with threshold energy of 20 MeV in the tungsten target-installed lead assembly for 500 MeV proton bombardment.22), 23) Here the letter r denotes the radius distance from the axis of cylidrical target/assembly. New version code (NMTC/JAERI94; solid line) with total cross section estimated by Parlstein's systematics reproduces experiment data better than the original version code (NMTC/JAERI ; dotted liine) with geometric total cross section. In Fig.6 e xperimental and calculated neutron yields are shown in the stopping length Au target bombarded by 68 MeV protons.24) The Bertini model with preequilibrium process overestimates by a factor of three to ten the experiment neutron yields at the forward angle of 15° and 30° above 30 MeV but reproduces well at the angle of 60° and 120° due to the preequilibrium contribution. The ISOBAR code with free space NN cross section reduces the neutron emission at all angles and underestimates the experimental data relatively low. The ISOBAR code with the in medium N-N cross section parameterized by Cugnon reproduces the experimental data quite well. Especially at angles over 30° the calculation is in excellent agreement with the experiment and even at the angle of 15° still gives the lower neutron yield by 30 % than experiment. The applicability of The INC model foe both thin and thick target calculations was extended down to the incident energy of 68 MeV by the inclusion of the medium effects in terms of the relection and refraction and the in-medium NN cross sections.

The cascade code calculates also the high energy fission (HEF) reaction as competing process for particle evaporation reaction in an excited nucleus at equilibrium state after intranuclear cascade ceased. We have done the modification of HEF (Nakahara) model by adjusting the parameter in the the model and adopting of total reaction cross section revised by the systematically estimation. Experimental data of fission cross sections for ^{238}U(p, fission) in the high energy from 10 MeV to 10 GeV were surveyed and compared wtith predictions by both cascade codes of NUCLEUS and HETC/KFA2 as seen in Fig.7. Although experimental data are relatively distributed among authors, both code's predictions agreed within 50 % to them. It was often pointed out that the original Nakahara HEF model gives flatter mass distribution of residual nuclides than other model such as Atchison's one in HETC/KFA2[25]. In this model the mass distribution in actinide fisson reaction is calculated by superposing three Gaussian ones with different central values and a half width$<W_{1/2}>$(= E - E_f+ 7, E; excitation energy, E_f; fission barrier height). The adjustable parameter $< W_{1/2}>$ has been searched by changing E to be fitted to experimental data for ^{238}U(p, fission) reaction with a 300 MeV proton [26] as shown in Fig 8 and fixed to be $< W_{1/2}>$(E_{max}=15 MeV) for E>E_{max}. The mass distribution for ^{238}U(p, fission) reaction with proton energy of 2.9 GeV was calculated by this adjusted model in NUCLEUS to compare the adjusted result with experimental data [27] as seen in Fig.9 and both distributions showed farely the good agreement. This fact suggests that the excitation energy stored in an actinde has the maximum value, at least for high energy fission process. In calculating the Fission probability from the statistical theory, the level density parameter a_0 makes an important role. The a_0 dependent on mass number A was replaced from the value proportioal to A to Baba's estimated data. [27]. Figure 10 shows e xperimental fission cross sections from 1 MeV to 10 GeV for ^{235}U(n, fission) and ones predicted with Baba's data.

2.2 Benchmark calculations of cascade codes

For the OECD/NEA code intercomparison task force[28], proton-induced nuclide production cross sections in the targets made of mono nuclide such as ^{16}O, ^{27}Al, ^{nat}Fe, ^{59}Co, ^{nat}Zr and ^{197}Au were calculated at incident energies of 20 MeV to 5 GeV with the HETC-3STEP code[29]. This code was improved at JAERI to include the preequilibrium process in the original HETC code and newly to employ the level density parameter introduced by Ignatyuk[30], the atomic mass table of Audi and Wapstra[31] and the mass forrmula given by Tachibana et al.[32]. The HETC-3STEP code cannot estimate the production of isomar because the level structure of a nucleus is not considered in the evaporation model. Consequently the isomar production was not calculated.

The cross sections were calculated at 39 incident energies. The number of incident protons was chosen as 250,000 for ^{16}O, ^{27}Al and ^{nat}Fe, 200,000 for ^{nat}Zr and ^{197}Au, and 50,000 for ^{59}Co, respectively. In Figs 11 to 13 the cross sections calculated with HETC-3STEP are compared with the experimental results for the productions of ^{7}Be, ^{89}Zr, ^{190}Ir and ^{197}Hg in the ^{197}Au target. As seen in Fig. 11, this code cannot estimate the ^{7}Be production because of lack of the fragmentation process model. The similar discrepancies are observed for all the other tragets. For the ^{89}Zr production, the HETC-3STEP reproduces the experimental result quite well in Fig 12. Aithough the high energy fission of sub-actinide nuclides is taken into account in HETC-3STEP, the calculated result indicates that the present fission model is applicable to the estimation of the medium-mass nuclide production by the spallation reaction of a heavy nuclide. It is observed in Fig. 13 that HETC-3STEP's results also agree well with the experimental ones of the ^{190}Ir production. Even for the other nuclides in the mass range of 177 to 197, a fairly good agreement with a factor of two to three is obtained between the calculated and experimental results. For the other target, however, ligher the mass of produced nuclides become, worse the agreement between the calculated and experimental results becomes. Therefore the accuracy in the HETC-3STEP calculation for the other targets is poorer than that of ^{197}Au even in the production of a nuclide with mass number close to the target one.

As the present intranuclear cascade/evaporation model calculates the nuclear reaction in a very classical manner, the accuracy of a factor of two to three for resultant production seems to be acceptable. In order to improve the accuracy of the nuclide production cross section in the framework of the intranuclear cascade/evaporation model, the following physical aspects should be taken into account: (i) the accurate treatment of the threshold energy of the nuclear reaction, (ii) the inclusion of the fragmentation reaction, (iii) more accurate estimation of the charged particle emission in both the INC and preequilibrium processes, (iv) more precise simulation of the statistical decay process by the use of the Hauser Feshbach model.

There are many light nuclei such as Cl in active fluid tagrets in the JAERI-type liquid transmutation system as described later. In the target region the spallation reaction of MA and light nuclei occur simultaneously in addition of fast fission transmutation of MA. So cascade calculations for the high energy hadron nuclear rection in a light nucleuus and the transport process in the dilute fluid target/core should be analyzed with the higher precision.

2.3 Development of the entire code system ATRAS

The design code system ATRAS (Accelerator-based Transmutation Reacotr Analysis System) has been developed to carry out the accelerator transmutation calculation more accurately and effectively. This system consists of the cascade code NMTC/JAERI94, Sn transport code TWODANT, Monte Carlo one MCNP4A, and the burnup code BURNER linked with TWODANT as seen in Fig. 14 . The 73 groupwised cross section set below 20 MeV was newly developed on base of the JFS-3-J2 set from the cross section library JENDL-3.2, where the connecting energy between cascade and transport codes was changed from 15 MeV to 20 MeV corresponding to the available energy in the JENDL 3.2.

The ACCEL has been revised and named the ATRAS, which is the integrated code system which can perform following calculations

1) spallation, evaporation and high energy fission processes above 20 MeV,
2) neutron transport process below 20 MeV and
3) core burnup process below 20 MeV.

The first process was calculated with the NMTC/JAERI4 code in which many modifications were applied as described above. For the calculation of the neutron transport below 20 MeV, the two-dimensional Sn transport code TWODANT[33] and the three-dimensional Monte Carlo transport code MCNP-4A[34] were selected. Both codes were modified to apply the non-isotropic volume source of spallation neutrons emitted from the target for the fixed source calculation. The effective microscopic and macroscopic cross sections needed for the transport calculation can be generated by the CSASI module from the SCAL-4 code system[35]. For the third burnup process the ATRAS code is capable for the burnup analysis of the accelerator-based transmutation systems. The BURNER code, burnup calculation module in the VENTURE[36] code system, was used for this purpose. The code was used, linking with TWODANT code to include the effect of the changes of core-averaged neutron spectrum corresponding to each burnup step.

The new file of groupwised cross sections was also prepared for the ATRAS code from the latest JENDL-3.2 library. Structure of this groupwised cross section file is almost the same as the fast reactor group constants set JFS-3-J2. Three groups were added to the energy range from 10 MeV to 20 MeV to connect smoothly the transport code with the NMTC/JAERI code. About 30 nuclides, which are necessary to calculate the JAERI-proposed accelerator-based transmutation systems, were selected from the JENDL-3.2 library. Higher actinides such as Bk and Cf and long-lived fission products (Tc-99, I-129) were also selected for this groupwised cross section file.

By using these codes we could calculate the nuclear characteristics about the accelerator driven transmutation systems in the whole energy range more efficiently and accurately. The ATRAS will be opened for outside use in the near future.

3. Design Studies on accelerator-driven transmutation system

The design studies for two different types of accelerator-driven transmutation system, that is, solid fuel type and liquid fuel type, are being carried out at JAERI.[37),38)] In the present paper the MA nitride fuelled core and the Pb-type chrolide molten salt core were researched respectively as the anouther promising option.

3.1 Nitride fuel system

The JAERI system concept with TRU-nitride fuel is now under study to take full advantage of the excellent thermal performance of nitride fuel. Neutronics analysis of the accelerator-based transmutation system with solid disk tungsten target and TRU-nitride fuel was performed using the ATRAS codes. This core is a cylinder in dimensions of 80 cm height and 40 cm radius, surrounded by the stainless steel reflector with thickness of 50 cm. The fuels with composition of TRU (90{Np, Am, Cm}:10Pu) -^{15}N are arranged as pin-bundle type assemblies around tungsten target in the core and cooled by sodium flow. Specifications of the core are listed in the Table 1. The target was divided into the thin-disk part and thick-part to flatten the released neutron distribution from the side surface of the target. Results of neutronics calculation are summarized in Table 2. The calculation results showed that this system with k_{eff} of 0.927 and average neutron fluxes of 2.67 x 10^{14} n/cm^2/mA can transmute about 5.18 kg/mA of TRU per year and generate a thermal power of 15.7 MW/mA in operation with the 1 GeV proton beam. TRU inventory in the core is about three times lower than the metallic fuel system because of higher density of heavy metal in the nitride fuel. Much higher power density, enough trasmutation ratio and neutron spectrum as hard as in the metallic fuel system have been gotten also. The specification of the system was proposed for a benchmark problem in the " OECD/NEA/NSC Benchmark on Physics Aspects of Different Transmutation Concepts".

3.2 Molten salt system [39]

The system with the liquid fuel such as the molten salt is being developed as the long term project in the OMEGA plan. In the liquid target/core transmutation system using MA molten salts, there are no limitations for preventing the melt down accident of fuel and target due to local power peaking. The minor actinides (MA) concentration in molten salt fuel is lower than one in solid fuel. The power peaking around the beam-irradiated zone is not a critical factor because of the relaxed heat condition due to liquid fuel. So the high energy proton beam can be injected directly on the center of the MA fuelled zone where there is no target region with the definite boundary. The concept of an advanced MA molten-salt system has been studied to apply the advantages of a simple fluid target/core to the transmutation system. The flowing MA molten salt forms the subcritical core and itself acts also as spallation target and primary coolant circulating between target/core and heat exchanger in the calculational model in Fig. 15. This system has the reaction vessel with 170 cm in height and 105 cm in radius and the stainless steel reflector with thickness of 20~40 cm. The compact heat exchanger and molten salt pump are installed inside the vessel to reduce TRU molten salt inventory. Nuclear characteristics in the subcritical core filled by the promising chloride molten salt 70PbCl_2-30TRUCl_3, with hard neutron spectrum, has been examined for the same configuration in reaction vessel to one with 64NaCl-36TRUCl_3 which was already studied as the previous case. The flouride molten salt NaBF_3-NaF was selected as the secondary coolant. The plotting curves in Fig. 16 (a) and (b) represent the neutron hard energy spectra in both molten salt target/core irradiated by 1.5 GeV protons and the outer torus region where the primary cooling system (heat exchangers and pumps) is installed. These regions are separated by an inner cylidrical reflector to protect the heat exchangers and pumps against high neutron flux. The neutron flux in the outer region is kept to be lower by about two orders of magnitude than one in target/core. The power density distribution in the target/core region irradiated with 1.5 GeVprotons is shown in Fig.17, where the maximum power density is about 70 MW/m^3 at proton current of 1 mA. This system with k_{eff} of 0.93 and 1.5 GeV-20 mA proton beam can transmute 238 kg of MA per year and produce the electricity of 228 MW sufficeint to power up the accelerator. The operating condition is compared with one in the case of 64NaCl-36TRUCl_3 fuel salt in Table 3.

As reported already, the molten salt transmutation system is expected to have the possibility of removing reaction products from the system through He purge method, Cd metal extraction method, cold trapping method and oxidation method respectively.

3.3 Burnup calculations of the JAERI accelerator-driven transmter

To examine the transmutation capability and the nuclear safety the burnup calculations were carried out in the energy region below 20 MeV for the accelerator-driven MA transmutation systems composed of tungsten target and solid nitride-fuelled core, using the BURNER-TWODANT code in the ATRAS. The one group cross sections were made by averaging the microscopic cross sections produced from JENDL 3..2 with the neutron spectrum calculated by the TWODANT and with the decay data library. From the burnup calculation, it revealed that the transmuted amount is about 300 kg of TRU annually at the constant thermal output of 800 MW through the burning period by adjusting the proton beam current. The evolution of actinide compositions with burnup time was calculated up to 300 days. Figure 18 represents the dependences of the minor actinide compositions. It is apparent that main minor actinide ^{237}Np is reducd to about three fifth of initial inventory at 300 burningdays. The fact implies the transmutation ability of about 200 kg per year. The BURNER code in the latest version has been improved to include the fission product effects, based on the lamped FP method. Figure 19 shows the comparison of dependences of k_{eff} on the burnup time calculated by the BURNER without and with the fission product effects. The k_{eff} was recalculated by the code TWODANT for the renewed compositions of actindes on each step. It has been recognized that the system for both cases always keeps the subcritical state with sufficient margin ($k_{eff} < 0.96$) although k_{eff} swings to the positive side over this stage. It seems that the FPs produced in core due to actinide burning makes the good role to supress thereactivity swing for this short burning period.

3.4 Dynamics calculations of the JAERI accelerator-driven transmter

The dynamic analysis on the proton beam-driven subcritical transmutation system is very improtant from a point of view of nuclear safty at normal operation and accidental stiuation. The prelimary dynamic calculations were done for the metal fuelled fast reactor at subcritical stste simulating the proton beam-driven subcritical core, using the EXKARS code. The transient process of the fast reactor with the effective neutron multiplication factor k_{eff} of 0.96~0.99 and neutron source was calculated by solving the standard dynamic equation including only Doppler and coolant's temperature reactivity coefficients for some reactivity insertions.

4. Summary

The ACCEL code system calculating the nuclear process in the accelerator-driven transmutation system in the wide energy range of eV to GeV has recently been upgraded as ATRAS. For the energy region above 20 MeV the cascade codes NMTC/JAERI and NUCLEUS have been upgraded by taking into accounts the in-medium nucleon-nucleon cross sections, the effects of reflection and refraction of nucleon in nucleus, the preequilibrium process, modification of high energy fission model and Baba's level density parameter. These improvements have increased the accuracy of prediction of main parameters estimating the transmutation ability. For Neutronic calculations below 20 MeV in the transmutation system more excellent transport code such as Sn code TWODANT and Monte Carlo code MCNP 4A have newly been prepared respectively. The 73 groupwised cross sections below 20 MeV were produced for the TWODANT calculation from the JENDL3.2 library to calculate effectively the nuclear process of system with spallation neutron spectrum harder than fission neutron one. To examine the transmutation capability and the nuclear safety the burnup code BURNER linked with the TWODANT code was prepared well. By using these excellent and powerful codes we could calculate more efficiently the nuclear characteristics about the accelerator-driven transmutation system in the whole energy range.

The conceptual design studies were also carried out on two promising transmutation systems, MA nitride fuelled and MA Pb-type chrolide molten salt cores, using the ATRAS codes. The proposed prototype nitride fuel accelerator transmutation system with $k_{eff} = 0.927$ can transmute 5.18 kg/mA of MA per one year and generates the thermal power of 16 MW/mA when operated by the proton beam with the energy of 1 GeV. From the burnup calculation, it was shown that the TRU amount transmuted in the nitride fuel system is about 300 kg annually at the constant thermal output of 800 MW. The evolution of actinide compositions with burnup time was calculated up to 300 days. It was also recognized, from the calculation considering the FP production in the core, that the system always keeps the subcritical state with sufficient margin ($k_{eff} < 0.96$) upto 300 days. The Pb-type chloride molten salt has been chosen due to better nuclear characteristics for studying the new MA molten salt transmutation system. This system with $k_{eff} = 0.93$ can transmute about 238 kg/y of actinides and generates the electric power of 228 MW in operation with a 20 mA proton beam with an energy of 1.5 GeV. The prelimary dynamic calculations was started for the subcritical fast reactor simulating the proton beam-driven subcritical core.

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References

1) Yoshida, H., et al., " A Strategic Study of the Partitioning and Transmutation System being Developed at JAERI," OECD/NEA/P&T REPORT No.7, pp.79 (1992) .
2) Takada, H., Kanno, I., Hasegawa, K., and Sasa, T., "Production of Radioactive Nuclides in a Lead Assembly wirh 500 MeV Protons," PSI Proc. 92-02, ISSN 1019-6447, pp.568 (1992).

3) Takada, H. et al., JAERI-M 93-181, pp. 210 (1993).
4) Mizumoto, M., et al. : "High Intensity Proton Accelerator for Nuclear Waste Transmutation," Proc. 16th Int. Linear Accelerator Conf. LINAC-92, (1992) (Ottawa).
5) Nakahara, Y. and Tsutsui, T. , " NMTC/JAERI : A Simulation code system fot High Energy Nuclear Reactions and Nucleon-Meson Transport Processes", JAERI-M 82-198, (1982)(in Japanese).
6) Nishida, T. ,Nakahara, Y. and Tsutsui, T. , "Development of a nuclear spallation simulation code and calculation of primary spallation products", JAERI-M 86-116, (1986)(in Japanese).
7) Nakahara Y., et al. : " ACCEL : Code System for Analizing the Nuclear Characteristics on Acclerator-Target/Blanket System," (1981) (Private communication in Japanese).
8) Atchison, F., 1980, Jul-Conf-34.
9) Nishida T., Takada H., Nakahara Y., Takizuka T., Yoshizawa N. and Iwai S. : "Benchmark Study On the Computational Model in Accelerator-Based Transmutation Simulation Code," PSI Proc. 92-02, ISSN 1019-6447, pp.535 (1992).
10) Prael, R.E., Lichtenstein, H. : " USERS GUIDE TO THE LAHET CODE SYSTEM," LA-UR-89-3014.
11) Bertini, H,W.,, Phys. Rev. 188, pp.1711 (1969).
12) Nakahara, Y., J. Nucl. Sci. Technol., 20, pp.511 (1983).
13) Suetomi, E., Kishida, N. and Kadotani, H., Phys. Lett., B333, pp.22 (1994).
14) Li, G.Q. and Machleidt, R., Phys. Rev. C49, pp.556 (1994).
15) Takada, H., J. Nucl. Sci. Technol., 33, pp.275-282 (1996).
16) Cowley, A.A., et al., Z. Phys..A336, p.p. 189 (1990).
17) Amian , W.B. et al , Nucl. Sci. Eng. 112, pp.78 (1992).
18) Cugnon, J., Lemaare, M.-C., Nucl. Phys. .A489, (1988) and Private Communication.
19) Tuma, J.J., " Handbook of Physical Calculation," McGraw-Hill Book, p.p. 251 (1976).
20) Yoshizawa, N., Ishibashi, K. and Takada, H., J. Nucl. Sci. Technol., 32, pp.601(1995).
21) Feshbach, H., Kerman, A. and Koonin, S., Ann. Phys. 125, , p.p. 429 (1980).
22) Takada H., et al. : "Production of Radioactive Nuclides in a Lead Assembly wirh 500 MeV Protons," PSI Proc. 92-02, ISSN 1019-6447, pp.568-583 (1992).
23) Takada H., et al. : J. Nucl. Sci. Technol. 31(1), p. p. 80-82 (1994).
24) Meigo, S., Private communication (1995).
25) Nishida T., Takada H., Nakahara Y., Takizuka T., Yoshizawa N. and Iwai S. : PSI Proc. 92-02, ISSN 1019-6447, p.p. 535-553 (1992) .
26) Friedlander, G., et al., Phys. Rev.,129, p.p.1809 (1963).
27) Baba H. : Nucl. Phys. A159, p.p. 625 (1970).
28) Michel R. and Nagel P. : NEA/NSC/DOC995) 8, "Specification for an International Codes and Model Intercomparison for Intermediate Energy Activation Yields," (1995).
29) Yoshizawa N., Ishibashi K. and Takada H. : J. Nucl. Sci. Technol., 32, p.p. 601 (1995).
30) Ignatyuk A. V., Smirenkin G. N. and Tishin A. S. : Sov. J. Nucl. Phys., 21, p.p. 256 (1975).
31) Audi G. and Wapstra A. M.: Nucl. Phys. A, 565, p.p. 1(1993).
32) Tachibana T., et al.: At.Data Nucl. Data Tables, 39, p.p. 251 (1988).
33) Alcouffe R.E. et al. :" Usersguide for TWODANT : A Code Package for Two-Dimensionnal, Diffusion-Accelerated, Neutral-Particle Transport," LA-10049-M, (1990).
34) Briesmeister J.F. (Ed.) : " MCNP-A General Monte Carlo N-Particle Transport Code, Version 4A," LA-12625, (1993).
35) " SCALE-4 A Modular Code System for Performing Standarised Computer Analysis for Licensing Evaluation," CCC-545, (1990).
36) " BOLD VENTURE IV, A Reactor Analysis Code System, Version IV," CCC-459, (1980).
37) Takizuka, T., Nishida, T., Takada, H., Meigo, S. and Mizumoto, M., :" Conceptual Design Study of an Accelerator-based Actinide Transmutation Plant with Sodium-cooled Solid Target/Core," OECD/NEA/P&T REPORT No.7, pp.397 (1992).
38) Takizuka, T., Nishida ,T., Mizumoto, M. and Yoshida, H., : " Present Status of Accelerator Based Transmutation Research," Proc. 8th Journees SATURNE, LNS/Ph/94-12, pp.109-113 (1994) (Saclay).
39) Kato, Y., et al., PSI Proc. 92-02, ISSN 1019-6447, p.p. 133 (1992).

Table 1 Design specifications for the nitride fuel transmutation system

Proton beam	1.0 GeV
Proton beam radius	15 cm
Proton beam profile	uniform
Beam duct radius	15 cm
Target/Core	concentric cylinders with a height of 1 m
Radii	15 cm / 40 cm
Target	tungsten (disk layer type)
Upper target	height 26 cm, disk thickness 1.5 cm
Lower target	height 54 cm, disk thickness 13 cm
Fuel	
(90MA-10Pu)N (MA:Np, Am, Cm)	
Fuel pin outside diam.	7.3 mm
Pin pitch	9.9 mm
Effective fuel height	80 cm
Fuel pellet diam.	6 mm
Sodium bond	thickness 0.35 mm
Cladding	thickness 0.3 mm (HT-9 SS)
Reflector	stainless steel
Sodium Volume Fraction	
Upper target	86 %
Lower target	37 %
Core	62 %
Reflector	41 %
TRU initial inventory	1150 kg

Table 2 Neutronics charateristics in the nitride fuel transmutation system

k-effective	0.927
Thermal output	15.7 MW/mA
TRU disappearance	
Np	1.93 kg/mA/year
Pu	1.00 kg/mA/year
Am	1.96 kg/mA/year
Cm	0.29 kg/mA/year
Average neutron energy	840 keV
Neutron fraction above 1 MeV	24.6 %
Average neutron flux	2.67×10^{14} (n/cm^2/mA)
Maximum reactivity swing	+ 6.7 % delta-k/k at 210 days

Table 3 Comparison of operating conditions for both molten salt system

	$PbCl_2$ type	NaCl type
Target/Fuel/Primary coolant	Chroride molten salt	
(composition)	$70PbCl_2$-30(Pu+MA)Cl_3	64NaCl-36(Pu+MA)Cl_3
Secondary coolant	Fluroride molten salt	
(composition)	$92NaBF_3$-8NaF	
Actinide inventry	4932 kg	5435 kg
k_{eff}	0.92	0.91
Proton beam	1.5 GeV-28 mA	1.5 GeV-25 mA
Thermal power	800 MW	800 MW
Transmutation	250 kg/y (5.1 %/y)	250 kg/y (4.6 %/y)

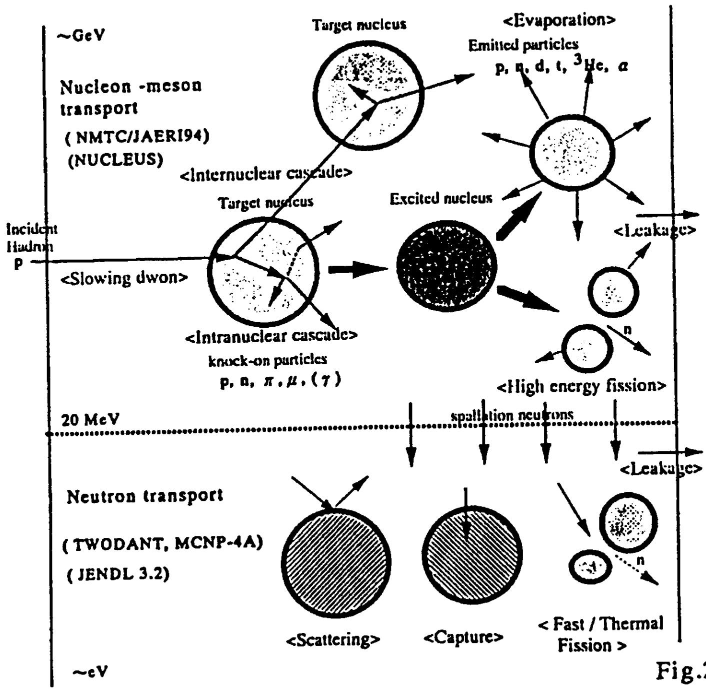


Fig.1 Schematic illustration of spallation & cascade and neutron transport

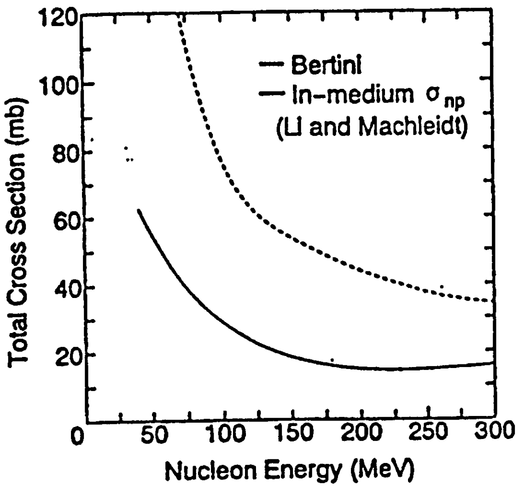
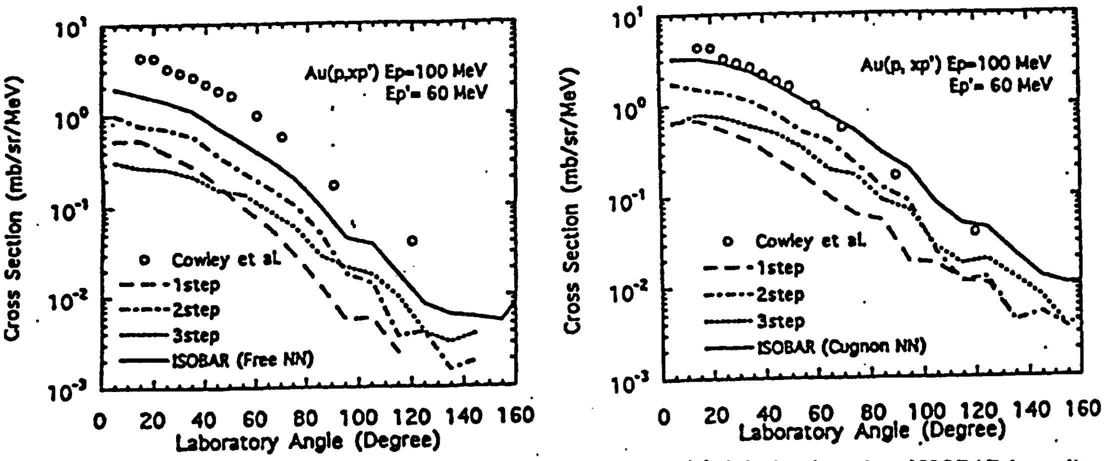


Fig.2 Comparison betweem the free nucleon-nucleon cross section and in-medium ones parameterized by Li, Machleidt and Zhuo



(a) Calculated results of ISOBAR-free

(b) Calculated results of ISOBAR-In-medium

Fig.3 Angular distributions of protons emitted with 60 MeV in the Au (p,xp') reaction for 100 MeV ptoton incidence

The open circle indicates the experimental data. The solid, dashed, dot-dashed and dotted lines represent the total, 1st step, 2nd step and 3rd step contributions of multi scatering in the calculation with ISOBAR respectively.

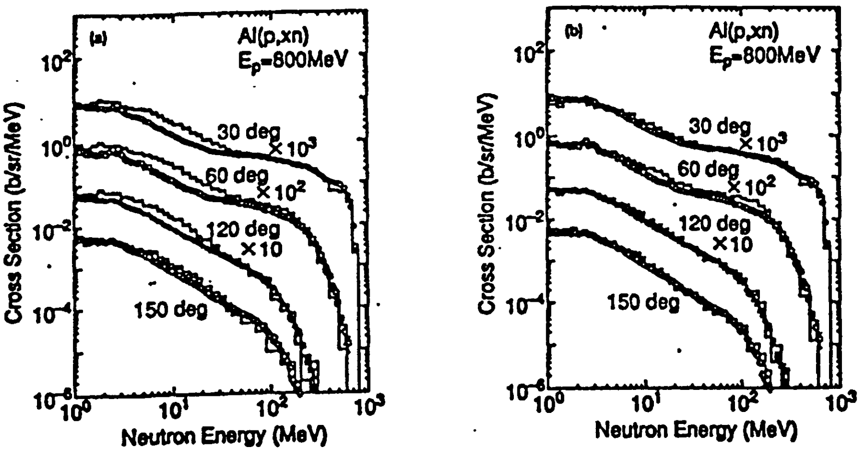


Fig.4 Calculated neutron production DDX in thin Al target bombarded by 800MeV protons are compared with the experimental data The solid linees stand for the calculated values with statistical error $\pm\sigma$ (a) free space NN cross sections and (b) in-medium cross section. The open symbols show the experimental data by Amian.

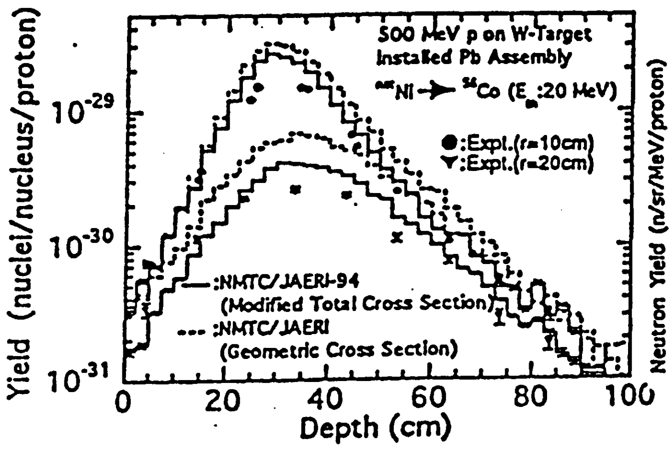


Fig.5 Experimental and calculated yields of ^{56}Co in ^{nat}Ni samples in the tungsten target-installed lead assembly for 500 MeV proton bombardment.

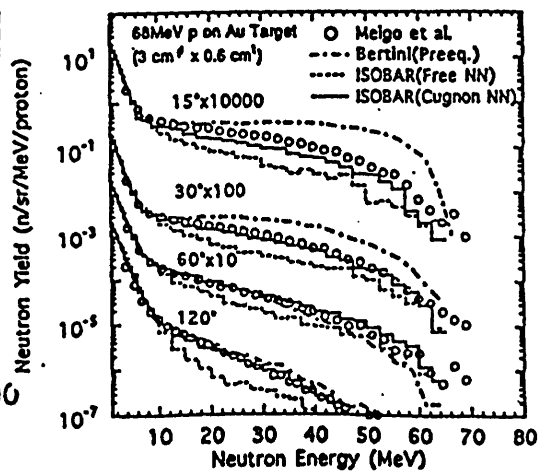


Fig.6 Experimental and calculated neutron yield from the stopping length Au target bombarded by 68 MeV protons

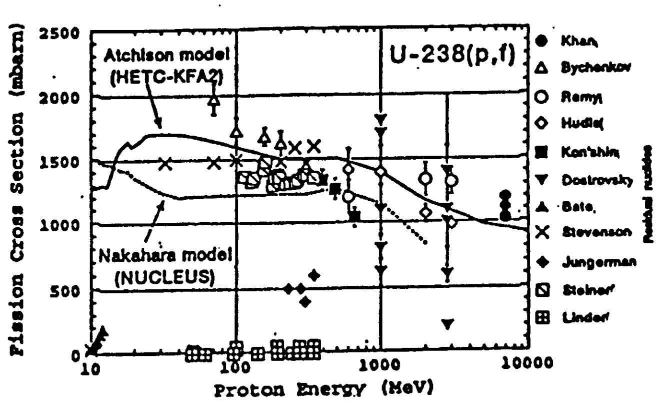


Fig.7 Experimental fission cross sections for ^{238}U(p, fission) from 10 MeV to 10 GeV and ones predicted by both cascade codes of NUCLEUS and HETC/KFA2

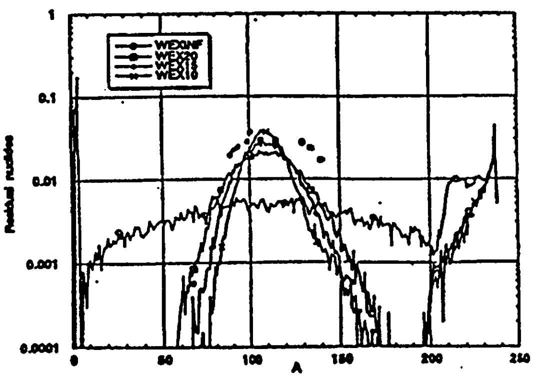


Fig.8 Adjustment of width in the FP yield curve in Nakahara's HEF model to be fitted to experimental data for ^{238}U(p, fission) reaction with proton energy of 300 MeV

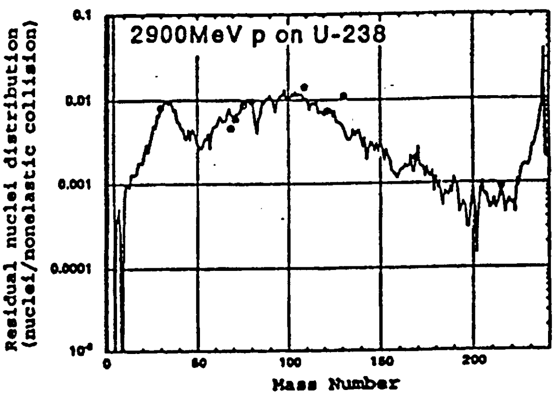


Fig.9 Comparison of the mass distribution of products of calculated by the modified Nakahara model with experimental data for ^{238}U(p, fission) reaction with proton energy of 2.9 GeV

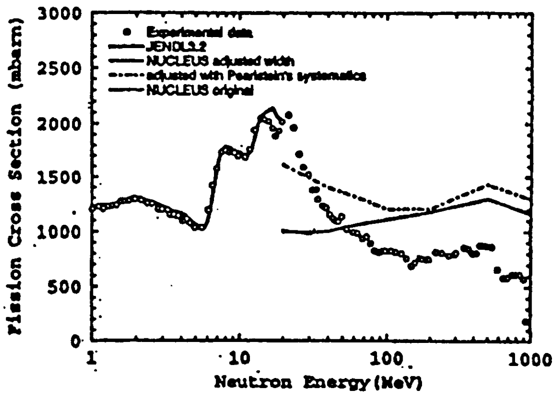


Fig.10 Experimental fission cross sections from 1 MeV to 10 GeV for ^{235}U(n, fission) and ones predicted by the NUCLEUS with Baba's data and Ignatyuk formula

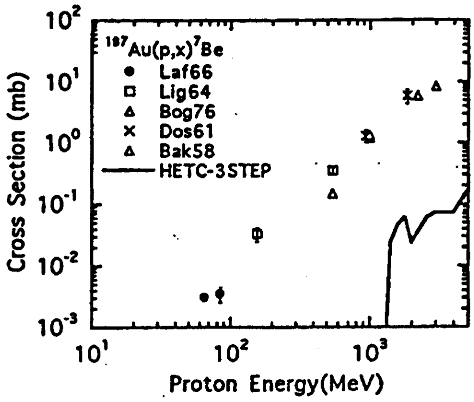


Fig.11 Cross sectons of the $^{197}Au(p,x)\,^{7}Be$ reaction The marks indicate the experimental results. The solid line stands forthecalculatedresulsofHETC/3STEP.

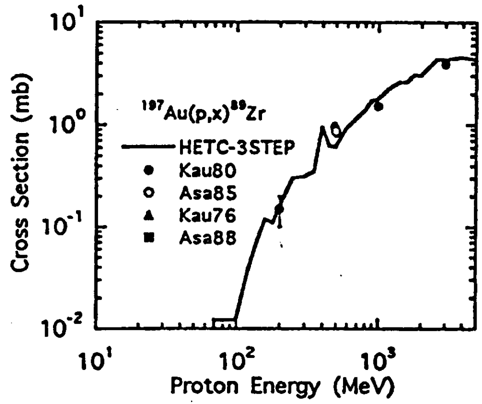


Fig.12 Cross sectons of the $^{197}Au(p,x)^{89}Zr$ reaction

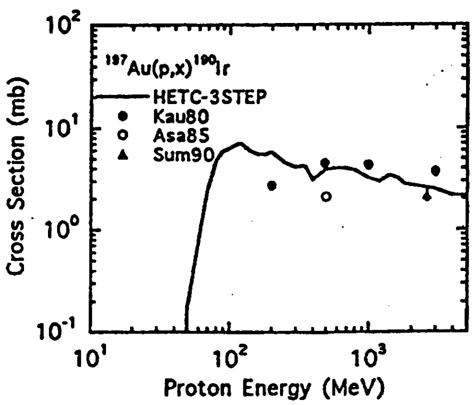


Fig.13 Cross sectons of the $^{197}Au(p,x)^{190}Ir$ reaction

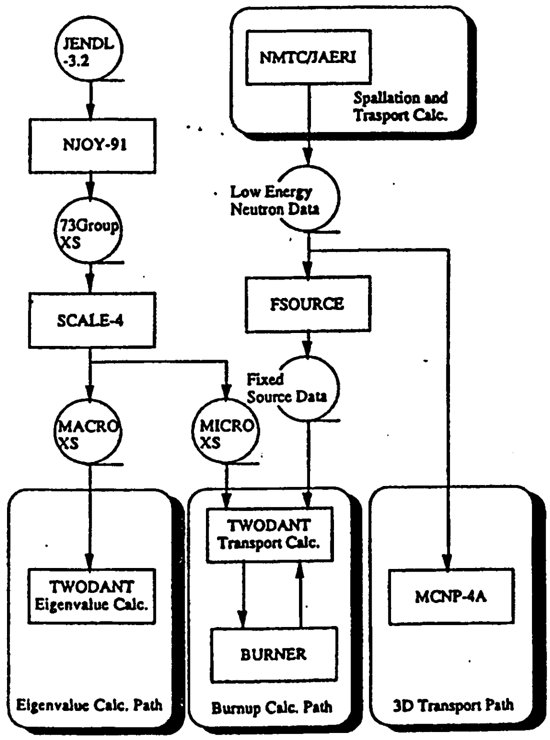


Fig.14 Flow chart of the ATRAS code system

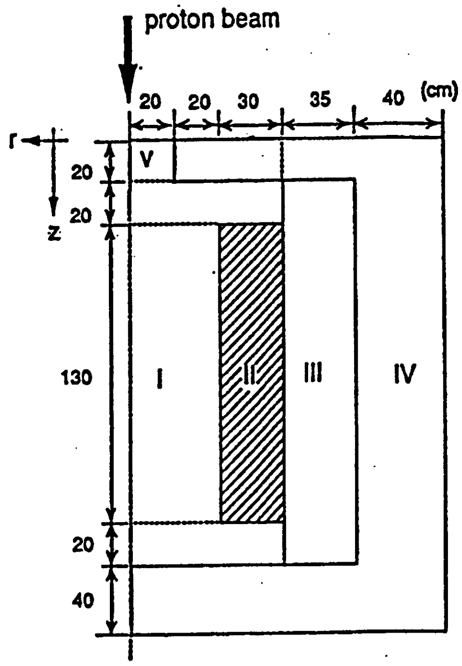


Fig.15 Calculational model of MA molten salt target/core transmutation system

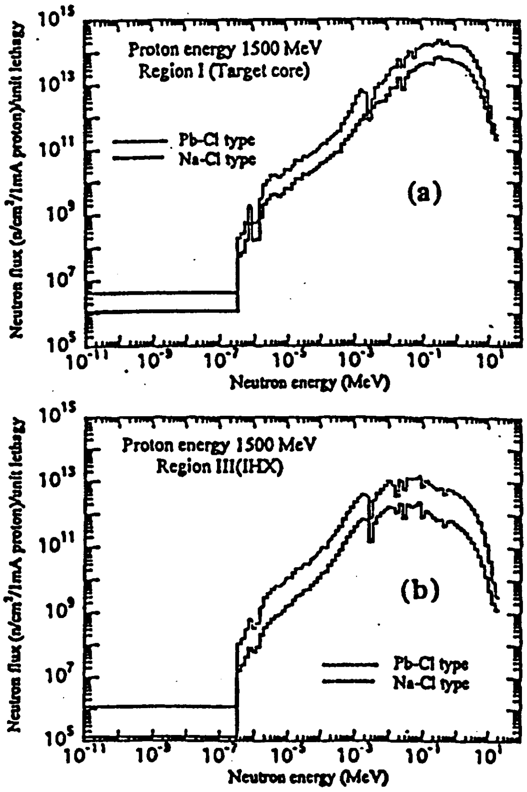


Fig.16 Calculated neutron energy spectra in the target/core filled with TRU chloride molten salt and the outer porus region for heat removal

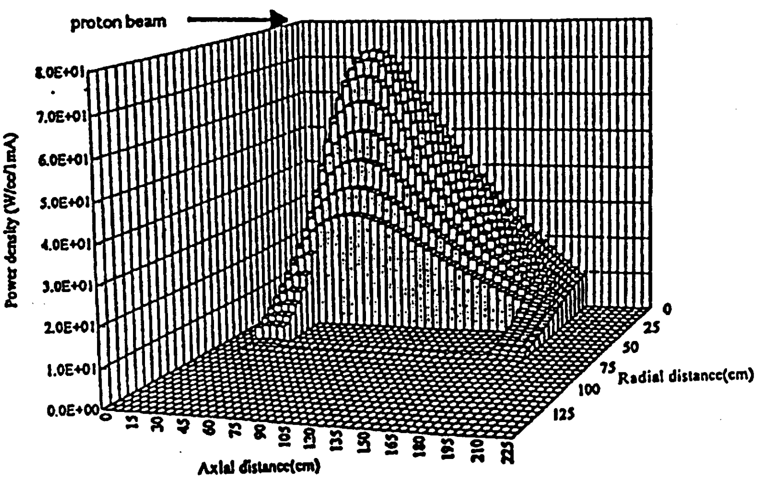


Fig.17 Power density distribution in the target/core region

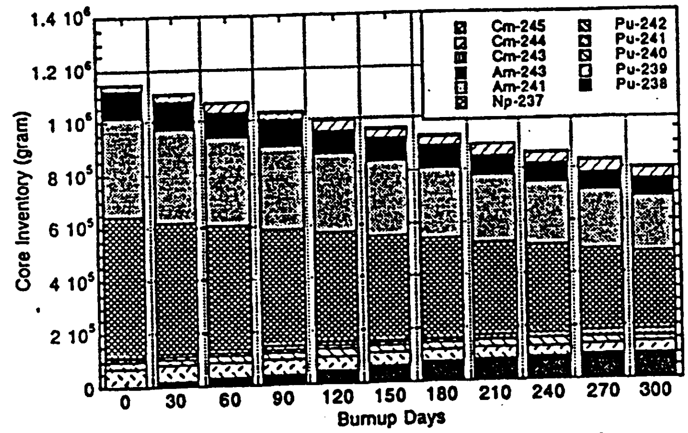


Fig.18 Variation of minor actinides in the nitride fuel core upon burnup

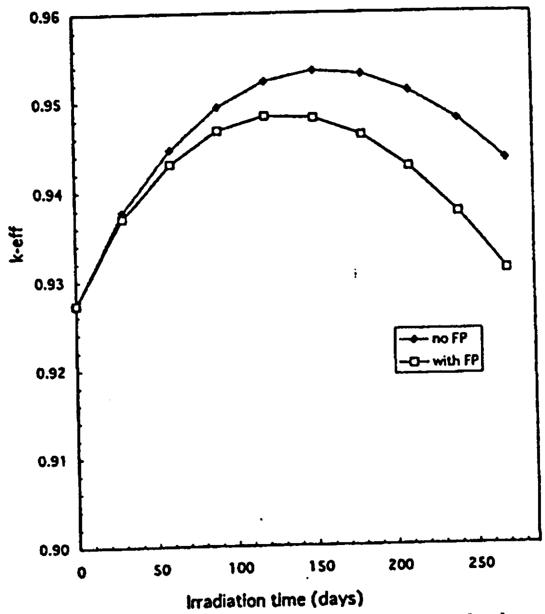


Fig .19 Comparison of dependences of k_{eff} on the burnup time without and with the fission product effects.