The Utilization of Accelerators in Subcritical Systems for Energy Generation and Nuclear Waste Transmutation - the World Status and a Proposal of a National R&D Program

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A summary of the world status on R&D related to the utilization of Accelerator Driven System (ADS) for energy generation and mainly for transmutation of long lived nuclear waste is presented. A proposal to start a Technical Working Group (TWG) in Brazil to prepare a Road Map having as a final goal an experimental facility to utilize an accelerator in basic and applied research; products and services and R&D in energy generation and transmutation are presented.

I Introduction

The present time commercial thermal reactors (LWR; CANDU) operate in once-through fuel cycles, and are producing a large stockpile of radioactive waste, mainly long lived transuranics elements (TRU) (Pu, minor actinidies (MA) such as Am, Np, Cm, etc.), and long-lived fission fragments (LLFF), such as Tc-99 and I-129, which will require final underground geological repositories for thousands of years.

The build up of radioactive stock piles, besides the concern of waste disposal, also brings the issue of proliferation, mainly due to Pu isotopes. To overcome these issues, the next generations of nuclear reactors are considering concepts which, coupled with a closed fuel cycle, aim at reducing the burden on geological storage and non-proliferation. The idea of partition and transmutation (separation of TRU, and LLFF from the spent fuel and incineration in a dedicated reactor) is fostering new international initiatives to develop these new concepts, such the USA Generation IV forum, and the IAEA INPRO.

Transmutation is more favorable in fast neutron systems, taking into account that the fission cross section for the TRU elements is higher than the capture cross section, and thus in a fast environment these elements will be transmuted by fission into short and medium-lived fission fragments. Therefore, fast reactors and hybrid systems in which ultra fast neutrons coming from a spallation reaction induced by protons from accelerators are coupled with a subcritical core (Th, U, TRU), the so-called accelerator driven system (ADS), are being considering as a dedicated waste burner reactors, preferably in a double strata fuel cycle. Besides, an ADS operates in a subcritical mode and can more easily address safety issues associated with criticality (although there is an still open research field in the dynamics and kinetics of ADSs). They also offer substantial flexibility in overall operation. ADSs can provide more excess of neutrons compared with critical systems, may be utilized for breeding purposes, and having a positive energy gain (net energy/energy to operate the accelerator), serve also as energy generators.

All these innovative features have motivated international R&D programs in several fields and several ADS concepts. This work intends to review briefly the technical issues associated with ADS, as well as the world status regarding the main programs under development. Finally, a proposal to start in Brazil a R&D program related to of accelerator applications, including ADS is presented.

II Conceptual bases and historical background

The basic idea of ADS is to bombard a subcritical fuel mixture (U, Th, Pu, TRU), with ultra fast neutrons coming from a spallation source induced by an accelerator (Linac or Cyclotron). Spallation is a well-known nuclear reaction in which energetic particles (e.g. protons) interact with the atomic nucleus. Given the energy of the incident particles they interact in a first stage with the individual nucleons instead of the formation of a compound nucleus as in low energy nuclear reactions. The initial collision leads to the ejection of nucleons and pions, which have enough energy to start a cascade reaction (inter-nuclear cascade). After this point, the nucleus is left in an excited state and goes to a ground state by evaporation of nucleons. Fragmentation (fission) of the nucleus may occur, as well as (n,xn) reactions in secondary stages. In short, the spallation reaction pro-

duces high energy secondary particles (neutrons, protons, mesons, gammas). These particles, besides depositing a large amount of energy and generating spallation products in the target, are the external source (mostly of neutrons) in the sub critical media when they leave the target.

The most commonly used targets are Pb and Bi. Although the theory of proton spallation (e.g. Cascade Excited Model, CEM) provides a good description, and is incorporated in codes like LAHET [1] and FLUKA [2], as well as included in Nuclear Data Libraries, such as EXFOR, NUCLEX, to enhance the quality of such data is still an open field of research, both experimental and theoretical. Table 1 illustrates the neutron yield for spallation processes induced by high-energy protons calculated by LAHET.

Table 1. Neutron Yield for Spallation process induced by high-energy protons calculated by LAHET.

Proton Energy (MeV)	Multiplicity n/p	Integrated Yield (n/sec.m A)
100	0.321	2.00E15
150	0.835	5.21E15
200	1.627	1.02E16
250	2.664	1.66E16
300	3.883	2.42E16
350	5.272	3.29E16
400	6.784	4.23E16
1000	28.76	1.79E16

The utilization of accelerators in fission media is not new. The first man-made Plutonium was produced by Glen Seaborg using a cyclotron, in 1941. During the fifties, at Lawrence Livermore and in Canada, investigations on the use of accelerators to produce fissionable material were performed, and spallation yield measurements were made. A material production accelerator - The Electro-nuclear Reactor - was patented in 1960 by Lawrence to provide adequate quantities of artificial materials (TRU). At Chalk River the Intense Neutron Generator concept was proposed. The Fertile-to-Fissile Conversion Program (FERFICON) during the 70s and 80s involving several national laboratories investigated the energy dependence of the fertile to fissile conversion using accelerators.

The concept of ADS where safety and transmutation were considered was developed in the late eighties in the Brookhaven National Laboratory. However, the first conceptual molten-salt thermal energy amplifier and transmutation system was proposed in Los Alamos in 1991 (C. Browman) where the terminology by which ADS are known in the USA was introduced: Accelerator Transmutation of Waste (ATW). In 1995 a group from CERN led by Carlo Rubbia [3] presented the basic concept of the Fast Energy Amplifier, a sub critical nuclear system based on the U-Th fuel cycle, imbibed in melted lead (coolant), with an external source of neutrons coming from spallation in Pb induced by protons from a 3-stage cyclotron (1 GeV, 12.5 A), which actually raised the international interest on Hybrid Systems. Later on, several experiments were conducted at CERN and several reports were published with some variants from Rubbia's idea. Figure 1 illustrates the basic concept of the Rubbia Fast Energy Amplifier.

III World status

Besides the Rubbia's concept, several R&D initiatives related to ADS, partition and transmutation (P&T), etc., were developed in the world. Here we wish to briefly describe these initiatives and programs, mainly those discussed in the IAEA Technical Working Group meetings, in which one of the authors (José Rubens Maiorino) is a Brazilian representative [4]. The world status is going to be presented by countries, or international organizations:

Belgium: The most important project conducted at SCK is the MYRRHA project, which started in 1996 as a conceptual study aiming at development of a multipurpose neutron source based on ADS. The accelerator is being developed by Ion Beam Applications (IBA), and consists of a 25mA proton source, an accelerator of 25-30 MeV (injector), and a proton cyclotron (250 MeV, 2-10 mA). The facility will be used in material's research, radioisotope production (Mo-99), proton therapy, and studies of spallation-induced transmutation of long-lived nuclides. The total energy will not exceed 30 MWth, and the fast flux 1.5×10^{15} n/cm^{2}s. The spallation source will be a windowless liquid lead-bismuth target surrounded by a fast and a thermal subcritical units. The MYRRHA system is a prototype research facility and could be a replacement for the present time research reactors. This development is in the ADS European Road Map.

Belarus: R&D on physics of ADS are carried out in STC Sosny along the following main lines: evaluation of nuclear data for nuclides of the Th-U and U-Pu fuel cycle; nuclear model theory and code development for high energy file evaluation; development of calculation methods for ADS and experimental investigation of neutronics of sub-critical systems driven by accelerators (YALINA, a subcritical UO2 facility driven by a 14 MeV neutron generator). The YALINA facility is providing data for IAEA CRP benchmarking data on ADS.

Brazil: At present only ADS calculation methods and conceptual proposals are underway at IPEN. The proposal of a program is present in this work involving the nuclear physics and engineering communities, similarly to the Belgian program.

China: Presently a multipurpose system consisting of a low energy accelerator and a sub-critical facility pool is underway. A conceptual experimental facility has been proposed for R&D. Calculation methods and theoretical models (Cascade Evaporation Model) are being studied at several Chinese Institutions (CIAE, IHEP, PKU-IHIP). Also accelerator technology development is being conducted by China

aiming at a high energy proton accelerator.

Chech Republic: The Chech Republic started a national R&D program on ADS. A project LA-0 is proposed for testing a modular sub-critical assemblies with fluoride salts on the experimental reactor LR-0.

France: The R&D activities in France are conducted by CEA. A program called ISAAC has been set-up to investigate the physics of ADS. The main activities include the MUSE experiment at MASURCA in Cadarache, which is a lead sub-critical facility driven by neutrons (14 MeV) from an accelerator and will provide benchmark data for the ADS European Road Map. Another activity is spallation experiments (SATURN). France is participating in several European experiments. Recently, a research group GEDEON, made up of CEA, EDF and Framatome has been set up to intensify and co-ordinate research in these areas. An experimental fast system called HADRON has been proposed for experimental validation and demonstration of an ADS. The concept is based on a sub critical core with thermal power of 50-100 MW. France is involved in international cooperations on P&T studies (MA separation, pyro-reprocessing, etc.).

Germany: At the Technical University Munich the design of a separated-orbit cyclotron with superconducting channel RF cavities for a 1GeV proton beam of up to 10 MW beam power is under development (TRITON). At FZK-Karshrure a Lead-Bismuth Thermal Hydraulic Loop (KALLA) develop R&D in the framework of the European ADS road map. Several programs related to scenarios to prevent and minimize nuclear waste and to reduce the radio toxicity, such as P&T are underway, as part of the environmental German policy. Exploratory ADS investigation (calculation methods) are under way at FZJ Julich and FZK Karlshure.

India: Although there is no official program on ADS in India, some investigation on concepts using U-Th fuel cycle are underway.

Italy: ENEA and INFN set up a basic R&D program TASCO aiming at the study of Physics and technologies needed to design an ADS for transmutation, consisting of subprograms on proton accelerators, neutronics, thermal hydraulics, beam window technology and material technology and compatibility with Pb and Pb-Bi. Also ANSALDO is conducting an industrial program to design an European ADS prototype (XADS). Besides, ENEA is carrying out studies on Metal corrosion (Pb, Pb-Bi), using LECOR and CHEOPE facilities at Brasimone. The thermal hydraulic loop CIRCE, a large facility for metal technology is being used for full scale demonstration of the critical components of the spallation targets. A coupling of a TRIGA operating at sub-critical condition with an accelerator is planned to be conducted at ENEA Cassacia Center.

Japan: The project called OMEGA is underway to conduct long term R&D on Partition and Transmutation. The Program includes studies of the physical and chemical properties of MA and LLFF; partition of radioactive elements from high level liquid waste of reprocessing process, and ADS transmutation (nuclear and fuel properties data of MA, system design studies, reactor fuel and accelerator target development, development of a high power accelerator, 1.5 GeV - 5.3 mA).

Korea: KAERI has initiated a study on transmutation since 1992 and now is setting up a long term R&D program called HYPER to construct an ADS (1000 MWth, Pb-Bi coolant, 1 GeV TRU-Zr metallic fuel, Tc99 and I129 burner). A multipurpose linear proton accelerator called KOMAC is on planning.

Russian Federation: Several research institutes in Russia are involved in P&T and ADS programs directed by MINATON. Russia has a large experience and facilities using accelerators and liquid metals (Pb), mainly from its nuclear submarines program, which can be used in research related with ADS. Some experiments are being conducted at Dubna (SAD, Sub critical assembly in combination with proton accelerator), and Russia is participating in cooperation with Japan, USA and the Europeans in several R&D.

Spain: CIEMAT started in 1997 a research program to study the physics of ADS. Also a private enterprise, LAESA (Laboratorio del Amplificador de Energia), has been created in 1997 to built a laboratory for research, development and demonstration of the Rubbia's Energy Amplifier Concept. Also a study of CERN was performed to demonstrate the possibility to eliminate the need for a final repository in Spain by using a Fast Energy Amplifier as a waste transmutation (spent fuel from Spain PWR).

Sweden: Research on P&T is mainly supported by the Swedish and Waste Management Co. (SKB). The main activities occurs at the Royal Institute of Technology in Stockholm, where the physics, safety and concepts of ADS are studied (concept of a multisource, burn-poisoned nitride fuel ADS, Sing-Sing Concept). The Svedlberg Laboratory at Uppsala has recently started a project for cross section measurements of interest for ADS.

Switzerland: The activities of R&D are conducted at PSI, where they are investigating high-current cyclotron development, supporting development for spallation source, material technology development work for spallation targets, experimental validation of models in nucleon-meson transport codes by means of the ATHENA irradiation experiment using actinide targets, reactor physics and safety analysis of ADS, and comparison studies of different reactor fuel cycles with emphasis on the impact of alternative transmutation strategies on the long-term risk of the radioactive waste. PSI operates one of the largest cyclotron facilities (590 MeV, 1.6 mA) which is used for the spallation source SINQ. Also, together with CEA and FZK, it is conducting the MEGAwatt Pilot Experiment (MEGAPIE) with a view to demonstrate the feasibility of a liquid Pb-Bi spallation target at power level relevant to ADS and to gain experience in design, operation and disposal of such targets.

United States: The main activities in the USA are con-

ducted by LANL, which is developing a LINAC with a beam power of hundreds of MW under a program called APT (Accelerator Production of Tritium). The LANL ATW (as ADS is denominated in the USA) aims to reduce the amount and long-time hazard of the spent fuel from the US nuclear reactors. The proposed ATW would incinerate the TRU waste and transmute LLFF such as Tc99, and I129. The ATW system is a fast spectrum liquid lead-bismuth cooled device coupled to an APT Linear proton accelerator (1Gev, 100 mA). The fuel fabrication and the fuel cycle rely on pyro metallurgical processing. The ATW being proposed (8 target) will in theory be capable of incinerating 10,155 tons of spent fuel (169 ton/year during 60 years), and since the total projected US spent fuel inventory is around 86,000 tons, around 8 ATW could incinerate all high level US waste. Besides one ATW also generates 2110 MW (discounting the 380 MW needed to drive the accelerator). LANL is proposing an experimental program called LIFT for using 1 MW proton beam at LANSCE on a Pb-Bi loop. Recently the DOE presented to the US Congress a Road Map for Developing Accelerator Transmutation of Waste Technology, which merges ATW with APT program, in a program called AAA (Advanced Accelerator Application), identifying R&D and funds. In 2000, the US Government approved US$ 65 M for this program. Also in the USA, at Oak Ridge, a MW spallation source is being developed.

Besides National Programs, after the Rubbia's concept proposal, the European Community created a working coordinated group to establish a common base for design, and R&D needs, synergies, cooperation, follow up, etc. This group produced the European Road Map, which has as an objective the construction of a 100 MW ADS prototype in Europe in the next 10 years [5]. This program has been funded within the European Frame Work Program. The total budget estimated to build (including R&D) a 100 MW prototype ADS in 10 years, is 980 MEuro, and for fuel development an additional 180 MEuro, i.e., around 1 billion Euro. Also OECD-NEA are giving a great support to ADS, publishing reports, promoting a series of conferences, Workshops, and inter comparing calculation methods, nuclear data and benchmark activities. Information on OECD-NEA activities and references may be found at www.nea.fr/html/pt. The IAEA has promoted a series of Coordinated Research Programs (CRP), Technical Committee Meeting (TCM) and published a series of technical documents (TECDOC) related to P&T and ADS. A Technical Working Group (TWR-FR-ADS) provides a forum for scientific and technical information, international cooperation and advises the IAEA on the status and achievements on Fast Reactors, Accelerator Driven Systems, as well as associated advanced fuel cycles for transmutation and thorium utilization. Also an INIS-TWG-FR web site contains all documents, and information related with these topics, www.iaea.org/inis/fnss/index.html and for ADS www.iaea.org/inis/ws/fnss/ads.html. Since one of the authors (José Rubens Maiorino) is the Brazilian representative at this forum, it would help to conduct activities of R&D related with these topics.

IV Proposal of a R&D program, or a network for basic and applied nuclear physics and engineering

Brazil, although still a developing country, has an Industrial Nuclear Program, that, despite being small, has already achieved a good standard (2 NPP in operation, ANGRA I, II, generating 2%(2 GW) of the total installed electricity, a third unity under construction, a fuel cycle industry, INB, with capability to provide fuel elements for the NPPs. The Brazilian Navy develops a nuclear program for nuclear propulsion which includes all the front end of the fuel cycle including the enrichment by ultracentrifuge that is being transferred to the industrial sector (INB). R&D is conducted by Research Institutes of the Brazilian Nuclear Energy Commission (CNEN), IPEN, CDTN, and IEN, which have technical capability and facilities (Research Reactors and Accelerators) appropriate to support the industrial nuclear program, provide products and service for the Brazilian society (e.g. Radio Isotopes), as well as to develop new technologies. However, it needs some new challenges, mainly to motivate and to stimulate young engineers and scientists in Nuclear Energy R&D.

Brazilian science, being developed mainly at Universities, already produces a significant contribution to the human scientific knowledge and is forming thousands of Ph.D.'s. In spite of such scientific potential, although with exceptions (e.g. the GENOMA projects), it still needs to transfer to the Brazilian society the benefits of scientific knowledge. In particular, the physics developed by Brazil is traditional and outstanding even by first world standards, by the number of publications in international journals, scientific citations, number and quality of scientists. etc. However, Nuclear Physics needs, besides good brains, big machines such as accelerators, to be competitive with the physics being developed all over the world, although with international cooperation a good level of work is being produced.

This scenario calls for a program which identifies synergies, society needs, be in consonance with the international science and technology, have sustainability, be adequate to the Brazilian reality, and give an umbrella for the activities already underway in Nuclear Physics and Engineering, and preserve the identities and focus of the involved parts. Of course, to develop such a program relying only on ADS, is out of question, since the Brazilian society has another set of priorities in the short and medium time scales, and it could be done through international cooperation. On the other hand, to develop a R&D program in Nuclear Physics relying only on basic research and without any relation with the needs and expectation of the society is also out of ques-

tion and will not have government support, mainly in view of the new models and policies of financing adopted by the Science & Technology supporting foundations (CNPq, FINEP, FAPESP), based on the governmental policy of financing projects by social demands or economic and technology transfer for the private sector (Funds by Sectors, such as energy, oil, aerospace, health, etc). As a consequence, the nuclear community must find a new approach to continue its activities, attract new talents and, mainly, get governmental support. This is the main appeal to propose an integrated R&D Nuclear Program, preserving the individuality of the participants (synergies, spin off etc.). Taking into account the status of R&D on ADS, Partition and Transmutation presented in this work, and by the following points:

- IPEN has already initiated some conceptual studies on ADS, has nuclear codes and data libraries to design a subcritical fast facility. Also operates cyclotrons, one from IBA (30 MeV), dedicated to Radioisotopes Commercial Production, another CV-28 presently not in operation but which could be fixed and utilized for R&D. Besides IPEN operates two industrial electron accelerators and has a critical facility (which could be made sub-critical) and a 14 MeV Neutron Generator which could be used as an experimental hybrid system for R&D.

- The IFUSP operates and performs R&D with accelerators, such as the PELETRON, LINAC, and MICROTRON, and has a high quality staff in Theoretical Nuclear Physics.

- IEN has also a cyclotron, and CDTN has also Nuclear Calculation capability.

Certainly there are several universities, Research Institutes, like CBPF in Rio, which have international cooperations with CERN, and a good R&D on Applied and Theoretical Nuclear Physics. Of course they need to be identified.

In preliminary meetings between IPEN and IFUSP, both organizations hae converged to a program which employs accelerators in research (basic or applied), products and services (radioisotope production, irradiation for industrial applications), and in energy generation and transmutation (ADS). The long range idea is to define a proton accelerator (cyclotron or linac), with beams dedicated to these applications, and to have an experimental facility for accelerator applications. Of course, it would be out of the Brazilian reality to think of an extremely powerful machine, but following the Belgium example, consider an accelerator in the range of 300-350 MeV, a few mA (3-5), powers of 30-50 MW. The subcritical facility would be a thermal and fast one which would allow, besides R&D in ADS topics, would also be used to produce radio isotopes (Mo99) and other applications (e.g. cold neutron source). Notice that this proposal is in synergy with the fact that the only Brazilian Research Reactor, the IEA-R1 capable of some multipurpose applications, such as radioisotope production, is already almost fifty years old, and probably in the next 10-15 years will need to be decommissioned. Thus the subcritical accelerator driven facility would be the replacement of the research reactor. Moreover, a proton accelerator could be used to produce radioisotopes as well as cancer therapy by proton beams. A proton or heavy-ion bean from the accelerator would also be useful for Nuclear Physics research. Finally, the subcritical driven facility could be used to generate the energy to drive the accelerator, thus developing in the country a new energy R&D.

The strategy to define a R&D program as proposed would be to form a technical working group which, in the following activities, would identify:

- Accelerator technology: world and national status, and define the more suitable machine for the program.

- Subcritical facilities: world status, conceptual design and criteria, capability to construct, R&D needed, etc.

- R&D: present time R&D in development, competence, future potential R&D programs, synergies among the groups.

- Financial Support and logistic: cost of such facility, and strategies to obtain financial support as well as the logistic.

- Application (Products and Services): the present applications being done, and potential for the future .

This information should result in a Brazilian Road Map to a Program on Accelerator Applications, and should give to the Nuclear Physics and Engineering communities a common umbrella and a direction, which will allow to overcome the difficulties exposed before, get governmental support, and be in consonance with the international R&D programs.

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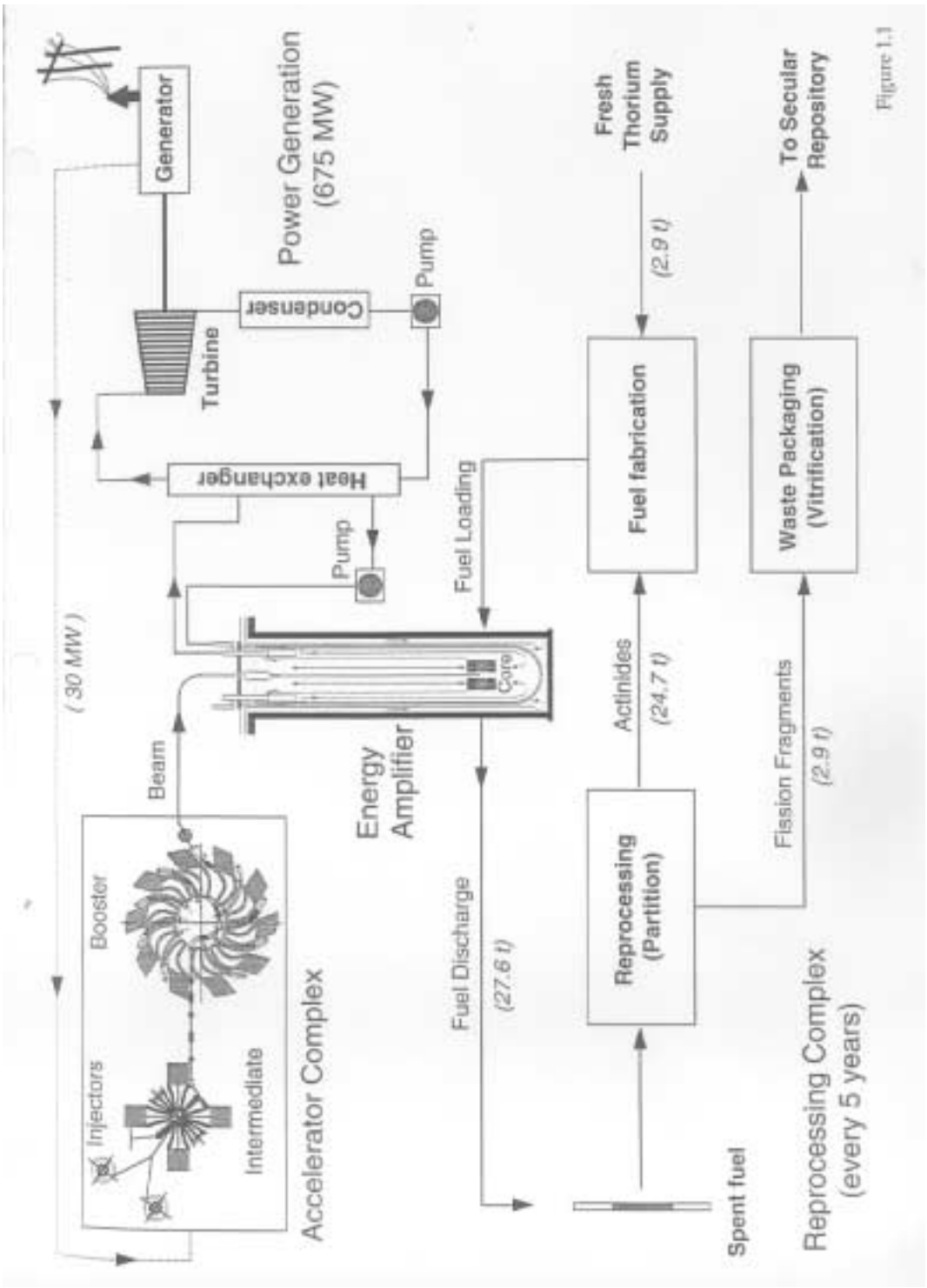

Figure 1. Rubbias Fast Energy Amplifier.