THORIUM UTILISATION IN GAS COOLED REACTORS

by

Barbaros ÇATKAN

B.S. in PHYSICS Middle East Tecnical University, 1982

Submitted to the Institute for Graduate Studies in Science and Engineering in partial fulfillment of the requirements for the degree of Master of Science in Nuclear Engineering

Boğaziçi University

1986

ACKNOWLEDGEMENTS

I would like to thank the Department of Nuclear Engineering for the understanding and supporting attitude shown to me.

My special thanks are directed to my Thesis Supervisor Prof. Dr. Turan B. Enginol and Doç. Dr. Vural Altın as it would not be possible to complete my study without their help.

I would like to mention Doç. Dr. Vural Altın for his invaluable support and patience that he showed during the completion of my study.

Mr Levent Akın and Mr Cemal Yardımcı have also supported me with their friendly attitude.

Finally, I would like to state my appreciation for Mr Demiray Hacıibrahimoğlu's help and understanding.

Barbaros Çatkan

ABSTRACT

Intensive effort for energy production through alternate paths is being spent in developed countries because of the limitedness of resources such as natural gas, petroleum, coal that make great contributions to energy production.

Nuclear energy can be noted as one of the most important amongst these alternatives. R & D work is being carried out for the purpose of reducing the reliance on the U-235 isotope and for more economical utilisation of known Uranium resources because of the limitedness of the isotope U-235 which plays an important role on today's reactor technology.

In this study, the Thorium fuel cycle which offers the possibility of reducing mined Uranium ore requirements is examined in connection with the High Temperature Gas cooled Reactor concept which supports the Thorium cycle. The Pebble-Bed core which employs spherical fuel elements is particularly interesting as it provides On-Load Refuelling.

As a prelude, the first chapter provides the basic information to introduce the concept to the reader. The properties of Thorium material are described in chapter 2. Several Thorium fuel cycles are discussed in chapter 3 and the technology associated with Thorium fuel along with HTGR designs is introduced in chapter 4. A possible strategy utilising Thorium is examined in chapter 5. The sixth chapter covers information about current HTGR models under active development.

ÖZET

Enerji eldesine önemli ölçüde katkısı olan doğal gaz, petrol, kömür gibi kaynakların sınırlı olmaları nedeniyle, gelişmiş ülkelerde alternatif yollardan enerji eldesi için yoğun bir çaba harcanmaktadır.

Bu seçenekler arasında en önemlilerden biri olarak nükleer enerji gösterilebilir. Günümüz reaktör teknolojisinde çok önemli bir yeri olan Uranyum-235 izotopu miktarının da sınırlı olması nedeniyle gerek eldeki kaynakların daha verimli olarak değerlendirilmesi, gerekse bu izotopa olan bağımlılığın azaltılması yolunda araştırmalar yapılmaktadır.

Bu çalışmada, U-235 izotopuna olan bağımlılığı azaltıcı nitelik taşıyan Toryum yakıt çevrimi ile bu kavramı destekleyen Yüksek Sıcaklıklı Gaz Soğutmalı Reaktörler ele alınmıştır. Bu reaktör tipi içinde özellikle Pebble-Bed olarak tanımlanan ve küresel yakıt elemanı kullanan model, On-Load refuelling olanağı sağlaması bakımından dikkat çekicidir.

Çalışmanın ilk bölümü, okuyucuya temel kavramları tanıtıcı bir giriş niteliğindedir. Bölüm 2'de Toryum materyalinin özellikleri tanıtılmış ve Türkiye'de önemli miktarda olan Toryum kaynağı üzerine yapılan bir çalışma sunulmuştur. 3. bölümde önerilen Toryum yakıt çevrimleri gözden geçirilmiş ve 4. bölümde bu alana özgü teknoloji tanıtılmıştır. 5. bölümde Uranyum cevherine olan gereksinmeyi azaltıcı bir tasarım olarak görülen bir Toryum yakıt çevrimi gözlenmiştir. 6. bölüm, Yüksek Sıcaklıklı Gaz Soğutmalı Reaktörler konusunda üzerinde çalışılan modelleri içermektedir.

TABLE OF CONTENTS

Page

LIST OF FIGURES

LIST OF TABLES

NOMENCLATURE

Δ	Mass Defect
Z	The number of Protons in a nucleus, the Atomic Number
N	The number of Neutrons in a nucleus
m_p	Mass of the Proton
m_n	Mass of the Neutron
m_A	Mass of the nucleus represented by A
D	Deuteron
T	Tritium
p	Proton
MeV	MegaelectronVolt
n	Neutron
A	Mass number
LWR	Light Water Reactor
PWR	Pressurised Water Reactor
BWR	Boiling Water Reactor
HWR	Heavy Water Reactor
OCR	Organic Cooled Reactor
LMCR	Liquid Metal Cooled Reactor
GCR	Gas Cooled Reactor
FBR	Fast Breeder Reactor
HTGR	High Temperature Gas Cooled
UOT	Uranium Once Through
TOT	Thorium Once Through
MOR	Mixed Oxide Recycling
RSE	Recycled in Separate Elements
SFB	Separate Feed and Breed
PB	Pre-Breeder
NB	Near-Breeder
LEU	Low Enrichment Uranium
HEU	High Enrichment Uranium
THTR	Thorium High Temperature Reactor
g	gramme

HM	Heavy Metal
N_C/N_{HM}	Moderation Ratio, Ratio of Carbon content to HM
η	Average number of neutrons emitted in fission per neutron absorbed in the fissile isotope
ν	Average number of neutrons released per fission
Σ_a	Macroscopic Absorption Cross-Section
σ_a	Microscopic Absorption Cross-Section
σ_f	Microscopic Fission Cross-Section
α	Capture-to-Fission Ratio
HTR	High Temperature Reactor
PCRV	Pre-Stressed Concrete Reactor Vessel
HDI	High Density Isotropic
PyC	PyroCarbon
WAR	Weak Acid Resin
BOP	Balance Of Plant
NSSS	Nuclear Steam Supply System
NI	Nuclear Island
GCRA	Gas Cooled Reactor Associates
AVR	Arbeitsgemeinschaft VersuchsReaktor
OTTO	Once-Through-Then-Out
GCFR	Gas Cooled Fast Reactor
HTGR-GT	High Temperature Gas Cooled Reactor with Gas Turbine
VHTR	Very High Temperature Reactor
LMFBR	Liquid Metal Fast Breeder
HTI	High Temperature Isotropic
LTI	Low Temperature Isotropic
FSV	Fort-St-Vrain
GWD	GigaWattDays
MWD/T	MegaWattDays/Tonne
$\lambda(x)$	Decay Constant of the species 'x'
N(x)	Concentration of the species 'x'

1. INTRODUCTION

1.1. Nuclear Energy as an alternative to world's energy supply paths

The main resources that have been extensively utilised in satisfying the energy needs are coal, oil, hydraulic, gas and nuclear reserves.

Nuclear Energy appears to be promising when the fact that other reserves are limitted is considered, as the energy demand in accordance with population and technological advances increases. The hydraulic potential is being almost completely utilised in developed countries and it bears particular importance for the developing countries in South Asia, South America and Africa. The known coal reserves are estimated to be sufficient until 2050, whereas the natural gas and oil until 2000 or 2010 only (1).

The world trend, due to familiar reasons, is in the direction of decreasing the contribution of oil to the total energy consumption in contrast with increasing the contribution of nuclear energy, while new energy sources are being developed. The contribution of nuclear energy in the year 2000 is forecast to be approximately 20%, whereas it was 6%, 9% and 12% in the years 1975, 1981 and 1983 respectively.

According to data gathered in 1985, a total of 345 nuclear react are in operation in 32 countries. The contribution of nuclear energy to the total electricity generation in some of these countries is shown in Table 1.1.

Table 1.1. Countries with the highest nuclear shares of total electricit produced during 1984. (2)

Country	Nucl. Shr. %	Country	Nucl. Shr. %	Country	Nucl. Shr. %
France	58.7	Bulgaria	28.6	UK	17.3
Belgium	50.8	Switzerland	36.5	Canada	11.6
Finnland	41.1	Japan	22.9	USA	13.5
Sweden	40.6	FRG	23.2	GDR	11.0

1.2. Basic Concepts

In order to get familiar with the subject and determine the terminology, a brief review of nuclear energy production and extraction processes is appropriate.

1.2.1. Binding Energy

A nucleus consists of protons and neutrons. The measured mass of a nucleus is, however less than the sum of the individual protons and neutrons that constitute the nucleus. This difference is called the Mass Defect and is given as

$$\Delta = Zm_p + Nm_n - m_A$$

By the Special Theory of Relativity which states the equivalence of mass and energy, the Mass Defect when expressed in energy units gives the amount of energy that must be supplied to break a nucleus into its constituent nucleons. Therefore, the Binding Energy is defined as the energy equivalent of the Mass Defect. The Binding Energy as a function of Mass Number A is plotted in Fig.1.1.

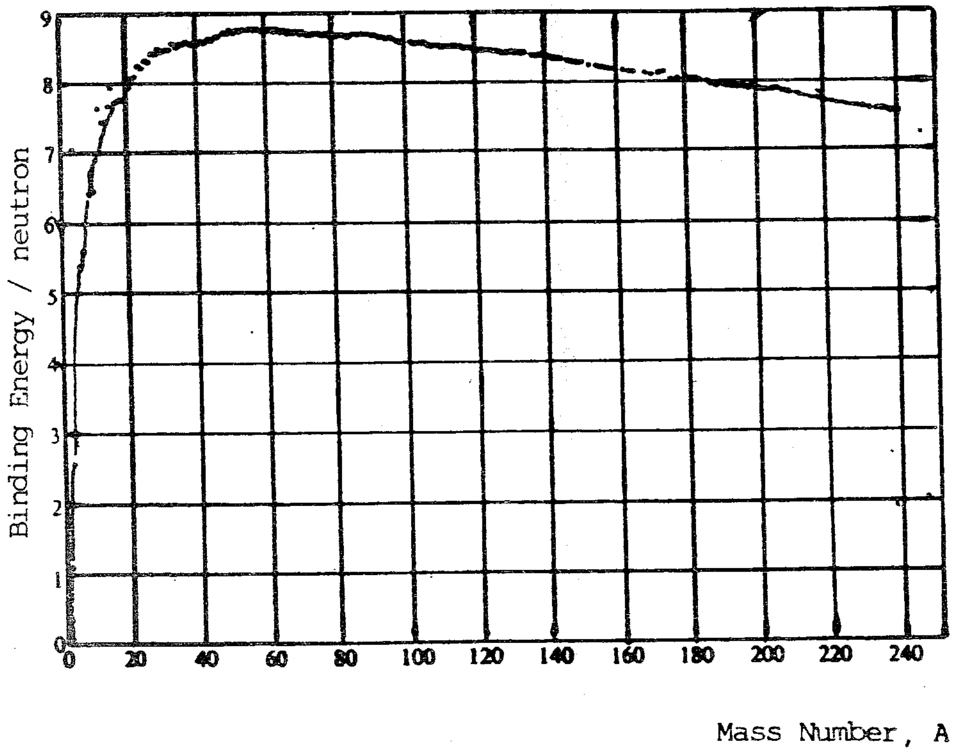


Fig.1.1 Binding Energy / neutron vs A

1.2.2. Fusion

Fusion process involves two very light nuclei combining to form a more stable nucleus increasing the Binding Energy per Nucleon as can be seen from Fig.1.1. The implication that the nuclei of very low A which will fuse to form a heavier nucleus are relatively less stable than the product can be explained through the Liquid Drop Model of nuclei in terms of the large surface to volume ratio in the light nuclei.

Because much of the earth is covered by seas containing the Hydrogen isotopes Deuterium and Tritium, ${}_1H^2$ and ${}_1H^3$ respectively, the fuel supply of low A nuclei seems to be almost inexhaustible. Several potentially useful fusion reactions are listed below :

$${}_1H^! + {}_1H^1 \longrightarrow {}_1H^2 + p + 4\ \text{MeV}$$

$${}_1H^1 + {}_1H^2 \longrightarrow He^4 + n + 17.6\ \text{MeV}$$

$${}_1H^! + {}_1H^1 \longrightarrow He^3 + n + 3.2\ \text{MeV}$$

$$He^3 + {}_1H^1 \longrightarrow He^4 + p + 18.3\ \text{MeV}$$

Although the fusion process is very promising, it seems not to be possible in the twentieth century to build a fusion reactor for commercial use. The problem lies in the repulsive Coulomb Barrier acting between two nuclei that must be overcome or at least penetrated to allow the short range nuclear forces to be active and fuse the two nuclei together. No material at present is available to confine the very high temperature nuclei so as to induce and sustain fusion. Attempts to use magnetic fields acting on the charged nuclei to contain them, the magnetic confinement, are being done, but still, obtaining thermal fusion for energy production requides further development. Another way to overcome the Coulomb Barrier is to load the incident nuclei with sufficiently high kinetic energy in cyclotrons but this method is still being developed

1.2.3. Fission

In Fig.1.1, in the region of high A, an exothermic nuclear reaction becomes one of breaking up large nuclei and this is called Fission. The heavy nucleus involved is split into two unequal nuclei by the absorption of a neutron. The mechanism is indicated in Fig.1.2

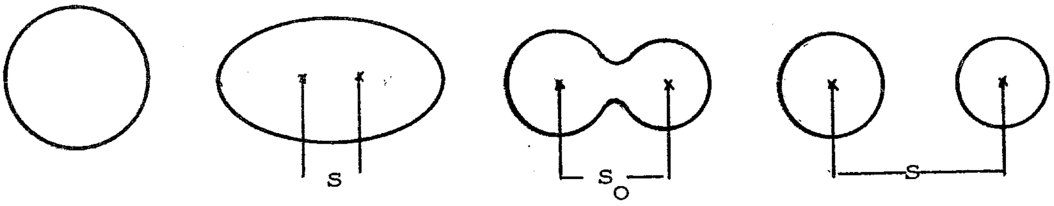


Fig.1.2 Interpretation of the fission mechanism

The surface area increases as s, which specifies the elongation of the fissioning nucleus, increases. The Liquid Drop Model implies that the increase in the surface area results in an increase in the surface energy. As s increases further, a surface tension effect produced by the surface energy causes the nucleus to assume a dumpbell shape. Once this state is achieved, the nucleus eventually splits into two. The resultant lighter nuclei are called the fission fragments and they move in the opposite directions in order for the momentum to be conserved.
The amount of energy that is required to put the nucleus into the dumpbell shape is called the Critical Energy. Fig.1.3 is a plot of the critical energy as a function of the separation parametre s.

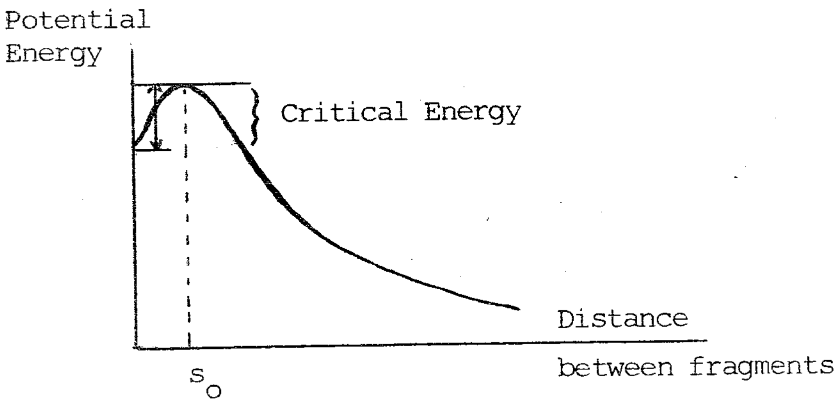


Fig.1.3. Critical Energy for fission

Table 1.2 presents the critical and binding energies of some important nuclei.

Table 1.2. Critical and Binding Energies of some nuclei.

Nucleus	Critical En. (MeV)	Neutron Binding En. (MeV)
Thorium-232	5.9	5.1
Uranium-238	5.9	4.8
Uranium-235	5.8	6.4
Uranium-233	5.5	6.7
Plutonium-239	5.5	6.4

For the species U-233, U-235 and Pu-239, the neutron binding energy is greater than their critical energy for fission. For this reason, these species can undergo fission by the capture of a neutron even with zero kinetic energy. These nuclei are called Fissile. Th-232 and U-238 can fission only if the incident neutron has sufficient energy. Because of this reason, these nuclei are called fissionable. However, they can be converted into fissile U-233 or Pu-239 by neutron absorption which may not induce fission. They are called, therefore, fertile nuclei as well.

The fission process can be described as

$$\begin{matrix} \text{U-233} \\ \text{U-235} \\ \text{Pu-239} \end{matrix} \quad + {}_0n^1 \longrightarrow \text{Fission products + Approx. 200 MeV}$$

The fission products cover the fission fragments that are the daughter nuclei formed by the splitting of the fissile nucleus. The splitting is almost asymmetric, ie, the fragments have unequal masses.

The fission fragments are excessively rich in the number of neutrons they contain. Therefore, they tend to decay into stable species through one or more Beta or Gamma emissions. A possible chain is the formation of Barium :

$$\text{Te-135} \xrightarrow{\beta^-} \text{I-135} \xrightarrow{\beta^-} \text{Xe-135} \xrightarrow{\beta^-} \text{Cs-135} \xrightarrow{\beta^-} \text{Ba-135} \quad \text{(stable)}$$

Prompt neutrons comprise another kind of fission product. This is important in the sense that the fission chain reaction is sustained by them. Prompt neutrons appear within 10^{-17} sec in a fission event with high kinetic energy.

Besides prompt neutrons, some neutrons are released following the Beta decay of certain fission fragments. These neutrons are called Delayed Neutrons and play an important role in reactor control and operation although they amount to less than one percent of all the neutrons emitted as a result of fission. The fragments which later cause the release of a delayed neutron are called Delayed Neutron Precursors. These precursors can be divided into six groups according to their half-life as displayed in Table 1.3.

Table 1.3 Delayed Neutron Precursors induced by thermal neutrons (3)

Precursor	Half-life (sec)	Group
Br-87	54.5	1
I-137	24.4	2
Br-83	16.3	
I-138	6.3	3
Br-89	4.4	
Rb-93, Rb-94	~6.0	
I-139	2.0	4
Cs, Sb or Te	1.6-2.4	
Br-90, Br-92	1.6	
Kr-93	~1.5	
I-140	0.5	5
Br, Rb, As	0.2	6

The emission mechanism of a Group-1 delayed neutron is presented in Fig.1.4.

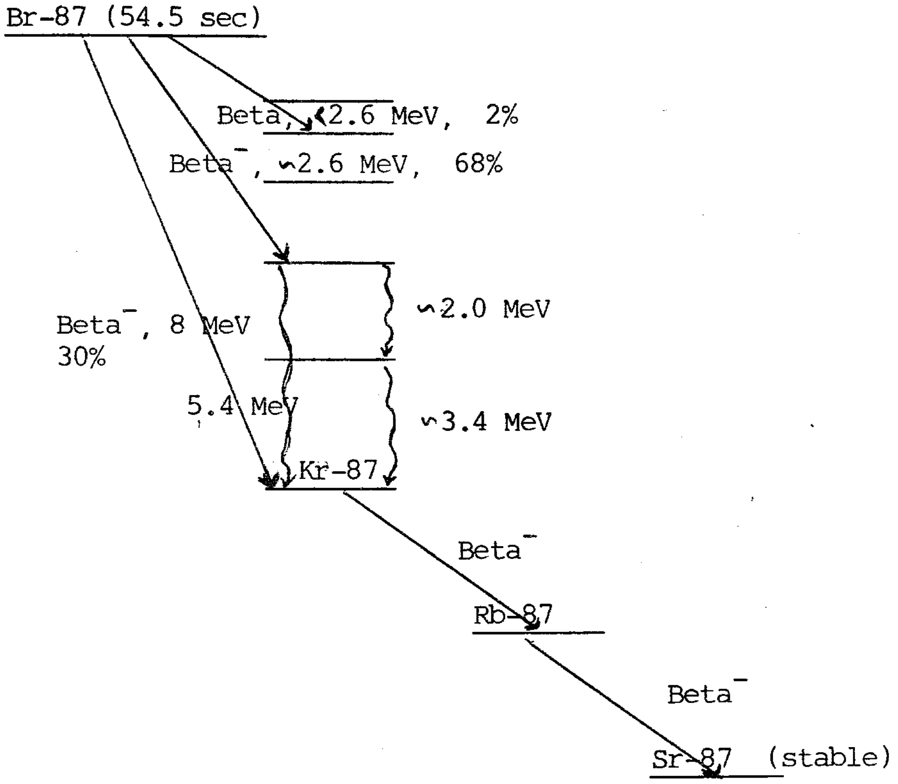


Fig.1.4. The emission mechanism of a Group-1 Delayed Neutron (3).

Similar to the emission of neutrons, the emission of Gamma-radiation is also either prompt, appearing immediately in the fission process or delayed, accompanying the decay of fission products.

The energy released in the fission process mostly appears as the kinetic energy of the fission fragments. The distribution of energy in the fissioning of U-235 is displayed in Table 1.4. By the recoverabil of the energy, it is meant that it remains within the core. Neutrinos escape from the core without any interaction, therefore, that amount of energy associated with the neutrinos is always lost being carried away with them, ie, is non-recoverable. But the other means of fission energy eventually shows up as heat in a fuel matrix and can be extracted

Table 1.4. Distribution of energy in the fissioning of U-235 (3).

Form	Emitted En. (MeV)	Recoverable En.(MeV)
Fission Fragments	168	168
Fission Product Decay		
Beta-Rays	8	8
Gamma-Rays	7	7
Neutrinos	12	-
Prompt Gamma-Rays	7	7
Fission Neutrons (Kin.En.)	5	5
Capture Gamma-Rays	-	3-12
Total	207	198-207

1.3. Nuclear Power Plants

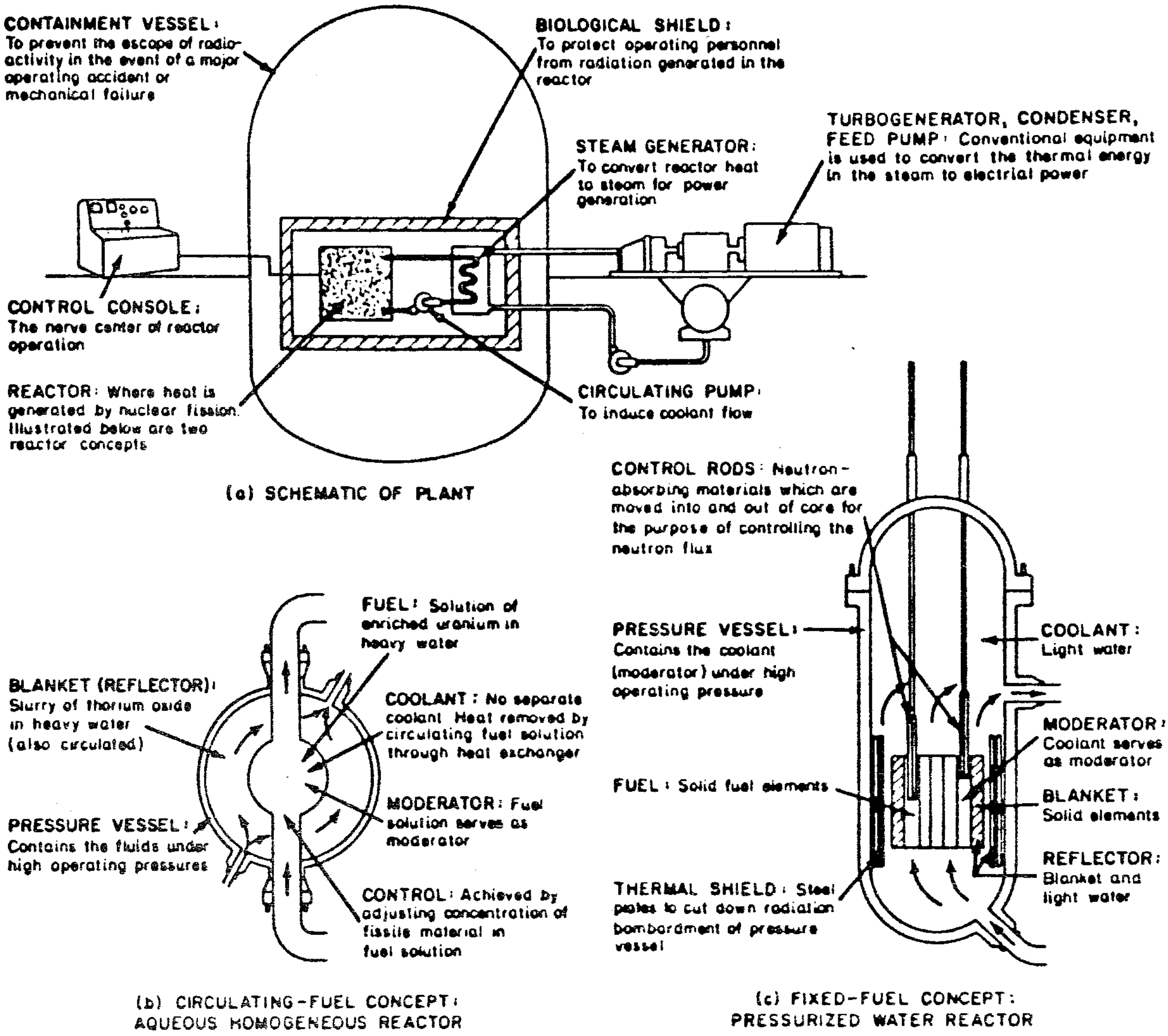
In the present section, nuclear systems devised for electricity generation are overviewed. First the terminology that applies to nuclear reactors is explained and then the plant types are briefly described. Some of the designs have found wide-spread applications, whereas some are still preserving their conceptual nature partly because of technological drawbacks and partly due to economic reasons.

1.3.1. Engineering Principles

Fig.1.5 shows the general features that nuclear power plants possess in common. These features are explained below :

CORE : That part of the plant in which the fission reactions take place. It contains the nuclear fuel which consists of fissile and usually ferti material. It has openings, as well, through which the coolant flow or control rod movements are allowed.

MODERATOR : The moderator is the material that is used to slow down fast neutrons emerging from fission to thermal energies as most of the fissioning is induced by slow neutrons. Materials that show little tendency to capture neutrons are good moderators. Examples are Ordinary or Heavy Water, Beryllium, Beryllium-Oxide, Graphitic Carbon or Hydrocarbons.



(a) SCHEMATIC OF PLANT

(b) CIRCULATING-FUEL CONCEPT: AQUEOUS HOMOGENEOUS REACTOR

(c) FIXED-FUEL CONCEPT: PRESSURIZED WATER REACTOR

Fig.1.5. General Features shared by Nuclear Power Plants

REFLECTOR : The reflector serves the purpose of returning back those neutrons that leave the core, to increase neutron economy and hence to reduce the critical fissile mass. The reflector can be of the same material as the moderator unless fissioning by highly energetic neutrons is employed in which case the moderator is not used.

COOLANT : The substance that is used to remove the heat generated in the core is the coolant. It is circulated at a rate determined by the operating conditions and it transfers the heat to a secondary working fluid which runs the turbo-generating system. The coolant can be Liquid Water, Liquid Sodium, Sodium-Potassium alloy, organic, air, CO_2 or Helium.

CONTROL RODS : Control rods consist of a neutron poison, ie, a material with high neutron capture cross-section, to adjust the neutron density in the core volume so as to control power generation. Control rods are dynamically inserted into or withdrawn from the core where/when appropriat

1.3.2. Nuclear Power Plant Types

Nuclear reactors can be classified according to various criteria such as operational goal or neutron energies at which fission is induced or the type of coolant adopted. These criteria, however, are not completel disjoint and there is considerable interplay because adoption of one purpose brings limitations on the selection of materials or brings further advantages.

So far as the species used as fuel is concerned, those that utilise U-235 as the driving fuel are restricted to be Thermal Reactors in which fissioning with the aim of neutrons at thermal energies is employed. U-235 is usually used in combination with U-238 to produce fissile Pu-239 that can later be used as fuel in some other reactors. Reactors of this nature are Converter Reactors. Another design for converter reactors uses Th-232 as fertile species. Burner reactors are compact reactors for special use such as in a submarine in which case only fissile U-235 is consumed and the fuel consists of almost purely U-235, ie, depleted in fertile material to a great extent. Reactors in which neutrons of high energies are used for fissioning are called Fast

Reactors. Pu-239 is used as fuel with fertile U-238 which implies that it produces Pu-239 in time. With appropriate design, one can achieve a conversion ratio, which is an indicator of the amount of fissile species to be obtained, eg. Pu-239, at the expense of fissile species consumed, eg. U-235, greater than unity, hence, these reactors can also be made to be Breeder Reactors.

According to the coolant or moderator material used, the following classification can be made :

i. LWR

Ordinary water is naturally abundant, hence, readily obtainable and the thermodynamic properties of ordinary water are well known. Its nuclear properties make it a suitable moderator material and ordinary water is widely used both as coolant and moderator. There are two concepts employing ordinary water and nuclear superheating as described below :

i.a. PWR

As its name implies, ordinary water, which serves both moderation and cooling purposes, is circulated through the core under pressure and hence, heat extraction is carried out without bulk boiling occurring. For this reason, heat transfer system consists of two circuits, a primary loop which contains the reactor and one side of the steam generator and a secondary loop which contains the steam side of the steam generator and the turbine generator. That the water coolant is kept under pressure is achieved by two designs. In the Pressure-Vessel design, the reactor vessel bears the full pressure of the coolant whereas it is kept at a lower pressure in Pressure-Tube design in which case coolant under pressur is confined to pressure tubes that bear the full pressure of the coolant. In the Pressure-Tube concept, the coolant and moderator are separated, therefore, the moderator can be of a different substance to be kept at a lower pressure.

Since bulk boiling is not allowed, the temperature of the water coolant is limited by its saturation temperature at that operating pressure, which is about 320°C. This limitation then determines the thermal efficiency of this type of reactor. Low degree of superheat in the steam requires special blading in the turbines and moisture separators in the steam circuit.

i.b. BWR

The water coolant is allowed to boil in the core so that boiling heat transfer is employed as is not the case with PWR. BWR concept has the following potential advantages over the PWR concept :

System pressure is low which reduces Pressure-Vessel construction costs. The intermediate heat exchanger boiler can be eliminated and the reactor heat is used in a straightforward manner as the steam generated is directly fed to the turbine. The elimination of the intermediate boiler conserves the temperature drop encountered in it and the efficiency is raised at the same fuel temperature.

The corrosive properties of water limit structural materials to stainless steel and Zirconium. Activity induced in the coolant because of the impurities present introduces problems. Another source of radioactivity in water results from the neutron reaction with O-16 producing radioactive N-16 which limits access to the coolant circuits during reactor operation. This activity of N-16 is unavoidable but it decays very soon after reactor shutdown with half-life of 7.5 seconds.

ii. HWR

Heavy Water, D_2O, has very good moderation and neutron economy characteristics. Heavy Water can serve both as coolant and moderator but designs using gaseous or Na coolant with Heavy Water moderator have been proposed. Heavy Water shows very little tendency to parasitic neutron absorption and this property permits the design of small reactors using natural Uranium fuel. The leakage of HeavyWater, on the other hand, presents health hazard because of Tritium produced from Deuterium by neutron absorption. The Canadian design CANDU is to be recognised in this concept.

iii. OCR

OCR reactors are very similar to PWRs in that the moderating properties of organic liquids are about the same as those of water and coolant cycles are nearly identical to those of PWRs. They are advantageous in the following features :

iii.a. System pressures at 300°C to 450°C range are low,
iii.b. Corrosion rate is negligible,
iii.c. No chemical reactions between the coolant and fuel, cladding or water take place.

Organic coolants, on the other hand, decompose under long irradiation or above 400°C. The thermal efficiency is, therefore, low. Diphenyl or terphenyl isomers and their mixtures are mostly used both as coolant and moderator.

iv. LMCR

Liquid metals show very good heat transfer properties such that they can be used for both thermal and fast reactors. Liquid Sodium, which is a widely used coolant, has its boiling point near 880°C and this permits operations at low pressure and high temperature. The pumping power requirements are, therefore, rather low. These systems are usually moderated by Graphite.

Liquid Sodium is activated by neutrons as it passes through the core. Another disadvantage associated with Liquid Sodium is the deposition of the corrosive Sodium-Oxide. The primary system must provide Sodium purification facilities to control the Oxygen content.

The reaction of Sodium with water necessitates that they must be kept separate in the steam generator.

v. GCR

Gases such as CO_2, air or in later advanced systems, Helium are used as coolant. Graphite is usually used as moderator but Heavy Water moderation has also been proposed. This reactor concept is investigated in detail at Chapter 4.

vi. FBR

These reactors are intended to breed nuclear fuel, mainly Pu-239 and they operate at the fast region of neutron spectrum. They are generally cooled by Sodium.

vii. Fluid Fuel Reactors

This concept is interesting as the fuel is not stationary as in other concepts but mobile. It is usually in solution or in suspension. Therefore, continuous addition or removal of fuel is eased and fuel element fabrication is avoided.

1.4. Burnup, Conversion and Breeding

Burnup is a measure of the amount of fissile material consumed (or power produced) before a fuel element is removed from the reactor. It is widely expressed in terms of MegaWatt-Days/Tonne of fissile isotope which is related to the number of fissions per cm^3 of fuel measuring the extent of the depletion of fuel. The average fuel burnup in thermal reactors is about 10,000 MWD/T with the exception of HTGR where very high degrees of burnup, exceeding 100,000 MWD/T can be achieved.

Conversion is a process in which some of the neutrons in the core are used for a purpose different from sustaining the chain reaction, such as producing fissile species from fertile nuclei. As has been already noted, neutrons below a certain energy do not induce fission in U-238 and Th-232. By neutron absorption, these species are converted into Pu-239 and U-233 respectively through a series of decays :

$${}_{92}U^{238} + {}_{0}n^{1} \longrightarrow {}_{92}U^{239} + \text{Gamma}$$

$${}_{92}U^{239} \longrightarrow {}_{93}Np^{239} + \text{Beta}^{-}$$

$${}_{93}Np^{239} \longrightarrow {}_{94}Pu^{239} + \text{Beta}^{-}$$

and

$${}_{90}Th^{232} + {}_{0}n^{1} \longrightarrow {}_{90}Th^{233} + \text{Gamma}$$

$${}_{90}Th^{233} \longrightarrow {}_{91}Pa^{233} + \text{Beta}^{-}$$

$${}_{91}Pa^{233} \longrightarrow {}_{92}U^{233} + \text{Beta}^{-}$$

The conversion ratio is defined as the average number of fissile atoms produced in a reactor per fuel atom consumed either by fission or absorption. If more fuel is produced than is consumed in a reactor, ie, if the reactor has a conversion ratio greater than unity, the reactor is said to be a Breeder and the process is called Breeding.

2. THORIUM

Thorium is an element of interest to nuclear reactor technology because of several reasons. The primary reason is that Thorium is a fertile material and can be converted into the fissile Uranium isotope U-233 by neutron absorption. The nuclear properties of U-233 can make Th/U-233 fuelled reactors produce energy probably at a lower price than from U-235 fuelled reactors. The high conversion ratio of Thorium systems can lower the Uranium ore consumption. Another reason that makes Thorium attractive is that its natural abundance in earth's crust is three times as much as that of Uranium. Although Thorium is not used extensively at present, it shows considerable potential for use in Light Water Breeder and High Temperature Gas Cooled Reactor concepts since thermal breeding is principally possible in Th/U-233 fuelled reactors.

2.1. Natural Abundance and Extraction of Thorium (4).

Thorium is found in nature as monazite in useful amounts and commercially obtained from sands bearing monazite. Monazite is a mixtur of rare-earth phosphates which contains 1 to 5 % Thoriumdioxide and a smaller proportion of Uranium.

Monazite is concentrated by applying standard ore-dressing methods to alluvial deposits that comprise Silica (sand). The abundances of Thorium and Uranium in the resulting product is 5 to 8 % and 0.15 to 0.25 % respectively. Silica, Titanum, Iron, Lanthanum and other metals are also present, though at varying amounts.

In the extraction process, the solid mixture containing Thorium is ground finely and digested with nitric acid. Following several steps of concentration, multistage solvent-extraction with Tributyl-Phosphate method is applied to separate Thorium in nitric acid solution from the associated rare-earths.

The Thorium is precipitated as the oxalate and the nitrate is back-extracted from this solution with water. Thoriumtetrafluoride (ThF_4) is formed by hydrofluorination of the dioxide that remains after heating. ThF_4 is reduced to Thorium metal by means of Calcium. Zinc is introduced in the reduction process to make an alloy of Thorium with low melting point since Thorium has a high melting point, 1750°C, by itself. Zinc is later removed by heating the alloy in vacuum.

Thorium assumes a face-centred cubic structure up to 1400°C. Above it, the stable phase becomes a body-centred cubic form. The radiation stability of Thorium is greatly superior to that of Uranium, since it is of the cubic system which implies that its properties are isotropic. The thermal conductivity of Thorium metal is 0.090 at 100°C and increases to 0.108 $cal/sec.cm^2.{}^\circ C$ per cm at 650°C.

Handling and processing of Thorium should be done with regard for the potential hazards that may be encountered. Thorium is a radioactive element and usually has radioactive daughter products, that can cause health hazards, with it. Thorium can also be a chemical poison in some forms but its hazard as a chemical poison is presumably less than its radiological hazard. Thorium is very active as a metal and as powder or any other form with a large surface area must be considered as a potent fire or explosion hazard (6).

2.2 Thorium as a Fertile Material

The applicability of Thorium as a power reactor fuel is based on an (n,Gamma) process on Th-232. The resulting nucleus, Th-233, is not stable and undergoes $Beta^-$ decay with half-life 22.1 minutes into Protactinium-233, Pa-233. This nucleus is not stable either and hence decays with half-life 27.4 days into U-233 through $Beta^-$ emission. U-233 is an Alpha emitter and has a half-life of $1.62X10^5$ years. The U-233 nucleus is capable of fissioning by thermal neutrons and the average number of neutrons emitted per absorbed neutron, the neutron yield η, is higher for U-233 than for U-235 at all significant neutron energies for reactors. The yield is even higher than for Pu-239 at neutron energies below 40 keV. The nuclear chain starting with Th-232 is presented in Fig.2.1.

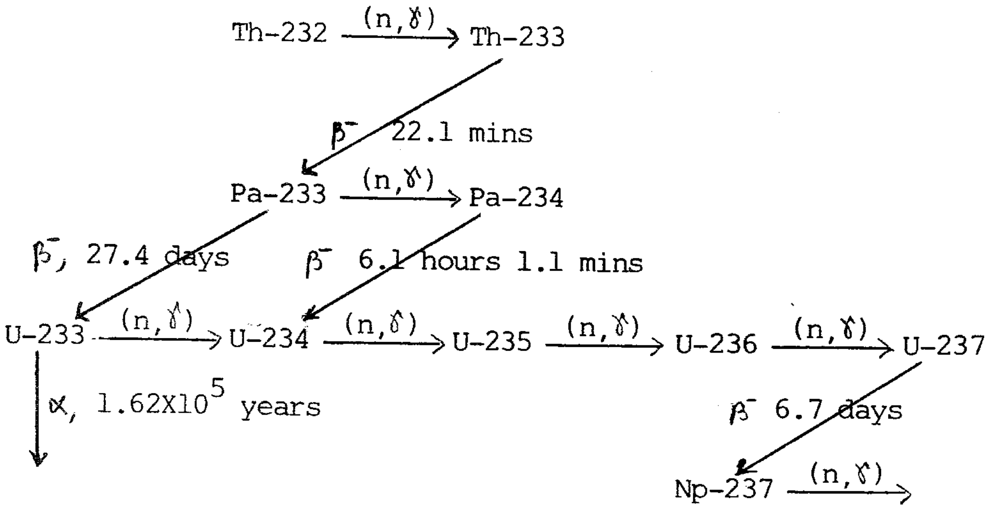


Fig.2.1 Nuclear reactions and decays starting with Th-232

2.3 Thorium Resources in Turkey

A large Thorium deposit is found in Turkey in the region Eskişehir-Beylikahır and has been coded as KTS-1. The deposit comprises 380,000 Tonnes of ThO_2 along with rare-earth elements as displayed in Table 2.1. The low Thorium content of the deposit, approximately 0.53%, should be noted and call for attention when compared with Canadian reserves exceeding 100,000 Tonnes of ThO_2 with 2 to 3 % Th content.

Table 2.1 The elements found in KTS-1 Thorium deposit (7).

Element	Abundance mg/g
Sc	0.015 ∓ 0.002
Fe	26.40 ∓ 6.20
Co	0.021 ∓ 0.003
As	0.19 ∓ 0.02
Sb	0.006 ∓ 0.001
Ba	118.4 ∓14.2
Cs	0.0075∓ 0.0007
La	3.62 ∓ 0.42
Ce	15.58 ∓ 1.85
Eu	0.33 ∓ 0.03
Tb	0.07 ∓ 0.01
Yb	0.03 ∓ 0.01
W	0.024 ∓ 0.006
Th	5.27 ∓ 0.057

3. THORIUM FUEL CYCLE

At present, the Uranium-Plutonium fuel system is strongly emphasised. However, the Th-232/U-233 system may provide an alternate path toward a solution to present energy requirements. The Thorium fuel is mainly advantageous over natural Uranium owing to the bred U-233 isotope, therefore, recycling the bred U-233 is an inherent requirement of Thorium utilisation. The principal operations and material flow in the Th-232/U-233 fuel cycle are presented in Fig.3.1.

The fuel cycle starts with the feed material. The standard starting material is the principal commercial salt $Th(NO_3)_4.4H_2O$, Thoriumnitratetetrahydrate. Fissile material is either enriched U-235 or U-233 produced elsewhere. In the fabrication step, the feed materials are converted into the desired chemical forms and fabricated into fuel elements. After being irradiated for a certain exposure time, the discharged fuel elements are directed into the Reprocessing Facility where the fission products, Uranium and Thorium are mechanically and chemically disassociated. Reprocessing is followed by waste disposal or storage stages, in the latter case the recovered actinides are put into a suitable form for refabrication.

Associated with the German Pebble-Bed design that is to be explained in Chapter 5, the following fuel cycles employing Thorium have been investigated :

i. UOT : Uranium Once Through cycle is an open cycle. Low enriched Uranium is used without recycling.

ii. TOT : Thorium Once Through cycle is again an open cycle.Highly enriched Uranium and Thorium are loaded as mixed oxides without recycling.

iii.MOR : Mixed Oxide Recycling scheme utilises the recycling of mixed oxide Uranium that is recovered from mixed oxide fuel.

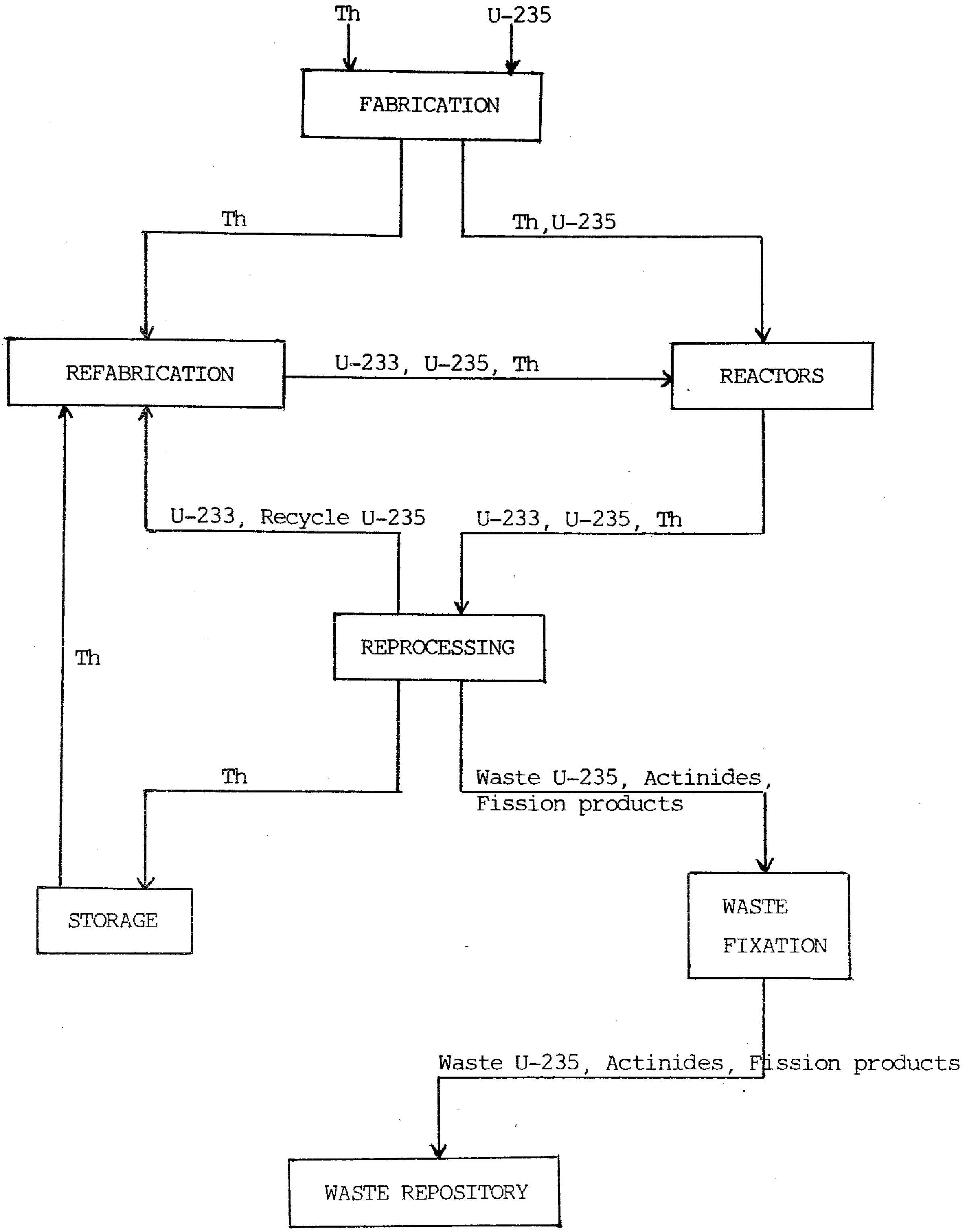


Fig.3.1 Principal Operations and Nuclear Material Flow in Th/U fuel cycle.

iv. RSE : Recycling in Separate Elements scheme makes use of recovered Uranium from mixed oxide in separate elements without Thorium. The recovered Uranium is recycled once to allow for the removal of U-236.

v. SFB : Separate Feed and Breed elements is a combination of MOR and RSE schemes. Highly enriched Uranium and Thorium are loaded into different fuel elements. Bred U-233 is repeatedly recycled as mixed oxide. Driver fuel is recycled only once in separate elements.

The characteristics of the cycles are outlined in Table 3.2.

Once through cycles of Uranium and Thorium rely on the most conservative fuel element design. Uranium enrichment is in the range 5 to 15 %. Gross Uranium ore demand is 13 % higher for the UOT cycle than for the TOT cycle. This is a consequence of the difference in the α-values, ie, capture-to-fission ratios, of bred U-233 and Pu-239 isotopes as can be seen in Table 3.1.

Table 3.1. Nuclear Properties of Fissile Isotopes (17).

Property :	$\eta = \nu \sigma_f / \sigma_a$		$\alpha = \sigma_c / \sigma_f$	
Moderation Ratio :	246	110	246	110
Isotope				
U-233	2.24	2.22	1.12	1.13
U-235	1.98	1.93	1.23	1.26
Pu-239	1.81		1.60	
Pu-241	2.10		1.42	

In either case, the disloaded fuel contains a very weak concentration of fissile material, so that the closure of the fuel cycle is not so urgent as for LWR.

Table 3.2 Characteristics of the Fuel Cycles considered (per 1000 MW_e)

		Non-Recycling		Recycling		
Fuel cycle		UOT	TOT	MOR	RSE	SFB
Average Moderation Ratio,	N_C/N_{HM}	357	325	325	325	278
Fuel residence,	days	1122	1217	1217	1209	1499
Conversion Ratio,		0.58	0.60	0.57	0.62	0.65
Maximum Power,	kW/Ball	2.8	2.9	3.3	3.5	4.4
Maximum Fuel Temperature,	°C	706	706	706	716	743
Fissile Inventory,	kg/GW	1022	914	1090	937	1208
Fuel Cycle Inventory,	kg/GW	1022	914	1340	1153	1436
U_3O_8 Demand,	kg/TJ	6.04	5.35	3.96	3.48	3.28

Important fuel cycle parametres and resource requirements are listed in Table 3.3. U_3O_8 and enrichment requirements provide a meaningful measure for the relative advantages and disadvantages of the fuel cycles. As can be recognised from the data in Table 3.3, Thorium cycle with lower moderation ratio yields the lowest resource requirement with recycling. The low enriched Uranium scheme is favoured in non-recycling case although the ore requirement is approximately 13 % higher than that of Thorium non-recycle. Thorium cycle HTGR is readily seen to be significantly advantageous over the standard Pressurised Water Reactor.

MOR, RSE and SFB schemes are proposed for the closure of the fuel cycle. In MOR, both make-up and recycle fuels comprise mixed oxide of Uranium and Thorium. For THTR design, Heavy Metal loading of 11.24 g/ball is used. The make-up fuel elements are of two enrichments loaded in two different radial core zones to achieve radial power flattening Recycling fuel elements comprise 28 % of the total charge. They require remote fabrication techniques. They are made of only one enrichment and equally loaded into core.

The major disadvantages of the MOR scheme are the continuous build-up of U-236 and its removal [1]. U-236 must be counter-balanced by fresh fuel supply at the rate of its continuous build-up to sustain criticality. In the life-span of the reactor, U-236 gradually builds up to the level of the fissile inventory in the core and lowers the conversion ratio. The influence of U-236 build-up is outlined in Table 3.4.

RSE scheme allows for removal of U-236 utilising separate elements. Use is made of mixed oxide fuel elements with 15 g of HM/ball for make-up. Recycling fuel elements are free of Thorium. After reprocessing, the recovered Uranium is used to form TRISO particles with UO_2 kernels. Beside these particles, the recycling

---

1. See appendix B.

Table 3.3. Comparison of LEU, HEU Th HTGR and PWR parametres

		LEU HTGR	HEU HTGR		PWR
Moderation Ratio,	N_C/N_{HM}	400	240	180	
Fuel lifetime,	years	3	4	4	3
Conversion Ratio,		0.50	0.66	0.76	
U-235 Enrichment,	%	11	93	93	3
U_3O_8 Requirement, Recycling		3.9	2.3	2.0	3.3
U_3O_8 Requirement, Non-recycl.		4.9	3.9	4.7	6.3

Table 3.4. Influence of U-236 Build-up in MOR

Number of recycling years,		6	29
Fuel residence time,	days	1348	1217
Average Burnup,	TJ/kg	9.59	8.64
Conversion Ratio,		0.59	0.57
U-235 Supply,	g/TJ	14.2	15.5
Inventory, kg/GW	U-233	582	585
	U-235	392	504
	U-236	514	998

elements contain 1.7 g/ball of Heavy Metal loading. The removal of recycled fuel is done through the weight difference between them and mixed oxide make-up fuel elements. Recycled fuel elements are reprocessed in a Purex Process for Uranium removal. The removed mixture contains 51 % of U-236 and 9.9 % of fissile Uranium isotopes.

In the SFB scheme, breed and make-up material are inserted into separate fuel elements which are recycled and reprocessed separately. The breed circuit is analogous to the MOR scheme and only Thorium is added. Heavy Metal loading in both cases is 20.1 g/ball. The make-up circuit is analogous to the RSE scheme. Fresh elements contain TRISO feed particles of highly enriched Uranium. Recovered Uranium is recycled once. 0.2 % of the initially loaded U-235 is spent for removing U-236 and the fissile enrichment of the removed Uranium is about 1.2 %.

SFB scheme permits the removal of bred U-233 for more advantageous use in the Near-Breeder cycle as well as U-236 removal. Weight differences of more than 7 g make quantitative separation of fuel spheres easy and any cross-over is thus avoided. SFB stream, however, requires four different streams in fabrication and two in reprocessing. Therefore, the necessity of extended commercial introduction of reactors using this scheme follows directly as a consequence.

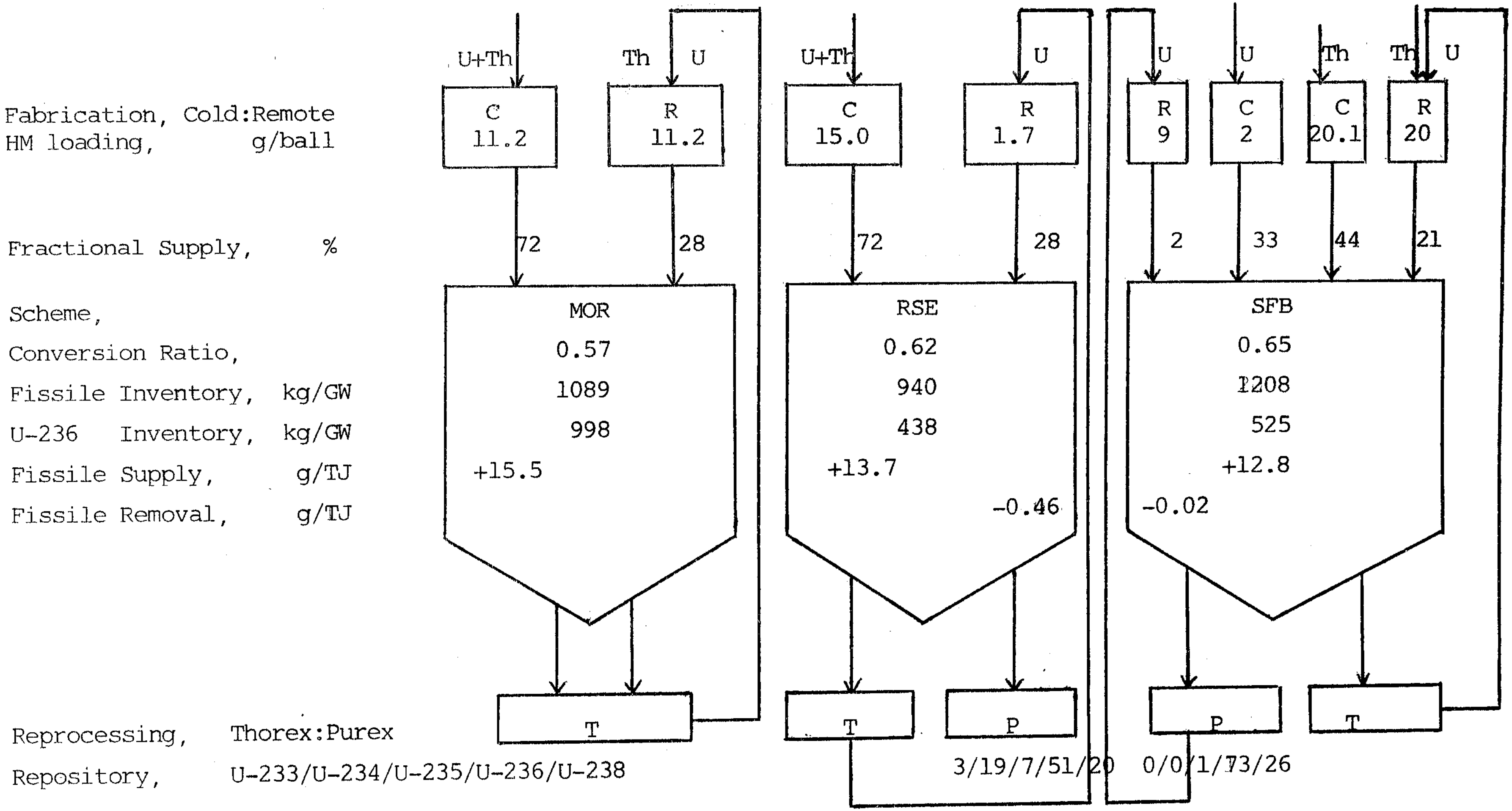


Fig.3.2 Comparison of Recycling schemes

Table 3.5. Characteristics of the fuel elements used in the Pebble-Bed HTGR fuel cycles

Fuel Cycle,	UOT	TOT	MOR		RSE		SFB			
Fuel, oxide	U	Th+U	Th+U	Th+U	Th+U	U	U	U	Th	Th+U
Particles diametre, μm	800	400	400	400	400	200	400	200	400	400
Fabrication, Cold/Remote	C	C	C	R	C	R	R	C	C	R
Fractional supply, %	100	100	72	28	72	28	2	33	44	21
HM loading, g/ball	10.2	11.2	11.2	11.2	15.0	1.7	8.9	2.0	20.1	20.1
Moderation Ratio, N_C/N_{HM}	357	325	325	325	242	2110	410	1879	180	180

4. GAS COOLED REACTORS

The first Gas Cooled Reactors were not intended to produce power but rather Plutonium from natural Uranium. Considering the facts that this concept did not place so high a demand on fresh water supply for cooling purposes as LWR and that the siting requirements were not so strict as for LWR, continuous effort was spent in UK, france and later in FRG and US to convert this concept into a power producing one.

Gas Cooled Reactors can be broadly classified into two groups according to the operating temperature level. The former types operated below 537°C (1000°F) whereas the later designs above it.

4.1. GCR using CO_2 as coolant

These reactors are fuelled with natural Uranium and cooled with CO_2. Their operation temperature lies below 537°C (1000°F).

Natural Uranium is an inexpensive and readily available kind of fuel when compared with other nuclear systems, but its use imposes certain restrictions on design. Because of the low excess reactivity in these systems, the neutron economy considerations require that the selection of the cladding material be made amongst Beryllium, Magnesium, Aluminum and Zirconium.

Table 4.1. displays the absorption cross-sections of some materials.

Zirconium is a poor absorber of thermal neutrons when free of Hafnium and has good corrosion resistance and fabrication properties.

Table 4.1. Absorption cross-sections of various materials (5).

Metal	Absorption Cross-Section (barns)
Be	0.01
Mg	0.059
Zr	0.18
Al	0.22
Nb	1.1
Fe,Mo	2.4

Although Zirconium has a high melting point, its elastic modulus is quite low and therefore, it loses strength rapidly with increasing temperature (9).

Beryllium has a very low absorption cross-section for thermal neutrons but is akin to swelling under irradiation and its brittleness introduces problems in fabrication.

Aluminum reacts slowly with Uranium. Therefore, a metallurgical diffusion barrier type of bond needs to be placed between the fuel and Al cladding (5).

Magnesium, in this respect, does not require a complex bond as such since it does not react with Uranium. Some of its alloys have low absorption cross-sections for thermal neutrons and good mechanical properties at low temperatures but lose their strength rapidly at high temperatures. Its alloys with Thorium have adequate strength at elevated temperatures but the absorption cross-section is then high enough to offset this property. British and french designs employ Magnesium alloys as cladding material but its use along with the creep strength of Uranium leads to special considerations for fuel element support. Its ability to withstand internal pressures exerted by the fission gases within the fuel element and its reaction with the CO_2-coolant limit the fuel element surface temperature to below 500°C and the usual operating temperature is between 400°C and 490°C.

Graphite is extensively used as moderator in thermal reactors It reacts with oxygen in air or in CO_2 but the rate of oxidation is acceptably low between 315°C and 400°C (0.5 by weight percent a year) (5). It undergoes dimensional changes and an increase in stored energy under irradiation.

4.2. Advanced Systems ; HTGR

The reaction of graphite with CO_2 at elevated temperatures, low excess reactivity because of natural Uranium fuelling and hence the necessity of Mg-cladding which permits operating temperatures below 500°C lead to advanced systems in which the coolant is other than CO_2 such as He or N and the fuel is enriched Uranium clad in stainless steel, Beryllium or Hast-alloy or is in the form of coated particles as in US and FRG designs.

4.2.1. Design Characteristics

Advanced systems primarily make use of the inert Helium as coolant for it does not interact with reactor materials. Its heat capacity is low since it is monatomic and it is second to Hydrogen in thermal conduction properties. Natural Helium comprises He-3 to $1.3X10^{-4}$ % and the rest is He-4. The absorption cross-section of He for thermal neutrons is 7.0 millibarns. Impurities in He, such as air, Hydrogen, Hydrocarbons and water vapour can be removed by passing the He through hot Zirconium chips or liquid Lithium. Another means of elimination is to pass the He under pressure through a charcoal bed at liquid air temperature (10).

The HTGR utilises an all-ceramic core, a graphite core strucţture and ceramic coated fuel particles. The use of core material that can withstand high temperatures combined with a single phase inert Helium coolant allows high coolant temperatures and results in a number of significant advantages including high tolerance for transient temperatures. Low power density and the heat capacity of the core imposes safety during operation and loss of forced circulation events eliminating the need for immediate operator action and the operator response time is long enough to be measured in hours.

The removal of decay heat through natural convection and the very low activity induced in the Helium coolant because of its nobleness combined with the unique design characteristic, the Pre-stressed Concrete Reactor Vessel, the PCRV, which confines the primary steam supply system further lowering the radiation exposure of operating and maintenance personnel ease reactor siting constraints reducing both cooling water requirements and the consequences of postulated accidents.

The current generation of HTGRs operates on a conventional steam cycle by wet-cooling though, but the thermal efficiency of 40.0 % has been achieved which is fairly high relative to the 30 % efficiency of LWR. When dry-cooling systems are developed and advanced systems employing direct-cycle Helium turbines are available, the efficiency will even rise beyond 40 % (11).

4.2.2. Coated Particle Fuel concept

Another innovation that the HTR brings besides PCRV is in fuel element design. Hence, it is necessary at this point to discuss the HTR fuel in detail.

The development of the HTR has proceeded in two directions :

i. The Pebble-Bed concept in FRG,
ii. The prismatic core in US and UK.

There is a substantial difference in the two HTR concepts though, but the characteristics in common is that the fuel is in the form of coated fissile and fertile microspheres. The graphite moderator is an integral part of the fuel element as it improves the neutron economy.

4.2.2.1. Characteristics of Coated Particle Fuels

These coated particles consist of either pure or mixed oxide or carbide Uranium and Thorium. They comprise, along with the fuel Kernels which range from 200 μm to 800 μm in diametre, coatings of 200 μm total thickness. One of the advantages of this design is that the small size of coated particles allows the collection of statistically sufficient and adequate data in testing.

The coating layers and to a lesser extent the fuel Kernels themselves serve as barriers to the release of the fission products. At elevated temperatures, these barriers are more or less effective depending on the material used and the type of fission product considered. The barrier expected to retain the fission products most effectively is Pyrolytic Silicon Carbide. The design of the coated particles with a SiC layer calls for additional layers to prevent the chemical reaction between the fuel Kernel and the SiC layer :

i. A low density pyrolytically deposited Carbon layer surrounding the microsphere, the buffer layer,

ii. Two layers of High Density Isotropic (HDI) Pyrolytic Carbon (PyC) between which the SiC layer is interlaid.

This design is known as TRISO coating. The earlier designs, the TRISO-I particles, did not contain an inner HDI PyC so that only a porous buffer layer was placed between the Kernel and the SiC layer.

Another type of particle coating which is in most common use with TRISO particles is the BISO coating. BISO coating comprises two layers of Pyrolytic Carbon. Seal coats of dense, relatively anisotropic PyC are sometimes used between the buffer and HDI layers in BISO particles. This is primarily for the purpose of minimising reactions between oxide Kernels and Carbon coatings during processing at high temperatures.

The coatings are formed by chemical vapour deposition in a fluidised bed. BISO particles are used with fertile Kernels, TRISO particles, however, with both fertile and fissile kernels with different particle diametres.

The coatings of TRISO particles and their functioning can be summarised as follows :

The Kernel of the particle consists of the fissionable fuel. Determined by the fuel cycle intended, it comprises either highly enriched Uranium together with fertile Thorium or low enriched Uranium or Thorium alone either in the form of oxides or carbides. The Kernel is coated with a buffer layer which attenuates fission recoils and provides adequate void volume to accomodate fission gases. It constitutes a barrier between the structural layers of coating and the Kernel.

The Isotropic Inner PyC layer provides impermeability to chlorine compounds during SiC layer deposition and restricts interactions between the fissile or fertile Kernel or fission products and the SiC layer. It helps in minimising the irradiation effects that lead to dimensional changes and stresses.

The SiC layer acts as a pressure vessel of a very small size and provides adequate support against internal fission gases and it is impermeable to metallic fission products. It is the primary means of structural support.

The SiC layer must be polycrystalline with nearly theoretical density of 3.21 g/cm^3 to prevent the release of solid fission products through the coating. According to the results of a series of experiments in which coated particles have been irradiated in a wide range of temperature, burn-up and fast neutron flux, SiC shows a distinct superiority in the retention of Silver isotopes Ag-110m and Ag-111.

The Isotropic Outer PyC layer adds to the structural support provided by the SiC layer. It acts as an additional barrier to the release of gaseous fission products in the event of SiC failure. It provides a bonding surface for the carbonaceous matrix material because the latter does not bond to the SiC layer. It also protects the SiC layer from external corrosion.

Both BISO and TRISO particles provide complete retention of gaseous fission products. Intact TRISO particles also provide nearly complete retention of metallic fission products at current peak HTR design temperatures. However, diffusional release of certain metallic fission products, particularly Cesium, Strontium and Silver does occur at elevated temperatures from BISO coatings. BISO coatings are generally preferred where fuel temperatures are relatively low or with relatively low burn-up oxide fuel where the Kernel provides sufficient retention of metallic fission products or with fertile Kernels.

The coating materials are almost always of PyC and SiC. PyC coating is dependent on its production process which determines its properties such as density, crystalline anisotropy. There are two kinds of PyC coatings :

High Temperature Isotropic (HTI), which is deposited from methane at high temperatures and Low Temperature Isotropic (LTI), which deposited from Hydrocarbons such as Propane, Propene, Propene mixed with Acetylene and Butane at low temperatures. LTI has a better irradiation stability than HTI but its retention capabilities for metallic fission products such as Cesium are weaker.

SiC is a much simpler coating material than PyC because its properties are not dependent on the production process. At operating conditions of current HTR designs, it provides complete retention and mechanical strength.

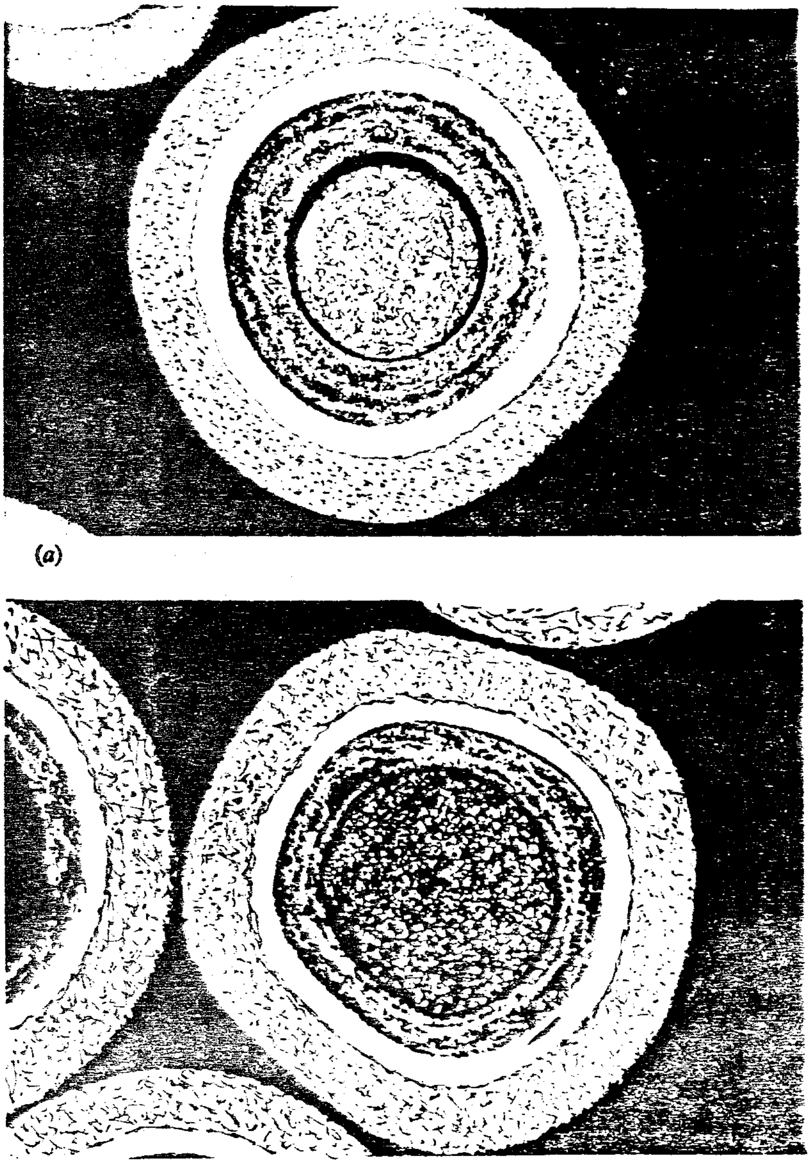


Fig.4.1 TRISO particles

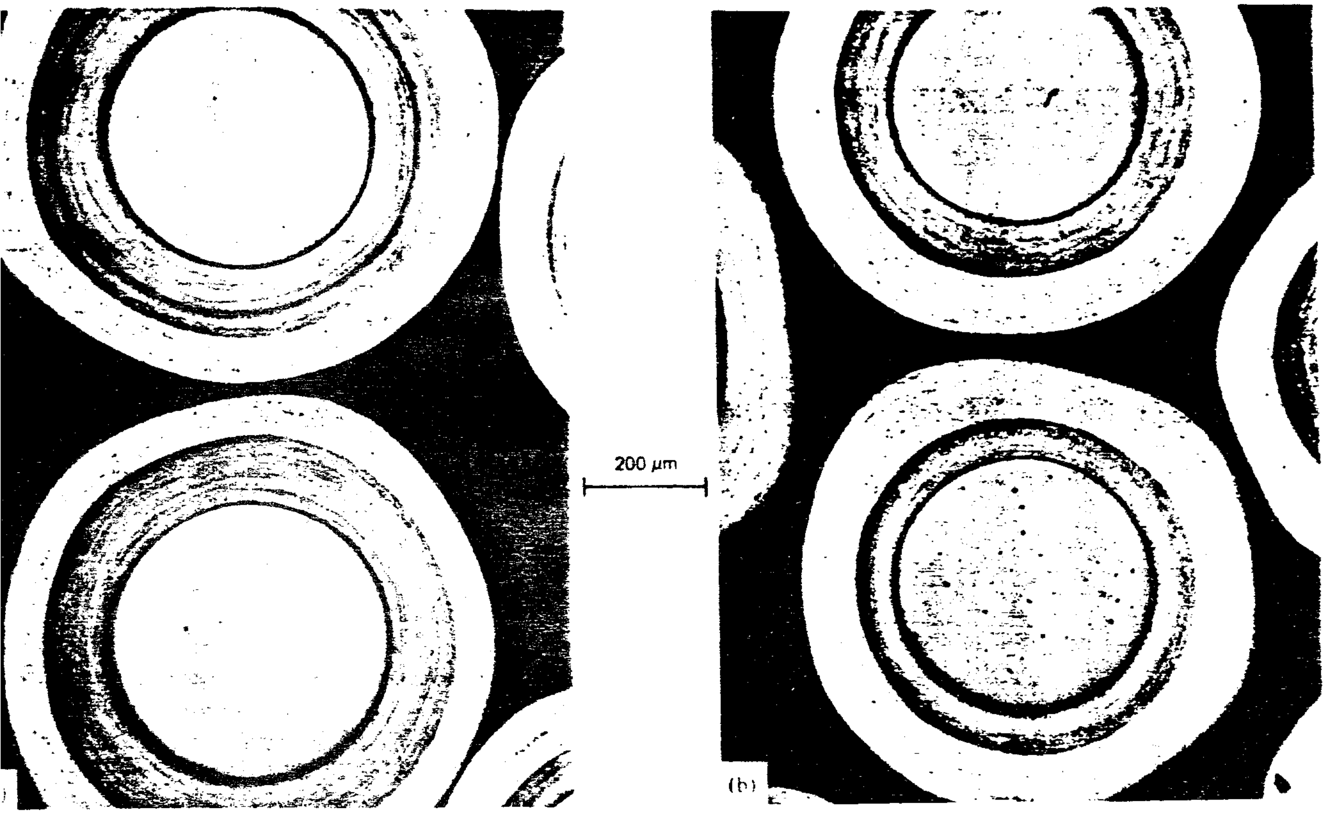


Fig.4.2 BISO particles

4.2.2.2. Failure

In general, there are mainly two kinds of failure that limit the performance of the coated particle fuel :

i. Mechanical failure,
ii. Chemically induced failure of the coatings because of Kernel migration or fission product attack.

Mechanical Failures :

Internal Pressure

The coated particles are designed as miniature spherical pressure vessels. With increased burn-up, the build-up of fission gases exert internal pressures that lead to tensile stresses which have the potential ability to crack the coating. Gases that contribute are the Xenon, Krypton and CO from oxide fuel Kernels resulting from increased oxygen availability. The moles of CO produced are equivalent to the moles of Oxygen generated by fissioning. This phenomenon can be avoided by proper design measures at the expense of increased fabrication costs and reduced Heavy Metal loading (12,13) .

Fast Neutron Effect

PyroCarbon failure induced by neutron dose is another example to mechanical particle failure phenomena. The anisotropic and inhomogeneous coating materials undergo dimensional changes leading to stress components that are able to crack the coating under irradiation. This is particularly observed in PyC layers. The Inner PyC and Outer PyC layers in TRISO-coated particles suffer from shrinkage as a result

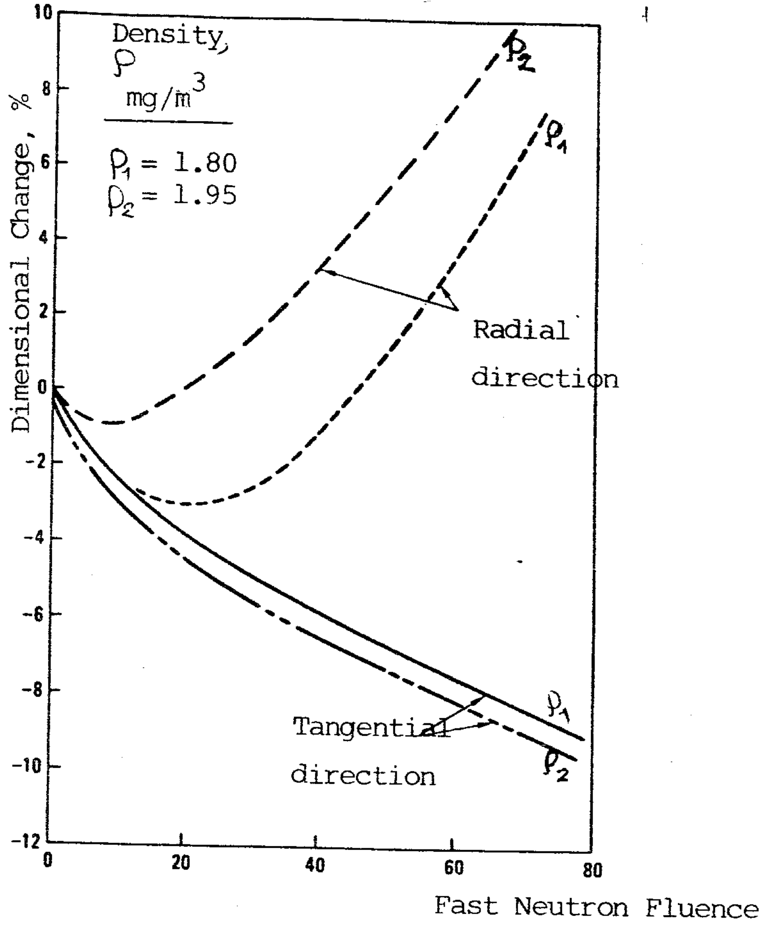


Fig.4.3. PyC Dimensional Changes

of long exposure to fast neutrons. As a consequence, the Outer PyC layer exerts compression on the SiC substrate counteracting the tensile stress in SiC arising from internal fission gas pressure. In the other direction, the Inner PyC layer tends to reduce the SiC tensile stress by contracting while maintaining contact with the inner SiC surface (12).

Chemical Failures :

Amoeba Effect

The Carbon transport and the unidirectional migration of the fuel into the coatings under the influence of a temperature gradient are other performance limiting factors which eventually may result in complete destruction of the coatings. This phenomenon is termed as the Amoeba Effect and is particularly observed in Uranium, Thorium and Plutonium fuels both in oxide or in carbide form. Results of recent experiments, however, suggest that WAR, Weak Acid Resin, UC_xO Kernels do not migrate in a thermal gradient as much (15).

Mass transport across or around the Kernel is possible in three different mechanisms such as the solid state diffusion of a species such as Carbon through the Kernel, gaseous diffusion of Carbon as CO around the Kernel and the gaseous diffusion of Carbon in some other compound.

When Oxygen is transported up the temperature gradient by some means, the Carbon transport occurs at the same time. This could be in the form of CO_2 by gaseous diffusion around the Kernel or in the form of O^{-2} by the diffusion of oxygen-ions through the Kernel.

Solid state diffusion occurs mainly in Carbide fuels since Oxygen availability is essentially high in Oxide fuels.

Gaseous diffusion of Carbon as CO assumes the following reaction and diffusion steps which are represented in Fig.4.4 :

i. Gaseous diffusion of CO_2,
ii. Diffusion of Oxygen ions,
iii. Decomposition of CO.

The Kernel migration was initially assumed to be controlled by gaseous Carbon transport in oxide fuels since Oxygen is released in fissioning. It is, however, the Oxygen diffusion mechanism that controls the migration rate whereas an overpressure of CO might trigger the oxide Kernel migration. One can deduce, therefore, that low Oxygen availability will suppress the Amoeba attack because of the lack of CO as Carbon carrier. This is known as Oxygen gettering.

Carbide fuel migration is better understood than the oxide Kernel migration mechanisms. In this case, Carbon is transported in solid form. Carbon diffuses through the Kernel as it is taken into solution on the hot side of the Kernel and deposited at the cool side or precipitated within the Kernel. As a result, the Kernel advances toward the hot side of the particle.

SiC Failure

This failure is characterised by a defective SiC layer that is incapable of bearing a tensile stress. Defective SiC layer results from either fabrication abnormalities or irradiation-induced thermo-chemical fission product or Kernel attack. The SiC fracture strength for these defects is determined by the extent of either as-fabricated or in-use induced cracks or the amount of thermo-chemical attack. The SiC corrosion is the terminal stage in coated particle deterioration and it leads to increased release of fission products.

Fig.4.5 schematically presents the SiC failure.

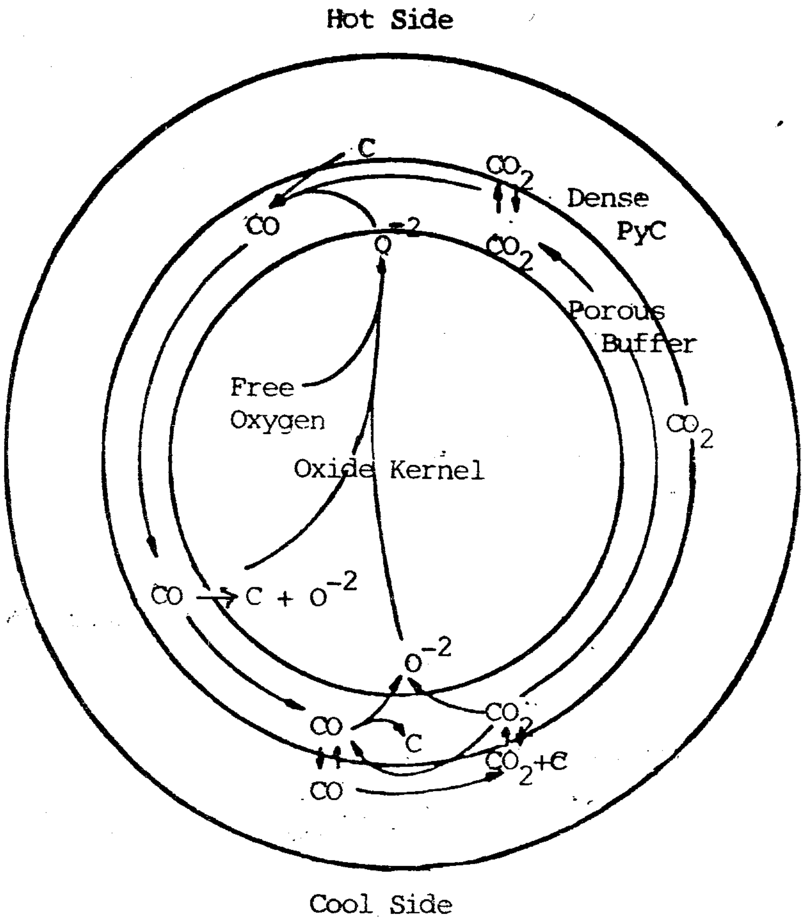


Fig.4.4. Reaction and Diffusion steps in Amoeba Migration

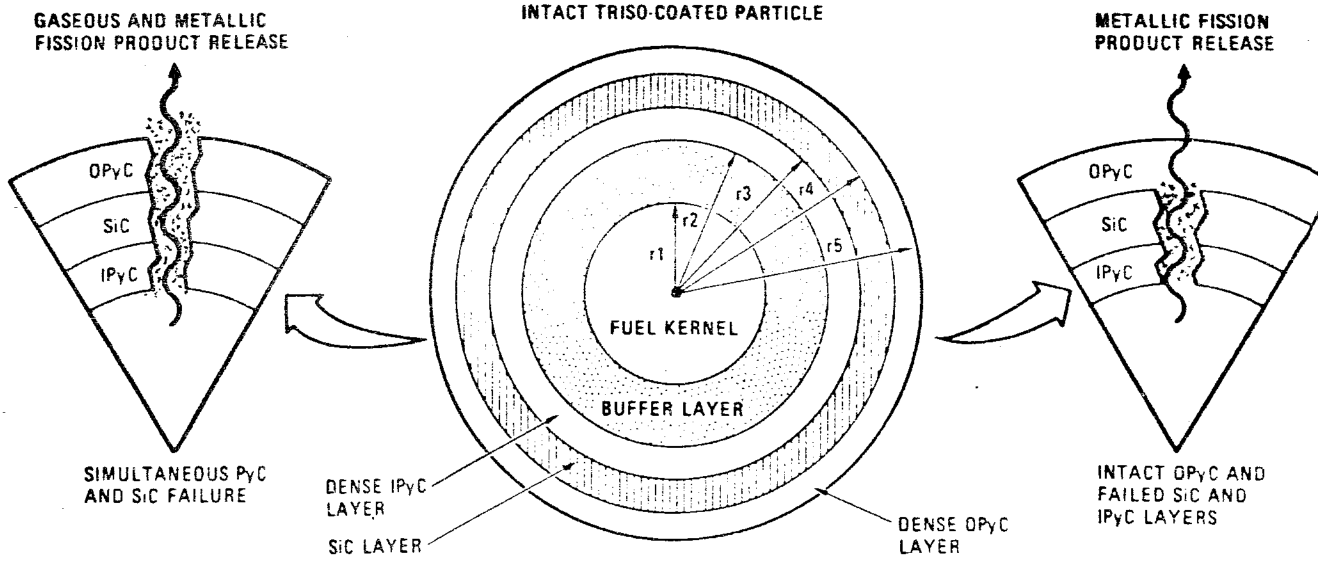


Fig.4.5. TRISO Particle failure

When both Inner PyC and Outer PyC layers are intact, the two PyC layers bear the internal gas pressure jointly and this reduces the tensile stresses in each layer. Following the failure of either of the layers, the entire load is borne by the remaining layer. Current TRISO designs rely upon the Outer PyC layer to bear the maximum stress arising from internal gas pressure.

Thermo-chemical attack of Kernel, the Amoeba effect, or of fission products result in the corrosion of SiC layer. One peculiar example is the reaction with the Lanthanide group of fission products such as Cerium, Neodymium, Lanthanum, Samarium, Praseodymium and Europium. These elements diffuse to the cooler side of the particle and concentrate near where the reaction takes place. This is a phenomenon mostly observed in carbide fuels. In oxide fuels, these Lanthanide fission products form stable oxides and are thereby retained within the oxide fuel Kernel. A similar phenomenon but with Palladium group of elements such as Ruthenium, Rhodium and Palladium occurs in oxide fuels with low enrichment. These elements concentrate at the Inner PyC interface.

The presence of Strontium and Chlorine seem to be correlated with the distribution of the fission products (16). Chlorine is in the form of Uraniumchloride which combines with Lanthanide metals, alkaline earth-metals and Zirconium as burn-up increases. Compounds that are thus formed are chemically more stable. These compounds ease fission product transport through the HDI PyC layer as far as the SiC layer which is corroded on the inner surface.

WAR-UO_2 particles show moderate attack to the inner surface of SiC at temperatures around 1250°C but the penetration is observed above 1500°C. It is in the form of oxidation and is not associated with the fission products. The oxidation of the SiC layer is explained through the gaseous mixture of CO-CO_2 which is produced at temperatures as high as 1500°C. Its origin is either the reaction of $(U,Th)O_2$ Kernel with carbon or the excessive Oxygen that is produced by the fissioning of Uraniumoxide. As a result, SiO is formed and transported from the hot to the cold particle zone under the influence of temperature gradient in which case the removal of SiC layer arises.

4.2.2.3. Fission Product Release

The coated particle Research & Development work is aimed greatly at the minimisation of Fission Product Release. The most important fission product isotopes cover the fission gases Krypton, Xenon and Iodine and the metals Strontium and Cesium. Silver is also recognised particularly in the low enrichment system as an important product. Neither BISO nor TRISO coatings allow fission gas and Iodine release when properly fabricated. Fission Gas release, therefore, occurs only from failed fuel. Fission Product Gases of radiological significance have relatively short half-lives. Therefore, it can be deduced that only those short-lived atoms lying very near the surface of the Kernel diffuse before they decay.

In the release of these relatively short-lived isotopes such as Kr-88, Xe-133 and I-133, the decay process is dominant over the diffusion process. The fractional release of these products is of the order of 1 % for failed fuel under reactor conditions. Fig.4.6 presents the release of these important short-lived isotopes from a 100 % failed fuel.

Another important release process is the fission recoil. Several percent of the fission product release from the fuel Kernel is by fission recoil. Recoiled atoms are stopped in the surrounding solid material such as Carbon coatings from which they are again released by diffusion. Diffusional processes are, therefore, to be considered

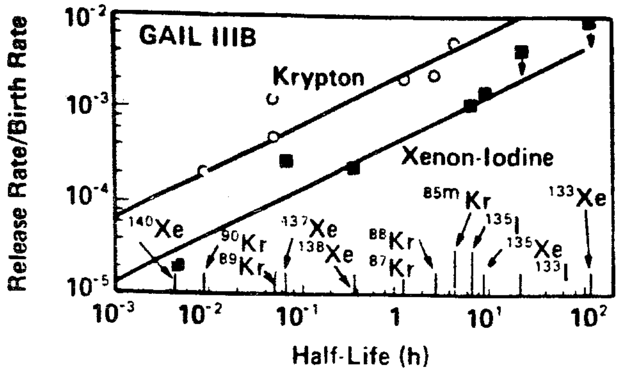


Fig.4.6. Release of short-lived isotopes

also for recoiled fission products.

The release of metallic fission products is more complex than that of the fission gases. The diffusional release through intact BISO coatings occurs at high temperatures. Even Ag-110m can diffuse through apparently intact TRISO coatings at high temperatures.

The radioactive decay and fission recoil processes are of less significance in the release of long-lived metallic isotopes such as Sr-90 and Cs-137. The diffusional release from the Kernel is apparently the dominant process.

The composition of the fuel Kernel plays an important role in the release of metallic as well as gaseous fission products. Strontium is gettered in oxide fuel since it forms a stable oxide which is strongly bound within the fuel kernel. This is not the case, however, with Cesium and Silver since they do not form stable oxides at reactor temperatures. These products are thus more mobile. Carbide Kernels have less retention capability for metallic fission products.

The diffusivity in PyC shows structural dependency such that HTI PyC is more retentive than LTI PyC. This can be understood because the properties of the PyC are dependent on its production process, as has been stated earlier.

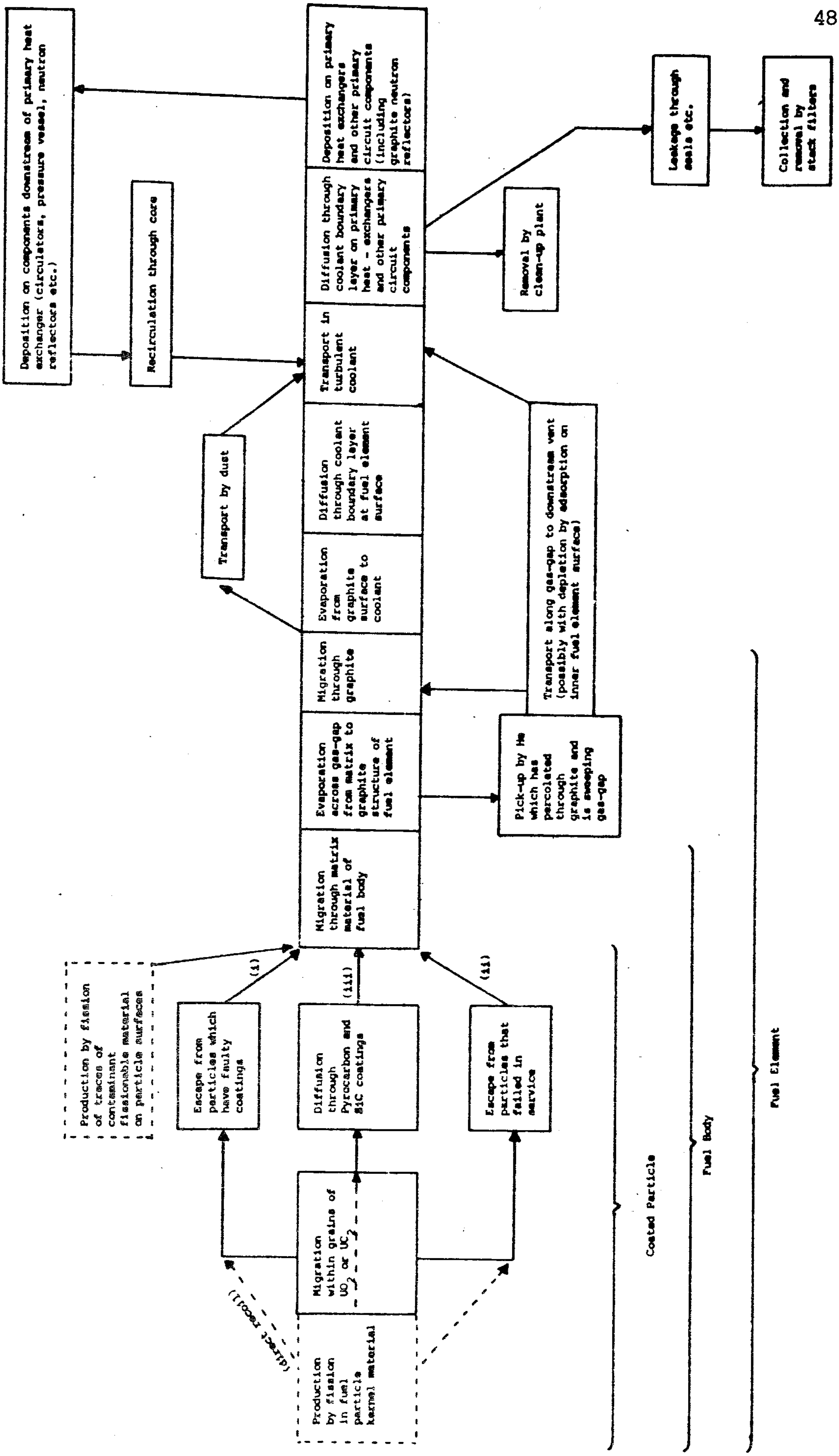


Fig.4.7.Pathways and Barriers for fission product release for an HTR

4.2.3. HTGR Designs with Prismatic Fuel Elements

HTGR designs in the US and UK utilise prismatic fuel elements. Interest in the Gas-Cooled Reactor programmes showed up with the successful operation of the DRAGON Project undertaken by OECD countries in UK. In the US, the Peach Bottom unit was operated as the prototype HTGR producing 40 MW_e of power. Relying on the experience gained over Peach Bottom, The first commercial steam-cycle HTGR, the Fort-St-Vrain, plant was built and has been operated since 1976.

FSV produces 330 MW_e of power and its measured efficiency is 39.2 %. It has several characteristics that make it unique in US reactor design and these features are summarised as follows :

Heat is generated in Carbide-based hexagonal fuel rods that contain ceramic coated (TRISO) particles and is transferred to compressed He at 700 psia. Graphite blocks are arranged in such a way that thermal growth differences are accomodated by sliding without disturbing the core geometry.

This plant is basically a converter and its fuel consists of a homogeneous mixture of fissile U-235 in full enrichment and fertile Th-232 particles. The prismatic fuel element consists of a machined hexagonal graphite block which is approximately 750 mm in length and 350 mm across the flats. Two sets of holes are drilled in the hexagonal array, one set for the Helium flow, the other for the stack of fuel rods with the fuel particles dispersed in a carbonaceous matrix. The graphite moderator is an integral part of the fuel element as it improves the neutron economy. Off-load re-fuelling is employed where 1/6 of the core is replaced each year in shutdown periods.

Steam generators are of Once-Through type and generate superheated main and re-heat steam at 538°C. Helium circulators are axial and driven by the steam turbines. One of the most important characteristics is the employment of a PCRV that contains the complete primary heat system with the Helium circulators and steam generators placed below the core. The PCRV has penetrations at the top through the core that are used for control rod movements and re-fuelling.

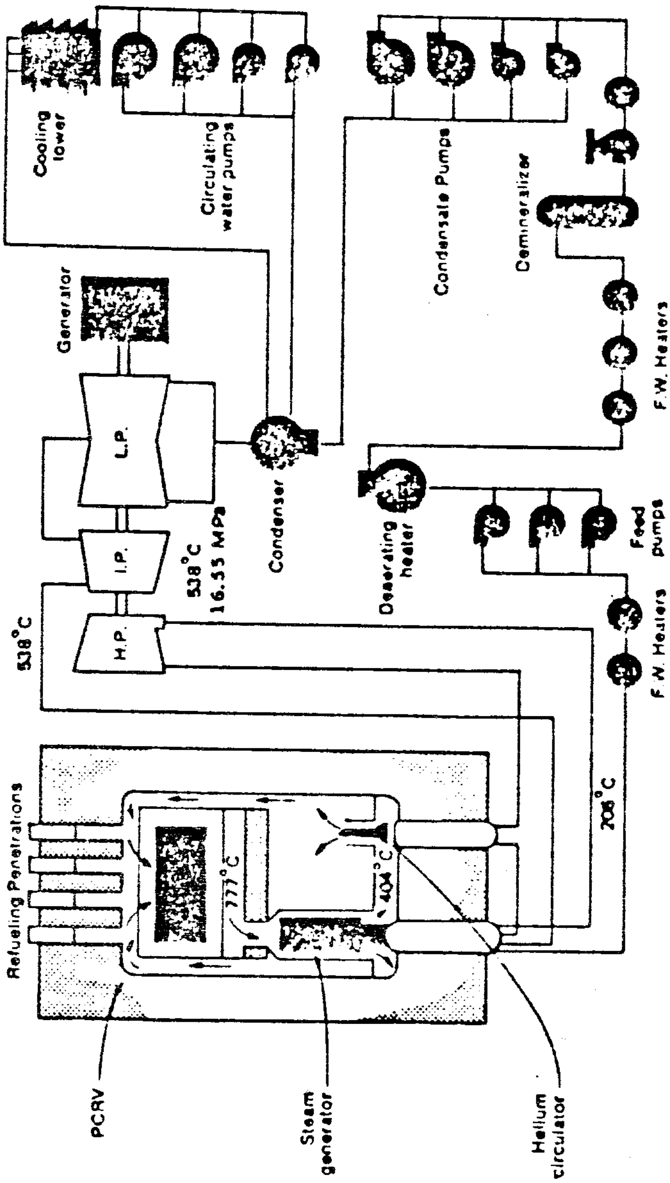


Fig.4.8. FSV Plant Layout

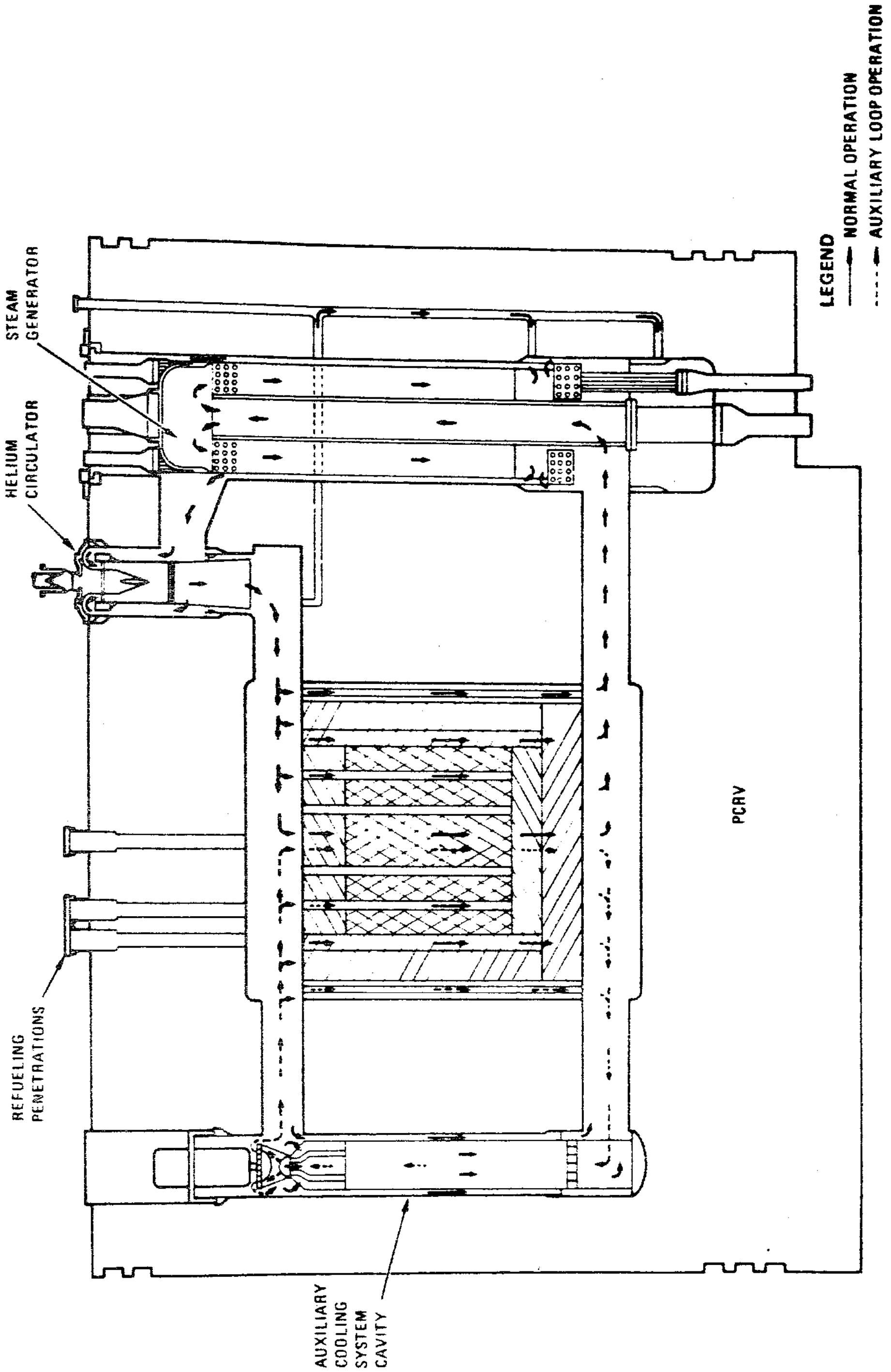
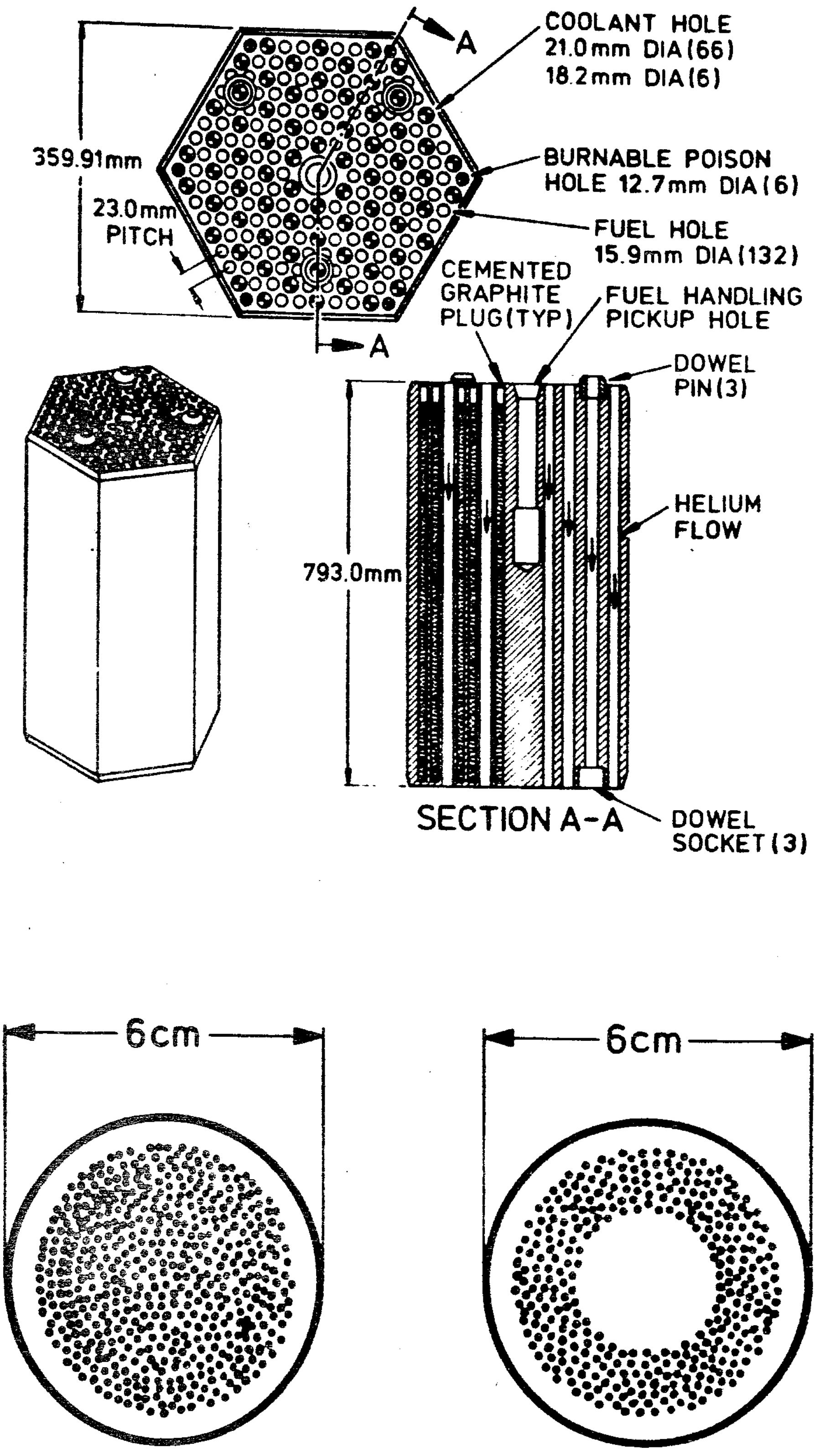


Fig.4.9. The PCRV and its interior



THTR-300 Element

Proposed 3-Zone Element for VHTR

Fig.4.10 Spherical and Block type fuel elements for HTGR designs

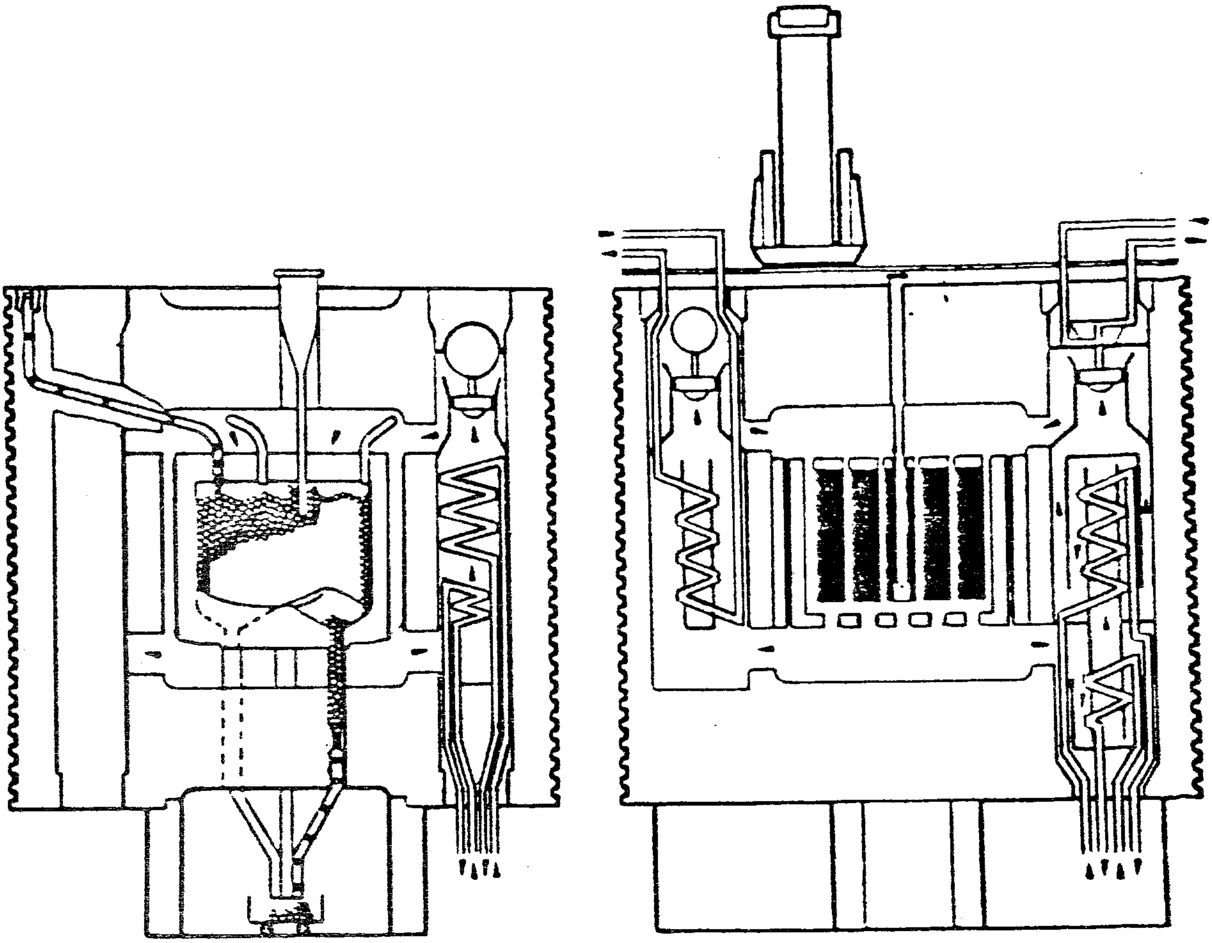


Fig.4.11. Basic Design differences in the Pebble-Bed and Block type cores

4.2.4. HTGR Designs with Spherical Fuel Elements

The HTGR development in West Germany started with the construction of the first Pebble-Bed reactor, AVR, Arbeitsgemeinschaft Versuchs-Reaktor. This plant has been in operation since 1967 contributing 15 MW_e of power to the electrical network.Extensive information about this design gathered through successful operation has led to the second HTGR, the THTR, the Thorium High Temperature Reactor. The construction of THTR started in 1971 in Schmehausen by Hamm. This plant is for commercial use and employs Thorium cycle operating at 300 MW_e since 1983. USA and People's Republic of China have also been involved in the development of the Pebble-Bed concept.

Pebble-Bed HTRs have a cavern in which fuel spheres are placed. This cavern is surrounded with graphite of 1 m in thickness. The core is cooled by He. The fuel spheres are continually changed by removing spent ones through an exit at the bottom and by adding fresh fuel spheres from the top conforming to the On-load refuelling scheme. For the controlled and temporary shutdown of the reactor, control rods through the lateral graphite block are used. For long-term shutdowns, the control rods above the core are driven in. A PCRV which can withstand 39 bars accomodates the whole assembly. The fuel pebbles are of graphite 6 cm in diametre. Approximately 40,000 coated particles, each measuring 1/10 of a mm are dispersed in each graphite sphere. Coated particles consist of oxides or carbides of Uranium and Thorium. Uranium is highly enriched but in more modern designs, enriched to 8 %.
Both BISO and TRISO particles are in use.

The characteristics attributed to HTGR associated with the use of all ceramic core and He coolant are again advantages of this system. The employment of spherical fuel elements and hence On-load re-fuelling capability avoids complete shutdown for fuel exchange and increases plant availability. The coated particle concept results in higher burn-up and efficient utilisation of fuel. The small size of fuel elements and their loading scheme introduce innovative design when the dimensions of the reactor are concerned. A large plant may employ one large reactor with several sphere loading and removal means or several small size reactors installed in a modular fashion and operated separately.

In the first two Pebble-Bed reactors, the AVR and the THTR, the fuel pebbles run through the core as many times as needed to reach the pre-dicted burn-up. The removed pebbles are tested on the exit path through a measuring system and the oldest ones are taken out from the cycle, directed to a dispose unit and fresh pebbles are added from the top. This scheme guarantees a mixture of fresh and old elements and a uniform power density distribution in the core volume which is achieved through placement strategies in other reactors.

In recent concepts, OTTO, Once-Through-Then-Out, loading is developed in which the pebbles run through the core very slowly and only once. The fresh fuel elements are placed at the upper portions of the core and the older ones towards bottom. The Helium flow is accordingly arranged in a direction from top to bottom so that local temperature increases are avoided. Fig 4.12 displays the power density and the temperature distribution in the OTTO-loading. The difference between the fuel element temperature and He exit temperature is less than 50°C in favour of the element. The OTTO loading, therefore, allows a higher heating of the He without exceeding the upper limit of the fuel elements' thermal withstanding.

An important problem associated with the OTTO-loading scheme is the achievement of a uniform fuel element flow through the core. The AVR core design with one exit introduces radial dependency to the

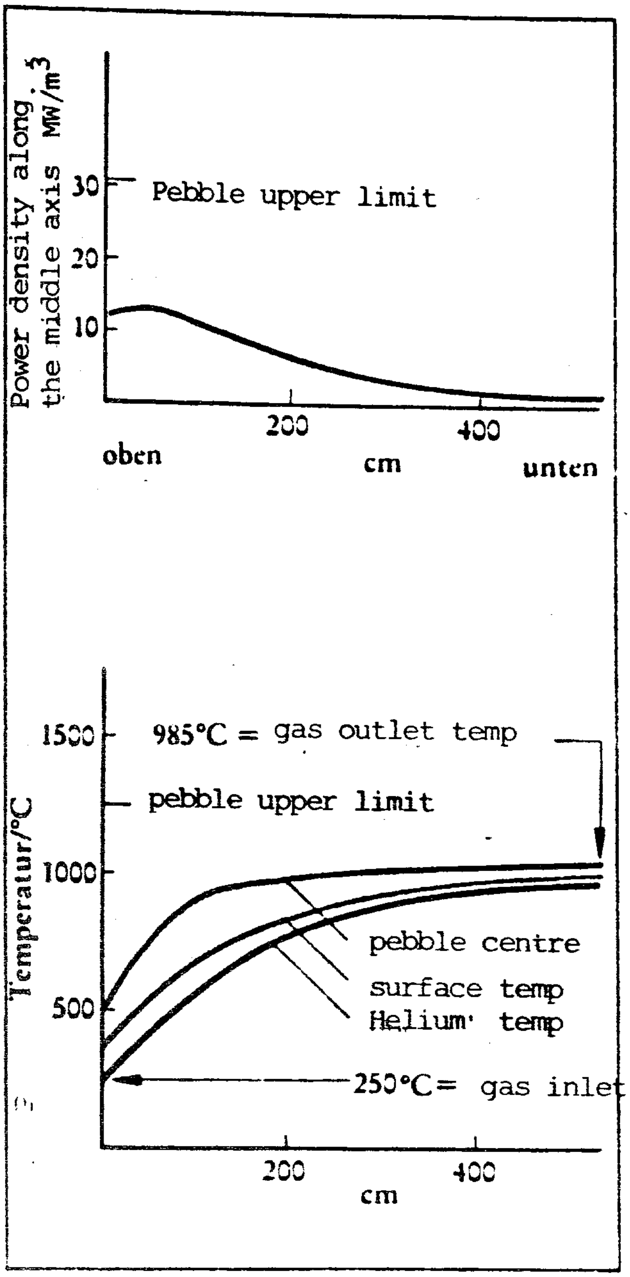


Fig.4.12 Power density and temperature distribution in OTTO loading

pebble flow as shown in Fig.4.13.

In larger units employing OTTO-loading scheme, particularly in those intended for high temperature industrial applications, this radial dependency would not be tolerated since it would tend to radially dependent burn-up and in turn, radially dependent power release. Uniform pebble flow further brings the advantages of flattenning the temperature profile of the outlet gas, lowest possible fission product release and achievement of a high conversion ratio. As a solution to this problem, increasing the number of exits has been proposed. The effect of several exits on the pebble flow is presented in Fig.4.14. Another design proposal is the pebble flow distributor which is shown in Fig.4.15.

The insertion of the control rod presents a problem which is peculiar to the Pebble-Bed concept only. The Pebble-Bed acts like a highly dense cloud and control rod insertion in large reactors with large core will require a great force. In order to circumvent this difficulty, a helical absorber is designed as is shown in Fig.4.16.

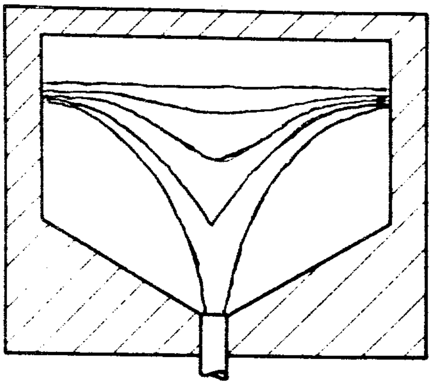


Fig.4.13 Pebble Flow in a core with one exit

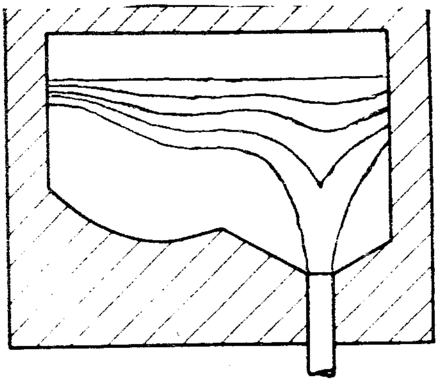
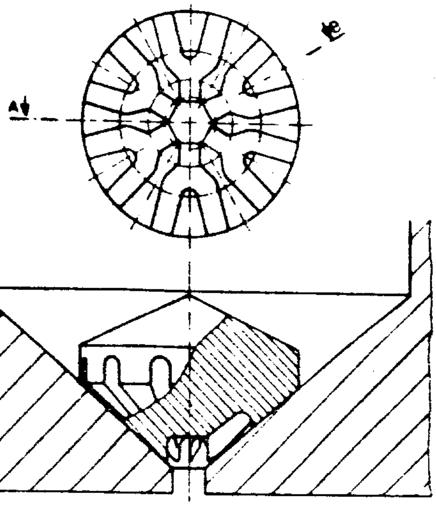


Fig4.14 Pebble flow in a core with several exits



CUT A-B

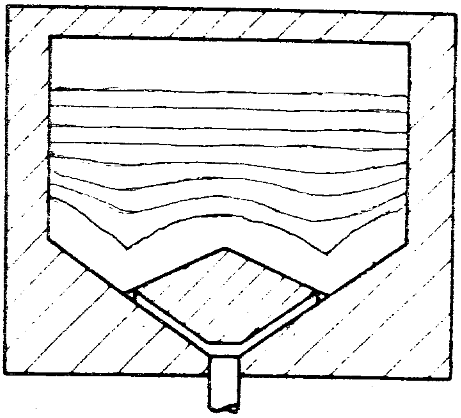


Fig.4.15 Pebble Flow Distributor and its Effect on Flow Profile

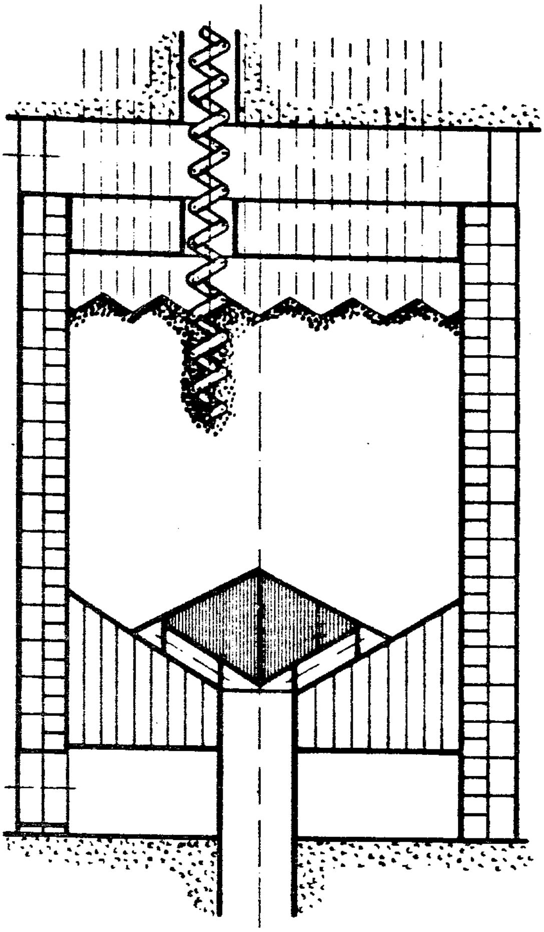


Fig.4.16 Helical Absorber Designed for Pebble-Bed core

Table 4.2. Technical Data about the AVR

Electrical Output	15 MW
Electricity Requirement	1.65 MW
Thermal Power	46 MW
Power Density	2.6 MW/m^3
Diametre	3 m
Height	2.8 m
Number of Fuel Elements	92,000
Helium Pressure	10.8 bars
Helium Temperature	
Inlet	275°C
Outlet	950°C
Number of Absorber Rods	4
Fuel Element Diametre	6 cm
Fuel	U and Th in Carbide or Oxide form as Coated Particles
U-235 Content	1 g
Enrichment	10 % ; 16.7 % ; 93 %
Thorium Content	0 ; 5 ; 10 g
Max Fuel Temperature	1230°C
Max Fuel Element Surface Temperature	1170°C

Table 4.2. Cont'd :

Steam Generator Inlet Temperature	115°C	
Temperature at Re-heater Outlet	505°C	
Steam Flow Rate	56 t/h	
Number of Pumps	2	
Number of Revolutions	400-4400 min^{-1}	
Gas Flow Rate	13 kg/s	
Pressure Vessel	Inner	Outer
Diametre	5.78 m	7.6 m
Thickness of the inner vessel	40 mm	30 mm
Height of the inner vessel	25 m	26 m
Design Pressure	12.3 bars	11.8 bars
Material	Steel	Steel

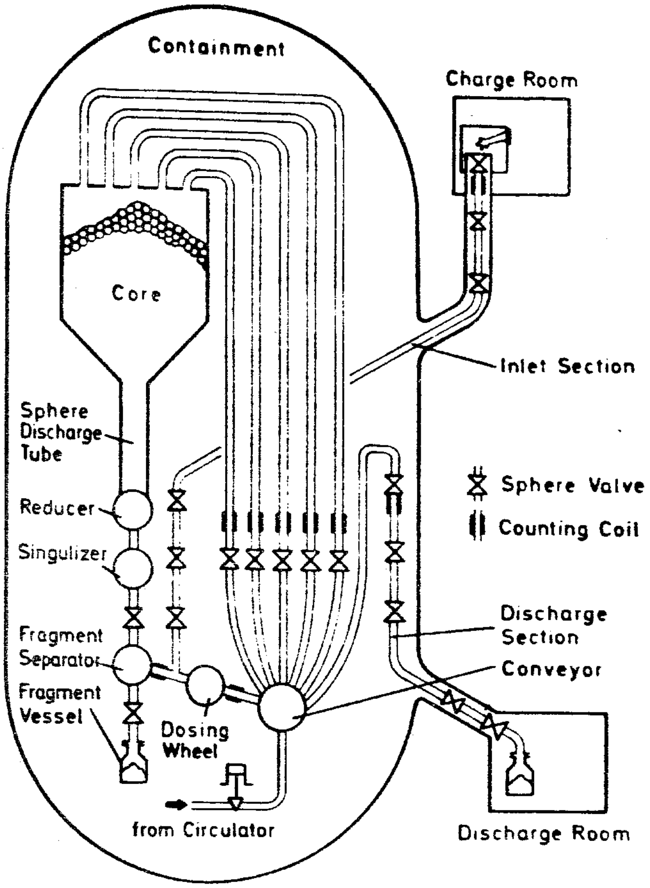
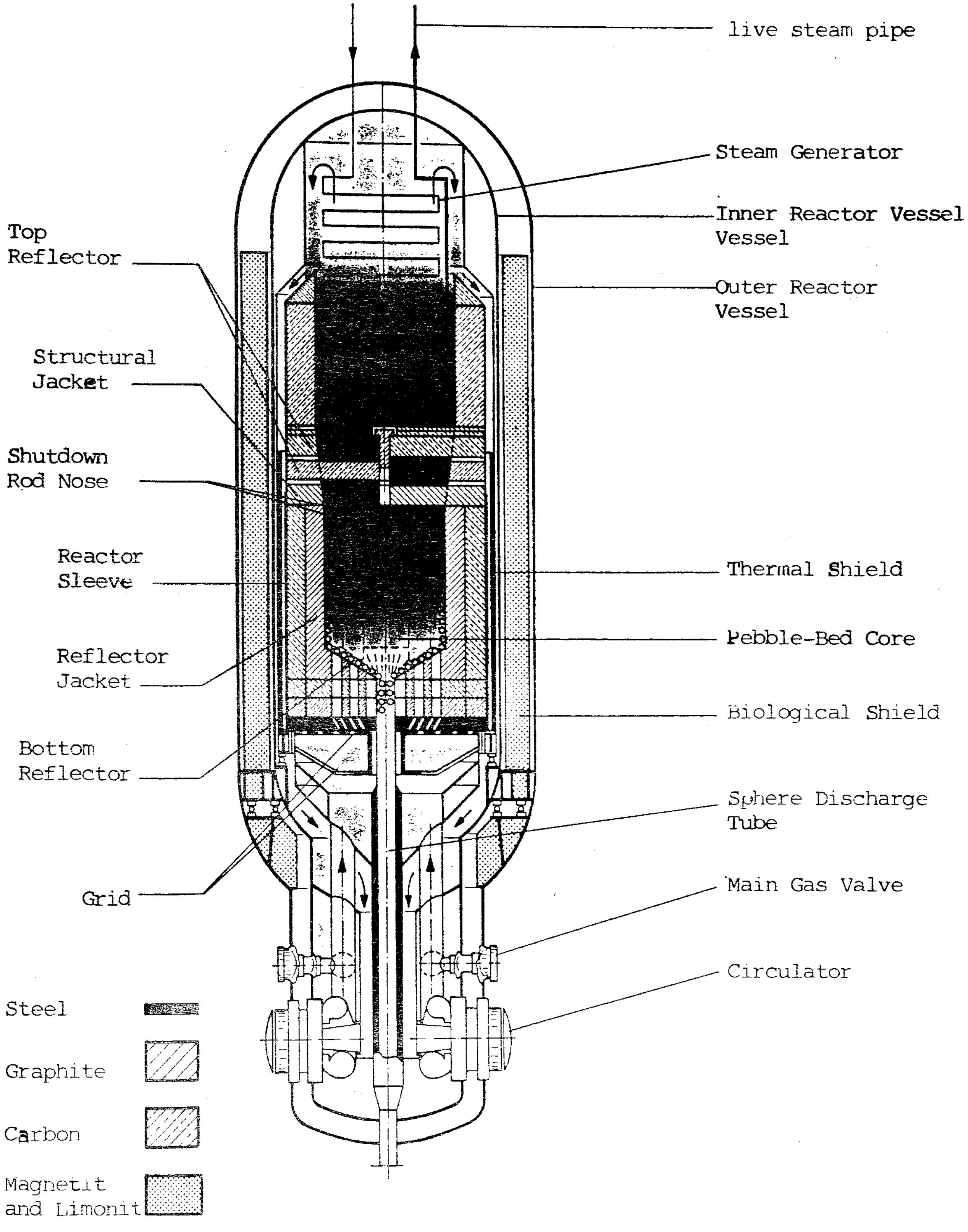


Fig.4.17 AVR Fuelling System

Fig.4.18 Vertical Cross-Section of AVR



4.2.5 Small Size Modular HTGR Designs

Large plants benefit from the economy of scale when evaluated in connection with the complexities in building and operation and the satisfaction of regulatory requirements.

In recent years, a new concept to give an economical answer to the demand in the fields of both electricity generation and process energy applications is being developed particularly in the USA and FRG. This concept involves modular plants with small unit size and is based on shop-fabricated standardised reactor modules optimising the BOP, Balance Of Plant, design which comprises the greater portion of the total plant cost with 60-80 % and the NSSS, Nuclear Steam Supply System, design which comprises 20-40 % of the total cost. The modular expansion capability of the system also answers the increase in the energy demand with time.

This NSSS package is relatively simple and offers the advantages of reduced shielding requirements, simple site installation and shorter construction schedule. Because of these advantages, this new HTGR design benefits from siting flexibility and reduced licensing difficulty.
The initial plant investment is also lower and this approach contributes significantly to the reduction of plant capital cost and construction time.

The NSSS package is embedded in a steel vessel. Therefore, heat dissipation by conduction through the steel vessel is enabled and the decay heat can be removed passively. Furthermore, the power density is very low and the reactor does not reach such high temperatures where significant release of fission products takes place. The reactor is, therefore, not enclosed in a pressure containment and this reduces

Table.4.3. Technical Data about small size modular HTGR

o Plant rating, nominal	197 MWe
o Plant efficiency, net	38.5%
o Number of NSSS modules	2
o Number of turbine-generators	1
o NSSS module characteristics	
- Thermal rating, nominal	256 MWt
- Average power density, nominal	4.2 w/cc
- Fuel material	low (< 10%) enriched uranium
- Fuel type	pebble
- Fuel management	recirculation
- Refueling	on-line
- Reactor coolant	helium
- Core inlet temperature, nominal	260°C (500°F)
- Core inlet pressure, nominal	70 bar (1000 psia)
- Core outlet temperature, nominal	730°C (1350°F)
- Steam generator inlet temperature, nominal	700°C (1290°F)
- Steam temperature, nominal	530°C (990°F)
- Steam pressure, nominal	190 bar (2800 psig)
- Reactivity control	with reflector rods for power level and scram; long term shutdown with absorber spheres
- Configuration	core and heat transport equipment integrated within steel pressure vessel, vertical in-line
o BOP characteristics	
- Fission product release control	filtered confinement
- Emergency decay heat removal	reactor enclosure wall cooling panels
- Reactor enclosure embedment	fully embedded
- Plant control	single control room
- Auxiliary power	2 gas turbines
- Construction approach	separated nuclear-grade from conventional field construction

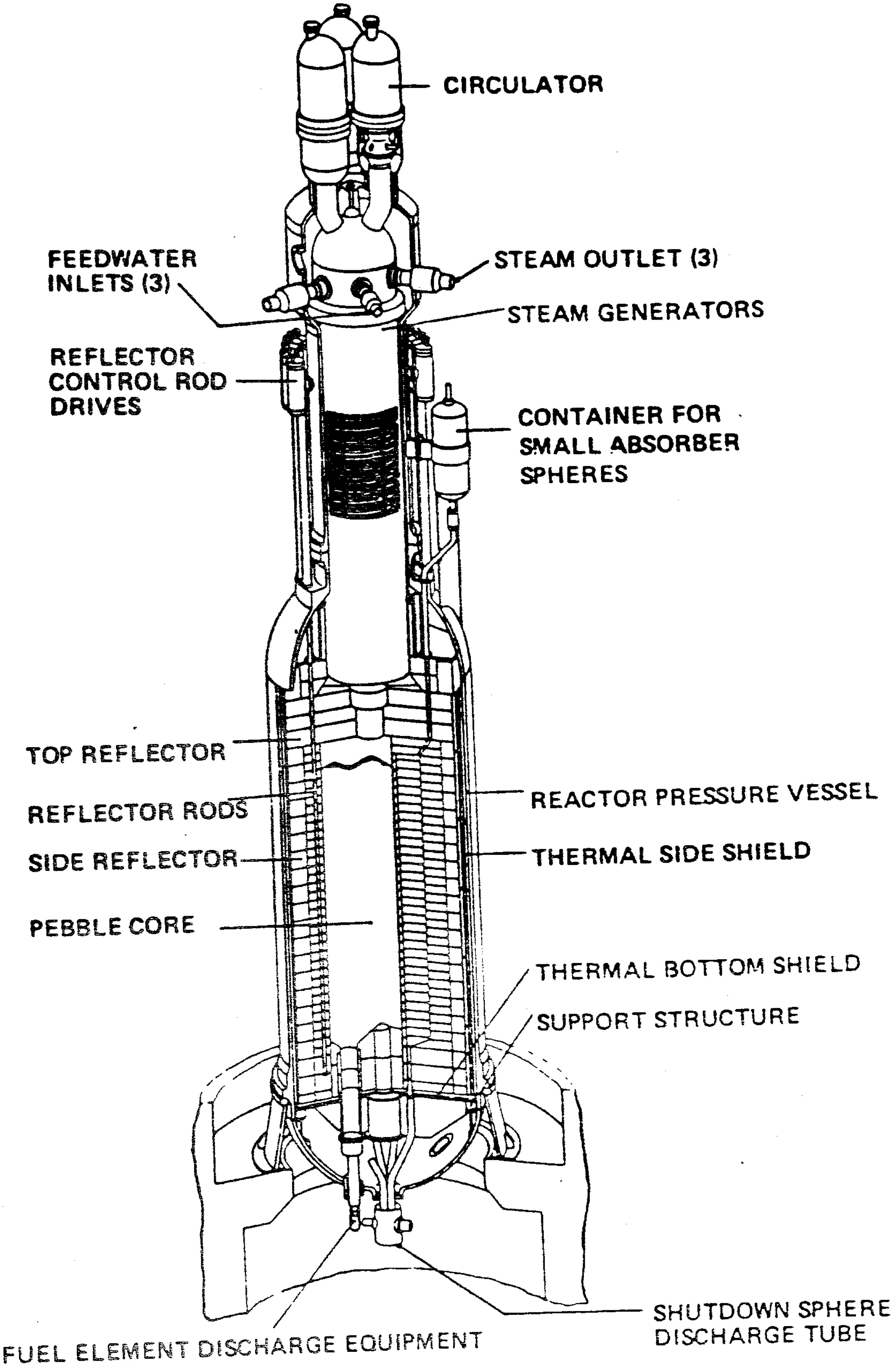


Fig.4.19 Small Size Modular HTGR cross-section

the complexity of the BOP design significantly.

The BOP design is further simplified by the Nuclear Island, NI, which contains the NSSS package and the associated safety-related portions of the plant. The NI introduces the separated construction approach in which the non-safety-related BOP portions such as the turbine generator facility are separated from the safety-related portions of the plant. This is reflected in low BOP costs because those parts outside the NI can be constructed according to the same standards as a conventional power plant (19).

4.2.6 The HTGR and Process Heat Applications

The HTGR provides high outlet gas temperatures that can be utilised in process heat applications. The Very High Temperature Reactor, under active development in Japan, the FRG and the US, is intended for this purpose.

Process heat or steam is largely consumed in industrial processes and the energy from nuclear fission needs to be applied directly in order to replace the primary fuels such as natural gas, coal or petroleum.

A significant fraction of these processes require that heat be applied at high temperatures and the heat of nuclear origin can be used in such processes to synthesise hydrogen which is either consumed in the process or can be made available as a secondary fuel. These processes involve the direct reduction of iron ore, the manufacturing of synthetic fuels from coal and water. If the widely employed steam-methane reforming process is used to synthesise the hydrogen, peak process temperatures in the range of 750°C to 850°C are required. The direct synthesis of hydrogen by thermochemical splitting of water will require process temperatures up to and beyond 1100°C.

In the thermochemical pipeline which is used to transport energy, a gaseous mixture of CO and Hydrogen is obtained by steam reforming of Methane and this mixture is transported to the consumer site where the Methane is synthesised from the gaseous mixture. This exothermic process gives the applied reactor heat back at temperatures of approximately 500°C to 600°C which can be used in district heating or in electricity generation.

The chemical and petroleum refining industries are the most promising application fields because of the large quantities of thermal energy required.

5. A POSSIBLE NUCLEAR STRATEGY USING THORIUM

Thorium has been introduced as fertile species before the introduction of High Temperature Reactor concepts and it has been used in converter Light Water Reactors, though not extensively. The primary reason is that U-238 is abundantly available in Uranium-fuelled reactors and the bred Pu-239 finds fields of application more readily.

The Thorium fuel is, however, advantageous over Uranium in that the bred fissile species U-233 is more suitably used in thermal reactors than Pu-239 because of its neutronic properties such as the higher neutron yield at thermal energies. Profitable utilisation of Thorium, therefore, implies recycling of U-233, namely, the closure of the Thorium fuel cycle.

The use of Graphite as structural core material and Helium as coolant in High Temperature Reactors result in good neutron economy and when Thorium fuel cycle is employed, the possibility of thermal breeding shows up.

Without U-233 make-up, the high conversion is hardly achieved. During the first years of operation a low burn-up should be adopted while the neutron economy could be improved by frequent re-fuelling. A further complication arises with the necessity of changing the moderation ratio as high conversion cycle is approached. Such a strategy is better suited to a concept which employs On-load Re-fuelling than an Off-load fuelled concept.

In the following section, a Thorium fuelling strategy is discussed in connection with the Pebble-Bed HTR concept which characteristically employs On-line Re-fuelling.

5.1. Pre-Breeder/Near-Breeder Schemes

An optimisation in fuel economics and resource conservation is achieved if the fuel is completely separable. Therefore, development in the SFB scheme, in which the breed and make-up material are inserted into separate fuel elements, helps in recycling and the Prebreeder and Nearbreeder schemes are devised.

The Pre-Breeder scheme is realised when the U-233 that is bred in an SFB scheme is not recycled but removed from the cycle for more profitable utilisation in the Near-Breeder scheme.

The Near-Breeder scheme is analogous to the MOR scheme. The U-233 that is bred in the Pre-Breeder is used as start-up fuel and as make-up. It offers high conversion ratio with increased coated particle loading and reduced Burn-up.

An explanation of the influence that Heavy Metal loading leaves needs to be made at this point. A high conversion rate is achieved through high Heavy Metal loading because it lowers the Moderation Ratio, N_C/N_{HM}, in which case parasitic neutron loss is reduced. By hardening the neutron spectrum, one achieves a reduced $\eta = \nu\sigma_f/\sigma_a$ value of U-235 whereas the η value of U-233 is not very sensitive as can be seen in Table 3.1. It is for this reason that the Near-Breeder uses U-233 as first core inventory and as make-up which has been produced in a Pre-Breeder with a softer neutron spectrum. Another possible way for U-233 accumulation is the conversion in an FBR.

As can be seen from Table 3.1, U-233/Th fuel cycle gives preference to the lower Moderation Ratio, ie, $N_C/N_{HM} = 110$, whereas the conventional U-233/Th fuel cycle uses a Moderation Ratio in the range between 180 and 250 since the effect of low Moderation Ratio, ie, high Heavy Metal loading, is offset by the poorer values of η and α .

As the conversion rate is below one, a certain amount of highly enriched Uranium is needed as make-up in the Pre-Breeder which is optimised for maximum U-233 discharge rate. In the reference design of Fig.5.1, the Pre-Breeder is fed with three types of fuel elements at different percentages :

36 % of total charge comprises fresh feed fuel with ThO_2. 14 % representing the second type is the recycle feed fuel. It requires remote fabrication techniques. The remaining 50 % of fuel is covered by the breed fuel balls which contain pure ThO_2. The breed fuel elements contain a relatively high Heavy Metal loading to achieve the high conversion rate.

The fuelling charges of the Near-Breeder consist of two different types of fuel elements. After disloading, they are reprocessed together. The recovered Uranium is mixed with fresh Thorium and then recycled. They can be concentrated into 25 % of the total number of fuel balls. The remaining 75 % is made of Thorium breed fuel. The average burn-up of these Thorium balls is approximately 7100 MWD/T and one layer of coating only is sufficient at this low burn-up.

The connecting link between the Pre-Breeder and Near-Breeder cycles is the balance of the U-233 isotope. With the reference design in Fig.5.1, the net discharge rate of fissile Uranium is 564 g/GWD and the net consumption of the Near-Breeder is 41 g/GWD. As a result, one Pre-Breeder is able to produce the make-up of 10.3 Near-Breeders with 1 % loss in the outer circuits. A total of 8.5 full-power operation years of the Pre-Breeder produces the first core loading and 1.5 year disloading at 0.7 capacity of a Near-Breeder (17,18).

A conventional HTR can switch over from Pre-Breeder to Near-Breeder in a period of two years. Fig.5.2 displays the life periods which the reactor undergoes. Equilibrium inventory of conventional operation is achieved within the first three years. According to Fig.5.2,a, the Switch-over period is achieved after 11 years of operation during which the sufficient amount of bred U-233 is accumulated.

Fig.5.2.b presents the supply and disloading rates. Recovered Uranium is re-inserted into the core as recycled fuel. The difference between the supply and disloading is removed by fresh fuel. This is the net consumption in the equilibrium period that follows the first three years of operation.

The breed material,almost pure U-233, is removed from the cycle and reserved in a storage. In the Switch-Over period, breed supply replaces the feed supply and breed disloading increases as feed disloading is ceased. In the equilibrium Near-Breeder phase, the difference between breed supply and breed disloading is covered by the reserve fuel from the storage.

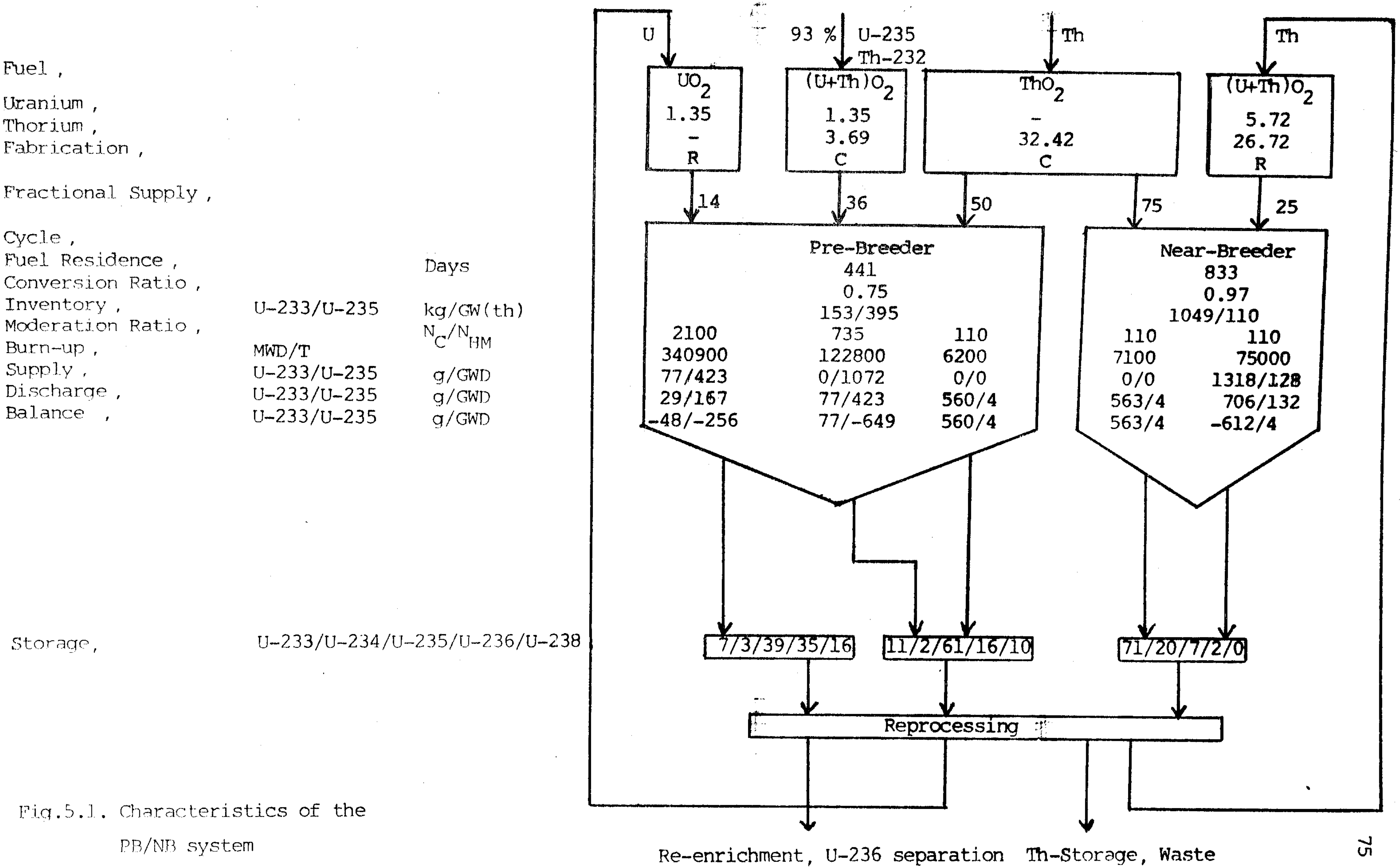


Fig.5.1. Characteristics of the PB/NB system

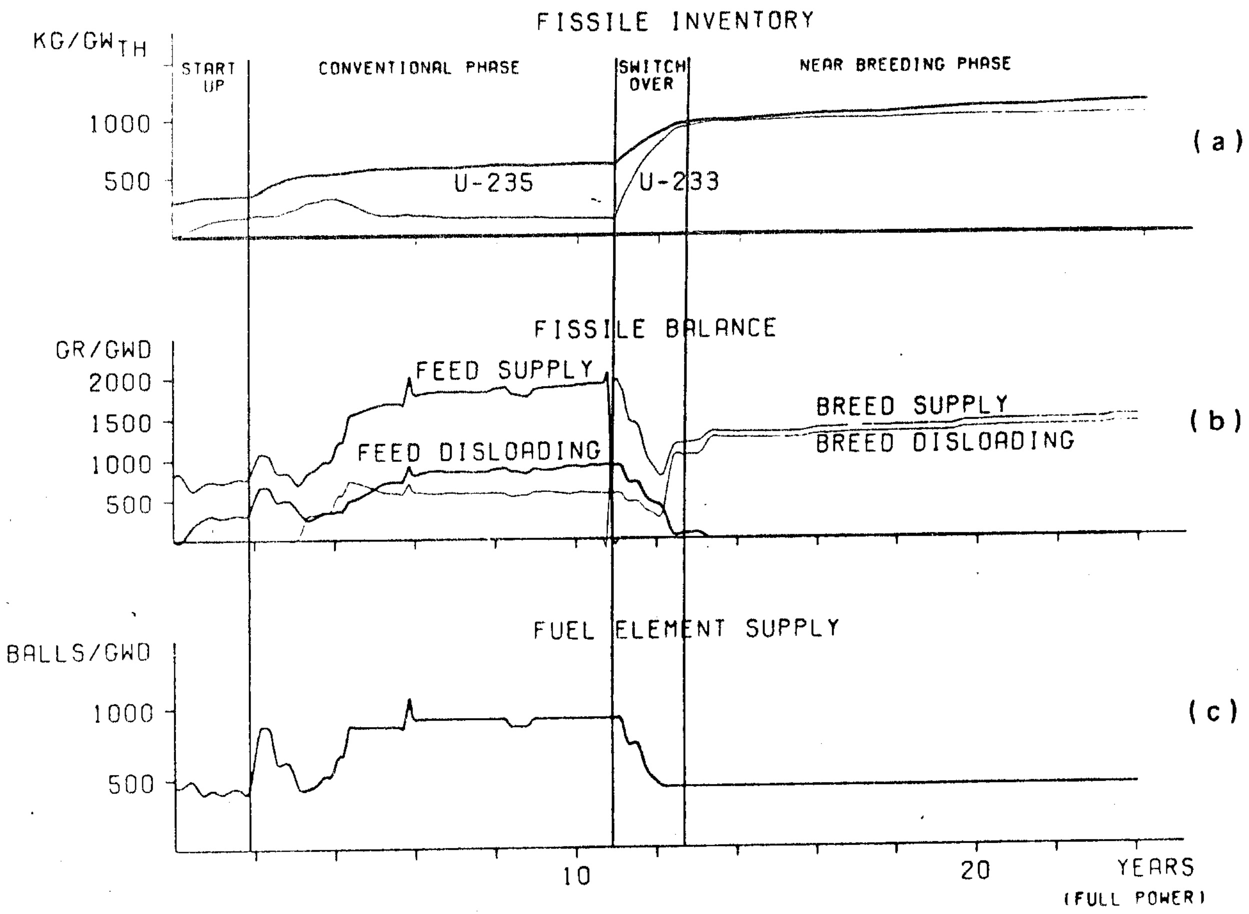


Fig.5.2 Fuel Management in the conversion from PB to NB

5.2 Analytical Study of the Pre-Breeder Scheme

In this section, the Pre-Breeder Scheme is examined analytically and the effect of the long exposure on the breed elements is discussed.

The following are the reactions involved in the production of U-233 :

$${}_{90}\mathrm{Th}^{232} + {}_{0}\mathrm{n}^{1} \longrightarrow {}_{90}\mathrm{Th}^{233} + \text{Gamma}$$

$${}_{90}\mathrm{Th}^{233} \longrightarrow {}_{91}\mathrm{Pa}^{233} + \text{Beta}^{-} \quad \text{in 22.1 mins}$$

$${}_{91}\mathrm{Pa}^{233} \longrightarrow {}_{92}\mathrm{U}^{233} + \text{Beta}^{-} \quad \text{in 27.4 days}$$

The flux is assumed to be constant and the breed spheres that contain Th only are assumed to reside for 441 days in the core.

$$\frac{d}{dt} N(\text{Th-232}) = - \Sigma_a(\text{Th-232})\ \phi = - N_o(\text{Th-232})\ \sigma_a\ \phi \longrightarrow N(\text{Th-232}) = N_o(\text{Th-232}) \exp\left[- \sigma_a(\text{Th-232})\ \phi\ t \right] \quad (5.1$$

$$\frac{d}{dt} N(\text{Th-233}) = \Sigma_a(\text{Th-232})\ \phi - \lambda(\text{Th-233})\ N(\text{Th-233}) \quad (5.2)$$

$$= \sigma_a(\text{Th-232})\ \phi\ N_o(\text{Th-232}) \exp\left[- \sigma_a(\text{Th-232})\ \phi\ t \right] - \lambda(\text{Th-233})\ N(\text{Th-233}) \quad (5.3)$$

which yields $N(\text{Th-233}) = A * \left(\exp\left[- \sigma_a(\text{Th-232})\ \phi\ t \right] - \exp\left[-\lambda(\text{Th-233})\ t \right] \right)$ (5.4)

where $N_o(\text{Th-233}) = 0$ at $t = 0$ and $A = \dfrac{\sigma_a(\text{Th-232})\ \phi}{(\ \lambda(\text{Th-233}) - \sigma_a(\text{Th-232})\ \phi\ )} N_o(\text{Th-232})$ (5.5)

$$\frac{d}{dt} N(\text{Pa-233}) = \lambda(\text{Th-233})\ N(\text{Th-233}) - \lambda(\text{Pa-233})\ N(\text{Pa-233}) - \Sigma_a(\text{Pa-233})\ \phi \tag{5.6}$$

$$\frac{d}{dt} N(\text{Pa-233}) + (\ \lambda(\text{Pa-233}) + \sigma_a(\text{Pa-233})\ \phi\ )\ N(\text{Pa-233}) = \lambda(\text{Th-233})\ N(\text{Th-233}) \tag{5.7}$$

$$d(\ N(\text{Pa-233}) \exp\left[B\ t\right]\ ) = \lambda(\text{Th-233})\ A\ (\ \exp\left[(B - \sigma_a(\text{Th-232})\ \phi)t\right] - \exp\left[(B - \lambda(\text{Th-233}))t\right])dt \tag{5.8}$$

which yields

$$N(\text{Pa-233}) = \lambda(\text{Th-233})\ A \left[\frac{\exp\left[-\sigma_a(\text{Th-232})\phi t\right]}{(B - \sigma_a(\text{Th-232})\phi)} - \frac{\exp\left[-\lambda(\text{Th-233})t\right]}{(B - \lambda(\text{Th-233}))} - \left[\frac{1}{(B-\sigma_a(\text{Th-232})\phi)} - \frac{1}{(B-\lambda(\text{Th-233}))}\right]\exp\left[-Bt\right]\right] \tag{5.9}$$

where $N_o(\text{Pa-233}) = 0$ at $t = 0$ and $B = \lambda(\text{Pa-233}) + \sigma_a(\text{Pa-233})\phi$ (5.10)

$$\frac{d}{dt} N(U\text{-}233) = \lambda(Pa\text{-}233)\ N(Pa\text{-}233) - \lambda(U\text{-}233)\ N(U\text{-}233) - \Sigma_a(U\text{-}233)\ \phi \qquad (5.11)$$

The term $(\lambda(U\text{-}233)\ N(U\text{-}233))$ in Equ.5.11 is negligible since $t_{1/2} = 1.62 \ast 10^5$ years for U-233.

then Equ.5.11 yields

$$N(U\text{-}233) = \lambda(Pa\text{-}233)\ \lambda(Th\text{-}233)\ A \left[\frac{\exp\left[-\sigma_a(Th\text{-}232)\phi t\right]}{(B-\sigma_a(Th\text{-}232)\phi)(\sigma_a(U\text{-}233) - \sigma_a(Th\text{-}232))\phi} - \frac{\exp\left[-\lambda(Th\text{-}233)t\right]}{(B-\lambda(Th\text{-}233))(\sigma_a(U\text{-}233)\phi-\lambda(Th\text{-}233))} - \left[\frac{1}{(B-\sigma_a(Th\text{-}232)\phi)} - \frac{1}{(B-\lambda(Th\text{-}233))}\right] \frac{\exp\left[-Bt\right]}{(\sigma_a(U\text{-}233)\phi-B)} \right]$$

$$+ C \exp\left[-\sigma_a(U\text{-}233)\phi t\right] \qquad (5.12)$$

where $N_o(U\text{-}233) = 0$ at $t = 0$ and C is given as (5.13)

$$C = \lambda(\text{Pa-233})\ \lambda(\text{Th-233})\ A\left[\frac{-1}{(B-\sigma_a(\text{Th-232})\phi)(\sigma_a(\text{U-233}) - \sigma_a(\text{Th-232}))\phi} + \frac{1}{(B-\lambda(\text{Th-233}))(\sigma_a(\text{U-233})\phi - \lambda(\text{Th-233}))}\right.$$

$$\left. + \left[\frac{1}{(B-\sigma_a(\text{Th-232})\phi)} - \frac{1}{(B-\lambda(\text{Th-233}))}\right]\frac{1}{(\sigma_a(\text{U-233})\phi - B)}\right] \qquad (5.13)$$

Evaluation of N(U-233) at $t = 441 * 86400$ seconds :

$$\text{Burnup} = 6200 \text{ MWD/T} = 6200 * 86400 \text{ Wsec/g}$$

$$= 6200 * 86400 * 3.1 * 10^{10} \text{ fissions/g} \qquad (5.14)$$

where 1 Wsec of energy is released by $3.1 * 10^{10}$ fissions

$$\text{Burnup rate} = \frac{6200 * 3.1 * 10^{10}}{441} \text{ fissions / sec.g}$$

$$= 4.35 * 10^{11} \text{ fissions / sec.g} \qquad (5.15)$$

$$\Sigma_f(\text{U-235})\phi = 10.5 * \frac{0.602 * 10^{24}}{235} * 582 * 10^{-24}\ \phi = 4.35 * 10^{11}$$

$$\phi = 2.78 * 10^{10} \text{ neutrons / sec.cm}^2 \qquad (5.16)$$

$$\lambda(\text{Pa-233}) = 0.29 * 10^{-6} \text{ sec}^{-1} \qquad (5.17)$$

$$\lambda(\text{Th-233}) = 0.522 * 10^{-3} \text{ sec}^{-1} \qquad (5.18)$$

$$\sigma_a(\text{Pa-233}) = 43 * 10^{-24} \text{ cm}^2 \qquad (5.19)$$

$$\sigma_a(\text{Th-232}) = 7.56 * 10^{-24} \text{ cm}^2 \qquad (5.20)$$

$$\sigma_a(\text{U-233}) = 576 * 10^{-24} \text{ cm}^2 \qquad (5.21)$$

$$A = \frac{\sigma_a(\text{Th-232})\phi}{(\lambda(\text{Th-232}) + \sigma_a(\text{Th-232})\phi)} N_o(\text{Th-232}) = 4.023 * 10^{-10} N_o(\text{Th-232}) \qquad (5.21)$$

$$B = \lambda(\text{Pa-233}) + \sigma_a(\text{Pa-233})\phi = 0.29 * 10^{-6} \qquad (5.22)$$

$$\exp\left[-\sigma_a(\text{Th-232})\phi t\right] = 0.999 \qquad (5.23)$$

$$B-\sigma_a(\text{Th-232})\emptyset = 0.29 * 10^{-6} \tag{5.24}$$

$$(\sigma_a(\text{U-233})-\sigma_a(\text{Th-232}))\emptyset = 1.58 * 10^{-11} \tag{5.25}$$

$$\exp\left[-\lambda(\text{Th-233})t\right] = 0 \tag{5.26}$$

$$B-\lambda(\text{Th-233}) = -0.522 * 10^{-3} \tag{5.27}$$

$$\sigma_a(\text{U-233})\emptyset-\lambda(\text{Th-233}) = -0.522 * 10^{-3} \tag{5.28}$$

$$\exp\left[-Bt\right] = 1.59 * 10^{-5} \tag{5.29}$$

$$\sigma_a(\text{U-233})-B = -0.29 * 10^{-6} \tag{5.30}$$

$$\exp\left[-\sigma_a(\text{U-233})\emptyset t\right] = 0.994 \tag{5.31}$$

With the numerical values from (5.14) through (5.31), one obtains

$$\frac{N(\text{U-233})}{N_o(\text{U-233})} = 6.56 * 10^{-5} \tag{5.32}$$

This result implies that the U-233 will be used up within the core during the residence time. This is reasonable so far as the large fission cross-section for U-233 is considered. The vanishing of the term (5.26) can be explained in the light of its short half-life of 22.1 min and the long half-life of Pa-233, 27.4 days into which Th-233 decays. This is the reason why fuel elements that contain fertile Th-233 for breeding purposes should be discharged after a short in-core time and accumulated in storage.

Decision must be made whether to remove the irradiated breed fuel elements or to recycle them with regard to the presence of the isotope U-232 the production of which is presented in Appendix A. Since the reactor medium contains a sufficient number of barriers against the associated Gamma-activity, its presence is problematic only in the storage and both the storage and reprocessing/refabrication media must accomodate the necessary protection and remote handling facilities.

6. OUTLOOK IN PRESENT HTGR TECHNOLOGY

The advantages offered by Thorium utilisation and especially the possibility of high conversion rates in thermal reactors combined with the features of He-cooled, Graphite moderated reactors such as high outlet gas temperatures, large safety margins are recognised and considerable efforts are being spent to overcome the drawbacks such as those in reprocessing and to introduce the concept for commercial use.

Relying upon the successful operation of the first three experimental/test reactors, the DRAGON, Peach Bottom and AVR, the latter still being in operation with 15 MW of electrical output, the commercial medium power prototype plants, the Fort-St-Vrain in the US and Thorium High Temperature Reactor in FRG have been built.

Other concepts that are being developed within the framework of HTGR are the HTGR with Gas Turbine, HTGR-GT and the Very High Temperature process heat Reactor, the VHTR. A He-cooled high temperature reactor operating with fast neutron spectrum is the Gas Cooled Fast breeder Reactor, GCFR.

The closure of the He gas-cycle in HTGR leads to HTGR-GT which is particularly useful where cooling water is limited. HTGR-GT requires dry cooling towers and offers higher thermal efficiency than HTGR. The primary intention behind HTGR-GT is elctrical power production, whereas low temperature process heat applications such as district heating can also be provided from the heat rejected from the turbines. although operating at higher pressure, it is based on HTGR technology, therefore its inroduction depends upon successful commercial operation of HTGR.

The VHTR is a logical extension of HTGR-GT and operates at higher temperature regimes. It is intended to supply high temperature heat to replace fossil fuel plants for process heat applications. The introduction of VHTR will result in fossil fuel conservation and reduced environmental, especially atmospheric, impacts.

GCFR is a high performance breeder to produce fissile fuel while producing power for electricity generation. Development work is being carried out in the USA, FRG and Switzerland involving primarily fuels and materials testing, heat transfer, plant and core design and shielding investigations. The GCFR benefits significantly from LMFBR fuel studies and HTGR component and materials tests. Its impact on resource utilisation is considerable reduction in mined ore requirements. This aim, however, can be achieved only if Thorium fuel recycling is realised.

Finally, the design of modular, shop-fabricated reactors is another field of application of HTGRs. In this design which is at present conceptual, Pebble-Bed core is adopted and the thermal output is 250 MW with approximate plant efficiency of 40 % .

7. DISCUSSIONS AND CONCLUSIONS

The general trend of industrialised countries is to reduce their reliance on the fossil fuel because of the limitedness of the resources. Amongst several alternatives such as solar, tidal or natural gas, the nuclear energy seems to be a promising choice in answering future energy demands and its contribution to the electrical networks is continually increasing while extensive Research and Development work is being carried out in technologically developed countries.

Today's nuclear technology is primarily based on U-235 utilisation in thermal reactors, with the LWRs being the most popular ones. However, world's Uranium ore deposits are also limited and shifting the energy source from fossil fuel to nuclear may not seem logical at first glance. Therefore, the nuclear fission can be a long-term solution only if optimal utilisation of known Uranium resources and breeding goals are achieved.

Optimal utilisation can be achieved by reducing mined ore requirements through either reprocessing spent fuel and thereby recovering fissile isotopes or using Thorium for fissile U-233 production. Another possibility is the conversion of U-238 into Pu-239.

Pu-239 is more suitable for use in a fast neutron spectrum and from another point of view, has strategic value as it is being used in nuclear weapons. Thorium and U-233 that is bred from it, on the other hand, are suitable for thermal reactors while the latter can also be used in fast reactors. That the natural abundance of Thorium is almost three times as much as that of Uranium, makes its utilisation for a long term possible. The adoption of Thorium, however, presents peculiar problems and considerable effort is being spent on the subject.

Thorium fuel cycle leads to a reduction by 25 to 30 % in Uranium ore requirements relative to LWR of comparable size.

Thorium fuel cycle steps are similar to those of Uranium-Plutonium cycle, but there are some problems that are not associated with the latter or that are of greater significance in the reprocessing and refabrication of Thorium fuel elements.

Of major importance is the U-232 isotope whose chain is presented in Appendix A. It leads to highly Gamma-active isotopes and this requires that the reprocessing of the spent fuel be made either immediately or after a sufficiently long cooling time.

More powerful dissolving agents such as nitric acid more concentrated than in Purex process, as well as a catalyst are required for Thorium fuel. The use of such agents, however, may complicate the following steps involving feed adjustment, solvent extraction and waste disposal. Thorium fuels are processed through a given size of equipment at a lower rate than processing Uranium fuels because of chemical properties of Thorium. These problems all tend to increase the cost of reprocessing.

The AVR experimental power station is being operated successfully with the operating temperature increased from 750°C to 850°C in 1972 and from 850°C to 950°C in 1974.This proven capability of high outlet gas temperature makes the concept unique amongst other reactor concepts and shows that the HTGR is potentially suitable for both electricity generation and industrial process heat applications. When dry cooling is employed, the direct use of nuclear heat will be attained and the HTGR can be used as nuclear heat source for applications such as thermochemical pipeline, thermochemical water splitting, for district heating or in steel industry.

The use of graphite as structural material and an inert gas, Helium, as coolant combined with the PCRV attributes to the HTGR concept a wide safety margin and hence offers good potential for adoption to very strict safety standards and licensing requirements.

Optimisations in fuel economics and in resource conservation follow when the bred U-233 that is generated in particles which contain Thorium can be separated from the neutron poison U-236 that is generated in the particles containing fissile U-235 in the Thorium fuel cycle. The achievement of completely separable Thorium cycle fuel and reduced fission product release are, therefore the objectives of current coated particle fuel development.

Gas Cooled Reactor concept operates under Brayton cycle and , therefore, offers higher efficiency than LWRs of comparable size. The amount of heat that is rejected from the cycle is less then the amount rejected in LWRs and this makes HTGR demand on cool water lower than LWR demand and results in less thermal pollution.

The introduction of modular, shop fasricated reactors of small unit size is dependent on commercial success of the Pebble-Bed concept and the evaluations of this concept show that plant capital costs can be lowered when serial production of them is started.

Gas cooled reactors seem to be promising in answering future energy demand. Except the GCFR concept, however, the other types operate in the thermal region of the neutron spectrum and when the optimal utilisation of world Uranium reserves is considered, the Thorium fuel cycle appears to be the best alternative because of the good neutronic properties of the bred fissile U-233. The HTGR concept offers the possibility of thermal breeding but the commercial introduction of the concept largely depends upon the closure of the Thorium fuel cycle which has not been achieved yet, although the very high burnup values attained in the prototypes yield effective fuel utilisation.

So far as the U-235 consumption is concerned, a Pre-breeder must switch over to a Near-Breeder at the expense of increased fuel cycle costs as the former is strictly dependent on U-235 for its make-up.

APPENDICES

The Appendices cover information about the formation of U-232 and the influence of U-236 build-up. Appendices A and B are self - explanatory whereas a brief introduction for the chemical processing is appropriate here.

One method to separate an aqueous solution into its constituents is the solvent extraction method. In this method, an organic acid is brought into contact with the aqueous solution in which one or more of the constituents is sufficiently soluble in the organic medium, whereas others are not so much. This leads to the distribution of the constituent substances into the organic or aqueous medium. The process is principally a diffusion problem in which a solute is transferred from one liquid to another and the operation is carried out in extraction columns or in mixer-settler devices in which the organic phase flows in one direction and the aqueous phase in the other.

A. PRODUCTION AND DECAY OF U-232

U-232 isotope is formed by an (n,2n) process on Th-232 or on U-233. U-232 is an Alpha-emitter with a half-life of 74 years and initiates a radioactive decay chain which ends at Pb-208. The main disadvantage associated with this series is the production of highly Gamma-active intermediates such as Bi-212 and Tl-208 and the gaseous isotope Rn-220 which can be transported through the filters.

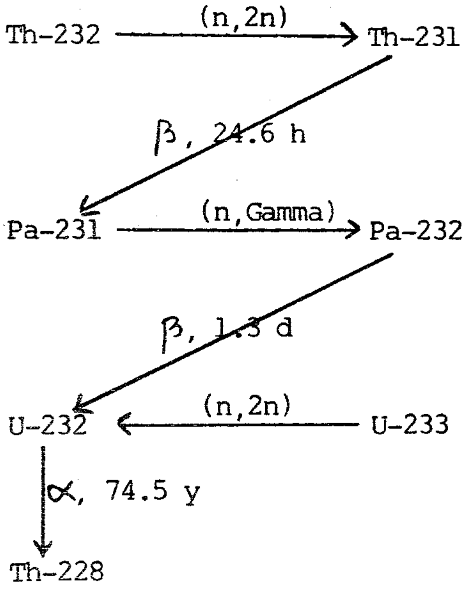


Fig.A.1. Production of U-232

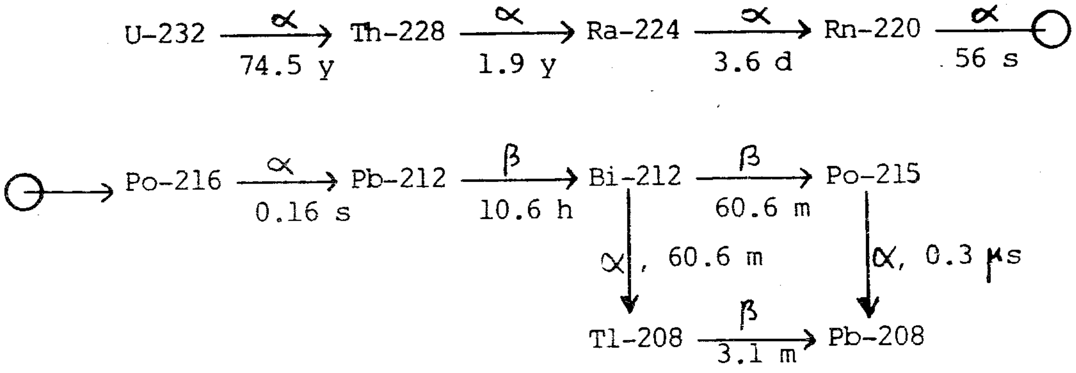


Fig.A.2. U-232 decay chain

B. THE INFLUENCE OF U-236 ON THE FUEL CYCLE

Neutron capture in U-235 results in the production of U-236, therefore, U-236 acts as a parasitic neutron absorber and depending on the exposure of Uranium fuel in the reactor, U-236 build-up may lead to significant amounts during subsequent recycling. Furthermore, by successive neutron absorption and decay of U-236, the production of U-237, Np-237 and Pu-238 follows.

The distribution of U-236 throughout the fuel is unavoidable when the recycled fuel is mixed with fresh fuel while enrichment and its effective concentration is determined by the ratio of fresh and recycled fuel mixing. During the enrichment process, enrichment in U-236 also takes place as well as in U-235. The relative increase in U-236 enrichment is about 80 % of the increase in the enrichment of U-235. The parasitic neutron absorption in U-236 and its nuclide chain products must be counter-balanced by extra enrichment in U-235.

C. PUREX PROCESS

The Purex Process is a typical solvent extraction method in the reprocessing of spent fuel. N-TributylPhosphate, TBP, is used as the organic solvent since Uranium and Plutonium nitrates can be extracted from the aqueous solution by the TBP, but the fission products to a much smaller extent.

Uranium, Plutonium and fission product nitrates are fed into the Scrub Extraction column at the middle. Plutonium is best extracted by TBP when it is in the tetrapositive oxidation state, ie, with four loosely bound outer electrons and this is assured by the addition of sodium nitrate. The organic solvent is less dense and enters at the bottom flowing upward. Uranium and Plutonium nitrate are thereby removed from the aqueous solution and pass into the organic solvent. The organic phase is scrubbed with nitric acid, which acts as a salting agent, in the upper part of the column. The use of a salting agent lies in that it prevents Uranium and Plutonium to be back extracted into the aqueous medium whereas the fission products that may have entered the organic medium are returned back to the aqueous medium. The output of the extraction column contains all of the fission products with little or no Uranium or Plutonium.

The second step involves the introduction of the organic phase with Uranium and Plutonium into the partitioning column. The organic phase flows upward and reacts with aqueous stripping solution that flows downward and contains a reducing material and salting agent. The reducing material is ferrous sulphamate in which the ferrous ion is the reducing agent and the sulphamate removes the nitrate which prevents the reduction. The Plutonium is reduced into tripositive valence state to be back extracted into the aqueous phase. As this flows downward, fresh TBP moves upward to return any Uranium back to the organic medium. The aqueous phase containing Plutonium nitrate leaves the bottom of the partitioning column.

The organic solution of Uranium separated from Plutonium and fission products is fed into a third column flowing upward to be stripped by dilute nitric acid that flows downward. The Uranium is back extracted to the aqueous phase and leaves at the bottom. The spent solvent is sent to a recovery plant for purification and subsequent re-use in extraction. For further purification, the aqueous solutions of Uranium and Plutonium can be re-cycled.

D. THOREX PROCESS

The Thorex Process is in general analogous to the Purex Process. It is based on solvent extraction with TBP. The products, Th-232 and U-233 are to be separated from contaminants and from each other. The first cycle of solvent extraction consists of three contactors as in Purex.

The usual auxiliary steps, such as solvent washing and inter-cycle evaporation are also included. Nitric acid may be used as the primary salting agent throughout the Thorex Process but at least some aluminum nitrate is required in the first contactor to complex the Fluoride ion that has been used as a catalyst for the dissolution of the Thorium. Otherwise, Fluoride could form a complex with Thorium.

Thorium is not so well extracted as Uranium, therefore, more moles of TBP are required for each gramme atom of metal extracted and this makes the capacity of the Thorex process smaller. Larger flows of solvent are needed. The net effect is that equipment of the same size has about half the capacity for irradiated Thorium that it does for irradiated Uranium.

Partitioning of Uranium and Thorium is more difficult than the partitioning of Uranium and Plutonium because no change of oxidation state for Uranium or Thorium analogous to that for Plutonium is available. The separation of Thorium and Uranium in the second contactor requires a delicately adjusted salting agent introduction.

Another problem in Thorex process is the radiological problem of Pu-233. Pa-233 has a relatively long half-life of 27 days for its decay to U-233 and this requires storing for more than 200 days after discharge. Th-234 is also present in irradiated Thorium with 24 days of half-life, hence, adds to the desirability of a long decay period before processing.

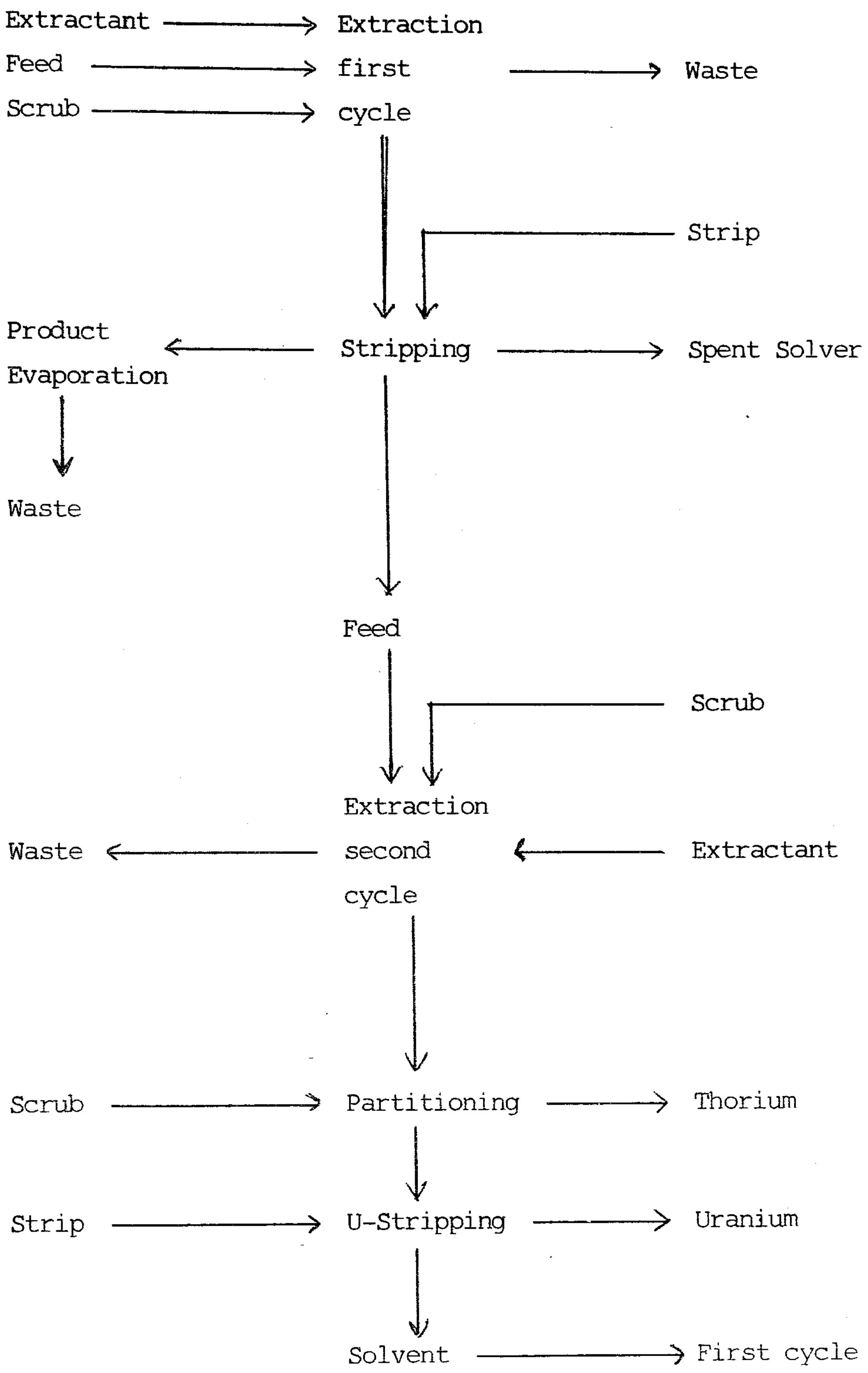


Fig. D.1. Thorex Process Flow Diagramme

REFERENCES

1. Özdemir, Ünal ; Elektrik Üretiminde Nükleer Santraller ve Ülkelerin Elektronükleer Programları, TC Başbakanlık AEK, Teknik Rapor No.3
2. IAEA Bulletin Vol.27 No.4 pp.71, 1985
3. Lamarsh, J R ; Nuclear Reactor Theory,
4. Sesonske Alexander, Glasstone Samuel ; Nuclear Reactor Engineering
5. Loftness, Robert l ; Nuclear Power Plants
6. Smith, J F, Carlson, O N, Peterson, D T, Scott, T E ; Thorium : Preparation and Properties, IOWA State University Press, 1976
7. Gülovalı, M Ç, Tarhan, M, Somer, Ş N ; Thorium Cevheri ve Çözütleme Çözeltileri Analizi,
8. Sesonske, Alexander ; Nuclear Power Plant Design Analysis, Technical Information Centre, Office of Information Services, USAEC,1973
9. Lustman, B, Goodwin, J G , Reactor Handbook, Vol.1, pp.708 Interscience Publishers Inc. 1960
10. Davidge, P C, Edwards, H S, Lyon, R H ; Reactor Handbook, Vol.1, pp.979, Interscience Publishers Inc. 1960
11. Kraemer, H ; Gas Cooled Reactors Vol.1, pp.11, IAEA 1976
12. Conrad, Rainer P, Merlini C, Mehner Alfred W ; Irradiation Performance of Advanced Coated Particles for HTGR Fuel Cycles, Nuclear Technology, Vol.61, No.3, pp.403, 1983
13. Hick H, Graham, L W, Nabieleck, H et al. ; Fission Product Retention and Fuel Development for Advanced HTRs, Gas Cooled Reactors, Vol.2
14. Wagner, Löffler, M ; Amoeba Behaviour of UO_2 Coated Particle Fuel, Nuclear Technology, Vol.35, pp.392, 1977

15. Smith, C L ; Migration of ThO_2 Kernels under the Influence of a Temperature Gradient, Nuclear Technology, Vol.35, pp.433, 1977

16. Grübmeier, H, Naoumidis, A, Thiele, B A ; SiC Corrosion in HTGR Fuel Particles, Nuclear Technology, Vol. 35, pp.413, 1977

17. Teuchert, E, Rütten, H J ; Near-Breeding Thorium Fuel Cycle, Gas Cooled Reactors, Vol.2, pp.491, 1975

18. Teuchert, E, Werner, H, Performance of Thorium Fuel Cycles, Nuclear Technology, Vol.38, PP.374, 1978

19. GCRA ; Evaluation of Small Modular HTGRs, 1984

20. The High Temperature Reactor and Process Applications, pp.45.1,,1974

21. Flagg, John f, Chemical Processing of Reactor Fuels, Academic Press, pp.210, 1961