

**Survey of codes relevant to design, engineering and simulation of
transmutation of actinide by spallation.
(The cost estimation of accelerator for incinerator and the problem
of radiation hazard)**

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Abstract

The summary of the survey of codes relevant to design of transmutation of actinide by **spallation** is presented. The accelerator actinide incinerator has been studied by using the codes of nuclear cascade calculation including a high energy fission. The validity of the codes has been examined by comparing the experimental data of the yields of neutrons, and of **spallation** and fission products by a interaction of medium energy proton and thin and thick uranium targets. The cost of accelerator for incineration was estimated using the data obtained from studies of accelerator breeder and accelerator tritium producer. The problems of radiation hazard which are shielding, beam loss, and the other radioactivity, are discussed.

1. Introduction

Studies have been made of the possibility of incinerating actinides in light-water reactors and in liquid metal fast breeders without processing the long-lived **nuclei**[Cl,82]¹. However, thermal neutrons and fast neutrons, whose spectrum is not sufficiently hard, do not incinerate actinide efficiently. To do so, a fast neutron spectrum is needed that is hard enough so that the fission reaction dominates the capture reaction.

This requirement might be met by using Np-237 elements that give a harder neutron spectrum in the target assembly than the spectrum from conventional LMFBR[MT,88]. However, the lifetime of neutrons in this hard spectrum is very short and the transient behavior of the reactor increases rapidly as it **approaches** the supercritical condition. Furthermore, the delayed neutron fractions of Np-237[Tm,89] fission are smaller than those of the uranium isotopes, which somewhat restricts their control.

To keep a minor actinide fuel reactor running, the choice of the composite materials of fuel, cladding structure, and coolant is limited: this limitation makes the safety problems associated with criticality much more severe.

The accelerator actinide incinerator can resolve the difficulties associated with criticality in reactor safety because it is a subcritical reactor, **assisted** by neutrons created by high-energy proton **spallation** and the high-energy fission reaction.

Studies of the incineration of actinide **nuclei**[BR,87], [Ta,85] that have very long **half-lives**, such as Np-237, suggest that the power from a small beam, in the order of **15-30MW**, can incinerate the actinide produced by about ten lGWe light water reactors. Furthermore, an incinerator with **900MW** thermal power can produce **270-240MWe** of excess **electricity**, as well as **100kg** of fissile materials, such as U or Pu, when its core is surrounded with fertile material such as Th or U, respectively.

¹ Only references with underline are cited in the reference section because of space limitation.

The concept of accelerator incinerator is new, and has not yet been studied extensively as much as the accelerator breeder which has been promoted in many laboratories. Because of the similarity between the accelerator breeder and the incinerator, many materials were taken from the studies of the accelerator breeder.

This report is a summary of my voluminous report prepared for OECD, for more detailed information please refer to the forthcoming report (Ta, 91).

2. Accelerator Actinide Incinerator

In the process of transmuting actinides, large numbers of fast neutrons generated by **spallation** processes incinerate minor actinides by fission. In quantitative terms, the total number of fissions, N_{fiss} , can be expressed by:

$$N_{fiss} = N_h + S_h \frac{k}{(1-k) \nu} \quad (1)$$

where N_{fiss} = total number of fissions, N_h = total number of fissions by high-energy proton reactions, S_h = number of neutrons produced by high-energy proton reactions (**spallation**, evaporation, and very high energy fission), ν = number of neutrons per 'regular' fission and k = multiplication factor for 'regular' fission neutrons. By increasing the k value, the proton current required to incinerate the minor actinides can be reduced. The k value of the incinerator should be considered from many aspects, such as safety, operational procedure, choice of materials, and the cost of the incinerator. Since these parameters have not been studied, the value of $k = 0.9 - 0.95$ has been arbitrarily chosen. When Np-237 captures a neutron it converts the fertile material to **fissile** material and the reactivity of the target increases during the operation of incineration.

2.1 Study at EURATOM, CERN, and BNL [BR, 87]

About 250kg of minor actinides are produced annually from 10 units of 3000MW(Th) LWR without uranium **isotopes**; and the thermal power generated by incinerating this amount of actinides is 900 MW. When this 900MW thermal power is generated with a specific power of 150W/gr HA, the total amount of actinide in the reactor becomes 6 tons.

The target lattice system we studied was made up of actinide-oxide fuel pellets clad by steel can, and cooled by sodium or helium. The V-shaped geometry target was surrounded by a thorium blanket to capture the large fraction of leakage neutrons that produce the **fissile** material of U-233; the target lattice was irradiated directly by a 1-GeV proton beam, which is spread by a magnetic field. Table 3 shows the results of these studies.

Table 3. Requirements for accelerator-driven sodium - or helium-cooled incinerators.

Coolant	Keff	Beam Power (MW)	Beam Current		Reactor Power (MWth)	u-233 produced in blanket (Kg)
			1GeV	3GeV		
Na	.90	27.9	27.9	9.3	900	85
He	.95	13.0	13.0	4.3	900	103

The requirement for a relatively low beam current favors the comparatively inexpensive "Multistage Cyclotron Technique".

2.2 JAERI Incinerator Study, [TK, 89]

An accelerator-driven TRU target system designed at JAERI was operated at a subcritical condition of $k = 0.99-0.95$. The target core was 2m long in the direction of the beam, and 1.0m in height and 0.85m in width. This core was surrounded by a 0.2mm thick stainless steel reflector. The beam window was located at a depth of 0.7m from the front face. Heat was removed by forced circulation of a liquid metal coolant of Na or Pb-Bi. The metallic alloy fuels of Np-Pu-Zr and Am-Cm-pu-Y give a much harder neutron spectrum than oxide fuel. These alloys are expected to have a sufficiently high phase stability, and with the addition of 20 wt% of Zr, the melting point of Np is expected to increase from 640°C to about 900°C. For Na cooling, the maximum thermal power is 390MW and the beam current required is 18.1mA. For Pb-Bi cooling, the maximum thermal power is 163MW, requiring a 5.4mA beam current.

2.3 BNL Large Scale Incinerator Study, [VT, 90]

In order to use the present technology, the incinerator target assembly was designed in very conservative way. Initially, a sodium-cooled oxide fuel lattice based on the FFTF reactor was chosen as the target. We chose to use the large linear accelerator, with a peak current of 250mA of 1.6GeV protons, to derive a subcritical lattice of $K \approx 0.9$.

After two years of operation at 75% capacity factor, the fuel reached an average burn-up of 8.6%, with an additional 12.7% converted to plutonium. The use of a two-year cycle assures that more than 80% of the plutonium is PU-238.

The combined inventory of neptunium and americium decreased by about 2.6 tonnes/year, with 1.55 tonnes/yr being converted to plutonium. Thus, one unit of the incinerator would transmute the minor actinide wastes from about 75 1000 MWe LWRs.

2.4 LANL Thermal Neutron Incinerator Study, [Bo, 90]

Another accelerator incinerator of actinide, as well as of fission products such as Cs-137 and Sr-90, was studied by LANL [Bo, 90]. Instead of using fast neutrons, the LANL incinerator creates a high-intensity thermal neutron flux, similar to that in our earlier study of a fission product incinerator using spallation neutrons [TM, 80]. However, the geometry of the target system is different and the high energy proton is injected vertically in the eutectic flowing target. The irradiating material is immersed into the D₂O moderator which surrounds the eutectic flow target. Using thermal neutrons of $4.8 \times 10^{15} \text{ n/cm}^2/\text{sec}$ maximum flux produced by a beam energy of 1.6GeV and proton current of 25mA, the minor actinides and fission products would be incinerated. The incineration of the actinide will occur by fission of these resulting nuclei. The processes of capture and the fission that follows are rather complicated; their assessment requires detailed calculations using burn-up codes. They estimated that the spallation neutron produced from a 400 Mw proton beam accelerator ($8.6 \times 10^{22} \text{ neutron/year}$) could destroy the minor actinide output of 84 PWRs!

3. Nuclear Cascade Calculation(Medium Energy Nucleon-Nucleus interaction)

When medium energy protons collide with the nucleus, a nuclear reaction occurs by a two-step process of spallation and evaporation of the residual nucleus. When the residual nucleus has a large mass and moderately high excitation energy then it might undergo fission in competition with evaporation. The third process is the emission [Br, 71] of a cluster and a particle before reaching the thermal equilibrium state, the so-called the pre-equilibrium emission of the particle.

When a medium-energy proton collides with the nucleus, the transport of the nucleon inside the nucleus can be treated as a classical particle, because the wavelength of the nucleon is smaller than the nucleon's average distance. A collision of a nucleon with a nucleon is treated as a two-body collision. The π mesons which are created in such a collision are also included in the calculation of the cascade process.

The residual energy after **spallation thermalises** the nucleus, and neutrons, protons, or other light nuclei are evaporated. When this energy surpasses the fission barrier in the heavy nucleus, fission competes with the evaporation particles of light elements.

The particles emitted from the nuclear collision travel until encountering the next nuclear collision (called an inter-nuclear cascade) ; then, a similar process as that described is repeated until the energy of the particle falls below the cut-off energy. When the particle emitted or scattered from the nucleus is a charged particle, its energy is lost by exciting the electron surrounding its tracking path; this process is called the inter-nuclear cascade process. The cascade process inside the nucleus is called the **intra-nuclear cascade**. The information provided by the **intra-nuclear cascade** calculation consists of the energy and direction of each emitted neutron, proton, d, t, and n-meson , as well as the excitation energy, recoil kinetic energy, charge, and mass of the excited residual nucleus.

Neutron and photon transport below the cut-off energy has been accurately calculated in conventional reactor calculations which are very familiar to nuclear engineers; therefore, these subjects are not discussed here.

4. Procedure to Calculate the Nuclear Cascade Reaction

4.1 NMTC and HETC codes

The NMTC and HETC codes are most commonly used to calculate the intra-nuclear cascade.

To calculate the nucleon's cascade inside the nucleus, a model is required for nuclear matter, which is described here as the degenerated nucleons in the Wood-Saxon type potential.

The collision of a nucleon with a nucleon inside the nucleus is treated as a two-body collision, which satisfies the law of relativistic energy momentum conservation. After the collision, when the energy of the particle is less than the Fermi energy, this collision event is discarded and another collision is performed. When the particle's energy after a collision becomes less than the cut-off energy, then the history is terminated.

When the kinetic energy of the nucleon scattered through the nuclear surface is above the binding energy of nucleon, the nucleon escapes from the nucleus, its kinetic energy being decreased by the value of its binding energy.

When the nucleon's kinetic energy inside the nucleus is less than the binding energy , the nucleon gives kinetic energy to the nucleus as excitation energy. This energy thermalizes the residual nucleus, and neutrons, protons, or other light nuclei are evaporated. When this energy surpasses the fission barrier in the heavy nucleus, fission events will be compete with the evaporation of light element particles.

The cascade of nucleon in the nucleus is calculated by the code MECC2, developed by **Bertini**; the evaporation process from the excited nucleus is calculated by **EVAP** developed by **Dresner**. The transport of particles in the heterogeneous medium is calculated by many subroutines developed in the 05R codes. Furthermore, many subroutines can be added to calculate the transport of charged particles and the nuclear reaction associated with pions. The **EVAP** subroutine was improved by **Guthrie**, and the evaporation reaction is presently treated by subroutine called by DRES; the MECC2 in the NMTC code is named the BERT subroutine. In the HETC code, which can treat a higher energy reaction than the NMTC code, the MECC7 subroutine is used.

In the commonly used nuclear cascade codes **NMTC[CA, 70, 71]** and **HETC[CA, 72]**, the data for the nucleon -nucleon collision is obtained from **Bertini's** evaluated **data[Be, 63]**, and the production of the meson is treated by using the Isobar model developed by Sternheimer and **Lindenbaum[SL, 58, 611]**.

The original NMTC and HETC codes have no capability for calculating the high energy fission which is very important for nuclear targets of high atomic number, such as the nucleus of uranium and of **actinides**.

By adding their own fission models to the NMTC code, the **NMTC/JAERI** and **NMTC/BNLF** codes have been generated. The LAHET code [Pr, 89] was made by adding the RAL (Rutherford Appleton Laboratory) and ORNL models to the HETC code.

In both the NMTC and HETC codes, the nucleon number density is calculated using **Hofslader's** formula, and the momenta of the **nucleons** are distributed as the degenerated Fermi momentum distribution with absolute zero temperature. The nuclear potential is described by assuming that the nucleus is composed of three segmented regions. The nucleon-nucleon cross section to calculate the two-body collision inside the nucleus are the semi-empirical fit of the data by Hess et al.

- A) Nucleon Distribution skipped.
- B) Momentum Distribution skipped.
- C) Distribution of Potential Energy skipped.
- D) Nucleon-Nucleon Cross Section skipped.
- E) Meson Production Model

For particle-particle reactions that lead to the production of n-meson, the (E1) Steinheimer-Lindenbaum isobar model account only for single- and **double-pion** production in nucleon-nucleon collisions and **single-pion** production in **pion-nucleon** collisions. The practical thresholds for ternary-pion production by **nucleo double-pion** production by pions are about 3.5GeV and 2.5GeV, respectively. Thus the energy ranges of incident proton and pion are limited to these energies.

F) To treat the higher energy reactions, the extrapolation model of Garthier is provided, as discussed in HETC. To estimate the particle production for higher energy ($E_p > 3\text{GeV}$) collisions, this extrapolation employs particle-production data obtained from an **intranuclear-cascade** calculation for intermediate-energy, (3GeV), nucleon-nucleus and **pion-nucleus** collisions, together with the energy, angle, and multiplicity scaling relations that are consistent with the sparse experimental data available for high-energy collisions.

G) Charged-Particle Energy Loss such as protons, charged pions, and muons due to the excitation and ionization of atomic electrons is treated by the well-established stopping power formula, based on continuous slowing-down approximation.

H) Multiple Coulomb Scattering of primary charged particles is treated by Fermi's joint distribution function for angular and lateral spread, and Rutherford's formula for a single-scattering cross-section.

4.2 ISABEL (Vegas) code

A code Vegas, similar to Monte Carlo Nuclear cascade code was developed by K. Chen et al. [CFF,68,CFM,68] at BNL. This code takes into account the refractive process which is neglected in the NMTC and HETC codes. However, the original Vegas code did not take into account meson production, as does the NMTC and HETC codes, because nucleon energy is limited to the rather low energy of 380MeV. Therefore, Vegas was extended to calculate meson production, by incorporating the isobar model and model for the nucleus-nucleus collision by Y. **Yarif** and Z. **Fraenkel** [YF, 79,81]. To calculate the collision between antiproton and nucleus the model by M.R.Clover et al. [CD,82] was used.

The code named ISABEL, which was developed from the VEGAS code, is used in the LAHET code [Pr.89] together with the HETC code, to calculate the nuclear cascade calculation at LANL.

4.3 The Cascade Calculation for Light Mass Nucleus

The original NMTC code does not taken into account the nuclei whose mass is between 2 and 7. This omission is due to the lack of accuracy in the model describing the nuclear reaction. In the NMTC/BNLF and **NMTC/JAERI** codes, this

limitation is simply relaxed to encompass a mass range between 2-5, so that Li-6 and Li-7 then can be handled. Deuteron is treated as two protons in the NMTC/JAERI code, because the binding energy of the deuteron is only 2.2Mev.

4.4 Fermi's Break-up Model

In the LAHET Code, Fermi's break-up mode 1, which was developed by T.S. Suberamanian et al. [Su,83], has replaced the evaporation model for disintegration of light nuclei; these replacement models apply only to residual nuclei with $A \leq 17$.

4.5 The Pre-equilibrium Model

It is appropriate to consider that after **spallation**, the residual nucleus is not in a state of equilibrium and emits particles. This **pre-** equilibrium model has been studied by M.Blann[Bl,71], A.V. Ignatyuk et al. [IS,75] E.D.Arthur[AR,88], R.E. Prael and M. Bozoian [PB,88], and many other authors [KA,85], [NN,86]. In those theories, the pre-equilibrium states are described by the exciton state, which is the many particle and hole state of the single particle state.

In the LAHET code, the multi-stage pre-equilibrium model (MPM) is used for the emission of neutrons, protons, deuterons, He-3, and α particles at each stage of exciton states. The MPM terminates upon reaching the equilibrium exciton number; then the evaporation model (or Fermi's break-up model) is applied to the residual nucleus with remaining excitation energy.

4.6 The Fragmentation of the Nucleus

When the incident proton energy is increased, the nucleus may fragment. This process produces heavier nuclei with mass number $A=20-40$, even though this probability is much smaller than the probability of emitting the nucleon, deuteron, tritium, He-3, and α particle. It has been conjectured that **multi-**fragmentation is the manifestation of a liquid-gas phase transition occurring during compression - expansion of nuclear matter. [Ai, 84]. Other models or theories are based on either a statistical and chemical equilibrium picture, or a fast break-up process in which only minimal statistical assumptions are made[Mo,85]. However the actual process of fragmentation is so complicated that none of the theories has succeeded in offering a convincing explanation. [cd,84]

4.7 Neutron spectrum and yields of spallation and fission products

The neutron spectrum in the **spallation** reaction, using a rather thin block target, has a small bump in the region of 20-80Mev. To explain this small rise, several models such as a multi-temperature model and a moving source model have been proposed. Some improvements are seen, but the model cannot completely explain the rise in the neutron spectrum.

The **spallation** products including fission products and evaporation products have been extensively studied by Nishida and Nakahara[NN,86], using Yamada's mass formula.

5. High Energy Fission Model

5.1 General

To evaluate the yields of neutrons and reaction products in a heavy-mass target irradiated by a high energy proton beam, it is very important to take high energy fission into consideration. Several models of high energy fission have been proposed and incorporated into the high energy nucleon meson transport code HETC by Atchison[AT,79,80] and Alsmiller et al.[Al,81] and into NMTC by Takahashi[Ta,84b] and Nakahara[Na,80]. Baklashenkov et al. [Ba,78] independently developed a high-energy fission model.

Theoretically, all of these models are based on the statistical theory of fission [Fo,69], but their computational schemes differ in practice, in physical assumptions, and in the data used to calculate fission probability, mass and

charge distributions of fission fragments, excitation, and kinetic energies of residual nuclei.

When there is a possibility of fission, it can be considered to occur in competition with evaporation. The fission process itself can be treated as a two-step process. At the moment of fission, a nucleus splits into two fragments, from which particles subsequently do or do not evaporate, according to excited levels. For branching ratio calculation, since there is possibility of emitting p, d, t, he-3 and a particle, these emission other than fission and neutron are so small that they can be neglected.

5.2 Branching Ratio of Fission to Neutron Emission

A) In the RAL (Atchinson) model, the branching ratio of fission to neutron emission is calculated using the systematic of Vandenbosh & Huizenga [VH, 58] for $Z \leq 90$ nuclei. It is assumed that no fission occurs for excitation energy $E' < 6$ MeV. For the sub-actinide region of $Z < 90$, the other statistical model is used, which fits the experimental data.

B) The ORNL model (Alsmiller) adopted Hahn and Bertini's fission model. [HB, 72], which is essentially same as the simple model developed by Sikkeland, Ghiorso, and Nurmia [SG, 68]. In this ORNL model, the fission of nuclei whose z number is less than 90 is neglected.

C) BNL and JAERI models

In the JAERI and BNL models, the branching ratio is calculated using the fission barrier heights evaluated by Il'inov et al. [IC, 80] based on the liquid drop model of Meyers-Swiatecki [MS, 66, 67] and Nix [NI, 69]. The JAERI model takes into consideration the possibility of fission only for nuclei with masses greater than 175. The level density parameter, a_n , is calculated from LeCouteur's expression:

$$a_n = \frac{A}{B_0} \left[1 + 1.5 \left(\frac{A-2Z}{A} \right)^2 \right] \quad (c.2)$$

where B_0 is a universal constant, and $B_0 = 8$ is used in NMTC and NMTC/JAERI. (Atchison [At, 79], and Alsmiller et al. [Al, 81] use different values for B_0). B_0 seems to range from about 8 to 20 MeV, but the best value has not been evaluated.

The level density parameter for a fissioning nucleus, a_f , was fitted to the experimental data compiled by Vandenbosh and Huizenga [VH, 58]. The neutron binding energy, B_n , is obtained in the same way as in the subroutine DRESS of NMTC.

5.3 The Mass and Charge Distribution after Scission

If it is decided that fission will occur, the masses, charges, and parameters such as kinetic and excitation energies have to be selected for the fission fragments. These parameters also are determined using the statistical model, in which it is assumed that the fission process is so slow that an instantaneous equilibrium state will be established every moment of the process.

A) In the RAL and JAERI models, these distributions are determined by statistical theory based on the fluctuation probability with Gaussian distribution and on the experimental data.

In the RAL model, the mass distribution after scission is determined for the actinide region by taking into account the two competing modes, asymmetric division (dominant at low excitations) and symmetric division (which takes over at high excitations). For nuclei in the sub-actinide region, the mass split is always assumed to be a symmetric, i.e., about $A/2$ [NF, 63]

The charge distribution function for one fragment (the heavy one in the case of asymmetry) is assumed to have a gaussian distribution with a two-charge unit for the width. The charge of the other fragment is determined from conservation of the number of protons.

The total recoil kinetic energy correlates well with the Coulomb repulsion parameter $Z^2/A^{1/3}$, and this RAL model takes the correlation of E from E.K. Hyde [HY,64] The excitation energy of the fragment is computed by assuming a uniform distribution of both excitation and binding energy in the fissioning nucleus plus conservation of energy.

In the JAERI model, Pik-Pichak and Strutinskii's model [PS,] is used to determine the mass and charge distributions of the fission fragments. The total kinetic energy, E_k , of the fission fragments is determined by the Coulomb repulsion at the moment of splitting. The recoil energies of fragments are determined by assuming that they are proportional to the masses of the fissioned fragments.

B) In the BNL [Ta,84] and ORNL [Al,81] models, the Fong's statistical model [Fo,69] is adopted instead of the empirical formula used in the RAL and JAERI models. According to the Fong's statistical theory, the probability of producing fragments of fission products (A_1, Z_1) and (A_2, Z_2) is expressed by the function of the following quantities; C (the mutual Coulomb energy of a fission pair at the moment just before scission), k (the total translational energy of the same), D (the total deformation of two fission fragments), and E (the total energy available to the compound system C minus k), with a given partition of excitation energy E_1, E_2 and with given angular momenta, J_1 and J_2 , for the two fragments, where it is assumed that $j=0$ and there is no orbital angular momentum.

Incorporation of this most general distribution function of fission product into the calculation to the intra-nuclear cascade code is very time consuming. Therefore in the BNL model, a simplified distribution function is derived for a few variables by carrying out the summation of j , and the integration of k and excitation energy, E.

In the ORNL model [Al,81], the statistical functions at the scission and evaporation times are derived according to Fong's theory, but more reliance is placed on the empirically derived function than on the above models.

5.4 Photo-Fission

In recent years, there has been growing interest in electromagnetic interactions of heavy nuclei at intermediate energies. Such information provides data on the nature of the nuclear force, and the mechanism of intranuclear cascade. A broad program of studies is being carried out on nuclear photo-fission and photo-fragmentation using back-scattered laser photons. At present, studies of photo-fission of both u-238 and Np-237 have been made by D.I. Ivanova et al. [11,89].

6. Accelerator Reactor Code System

To analyze the accelerator breeder and the high-intensity neutron source facility, code systems were developed combining the nuclear cascade code and the conventional neutron and photon transport code used for the nuclear reactor at BNL [BR,87], RAL [At,79], JAERI [NT,89], and LANL [Pr,89].

A. BNL code system [BR,87]

Fig.A.1 shows the code system developed at BNL for analyzing the accelerator breeder and accelerator tritium production. The BNL computer code system has six main programs: NMTC/BNLF, HIST3D, EPR, (DLC-2), TAPEMAKER, ANISN, TWOTRAN-II, and three auxiliary programs: FIND, SURF, MULTISUM. The nuclear cascade is calculated by the nucleon meson transport code (NMTC) with the BNL fission model; neutron transport is treated either by the two-dimensional Sn code, TWOTRAN II, or the three-dimensional Monte Carlo code, MORSE-CG. The neutron source distribution to be used in the TWOTRAN calculations is prepared by HIST3D, which was designed to analyse the collision events file created by NMTC/BNLF. FIND and SURF were prepared to provide graphic representations of the output from the TWOTRAN-II processing programs.

B. RAL Code System, [At,79]

Fig.B.1 shows the code system which was developed at RAL. This code is based around the HETC code, and contains two major codes: HET which, for RAL's

particular case, transports, the incident protons and the produced neutrons, pions, and muons. The second part is 05R which takes the neutrons from 77MeV to 0.14MeV. The low-energy neutrons which escape are passed onto the TIMOC code, a time based neutron transport code, which was used in the moderator calculation by A.Taylor. Energy deposition is very important because the target has to be cooled, in particular, with uranium.

The most important quantity in the RAL project is the first source of low energy neutrons, useable for the moderator calculations. Also, it receives a contribution to energy deposition and some more contributions to nuclide distribution. The Y-transport code is calculated by MORSE code.

C. The JAERI CODE System[NT,89], [NT, 82]

Figs. C.1 shows the Accelerator code system (ACCEL) code system developed at JAERI. This system originally was developed from the BNL code system, so that there is lot of similarity between them. The nuclear cascade is calculated by NMTC/JAERI, which uses Nakahara's fission model. In the NMTC/JAERI, the calculation of the neutron transport uses the ENDF/B-4 nuclear data file library. The neutron sources used for 1D-transport ANISN and 2D-transport TWOTRAN calculation are, respectively, prepared by using HIST3D/A and HIST3D/B processing codes of the neutron files created by NMTC/JAERI. This system also uses the Sn transport code TWOTRAN and the Monte Carlo code, MORSE-DD (the JAERI version of MORSE-GC) to calculate neutron transport below the cut-off energy 15MeV.

D. The LANL Code System (LAHET) [Pr, 89]

Los Alamos National Laboratory developed a code system based on the LANL version of the HETC Monte Carlo code by adding many new features. Fig.D.1 shows the LAHET code system [Pr, 89]. The geometric transport capability in LAHET is that of LANL's continuous energy neutron-photon Monte Carlo code MCNP. LAHET includes two models for fission induced by high-energy interactions: the ORNL and RAL models. The alternative intranuclear cascade model in LAHET was adopted from the ISABEL code, which allows hydrogen and helium ions and antiprotons as projectiles. The ISABEL intranuclear cascade model itself is derived from the VEGAS intranuclear cascade code. The HMCNP computation may be executed as a coupled neutron-photon problem; however, to obtain a photon source from the high energy interactions computed by HETC, it is necessary to execute the PHT code.

6.2 Burn-up codes

The code system described above do not include a burn-up calculation for the target lattice. The burn-up of actinide can be calculated with the conventional burn up code used in the nuclear reactor if the change in the neutron spectrum due to spallation neutrons[LH,69] is taken into account.

6.3 Monte carlo codes used often in the accelerator reactor calculation below cut-off energy (15 or 20 MeV) Skipped - see complete report

6.4 Sn transport codes Skipped - see complete report

6.6 Computer codes for electron cascade shower

We do not discuss the electron accelerator in this report. The following codes are useful for the shielding calculation of the electron accelerator.

A. The EGS4 code written by W.R.Nelson, H. Hirayama, and D.W.Rogers at SLAC, will calculate various parameters of the electron photon in the range of 10TeV down to few ten keV by using the Monte Carlo methods. The output of this code include particle flux distributions, and energy deposition. The advantage of this program is the QED process that is very well understood and for which there are many numerical examples.

B. The ITS (Integrated Tiger Series) code was written by J.A. Halbleib and T.A. Melhorn; it is kept at RSIC and Sandia National Laboratories. This code calculates time-independent coupled electron/photon radiation transport from 1GeV down to 1KeV including or omitting macroscopic electric or magnetic fields that

is calculated by the Monte Carlo method. Slab, spherical, cylindrical, or combinatorial geometries can be handled.

7. Analysis of the Experimental Data (code verification and its limitation)

Only few integral experiments have been carried out with non-fissile and fissile materials. To evaluate the accelerator breeder concept, Fraser's group made measurements in collaboration with ORNL. Similar experiments were repeated by LANL's and Fraser's groups, Vasilkov's group measured the production rate of Pu using large, block-size uranium.

7.1 Microscopic Analysis

A. Alsmiller's Analysis[Al,81] of the Fission Reaction Because of the spread of the experimental data of fission cross section, it is difficult to judge whether the calculated results agree with the experimental data; however, in the 100MeV to 1GeV energy range, they are in approximate agreement. The calculated value for the neutron yields are not so sensitive to the values of level density parameter of B, but this is not the case at high energies.

B. Armstrong and Fliege's analysis[AR,83] of fission cross section indicated that the values calculated using the RAL model are about 15-20 % lower than those for the ORNL model with beam energies below 1GeV, and the energy dependence of the cross section above 1GeV differs. The spread of the experimental data is too large to judge whether either model is correct.

C. The Neutron Spectrum in the Spallation Reaction

In addition to the data discussed in section 3.1, Prael[PB,88] analyzed the LANL experimental data for 113MeV and 256MeV proton on stopping-length targets and thick target using the MCM model described the above. He examined several options of combining the Bertini's and ISABEL INC (Vegas) codes with MPM and without MPM, the Fermi break-up model. He concluded that the motivation for introducing the MPM has been to improve the agreement with experiments at back angles.

D. Spallation products

In addition to the one discussed in section 4.7, the spallation products, including fission products and evaporation products, have been studied by Russel et al.[Ru,80]; good agreement between theory and experiments was found. Such work is important in evaluating the radiation level and radiation hazard to maintain the accelerator operational.

7.2 Integral Experiment

A. Chalk River TRIUMF Experiment (FERION Experiment) [Fr,80]

To obtain data for the high intensity neutron source, Fraser's group performed several experiments using a small block of uranium surrounded by a water bath in 480-MeV proton beam of the TRIUMF facility. The analysis indicated that the calculations with level density parameter of $B_0=8\text{MeV}$ give reasonable agreement with the experimental values.

B. Chalk River, ORNL Experiment[Fr,75]

In collaboration with ORNL, Fraser's group performed similar experiments long before the experiment described above (in A) using 540, 720, 970, and 1470 MeV energy proton beams taken from a 3GeV cosmotron at BNL. In a series of experiments they used different sizes and material of Be, Pb, and U. Despite the small size of the target these data are often referred to as the basic data for the accelerator breeder conceptual design. The target geometry is qualitatively similar to that described in experiment C below. Alsmiller and Takahashi have analysed these experiments on uranium target. All of Alsmiller's calculated values are smaller than Takahashi's calculated values and close to the experimental values supplied by Garvey, except in the case of 1470MeV. The values calculated by Takahashi using $B_0 = 10.\text{MeV}$ are close to the original experimental values. Neglecting the high-energy fission of nuclei ($Z \leq 90$) in Alsmiller's

calculation results in a smaller neutron yield. For such nuclei without high energy fission our calculation shows a reduction of 12% in neutron capture.

C. LANL experiment [Ru, 80], [Ru, 81]

Experiments similar to those of Fraser (A&B), performed by Russel's group for cluster type fuel rod, were analyzed with the LAHET code system. Calculation using the ORNL and RAL Models (without high energy fission) showed that either version could predict radiative capture in uranium or thorium to within 7% of the measured values. However, the models gave lower number of fissions by factors of - 1.5 for uranium and -3 for thorium.

Of these two targets, thorium provided the more stringent test of the theoretical models because over 60% of the fissions were at energies >20 MeV.

The LANL group could not determine the values of the level density parameter B_0 , (see sec.5.2) from the experimental data on fission product yields, the number of fissions, or the measured spallation products for either target. However, they concluded that Armstrong's suggested value of $B_0 = 8 \text{ MeV}$ for proton energies <1GeV, in the RAL model substantiates their general finding.

D. Vasil'kov et al.'s experiment (the Large uranium block experiment) [Va, 78]

So far, only one experiment has been performed by Vasil'kov et al using a target assembled from rectangular block of natural ($2 \times 4 \times 8 \text{ cm}^3$) and depleted ($8 \times 8 \times 16 \text{ cm}^3$) uranium. The total linear dimension of the target was $56 \times 56 \times 64 \text{ cm}$ and it was covered with a layer of lead 0 or 20cm thick. The experiment was carried out with an extracted beam of 300, 400, 500 and 660MeV protons.

The density distribution of (n, γ) capture was measured by Np-239, distinguished radio-chemically from uranium samples irradiated at various points in the target.

This experiment was analyzed by Takahashi [Ta, 84], Nakahara et al. [NT, 79], Garvey [Ga, 79], and Barashenkov et al. [Ba, 78]:

E. Calculation for an Infinite u-238 block was performed by Barashenkov [Ba, 78], Alsmiller [Al, 81] and Takahashi [Ta, 84].

The values calculated by Varashenkov, Nakahara, and Takahashi of neutron capture by u-238 for 600 MeV incident proton energy have a reasonable agreement with the experimental value but the calculated values for 400MeV are smaller than the experimental value except for Varashenkov's calculation. The values calculated by neglecting the high energy fission are 23%-38% lower than the values with the fission.

Takahashi's values are close to those of Barashenkov; Alsmiller et al's value is 15% less than those of Takahashi and Barashenkov.

Recently, Vasil'kov [Va, 90] compared the values reported by several authors for neutrons captured including the data discussed above. He found a large disagreement among them. Some of the results were obtained by extrapolating the calculated value for a finite uranium block. The calculated values for the yield of Pu-239 are scattered around two experimental values of Dubna and ORNL-CRNL at BNL; still the uncertainty is about $\pm 15\%$. The calculated values for the fission number/proton are about 30% smaller than those from Dubna's experiment.

To overcome the lack of the experimental data, Vasil'kov is constructing an experimental facility with a cylindrical target of depleted metal uranium, having a total mass of 21 metric ton, and a beamline for transporting protons or deuterons with momenta 1.4- 3.4GeV/c.

8. Cost analysis of accelerator for incinerator

The target reactor used in the accelerator incinerator is rather similar to the conventional fast reactor, thus data on the cost of fast reactor can be used for estimating the target cost. Since the fuel processing cost is not

well established yet, we will not **try** to estimate the cost of incineration using this accelerator incinerator: we will limit ourselves to estimating the cost of the accelerator used for the incineration of minor **actinides**.

8.1 Meson factory Accelerator and other accelerators planed[Ku,89]

The existing accelerators used in the facilities of TRIUMP, PSI, **LAMPF**, and INR Moscow Meson Factory are the closest to the accelerator which might be used for incinerator.

The cyclotron of SPI in Switzerland has been reconstructed to increase the beam intensity to **1.5mA**, and future plans are to increase the intensity to **10mA**. This is rather close to the segmented cyclotron discussed in the incinerator section. The 800MeV proton synchrotrons is a rapid-cycling, strong-focusing machine designed to provide an average 200 **microamps** at a repetition frequency of **50 Hz**.

8.2 High power linear accelerator for accelerator breeder[Ko,77] and incinerator

There are several estimate of cost for the accelerator breeder, which uses the high power **linac** of **300-400MW** proton beam power. In the **linac** studied for accelerator breeder at CRNL, the ion source is placed at high energy of 75Kv, and, after injection, protons are accelerated by radio frequency quadruple (RFQ) . The assembled protons are accelerated by drift tube **linac (DTL)**, that is called the Alvarez **linac**, up to 200MeV. Protons are further accelerated by coupled cavity **linac (CCL)** above 200MeV to reach a final energy of **1GeV**.

The accelerators studied by LANL and BNL are very similar to the CRNL accelerator. Recently, a Russian group designed a **linac** for an incinerator that also is similar to these except that both H⁻ and H⁺ beams are accelerated in the initial part (1P) , first, and second parts.

8.3 Cost analysis of the Linac

Table 3.1 shows the breakdown of the capital cost estimated by LANL ,BNL, and Chalk River. In addition to capital costs, we have to take into account the running costs of the accelerator hardware. The lifetime of the PEP klystron of 500KW CW with a frequency of 353MHZ and 65% efficiency is 20000 **hr** (experimental value). Thus, the cost of this klystron is roughly \$100 K, (in 1980 \$), and the consumption cost of the Klystron is 1 ¢ /RF Kwh. When the beam loading factor is 5.5 and the (ratio of RF generator to beam loading)**x** (control range **ratio**)=**.85**, then the total power of RF generator is 420MW, and the consumption cost for the Klystron for 420Mw is \$4.2K /hr=M\$32.8/year. If we take the same life time of 20000 **hr** for the accelerator's structure, this consumption cost calculated by BNL, is **M\$ 32.8 /year**.

Linac	Chalk River		LANL		BNL	
Current	300.	15.	300.	15.	300.	15.
Energy (GeV)	1.0		1.0		1.0	
Year M\$	1981		1979		1979	
Accelerator						
Structure	95.	47.5	54.	30.	75.	37.5
RF	261.	13.	160.	8.	350.	17.5
Total	356.	60.	214.	38.	425.	55.

Table 3.1 The cost estimations of **CRNL, LANL, and BNL linacs**.

In the cost estimated by LANL, BNL, and Chalk River, the capital cost of the accelerator's structure is 20-25 % of the total capital cost of the accelerator. In the case of Accelerator tritium producer (**ATP**) [**AT,90**] the accelerator structure is about half of the total cost. This cost can be reduced by lowering the proton energy, but a low energy proton gives a small neutron **yield**. Because the cost of the accelerator structure can not be greatly reduced, even when the beam current is small, in the order of 15-30mA, it has been suggested that the segmented cyclotron accelerator is used

instead of the linac. The accelerator structure of the linear accelerator is rather expensive, therefore when we use the linear accelerator to incinerate minor actinides, the high power accelerator is more economical. A 300MW beam power can incinerate all the minor actinides produced in a 200 LWR when the multiplication factor of the target is $k=0.95$. However, the beam must be segmented to irradiate a reasonable sized target, and it is not expected that there will be a large inventory of minor actinides in order of 120 tons in the near future.

8.4 Cost Analysis of the Segmented Cyclotron

The cost of the accelerator structure can be reduced substantially by using a cyclotron type accelerator. Recently, the economics of a 1.5 GeV and 10 mA proton cyclotron was studied by Odera [Od, 90].

This cyclotron was designed and its cost was estimated conservatively; Table 4.1 shows the cost of each components in 1990 10⁹¥.

	Ion Source	RFQ	Cycro-1	Cycro-2	Cycro-3	total
Source	2.					2.
Magnet			4	40.	60.	104.
accel.Cav.		1.5	5	18	36	60.5
RF(incl.DC)		2.0	6	18	36	62.
Vacuum.	2.	1.0	2.	4.	8.	17.
others	1.	(Dev.) 1.5	1.	3.	5.	11.5
sum	5.	6.	18.	83.	145.	257.

The costs of other components (again, in 1990 10⁸) are:

Beam transport system (Between accelerator. achromatic system between accelerator and target system)	25.
Diagnostic of beam and Safety system (Non	15.
Control and Operation system (Including . remote control maintenance apparatus)	30.
Cooling system (Ion, removal apparatus)	15.
	sum
	85.
Total sum	342

Table 4.1 Cost of segmented cyclotron (1.5GeV, 10mA)

8.5 Comparison Between Linac and Segmented Cyclotron

To make an approximate comparison between the cost of the linac and the cyclotron with small beam power, the following assumptions are made. The cost of the RF generator is proportional to the beam power and the cost of the accelerator structure is proportional to α power ($\alpha = 0.2$) of the beam currents. The estimated costs of the 15mA, 1GeV proton linac, calculated at various laboratories using the above assumptions, are shown in the table 5.1. The cost estimated from the LANL data is low compared the costs calculated from the data of BNL and Chalk River. The cost of the accelerator structure for 1 GeV and 15 mA is 40-50 M\$ (1980), and cost of a RF generator is about 13-18 M\$; that is, the cost of the accelerator structure is about three times that of a RF generator for a 15mA accelerator. This ratio can be reduced by increasing the beam currents.

In the table 5.1, the cost of a cyclotron is compared with the cost of the small beam power linac (1GeV, 15-30mA), which is calculated from the data from the ATP [AT, 90] accelerator, using the same assumptions as above. Because of the high cost of the accelerator structure in ATP, the cost of the accelerator structure is more than 10 times of the cost of RF generator for a 15mA beam current.

The cost in US M\$ (conversion ratio of 150 ¥ to 1 \$) of the segmented cyclotron is shown in column (a). The cost of accelerator structure includes the cost of the magnet which is almost twice that of the accelerator structure. Column (b) shows the cost calculated by normalizing the cost of RF generator to that of the ATP (15mA accelerator).

8.6 Cost of small accelerator for incineration

When we use the cost data of the **ATP**, a substantial part of the cost of the accelerator incinerator comes from the accelerator portion, even for a small beam power. However, this incinerator produces a large excess of electric power, and also of **fi ssile** material, **Pu**, u-233, or **tritium**, as by-products, When the high-power accelerator is used for incineration, this system earns more money by selling the excess electricity and **fi ssile** material: this excess power corresponds to 114 **M\$**, and 100 kg of the **fi ssile** material earns on the order of 5M \$. If we use the accelerator power of a 300 MW beam, we can incinerate 20 times more actinides than in the previous case when we use a target with $k=0.95$; the earnings from selling the electric power become 2.3B \$/year, and the earnings from the production of **fi ssile** material become 100 **M\$/year**. These amounts far exceed the costs of the accelerator and target.

	Accelerator of ATP			Cyclotron	
Current	15	30	250	10	
Energy (GeV)	1.0	1.0	1.6	1.5	
YearM\$		1989		1989	
Accelerator				(a)	(b)
Structure	320	368.	991.	165	73.45
RF	27.6	55.2	738.8	62.	27.6
Total	348.	423.3	1729.7	227.	101.

Table 5.1 The cost Estimation of ATP linac and Segmented Cyclotron

9. Problem of Radiation Hazard[IA,88], [Mc,83]

The problem of radiation hazard associated to the accelerator facility is somewhat different from that in conventional nuclear reactor plants. The target size may be similar to the nuclear reactor; however, the shielding problem in the direction of the high energy proton beam is unique.

9.1 Proton Energy vs Shielding

skipped - see complete report

9.2 Shielding

9.2.1 Shielding at proton energies energy less than 3 GeV

The principal concern in shielding proton accelerators of less than about 3GeV is the neutrons produced by the high energy proton. Most of the published experimental and theoretical data in this energy range concerns neutrons whose energy is less than 400MeV. It is difficult to treat theoretically the energy region between 400MeV and 3GeV because the hadron cascade process has not then stabilized. Experimental data are **scarce**, so we have to resort to interpolation from data between 400 MeV and the ' high energy limit' achieved at proton energies of several GeV.

At proton energies above 3GeV, the longitudinal shielding may be dominated by simple models. At these higher energies simplification is possible because the attenuation length of high-energy neutrons is independent of neutron energy above 100MeV, and the yield of high energy neutrons is roughly proportional to the primary proton energy E_p .

* Particle yields from the proton-nucleus interaction

Tesch[Te,85] reviewed the published information on the total number of neutrons produced per proton interacting in various target materials (C, Al, Cu, Fe, Sn, Ta and Pb) over the energy range from 10MeV to 1.45GeV.

* Transport of the mono-energetic neutron through shielding

Below 400MeV , neutron transport in the shielding can be treated by the

standard method of solving the Boltzman transport equation, especially by the method of spherical harmonics, the method of discrete ordinates, and the Monte Carlo method. Studies by O'Brien and Alsmiller et al. showed that these methods give essentially equivalent results [OB,70, Al,69a].

Discrete ordinate calculations of the penetration of neutrons in broad beam geometry through concrete in the energy range from 50 to 400MeV were reported by Alsmiller et al [Al, 69b], and from 1 to 100MeV by Wyckoff and Chilton [WY73]. There is agreement between these two sets; also both sets agree with the calculation of O'Brien, using the spherical harmonic method. [OB,70]

Paterson described some early shielding studies for 90 MeV neutrons [Pa,57], suggesting that the attenuation length λ was approximately given by the well-known relation [Pa,73].

$$\lambda = 1 / N \sigma_{inel}$$

where σ_{inel} is the inelastic cross-section of the shield material. Theoretical studies suggested that, at least for high energies, the effective attenuation length λ_{eff} would be somewhat greater than λ_{inel} . The experience of Sychev et al. [Sy 66a,b] at Dubna suggested that for broad beam geometry in the energy range between 350 MeV and 660 MeV the attenuation length was given by : $\lambda_{att} = (1.3 \pm 0.1) \lambda_{inel}$.

9.2.2 Shielding of proton accelerators at energies greater than 3GeV

At proton energies about 3GeV, calculation of the hadronic cascade is important to determine the shield thickness of the proton accelerator; above 10GeV, the production of muons must be taken into account for specifying shielding in the forward direction. Fairly detailed reviews of the early shielding studies in the GeV energy region are given in the references of [Li,61, Pa ,71, Pa,73, Ri,73].

9.3 Skyshine

Skyshine (air scattered) neutrons commonly contribute significantly to the radiation dose in uncontrolled areas. Measurements have verified that mathematical models used to calculate doses of neutron skyshine are in good agreement up to about 200ft. However, at distance of half a mile or more, the various models may disagree by at least an order of magnitude.

A summary of the skyshine phenomenon around the accelerators was discussed by Rindi and Thomas, who reviewed experiences up to 1973 [Ri,73].

9.4 Total radioactivity

The total quantities of radioactivity produced in an accelerator structure may be related to the total number of inelastic interactions produced by a proton in the materials of interest.

A simple, approximate relationship between the total saturated activity (A_{sat}) and the value of inelastic interactions per second, N, is expressed as:

$$A_{sat} = k N \quad (6.1)$$

where k is a constant to be determined.

9.5 Radioactivity in earth and water

The radionuclides that can be produced by hadron-induced spallation interactions in the oxygen of the cooling water are summarised by Christensen et al. [Ch 78].

9.6 Beam loss problem

The beam loss problem was discussed by D. Young [Yo.79] taking the example of the 300 ma in the Fermi lab 200 Mev linac.

He concluded "I maintain that beam loss problems are serious concern in a high intensity, high-energy **linac**, but that it should be possible to limit beam loss so that "hands on" maintenance and repair of accelerator components can be performed".

Jameson [Ja.90] also studied the beam loss problem in the **LAMPF** and **TPA** accelerators. By analyzing the experimental data of **LAMPF** accelerator, he concluded that the hands-on maintenance can be retained by lowering the fractional loss /m in the case of **ATp**. A linear accelerator is considered to be radiation free if the induced **γ -activity** does not **28 μ Gy/hour**. The corresponding level of beam losses amounts to:

$$Wq = 1 \text{ GeV nA / m} \quad (8.1)$$

Under this condition and with specific acceleration of 1 MeV/m in the second part of accelerator (i.e. 0.1-1.5 **GeV**), the total permissible beam current loses amount to **3 μ A**. With the beam current of the **300mA** the permissible relative losses are about 10^{-5} . A recent Russian study concluded that radiation-free accelerators can be achieved by the methods of beam phase volume filtering, suppression of coherent longitudinal and transverse oscillations, contact-less beam parameter measurement, beam diagnostics through the beam loss measurement, and residual gas limitation in the H beam channel.

9.7 Other Radiation Sources

There can be other radiation sources such as klystrons, experimental devices in other buildings, or **RF** tests. Such sources can be much harder to control because the health physicist may not know they exist.

10. Conclusion and Recommendation

At present, almost no nuclear data are available for minor actinide, so to design the accelerator actinide incinerator, the theoretical model used for studying the accelerator breeder with the u-238 target was used. However, the nuclear data for u-238 are also scarce and considerable uncertainties exist in the wide spread of experimental data for neutron yield and a fission cross-section as discussed above. It is highly recommended to make measurement of the neutron yield and neutron spectrum measurement for both uranium or plutonium thick and thin target because of the scarcity of the minor actinide at present time, and to make highly reliable theoretical model to simulate medium energy proton reaction for actinide materials. For the shielding problem of the target, the bump observed in the neutron spectrum should be studied to make a more predictable theoretical model.

At present, the neutron yield caused by injecting the medium energy of proton into minor actinide can be estimated in the error range of $\pm 20\%$. Thus we can make an approximate evaluation of the concept of an accelerator minor actinide incinerator. To make a more detailed evaluation of this concept it is required to find a more reliable theoretical **model**. As discussed in the section of the cost analysis, the linear accelerator is more economical for high power accelerator of **300-400MW**. The high power accelerator has too much excessive power to incinerate the actinide with the target which is near critical. By using the beam of H⁺ instead of proton, the beam can be easily segmented into many small beams before injecting the incinerator targets by using the foil or gas target. Thus this high power accelerator can economically run many subcritical actinide targets.

Recently a metal fuel fast reactor has been studied extensively at ANL. This has many interesting feature such as small reactivity change from initial phase to final phase in one burn up cycle. Because of this small reactivity change, the sub-criticality of the target can be maintained close to near criticality, and it makes a proton beam current small and can alleviate a radiation damage problem associated with medium energy proton. Furthermore it makes the power distribution flat and can reduce the power peaking factor. By providing a external neutron created by small intensity proton beam to the

subcritical fast reactor, it can be operated more safely and makes more flexible choice of structural and fuel materials to get higher breeding gain.

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