IAEA-TECDOC-412

# THORIUM-BASED NUCLEAR FUEL: CURRENT STATUS AND PERSPECTIVES

PROCEEDINGS OF A TECHNICAL COMMITTEE MEETING
ON UTILIZATION OF THORIUM-BASED NUCLEAR FUEL:
CURRENT STATUS AND PERSPECTIVES
ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
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# FOREWORD

A number of IAEA Member States have extensive thorium resources, in some cases bigger than uranium resources and they are interested in the development of thorium nuclear fuels for the reason of self-sufficiency. The world situation with uranium production could be also one of the factors endorsing R and D on further investigation of thorium fuels.

Until the present time considerable efforts have already been made in the area of fabrication, utilization and reprocessing of Th-based fuels for different types of reactors, namely: by FRG and USA - for HTRs; FRG and Brazil, Italy - for LWRs; India - for HWRs and FBRs. Basic research of thorium fuels and thorium fuel cycles are also being undertaken by Australia, Canada, China, France, FRG, Romania, USSR and other countries. Main emphasis has been given to the utilization of thorium fuels in once-through nuclear fuel cycles, but in some projects closed thorium-uranium or thorium-plutonium fuel cycles are also considered.

Thorium fuel utilization, technical status and development needs were considered in detail by the INFCE Working Group 8 (1977-1980). Some aspects of the thorium fuel cycle were considered at the IAEA Nuclear Power Conferences in Salzburg (1977) and Vienna (1982), as well as, the TCM on Improved Utilization of Water Reactor Fuel with Special Emphasis on Extended Burnups and Plutonium Recycling (Mol, Belgium, 7-11 May 1984), TCM on Advanced Light and Heavy Water Reactor Technology (Vienna, Austria, 26-29 November 1984) and others. In-depth consideration of the technology of some thorium fuels was given at the IAEA Advisory Group on Advanced Fuel Technology and Performance (Würenlingen, Switzerland, 4-7 December 1984).

The purpose of the Technical Committee on the Utilization of Thorium-Based Nuclear Fuel: Current Status and Perspective was to review the world thorium resources, incentives for further exploration, obtained experience in the utilization of Th-based fuels in different types of reactors, basic research, fabrication and reprocessing of Th-based fuels. As a result of the panel discussion the recommendations on future

Agency activities and list of major worldwide activities in the area of Th-based fuel were developed.

The Agency wishes to thank all those who participated in the panel discussion. Special thanks are due to the Sessions' Chairmen, Messrs. M. Peehs, I. Slesarev, C. Ganguly and the IAEA staff members, Messrs. F. O'Hara and M. Ugajin. The officer of the IAEA responsible for the preparation of this document is Mr. V. Onufriev, Division of Nuclear Fuel Cycle.

## *EDITORIAL NOTE*

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## PAPERS PRESENTED

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## AN OVERVIEW OF WORLD THORIUM RESOURCES, INCENTIVES FOR FURTHER EXPLORATION AND FORECAST FOR THORIUM REQUIREMENTS IN THE NEAR FUTURE

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### Abstract

Thorium occurs in asociation with uranium and rare earth elements in diverse rock types. It occurs as veins of thorite, uranothorite and monazite in granites, syenites and pegmatites. Monazite also occurs in quartz-pebble conglomerates, sandstones and fluviatile and beach placers. Thorium occurs along with REE in bastnaesite, in the carbonatites.

Present knowledge of the thorium resources in the world is poor because of inadequate exploration efforts arising out of insignificant demand. But, with the increased interest shown by several countries in the development of Fast Breeder Reactors using thorium, it is expected that the demand will increase considerably by the turn of the century.

The total known world reserves of Th in RAR category are estimated at about 1.16 million tonnes. About 31% of this (0.36 mt) is known to be available in the beach and inland placers of India. The possibility of finding primary occurrences in the alkaline and other acidic rocks is good, in India. The other countries having sizeable reserves are Brazil, Canada, China, Norway, U.S.S.R., U.S.A., Burma, Indonesia, Malaysia, Thailand, Turkey and Sri Lanka.

Considering that the demand for thorium is likely to increase by the turn of this century, it is necessary that data collected so far, globally, is pooled and analysed to identify areas that hold good promise.

### Introduction:

Thorium in association with uranium and Rare Earth Elements (REE) occurs in diverse rock types; as veins of thorite, thorianite, uranothorite and as monazite in granites, syenites, pegmatites and other acidic intrusions. It also occurs as an associated element with REE bearing bastnaesite in carbonatites. Monazite also occurs in quartz-pebble conglomerates, sandstones and in fluviatile and beach placers.

Prior to the second world war thorium was used widely in the manufacture of gas mantles, welding rods, refractories and in magnesium based alloys. Its use as fuel in nuclear energy, in spite of its limited demand as of now and low forecast, is gaining importance because of its transmutation to $^{233}$U. Several countries like India, Russia, France and U.K. have shown considerable interest in the development of fast breeder reactors (FBR) and it is expected that by the turn of this century some of the countries would have started commissioning large capacity units.

## 2. World deposits:

Present knowledge of the Thorium resources is poor because of the relatively low-key exploration efforts arising out of insignificant demand. For the same reason the possibility of discovering new finds in addition to increasing the known resources contained in the deposits already known is good.

The largest known reserves of thorium are contained in the beach and inland placer deposits of monazite, which is exploited so far for its REE content (55-60% REO).

Monazite placers are reported from Australia, Egypt, India, Liberia, Brazil, Malgachi and the United States of America (Florida). In Brazil, monazite occurs associated with ilmenite and zircon along the eastern and southeastern Atlantic coast [1]. It also occurs at Araxa in association with carbonatites. In Burma, placer deposits derived from the weathering of biotite granites occur in the southern Shan states. Considerable quantities of cassiterite and wolframite occurring in these placers are derived by the weathering of quartz veins and pegmatite dykes injected into the granites.

Malaysian deposits occur on the coast of Kedan and Perlis at Pulan Lankawi and Seremban. Monazite associated with columbite and xenotime occurs in Ulusempam area near Pahang. Cassiterite placers at Trengganu contain as much as 58% monazite. The occurrences of monazite in Thailand and Indonesia are similar to the occurrences in Burma [2; 3].

In Sri Lanka rich deposits derived by the weathering of schists, granulites and gneisses occur along the N and NW coast. Alluvial deposits also occur in the lower valley of Kaleganga in Ratnapura district. The largest placer deposit (12,000 t) near Pulmoddai extending over a distance of 3 km. and a width of 50 m. contains 3 million t of sand at 0.4% monazite, 18% rutile and 62% ilmenite [3].

In Bangladesh monazite is also reported to occur in the beach placers (derived from granites) near Chittagong. Similar deposits are also reported to occur in Southern China, between Chianhua Hsien in South Western Hunan province to Kung Chen Hsien in northeastern Kwangsi province. Extensive deposits are also reported from Kwangtung province.

In Canada, large Thorium reserves associated with Uranium (with Th:U ranging from 3.5 to 1 to 0.5:1) occur in Elliot Lake area and other Huronian complexes. Thorium is also reported from the granitic and syenitic rocks in the Bancroft area of Ontario. Many occurrences of this type are reported from the Grenville geological province, the Charlebois Lake and the La Ronge areas in Saskatchawan. It also occurs at Oka in association with uranium and niobium in the carbonatites in Qubec [4]. In Turkey, the Eskisehir-Sivrihisar area is estimated to contain 880 thousand t of Thorium.

In USA about 80% of the known Th reserves occur in vein type thorite deposits in the Lemhi Pass area of Idaho and Montana and in the Western mountains of Central Colarado. Large reserves of Th are also known to occur associated with bastnaesite in the massive carbonatite deposits of California [4]. The Florida beach placers wherefrom monazite could be recovered along with ilmenite are estimated to contain 0.3% to 1% monazite [4].

The details of world reserves and production are given in Table I and Figure 1.

### TABLE I

WORLD THORIUM RESERVES AND PRODUCTION
(Based on USBM Mineral Industry Survey, May 16, 1985)

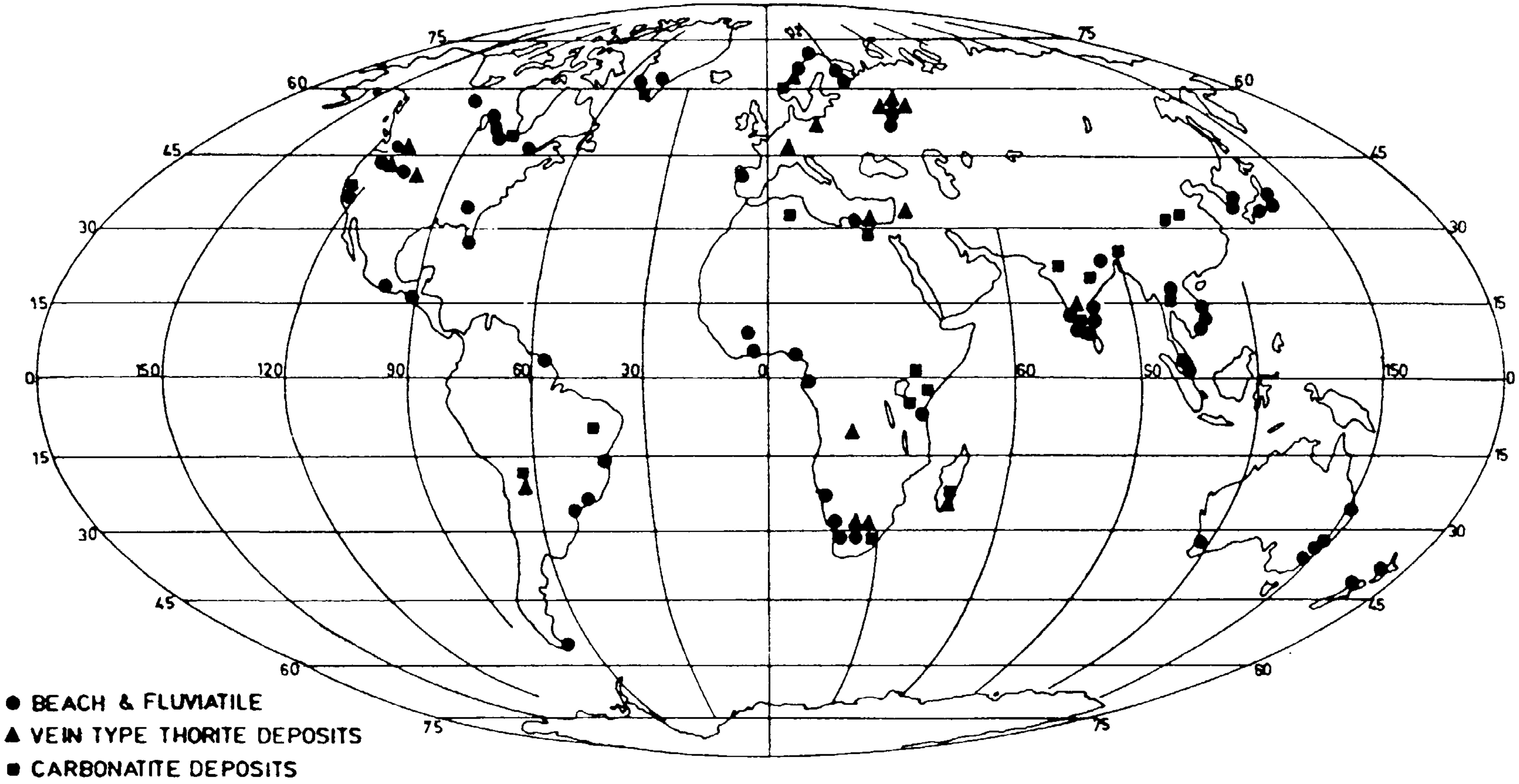
In thousands of tonnes

| | Name of the country | Reserve Base RAR | Production |
|---|---|---|---|
| 1. | United States of America | 200 + 520* | 17.03 |
| 2. | Australia | 40 | 8.0 |
| 3. | Brazil | 70 | 1.1 |
| 4. | Canada | 240 | -- |
| 5. | India | 360 | 4.0 |
| 6. | Malaysia | 10 | 0.2 |
| 7. | Norway | 150 | -- |
| 8. | OMEC | 55 | 0.2 |
| 9. | OPEC | 35 | 1.5 |
| 10. | China | @380* | 6 |
| 11. | U.S.S.R. | @120 | n.a. |
| | Total | 1,160 | 37.5 |

* Bastnaesite

n.a. - not available

@ excluded.



THORIUM DEPOSITS IN THE WORLD

FIG. 1.

## 3. Thorium occurrences in India:

Although many occurrences of uranothorite, thorite, thorianite and monazite are reported from in several intrusive pegmatites and younger granites of all ages, monazite occurring in the beach and inland placers remains the main thorium and REE bearing mineral source assessed by AMD (Fig. 2).

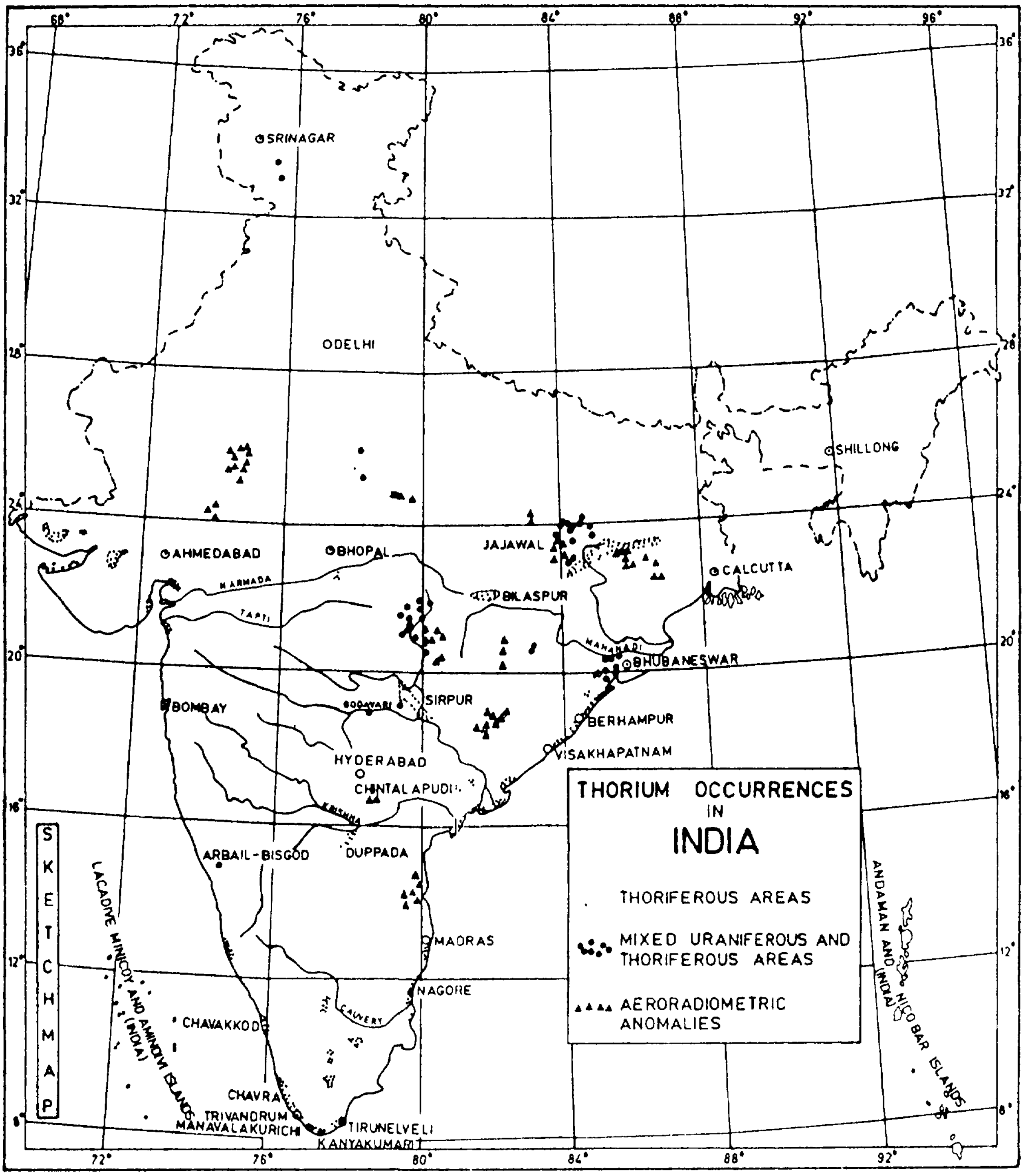


FIG. 2.

The occurrence of monazite in the placers (assessed so far at $4.5 \times 10^6$ t) may be divided into 4 types: (i) the Beach and Dune sands on the West and East coasts (42%); (ii) the Teris (16%); (iii) the sea bed (10.0%) and (iv) inland placers (32%).

### 3.1 Beach sands:

Although monazite occurs associated with ilmenite and other hm in the beach sands, skirting the entire Peninsular India, its economic concentration is confined to only some areas where suitable plhysiographic conditions exist. The west coast placers are essentially beach or barrier deposits with development of dunes where aeolin action is prominent in dry months. On the other hand, the east coast deposits consist of extensive dunes fringing the coast.

### 3.1.1. West coast

Of the several west coast deposits assessed so far the deposits at Chavara and Manvalkurchi in Kerala and Tamil Nadu respectively, are rich in hm content [5]. The other deposits occurring north of Chavara, stretching over a distance of 50 km. upto Ratnagiri are leaner with a hm content of $\approx$ 20% of which monazite forms 0.06% [6].

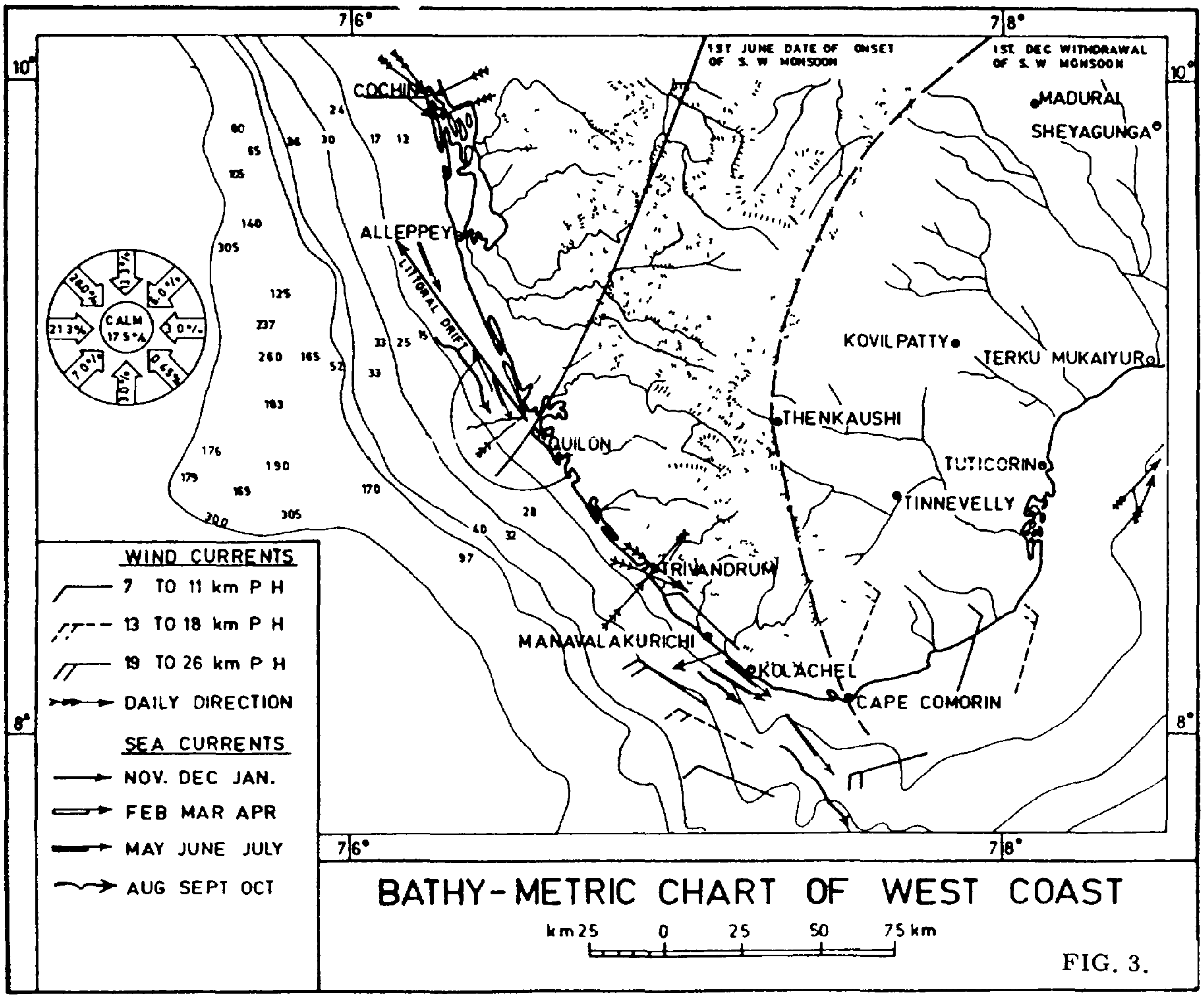
### 3.1.2 Origin of West Coast deposits:

Physiographically the peninsular India on the west coast may be divided into three divisions: (i) the 25-40 km. wide western ghats which rise steeply to an average height of 1200 m. above m.s.l. along a conspicuously straight fault (ii) the 20-30 km. wide narrow coastal strip (30 m. high) to the west, drained by 41 fast west flowing rivers and (iii) the narrow (45 km. wide) continental shelf with an average depth of 29 fathoms.

The western ghats receive about 3000 mm. of rainfall annually. All west flowing rivers are small and descend into the narrow coastal belt to finally empty into irregular, labile net work of estuaries and lagoons. The beaches along the coast are about 3 to 4 m. high.

Geologically, the western uplands consist of ortho and para gneisses of Precambrian age, charnockites of felsic to ultramafic composition and leptynites, intruded by swarms of pegmatites. Overlying these Precambrian rocks are the Tertiary Quilon-Warkala beds consisting of laterites, semi-consolidated sandstones and clays with lenses of lignite. They vary in thickness and dip 3° seaward. Overlying these beds asre the recent unconsolidated alluvial clays and estuarine sands.

A series of uplifts of western ghats shifted the strand line seaward resulting in the formation of bars, spits and lagoons [7]. The hm deposits are formed in four successive stages: (i) lateritisation of gneissic complexes, (ii) successive mountain uplift and simultaneous seaward shift of strand line., (iii) reworking of the beach sands by sea waves, which rise often to a height of 3 m. in 12 s. period and (iv) littoral drift caused by the breaking of the waves far away from the shore and consequent northerly movement of lighter minerals along the reflected waves. The



BATHY-METRIC CHART OF WEST COAST

FIG. 3.

wind and wave action causes erosion of Warkala beds during high wave period and suppliments the littoral drift (Fig.3) [8]. The barrier formed works like a riffle on the wilfley table and deposits the hm on the beach. The light minerals are carried forward [9]. The similarity in the size (Fig.4 and 5) and sphericity ( ($\psi \sim 0.8$) of the hm from the beach and sea bed ($\bar{D}_{80} = 100\mu$) indicates that there is very little fresh accretion to the sea bed. There is only an annual making and remaking of the coast line when the hm are enriched on the seaward side of the bar by a factor of > 9 [10]. The beach sands of Chavara bar (Kerala) contain (73% hm), 60-70% ilmenite (61% $TiO_2$). 4-7% garnet, 0.5% to 1% monazite (9.0% $ThO_2$), 5-8% zircon and 1.15% sillimanite.

In Manavalakurchi, Tamil Nadu, the deposit is formed by the "southerly tilt of the tip of the peninsula [9] aided by seasonal variation of sea currents, both in direction and magnitude [10]. It contains 64% hm with 45-50% ilmenite (with 54% $TiO_2$), 2-3% rutile, 3-4% monazite (9-10% $ThO_2$), 4-6% zircon and 56% garnet [12]. The higher percentage of monazite and garnet in this area is attributable to the high density of intrusion of the pegmatites and leptynites in the hinterland and its location on the sea side of the embayment of eroded laterite [13].

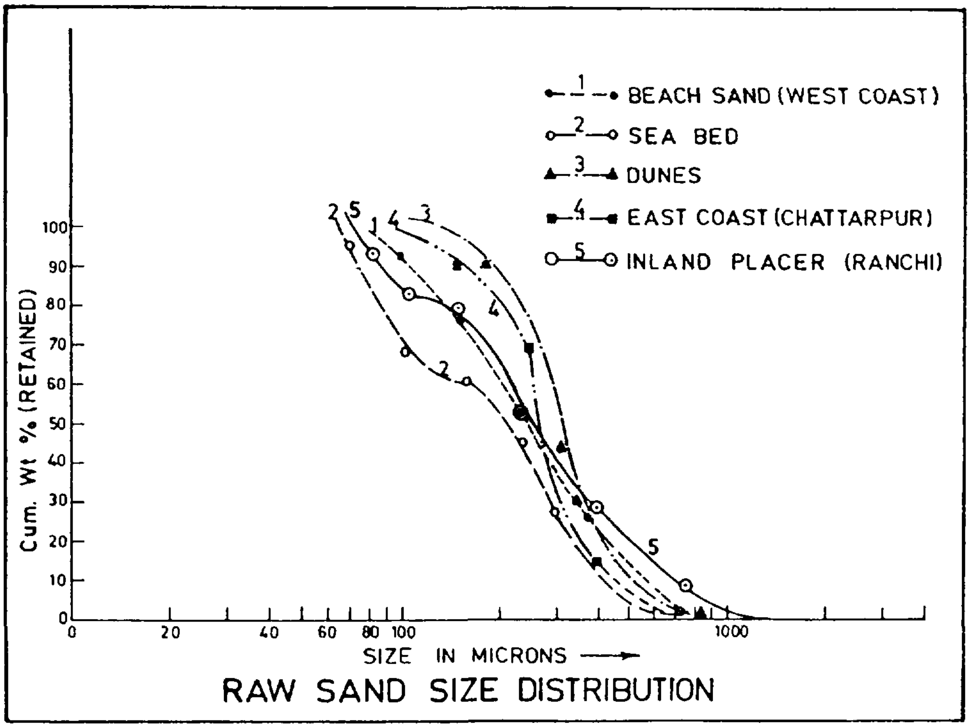

1 BEACH SAND (WEST COAST)
2 SEA BED
3 DUNES
4 EAST COAST (CHATTARPUR)
5 INLAND PLACER (RANCHI)
Cum. Wt % (RETAINED)
0
10
20
30
40
50
60
70
80
90
100
0
20
40
60
80
100
1000
SIZE IN MICRONS →
RAW SAND SIZE DISTRIBUTION

FIG. 4.

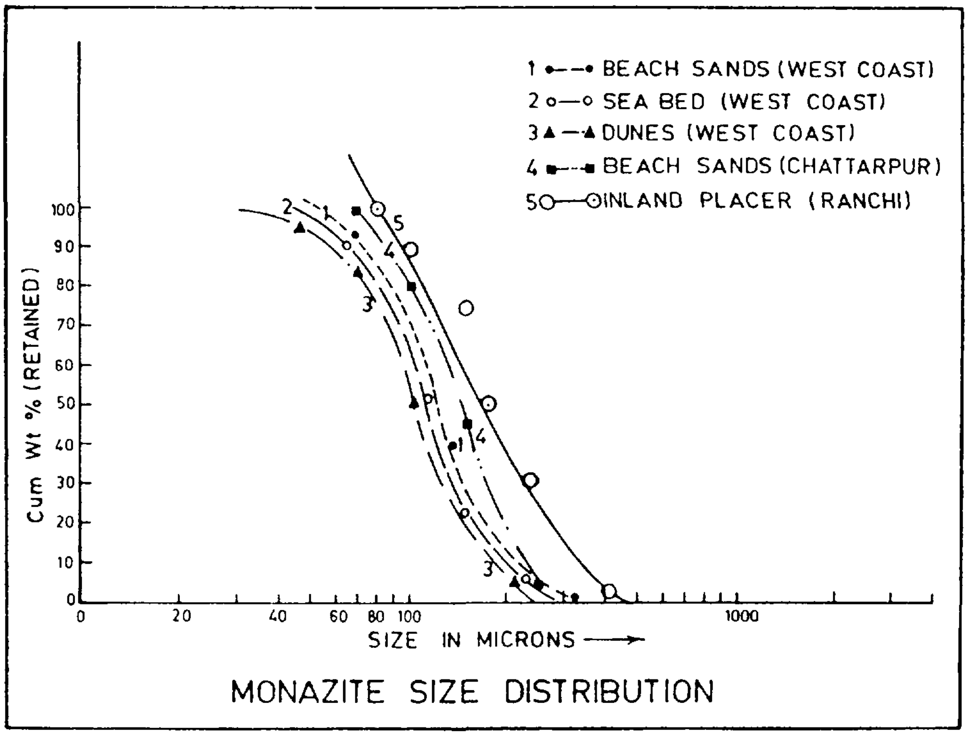

1 BEACH SANDS (WEST COAST)
2 SEA BED (WEST COAST)
3 DUNES (WEST COAST)
4 BEACH SANDS (CHATTARPUR)
5 INLAND PLACER (RANCHI)
Cum Wt % (RETAINED)
0
10
20
30
40
50
60
70
80
90
100
0
20
40
60
80
100
1000
SIZE IN MICRONS →
MONAZITE SIZE DISTRIBUTION

FIG. 5.

### 3.1.3 Dunes:

The dunes near Manavalakurchi situated at the mouth of Valliyar river are formed by aeolin action on the beaches during the dry season. They contain 50% hm of which 35% is ilmenite (55% $TiO_2$), 1.35% rutile, and 3.3% monazite (9-10% $ThO_2$) [14].

### 3.1.4 Sea bed off the Neendakarai-Kayankulam bar:

Investigations conducted about 600 m. off the Neendakarai-Kayankulam bar over a length of 22 km shows that the sea bed contain 5% hm of which 0.05% is monazite (9.5% $ThO_2$) [15].

### 3.1.5 The Teris:

The 'teris' are dunes formed by aeolin action in the arid, rain shadow plains east of western ghats. Typically, these dunes analyse upto 10% hm of which 3-4% is ilmenite and 0.06% to 0.35% is monazite [15].

### 3.1.6 The East Coast deposits:

The east coast beach placers and dunes are low grade with 8-20% hm. Their formation is influenced by the northerly long shore currents and the consequent annual littoral drift of about 2 million t of sand containing 4-5% hm. The sediments from eastern ghats, which consist of charnockites, khondalites and alkali rocks, are carried by east flowing rivers and deposited to form deltas and shallow beaches. During the low tide the beaches from the surf zone are exposed to strong, incessant wind action during summer months and cause accretion and landward migration. Of the several occurrence studied the Chatrapur deposit (Orissa) with about 20% hm and 0.5% monazite is important

The origin of these dunes may be traced to the initial building upof off-shore bars/spits resulting in the formation of frontal dunes and lagoons. Subsequently, the lagoons are filled by the deposition of fresh burden from local streams and spill over sand from frontal dunes and also the littoral drift. This results in the formation of low undulating inter dunes sandy area and finally the rear landward dunes wherever land barriers exist [17].

In addition to the Chatrapur occurrences several exploitable deposits (25-30% hm; 0.2% monazite) occur along the east coast, in Andhra Pradesh and Tamil Nadu.

## 4. Inland placers:

Significant occurrences of monazite-bearing inland placer deposits are located in the Ranchi plateau. The bed rocks in the region comprise of Archean metasediments profusely injected by aplo-pegmatites and finally by a porphyritic granite (18). The bed rock is covered by a 11/2 m thick mantle of coarse, yellow alluvium with thin discontinuous dark bands which contain 2-10% hm of which 0.45% to 1.0% is monazite.

## 5. Primary sources:

In addition to the placer deposits described above several primary occurrences of Th are noted at many places.

Of these, possible economic concentrations that might merit economic consideration in future are: (i) Proterozoic quartz-pebble conglomerates overlying the Archean basements., (ii) Jurassic sandstones of Kutch in Gujrat and the Gondwana sandstones in M.P., (iii) The igneous rocks of Dhabi, Jajawal (M.P.), Palamau (Bihar) along a 150 km long shear zone, (iv) The pegmatites of Salem District (Tamil Nadu) intruding biotite schists and (v) The alkali syenites and carbonatites of Sevattur, Pakkanadu (Tamil Nadu), Sung Valley (Meghalaya), Khammam & Visakhapatnam (A.P.), Kishengarh (Rajasthan) and several other occurrences.

## 6. Reserves:

The current knowledge of thorium reserves in the world is small. In Brazil, several monazite placers occur on the Atlantic coast. In addition large reserves (87%) are associated with bastnaesite in Araxa carbonatites. In Canada 240 x $10^3$ t of thorium resources are identified so far, bulk of which are associated with uranium (Th:U 3.5:1/0.5:1) in the quartz-pebble conglomerates of Elliot lake. In South Africa, of 12500 t of Th identified, about 77.5% is estimated to occur in the carbonatites, 11.2% in the sandstones of Karoo supergroup, and only 5% in the quartz-pebble conglomerates and the rest as veins in Archean complex.

China and USA have a large reserve base because of the association of Th (0.5% to 1% $ThO_2$) with REO in its bastnaesite deposits associated with carbonatites. The Chinese reserve associated with the bastnaesite in the carbonatites of Inner Mangolia are estimated at 380,000 t of Th [19].

According to USBM Industrial survey (1985), [20] the US reserves of thorium are estimated at about 200,000 t of which, about 70% occurs in vein type thorite deposits. The reminder are in the carbonatite deposits in California. Small amount of monazite (0.35%) are also recovered alongwith ilmenite, from the beaches of Florida.

The reserve base of other countries like Sri Lanka, Malaysia, Norway etc. are small and forms only 20% of the world total.

In India thorium reserves are estimated at 0.36 x $10^5$ t of Th forming $\approx$ 30% of the known world resources [21]. The details of Indian reserves are given in Table-II.

## 7. Process flowsheet:

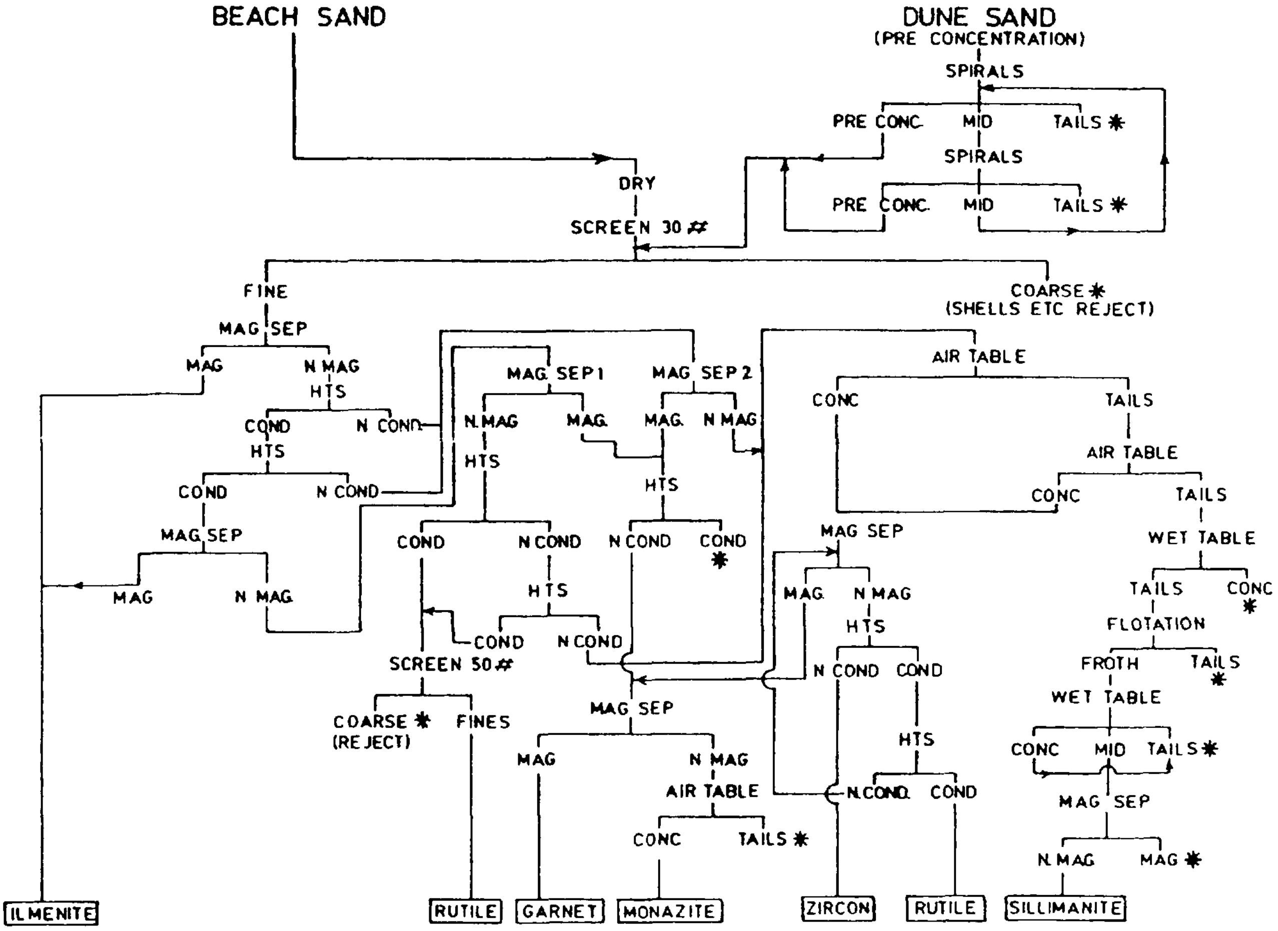
A generalised industrial process flowsheet for the separation of individual minerals is given in Figure 6. It comprises of five unit operations: (i) sizing, (ii) magnetic separation of varying intensity, (iii) high tension separation (HT), (iv) air/wet tabling and (v) flotation.

## TABLE II

RESERVES OF MONAZITE

In million tonnes

| | Nature | RAR | | EAR I | |
|---|---|---|---|---|---|
| | | Monazite | $ThO_2$ | Monazite | $ThO_2$ |
| 1. | Beach sands and dunes | 1.60 | 0.144 | 0.29 | 0.026 |
| 2. | Teris | 0.73 | 0.066 | -- | -- |
| 3. | Sea bed off coast | -- | -- | 0.43 | 0.038 |
| 4. | Inland placers | -- | -- | 1.44 | 0.130 |
| | Total | 2.33 | 0.210 | 2.16 | 0.194 |



GENERALISED FLOW SHEET WEST COAST BEACH SAND

FIG. 6.

The raw sand is trucked to the plant where it is dried and sized using 35 mesh screen. The oversize is rejected and the undersize is treated in HT separators. The conducting minerals are subjected to magnetic separation (light intensity) to yield a crop of ilmenite in the magnetics and rutile in the non-magnetics. The non-conducting minerals are subjected to a successive stages of light and high intensity magnetic separation to yield monazite in the magnetics which is cleansed by using air/wet tabling and/or flotation to the required grade. The non-magnetics of high intensity magnetic separation are cleaned on an air table to yield a concentrate of zircon and lights (tails) of silliminite. Silliminite is further cleaned using fatty acid flotation to obtain a marketable grade crop. Though ideally ilmenite and rutile must report as conducting magnetics and non-magnetics, several minerals, particularly garnet reports in these concentrates. Therefore, several steps of cleaning and scavenging are undertaken to yield and required grade of monomineralic concentrates and increase the recovery of individual minerals.

In case of low grade Dunes and east coast placer deposits, tests using several alternative equipment such as Tables, Trays, Cones and Spirals indicated that spirals are most satisfactory since they offer flexibility and also yield a rejectable tailing analysing 2.9% hm and a concentrate (75% hm at 90% recovery), which could be fed to the dry mill.

## 8. Discussion:

The reasonably assured resources of thorium in India, form about 31% of the world's estimated deposits. The reserves could have been several times more if systematic surveys are carried out for thorium in the geologically promising terrains such as we have in the acid intrusive belt of Salem in Tamil Nadu, the hinterland of Kerala and the alkali syenitic and carbonatite rocks of Tamil Nadu, Andhra Pradesh, Rajasthan, Madhya Pradesh, Bihar and Meghalaya.

Taking an overview, while intensive and extensive exploration identified $2 \times 10^6$ t of U in RAR category, the known thorium reserves, assessed so far in the same category indicate $1.2 \times 10^6$ t even when the inputs for locating thorium reserves are incidental to prospecting for hm like ilmenite and/or uranium. Therefore, globally speaking the possibility of increasing the Th reserves is high.

Long term projections of IAEA for the nuclear power growth for the period 2025 indicates an inadequate supply [22] of uranium even when a lower growth rate for nuclear energy (using LMFBR) and the planned growth of uranium production are considered. The gap will increase considerably, if the demand for nuclear energy increases and more reactors are commissioned.

While it is true that large stocks of available depleted U can be utilised, countries that have limited identified resources of U, may have to use Th to ensure continuous growth rate in their quest for power.

Thus, in the long term, it is necessary that the data collected globally is pooled, and prospective areas are identified for funding of surveys that might be considered rewarding.

## 9. Acknowledgements:

My grateful thanks to Dr. R. Ramanna, Chairman, AEC, India, for giving me this opportunity to represent India on this committee. Thanks to Sri T.M. Mahadevan, Director, AMD, for his valuable suggestions.

Thanks are also due to several geologists and mineral engineers who poineered the work and developed the techniques for assessing and processing the beach placers.

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# IAEA ACTIVITIES IN NUCLEAR FUEL TECHNOLOGY, PERFORMANCE AND UTILIZATION INCLUDING THE ROLE OF THORIUM-BASED FUEL

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**Abstract**

The IAEA activities in nuclear fuel technology, performance and utilization are presented with special emphasis on thorium-based nuclear fuel. Utilization of thorium fuels in once-through cycles is discussed, as well as, in closed thorium/uranium or thorium/plutonium fuel cycles.

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By the end of 1985 there were 374 power reactors in operation, with a total generating capacity of 249,6 GW(e) and about 90% thereof was generated by water-cooled reactors using $UO_2$ as fuel and zircaloy as cladding material.

Until now, large experience has been accumulated worldwide in nuclear fuel design, fabrication and utilization in water power reactors. Water reactor fuel fabrication plants are in operation in 15 Member States with licensed annual capacity of around 10,000 tons of heavy metal. It will meet the demand of utilities until sometime between 1990 and 1995, depending on the actual deployment of nuclear power plants and the degree of improvements in reactor and fuel performance.

For the actual parameters of fuel utilization in water-cooled power reactors the performance of fuel is satisfactory. Average failure rates are now as low as 0.01% failed fuel rods per reactor year or less. Thus, nuclear fuel cycle technology in the field of fuel fabrication and utilization is the proven technology today. It is important today and in the near future to reach the same or better level of fuel reliability under future operational and abnormal conditions.

Now it is also time to adapt nuclear plants to the requirements of the national and in some cases, multinational electricity grids, to improve economics under the level of safety, as necessary. Not the last target is the reduction of uranium consumption, plutonium and uranium recycling. In order to meet these requirements national and international projects are being performed with the aim to extend burnup (for PWRs from 33 MWd/kgU to 45 MWd/kgU by 1995), to improve fuel rod, assembly and active core design and to implement advanced fuel management schemes. Automation of fuel fabrication processes is directly linked with the production of mixed (uranium and plutonium) oxide fuels and recycling of fissile elements from used fuel. Further development of methods of characterization and Quality Control of nuclear fuel is performed, especially, regarding their feedback to fuel fabrication and behaviour.

In the current and near future water reactor fuel development is also primarily related to studies on water side corrosion, pellet cladding mechanical and chemical interaction, fission gas release, dimensional stability, the use of burnable absorber, etc.

The activity of the Nuclear Fuel Cycle Division of the IAEA in this area, has been performed under the guidance of the International Working Group on Water Reactor Fuel Performance and Technology (IWGFPT). This group comprises 21 countries with significant nuclear power programmes. Many developing countries constantly request that more efforts should be made by the Agency in order to provide them with technical advice and expertise in the development of nuclear fuel technology. Assistance has been given to Egypt, Indonesia, Republic of Korea, Romania, Yugoslavia in establishing their own fuel cycle facilities through projects performed by the Department of Technical Cooperation. The Agency financially supported participation of representatives from developing countries in meetings related to nuclear fuel fabrication and utilization.

Through Agency meetings, fellowships, technical contracts and agreements technical information was transferred to developing countries on the following subjects:

- water rector fuel fabrication,
- behaviour of water reactor fuel,
- water reactor fuel performance modelling,
- improvement of water reactor fuel utilization,
- alternate fuel and advanced reactor fuel technology,
- interim spent fuel storage of water reactors.

Developed countries have also profited from the exchange of technical information in these areas. The most helpful exchange of expertise obtained in different Member States has been performed through the initiation of three Coordinated Research Programmes, namely, on "Investigation of Fuel Element Cladding Interaction with Water Coolant in Power Reactors" (CCI), "Examination and Documentation Methodology for Water Reactor Fuel" (ED-WARF) and "Development of Computer Models for Fuel Element Behaviour in Water REactors" (D-COM).

The actual problems of reactor fuel design, fabrication and performance have been discussed, in depth, at International Seminars on "HWR Fuel Technology" (Argentina 1983), "Practical Experience in the Application of Quality Control in Water Reactor Fabrication" (FRG 1984) and "Remote Handling Equipment for Nuclear Fuel Cycle Facilities" (UK 1984). Specialists' and Technical Committee Meetings were held to discuss separate problems of fuel performance, including "Post Irradiation Examination and Experience" (Japan, 1984), "Improved Utilization of Water Reactor Fuel with Special Emphasis on Extended Burnups and Plutonium Recycling (Belgium, 1984), "External Cladding Corrosion in Water Cooled Power Reactors" (France, 1985), and "Fuel Rod Internal Chemistry and Fission Products Behaviour" (FRG, 1985).

Experience obtained by Member States in advanced fuel fabrication technology and its performance regarding all types of reactors, in order to meet the requirements in the near future, were discussed at the Advisory Group Meeting on Advanced Fuel Technology and Performance (Switzerland, 1984). At the meeting, areas which may centre main efforts and future IAEA involvement in the improvement of nuclear fuels, were defined. Due to the recommendation of the Advisory Group, the Agency is holding Technical Committee Meetings on "Utilization of Thorium Based Nuclear Fuels - Current Status and Perspectives", December 1985 and on "Properties of Materials for Water Reactor Fuel Elements and Methods of Measurement", October 1986.

A Symposium on "Improvements in Water Reactor Fuel Technology, Fabrication and Utilization" will be held in Sweden, September 1986 in order to analyse the developments in the related areas since 1978, when a symposium on a similar subject took place in Czechoslovakia.

Thorium fuel utilization, technical status and development needs were considered in detail by the INFCE Working Group 8 (1977-1980). Some aspects of the thorium fuel cycle were considered at the IAEA Nuclear Power Conferences in Salzburg (1977) and Vienna (1982), as well as, the TCM on Improved Utilization of Water Reactor Fuel with Special Emphasis on Extended Burnups and Plutonium Recycling (Mol, Belgium, 7-11 May 1984), TCM on Advanced Light and Heavy Water Reactor Technology (Vienna, Austria, 26-29 November 1984) and others. In-depth consideration of the technology of some thorium fuels was dealt with, as mentioned before, at the IAEA Advisory Group on Advanced Fuel Technology and Performance (Würenlingen, Switzerland, 4-6 December 1984).

Until the present time, considerable efforts have already been made in the area of fabrication, utilization and reprocessing of Th-based fuels for different types of reactors, namely: by FRG and USA for HTRS; FRG and Brazil, Italy - for LWRs; India - for HWRs and FBRs. Basic research on thorium fuel and thorium fuel cycles is also being undertaken by Canada, China, FRG, France, Romania, USSR and other countries. Main emphasis has been given to the utilization of thorium fuels in once-through nuclear fuel cycles, but in some projects closed thorium/uranium fuel or thorium-plutonium cycles were also considered.

The advantages in the use of U-233 and thorium in advanced thermal designs are dealt with and the potential for obtaining a self-sustaining equilibrium with perhaps a slight margin for breeding. Recycling of reprocessed U-233 from irradiated FBR blankets and HTR fuels is also considered as a worthwhile direction in the alternate fuel cycles. The utilization of U-Th oxide fuels in HTRs is a proven technology of today.

However, in spite of considerable uranium savings the utilization of U-Th oxide fuels is still limited. There are several major reasons explaining the situation. Technical concerns include the need for full remotisation of irrdiated Th fuel reprocessing and U-233 fuel refabrication due to penetrating gamma radiation from U-232 daughter products and the need for development of three stream reprocessing technology for extraction of uranium isotopes and Pu-239. The impact of these factors on the economics of the fuel cycle is also very essential.

The current situation on the uranium market seems at, first sight, unfavourable to the implementation of thorium nuclear fuels. The near-term uranium outlook is for oversupply and low prices, as in the past few years. As a consequence of the low prices (spot market price is now about $ 44/kg U in comparison with $ 112/kg U in 1977-79), uranium exploration continues to decline in WOCA. Correspondingly Reasonably Assured Uranium Resources following to uranium exploration expenses also continue to decline.

As a result, annual uranium production capability of WOCA from existing and firmly committed mines and mills is expected to be able to fill the annual reactor requirements before the end of this decade. Thereafter, additional mines and mills will be required. As lead times for uranium exploration and mining projects are 10 to 15 years and are still growing, higher levels of exploration uranium efforts will be needed if future reactor requirements are to be met. The alternative is the implementation of thorium fuel cycle.

Another aspect merit to be discussed is the potential for the reduction of uranium demand, due to improvements in reactor and fuel performance. In a paper by Stig Sandklef (Swedish State Power Board) presented at the Xth Uranium Institute Annual Symposium, 3-5 September 1985 this analysis is given for the year 1995. Advances in fuel management schemes include: extended burnup, low neutron leakage loading pattern, improved fuel designs, axial burnup shaping, natural uranium blankets, fuel reconstitution, initial core reinsertion, coast down operation, spectral shift operation, relaxation of safety constraints. Extended operating cycles and influence of capacity factors were also anlysed. It is seen that 18% uranium and 10% of separative work could be saved in 1995 if the above-mentioned advances in fuel managment are done. At the same time, implementation of extended operating cycles, increasing of capacity factor and increased power will lead to a certain increase in uranium demand and separation work. The total result shows that integral reduction in uranium demand and separative work will be very small.

This means that the world situation with uranium production and demand could be one of the factors endorsing R and D on further investigation of thorium fuel. Due to the delay in the construction of power fast breeder reactors, utilization of thorium/plutonium fuel in water cooled power reactors could be considered as one of the means of burning plutonium coming from reprocessing plants, thus, avoiding its long-term storage.

It is obvious that some uncertainty in scale of future development in thorium fuel cycles still exists and it influences the volume of national efforts in thorium resources exploration, in development of thorium fuel technology and basic research on physical, chemical and nuclear properties of thorium fuels.

Under these circumstances the Agency has contributed and will contribute to the collection and exchange of information in this area and to the co-ordination of national efforts in order to maintain and improve the knowledge of all aspects of utilization of thorium as an alternative nuclear fuel.

# THORIUM UTILIZATION IN PWRs: STATUS OF WORK IN THE COOPERATIVE BRAZILIAN/GERMAN PROGRAM

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**Abstract**

The major results of the program "Th-Utilization in PWR's" are presented and discussed: The investigations show that the standard KWU-PWR can accomodate (Th,U)$O_2$ and (Th,Pu)$O_2$ fuel without changes in the fuel element design, in 3 and 4-batch operation scheme, without penalties in the reactor performance. An advanced fuel fabrication scheme using direct pelletizing methods out of ex-gel microspheres has been developed on laboratory scale. The (Th,U)$O_2$ test fuel produced satisfies PWR-specification and is currently undergoing irradiation testing. Thermal and mechanical design of fuel pins with thoria-based fuel is validated by the current instrumented single rod irradiation test. Cold laboratory investigations indicate that the (Th,U)$O_2$ PWR-fuel can be reprocessed using presently known technology, including the chop-leach technique and modified THOREX extraction process.

## I. Introduction

The utilization of thoria fuel in thermal reactors can result, due to a high $\eta$-value of U-233, in higher conversion ratios and consequently in better resource utilization compared with

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the conventional uranium fuel. This holds particularly for advanced reactor types specifically designed for thoria application, as shown by numerous R+D activities in many countries, among others Brazil and Germany. But also in Light Water Reactors substantial advantages are anticipated in a case of a closed fuel cycle /1/. However the thorium fuel cycle technology was not as mature to permit well based feasibility statements in this area.
Therefore the cooperative R+D program on "Thorium Utilization in Pressurized Water Reactors" between NUCLEBRAS/CDTN on the Brazilian side and KFA, KWU and NUKEM on the German side aims to an improvement of our knowledge on this subject and contributes to fullfill in practice the "Governmental Agreement on Cooperation in the Field of Science and Technology" of 1969 and the "Memorandum of Understanding between KFA and NUCLEBRAS" of 1978 / 2 - 5 /.

The objectives of the program running since mid 1979 are:

- to analyse and prove the thorium utilization in pressurized water reactors,

- to manufacture, test and qualify Th/U and Th/Pu fuel rods and elements under operating conditions,

- to study the back end of thorium fuel cycle by reprocessing spent Th-containing PWR fuel elements where it seems to be reasonable, or by applying available storage techniques.

The program utilizes as much as possible the existing technologies and available know-how for fuel element and nuclear core design, fuel element fabrication and reprocessing. Therefore the existing techniques and equipment for the fuel cycle in High Temperature Reactors, i.e. manufacturing and reprocessing of fuel, as well as for the fuel cycle in Light Water Reactors, i.e. fabrication and head end treatment, have been applied.

## 2. Nuclear Core Design

The KWU standard 1300 MWe PWR is the reference reactor for the present study. The results of nuclear core design are based on Th-fuel cycles using highly enriched uranium (HEU) or LWR recycle Pu as initial fissile material / 1, 5 /. It was found that the present design KWU-type PWR can be operated without changes and restrictions with 3 and 4-batch loadings in open and closed fuel cycle modes with all types of fissile material investigated / 1, 6 /.

It is to be anticipated that the realistic way of introducing Th-fuel to a PWR would be via a partial and later complete Th-fuel assembly reload / 1, 7 /. Results gained so far in the cooperative program show that no changes of the Th/HEU fuel assembly design - in comparison with the standard U-fuel assembly - to be loaded into an U-core is necessary. The use of Pu instead of HEU reveals also some interesting feature. Fig. 1 shows the relative energy production of Th/Pu-fuel inserted in a standard PWR core. However the high fission cross sections of Pu combined with a high thermal flux of neighbouring U fuel assemblies would cause unacceptable power peaking / 1, 7 /. Thus, Th/Pu-MOX fuel assemblies - as used for Pu recycling via U/Pu-MOX assemblies - need 2 - 3 types of fuel pins with a different fissile material content as shown in Fig. 2, to avoid local power peaking when loaded adjacently to U-fuel assemblies / 1, 7 /.

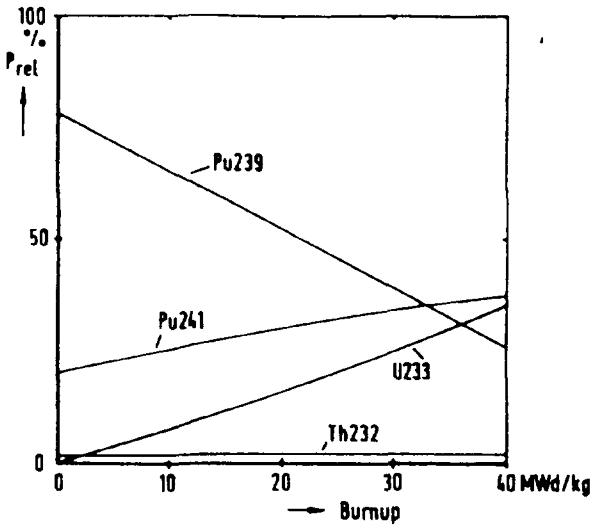


**Fig. 1** Relative Energy Contribution of the Isotopes of Th/Pu Fuel

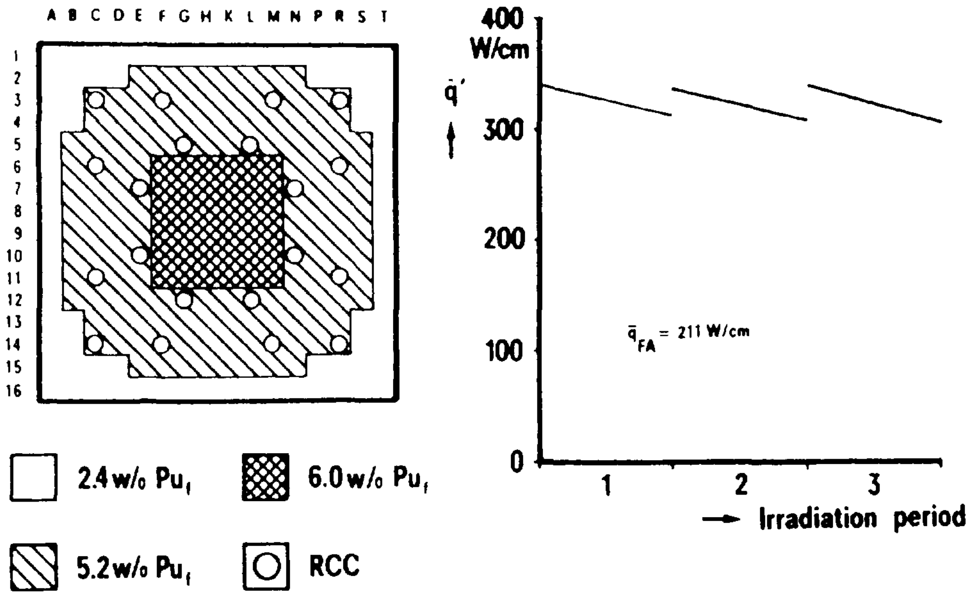


Fig. 2 Th/Pu Fuel Assembly with 3 Enrichment Zones

## 3. Strategy investigations

Strategy investigations are based on the need for natural uranium and separative work, calculated from the fuel cycle data of the nuclear core design / 1, 6 /. Recycling is found essential only for the Th/HEU to achieve substantial savings.

Recycling, however, may not be needed in the case of Th/Pu-cycles to realize savings in the need for natural uranium and separative work. Considerable saving come from Th/Pu-fuel via the replacement of U-fuel in existing reactors. The Th/Pu-fuel offers the potential of extending burn up to values where early closure of the fuel cycle by reprocessing Th-fuel could be avoided.

## 4. Fuel Technology

As reported above the present available fuel assembly design for KWU 1300 MWe PWR can be used to full extent to realize a thoria based fuel element. Thus, there is only a need for developing the oxide fuel manufacturing technology. This result from the nuclear core design is to be regarded as a major benefit for the Th use in commercials PWRs.

Since there are large experiences available for chemical conversion processes for (Th,U)$O_2$ fuel production from HTR work / 8 / and for direct pelletizing of $UO_2$ / 9 / from PWR technology it was emphasized to use, as shown in figure 3:

- the chemical ex-gel conversion process in combination with

- standard pelletizing processes for $UO_2$ ex-AUC.

Selecting this combination offers the unique possibility to use the existing manufacturing equipment and quality assurance programs from commercial PWR and HTR / 8, 10, 11 /.

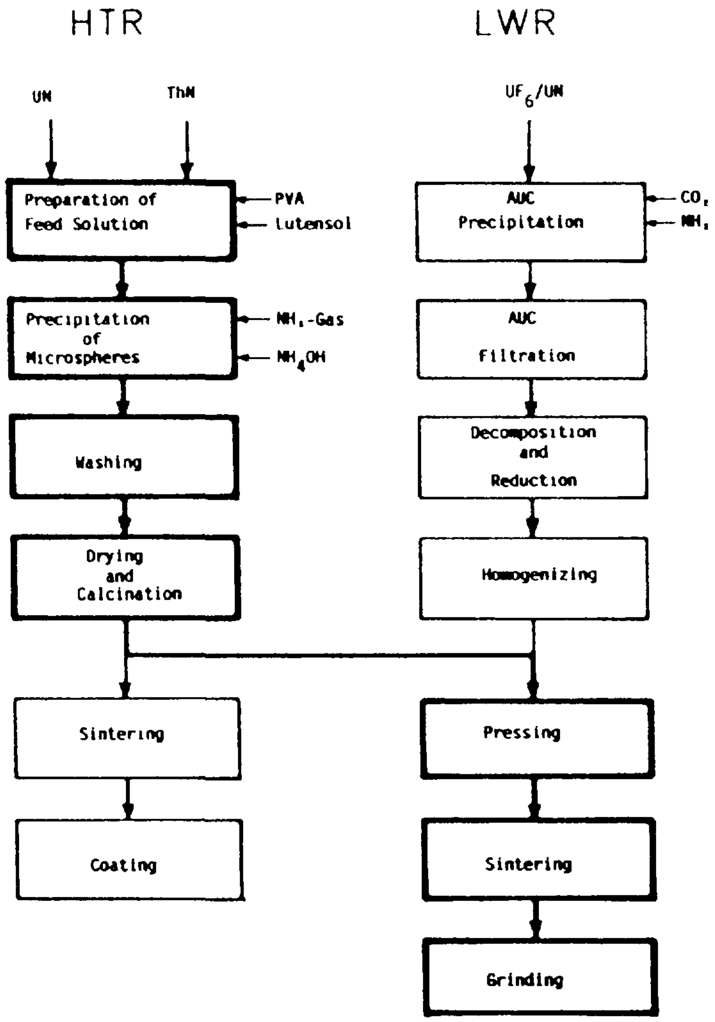


Fig. 3 Combination of Available Technologies for the Manufacturing of Mixed-oxide Fuel

Table 1 : Criteria for (Th,U)$O_2$-LWR-fuel for high burnup

| **Criteria** | | **High burnup targets for $UO_2$** | **High burnup targets for (Th,U)$O_2$** | **Status of (Th,U)$O_2$-fuel properties** | |
|---|---|---|---|---|---|
| Density | (g/$cm^3$) | (95 % td) 10.40 | 9.5 | 9.00 - 9.80 | |
| Open porosity | (Vol.-%) | 0.5 - 1 | 0.5 - 1 | 0.5 - 3 | low op.por. can be controlled |
| Grain size | (µm) | 10 - 20 | 10 - 2 | 10 - 15 | |
| Average pore diameter | (µm) | 2.5 at 10.40 g/$cm^3$ | not yet evaluated 1)<br>(2.0 - 3.0 preliminary) | 1.8 - 3 | |
| log. scattering factor of pore size distribution | ./. | 0.25 - 0.30 | 0.25 - 0.30 | 0.25 - 0.3 | |
| Shape factor 2) of pores | | > 0.7 | > 0.7 | > 0.7 | |

1) the results of FRJ-2 tests are necessary to confirm these values

2) $\frac{1}{F} = \frac{\text{Circumference of a pore in the microsection}}{\text{Circumference of a circle with the same area}}$

Table 1 summarized the target properties of the fuel with respect to the burn up defined in the nuclear core design and strategy studies / 1, 11 /. Two major areas of concern have been identified in the early stage of $(Th,U)O_2$-fuel development:

- the improvement of the fuel microstructure, especially to avoid the so-called black-berry structure.

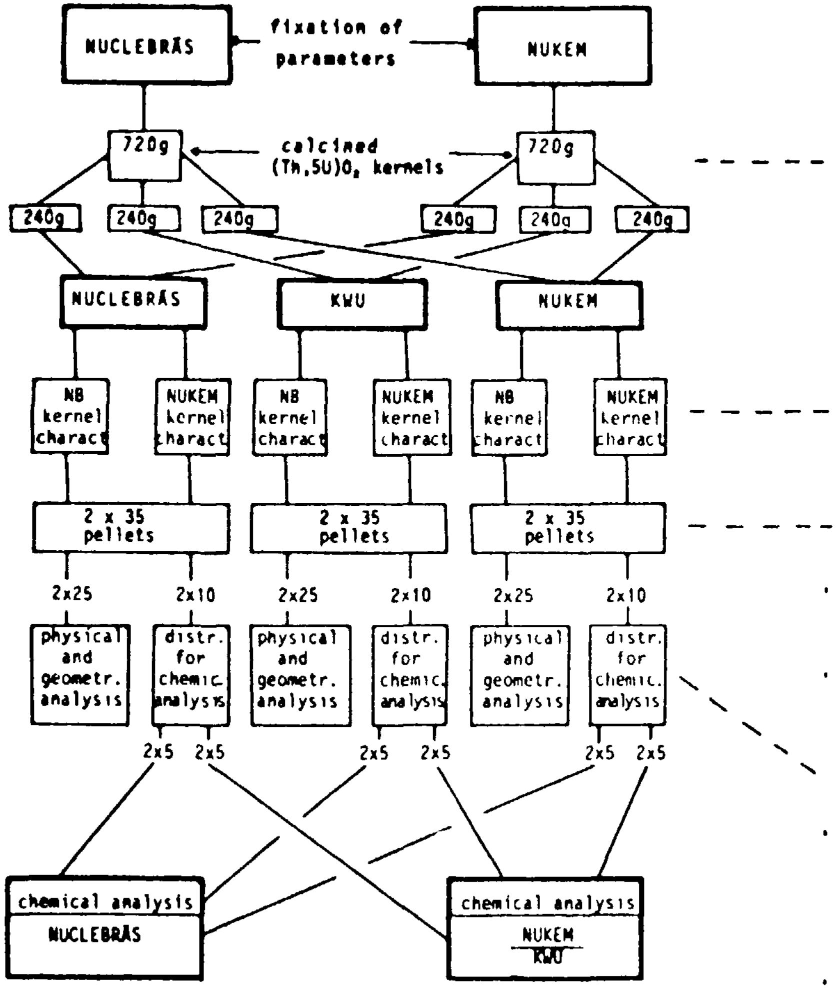
- the adjustment of the pore size distribution due to requirements given in table 1.

Details of the performed R+D-work have been reported earlier / 11, 10, 12 /. It turned out very soon that standard ex gel kernels are not suitable for the direct pelletizing process. However, the use of carbon black as additive to the feed solution permitted the adaptation of the ex gel kernels to the requirements for pressing and sintering, as well as to the control of density, microstructure and especially of the pore size distribution. The use of carbon black needs, of course a treatment for its removal after the kernel precipitation. The carbon black is completely oxidized during the calcination to $CO_2$ leaving the fuel kernel easily. Selecting its particle size distribution adequately, the burning carbon black leaves not only "press pores" behind improving the pelletizing, but also pores needed for the accomodation of swelling /shrinkage behaviour during burnup / 16 /. Table 2 compiles the results from test fuel production.

The reliability of the results from the R+D-program for $(Th/U)O_2$-fuel the fuel development is assured by a "Round Robin Test" involving a cross check of the manufacturing procedures performed at the different laboratories, as well as a cross check of the properties of the prematerial and the final pelletized fuel (Fig. 4).

Table 2 : Comparison of property-requirements and results from test fuel production

| **Properties** | **Unit** | **Specified** | **Observed** |
|---|---|---|---|
| (Th+U)-Content | w/o | min. 87.7 | 87.83 |
| U-Content | w/o HM | 5 ± 0.1 | 5.03 |
| Stoichiometry | MolO : MolU | 1.99 - 2.05 | 2.014 |
| $H_2$-Content | $Nmm^3/g\ (Th,U)O_2$ | max. 10 | 9.4 |
| Residual Gas Content | $Nmm^3/g\ (Th,U)O_2$ | max. 40 | 17.6 |
| Impurity Content: | | | |
| F | ppm (max.) | 10 | 3 |
| Cl | ppm (max.) | 15 | 6 |
| C | ppm (max.) | 100 | < 10 |
| S | ppm (max.) | 500 | 316 |
| N | ppm (max.) | 30 | < 14 |
| Ca | ppm (max.) | 100 | < 51 |
| Si | ppm (max.) | 100 | 51 |
| Ni | ppm (max.) | 50 | 10 |
| Fe | ppm (max.) | 100 | 23 |
| Boron equivalent (Gd, B, Sm, Eu, Cd, Li) | ppm (max.) | 1 | 0.911 |
| Density | $g/cm^3$ | 9.5 ± 0.15<br>- 0.2 | 9.47 - 9.58<br>adjustable |
| Mean grain size | µm | 4 - 15 | 8 - 15 |
| Pore structure | in line with requirements | | |
| Resintering Test | $g/cm^3$ | max. 0.20 | 0.08 |
| Surface Roughness | µm | 2 | 0.89 - 1.14 |



<u>Kernel fabrication</u> (2x5 batches with 720g each)

<u>Kernel characterization</u>
(each partner)

- diameter n=150
- crushing strength n=30
- particle density 1X
- BET 1X

<u>Pellet fabrication</u> (5x2 sinter batches with 35 pellets for each partner, totally 30 sinter batches)

- pressing of green pellets (each partner)
  - $\rho_{green}$ accord. to indiv. working point
  - $\Delta\rho_{green}$ should be as low as possible
- sintering of pellets (each partner)
  - sintering of pellets
  - $\rho_{sint.}$=9.50 g/cm³ nominal value
  - sintering parameter 1700°C, 2 h, $H_2$ or $H_2$ mixture

<u>Pellet characterization</u>

- physical and geometrical characterization of pellets (no exchange of samples)
  - geometry, density/open porosity, and roughness ... total
  - resinter test ... 2x
  - microstructure
    - pore structure ... 2x
    - grain size ... 2x
    - pore size distr. ... 1x
- chemical characterization: separate Round Robin Test (not all chemical analitical methods available at all partners, exchange of samples in these cases)

Fig. 4 Detailed Procedure of the Round Robin Test

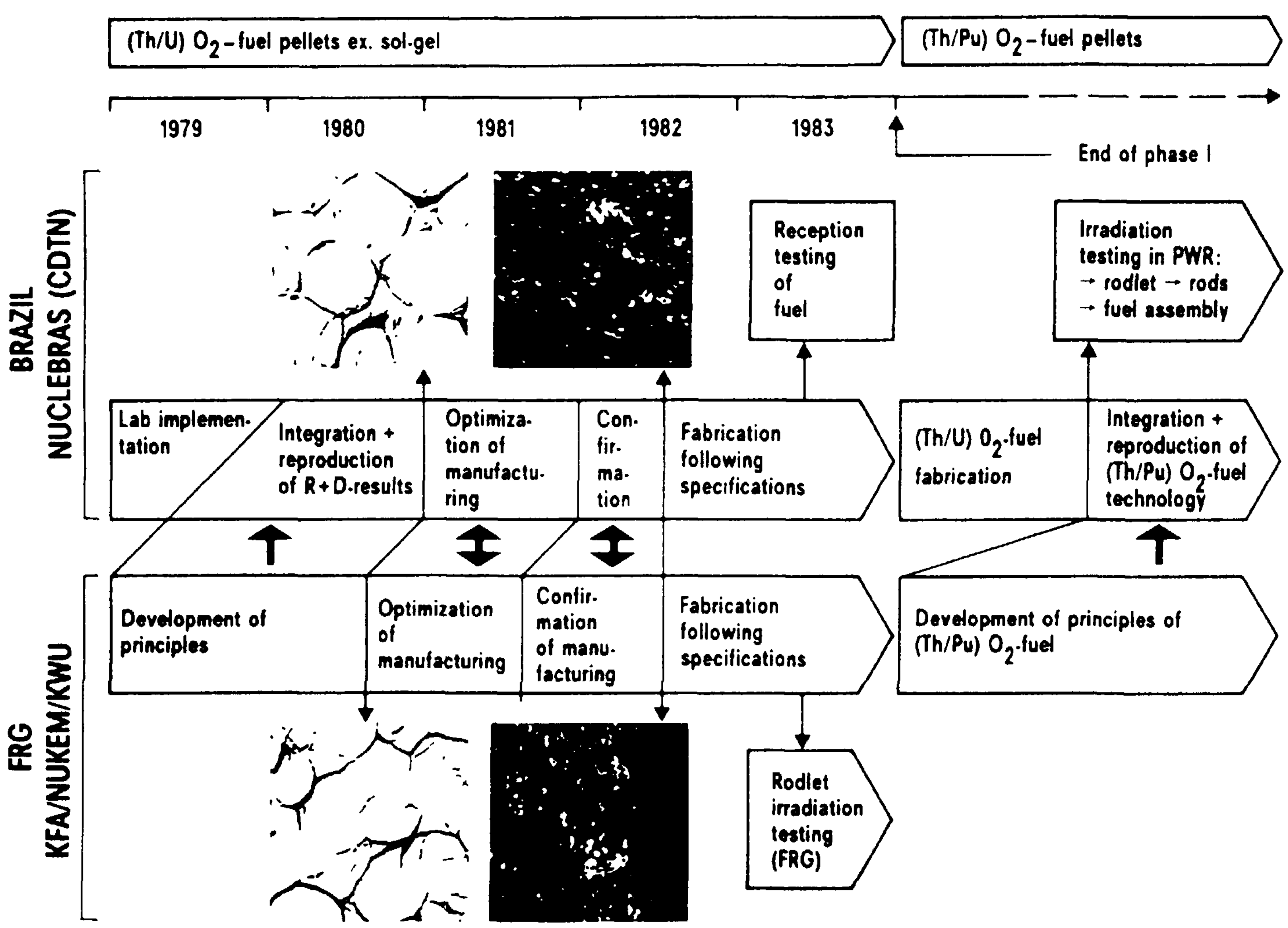


Fig. 5 Th-Utilization in PWR: Fuel Development and Fuel Fabrication

After completion of the $(Th,U)O_2$-fuel technology development the transfer of the gained knowledge to $(Th,Pu)O_2$-fuel production is initiated by the use of Ce as a Pu-simulator. Ce has proved to be an good Pu simulator under the aspect of its chemical, ceramurgical and physical properties / 13 /. The use of Ce as simulator allows for the development of the $(Th,Pu)O_2$ technology at a low cost basis. Only the final confirmatory investigation will use Pu. Fig. 5 comprises an overview on the strategy persued in fuel development and fuel fabrication.

## 5. Thermal Fuel Rod Design and Performance Prediction

Since the fuel assembly design and the linear heat rating of the Th-containing bundles are practically identical, thermal design work and performance prediction can be concentrated on the fuel rod analysis. The thermal conductivity for $(Th,U)O_2$ was determined also for the fuel with its typical microstructure. The results show that it is very near to the data known from $UO_2$ / 11 /.

To describe the fuel behaviour during burnup, the mechanistic densification-swelling model developed for $UO_2$ / 14 / has been adapted for thoria based fuel / 6 /. Also the CARO-code in its most recent version CARO D 5 / 15 / has been adapted also to describe in service fuel rod performance.

After completion of the code adaptation and having measured all necessary fuel properties, the thermal design and performance prediction were made for the test fuel rods currently under irradiation in the FRJ-2 at Jülich. The work resulted so far in a fairly good prediction for the beginning of life of the test fuel rods.

## 6. Irradiation Testing

The irradiation testing is performed in two complementary programs:

- an instrumented short length single pin irradiation in the experimental reactor FRJ-2 at KFA Jülich/FRG

- a segmented fuel rod and full length fuel rod irradiation in a host FA inserted into a commercial power reactor in Brazil.

The strategy behind these two branch program is to have a fast performing single rod irradiation, evaluating the burnup performance of the as-developed fuel and to generate the data base needed for licensing the pathfinder irradiation in the power reactor. The pathfinder irradiation provides the performance data under power reactor conditions. The single rods will be examined in a detailed PIE, whereas the pathfinder fuel rods will be inspected initially by available pool inspection techniques / 16 /. However, pool inspection is able to indicate the existence of any clad degradation, also from the inside, or the existence of defects if there are any. Thus, the results from both test series provide a reliable data base to assess the burnup performance of fuel pins with $(Th,U)O_2$ under power reactor conditions.

Table 3 gives an overlook on the single rod irradiation program and its actual status. The defect in the test LV 9.6-E 62 is due to a failure in the instrumentation.

Table 3 : Overview on FRJ-2 irradiation experiments
Values for E 61 in brackets were reached
till 30 Sept. 84

| Rig. No. | Test fuel rod (position) | Max. rod power | | Burnup | Irrad. time | PIE |
|---|---|---|---|---|---|---|
| | | from W/cm | till W/cm | GWd/tHM | d | |
| LV9.6-E60 | TT80 (top) | 167 | 346 | 6,82 | 116,1 | completed |
| | TDT81 (bottom) | 391 | 430 | 8,22 | | |
| LV9.6-E61 | TT82 (top) | 111 | 234 | 8,2 | | in progress |
| | TDT83 (bottom) | 278 | 308 | 10.0 | 176 | |
| LV9.6-E62 | TT84 (top) | 112 | 235 | 1,5 * | 34 | in progress |
| | TDT85 (bottom) | 276 | 300 | 1,26* | | |

* terminated after a defect
in the instrumentation

Table 4 summarizes the pathfinder irradiation program. In order to investigate the operational behaviour and the dimensional stability of the fuel at different temperatures, the clearances between the fuel pellet and the cladding shall be varied within the full tolerance scale, i.e. in the range of 120 to 220 µm, in the segmented rods. Thereby first priority shall be given to the medium clearance of 170 µm. A full-length rod shall be irradiated to show the operational performance of a standard fuel rod with that type of thoria fuel.

To get information on the fuel performance in the whole burn-up range to 45 GWd/kg HM segmented fuel rods shall be irradiated for 1 to 4 operational cycles. This means, one segmented fuel rod shall always be un-loaded during the refueling shutdowns of the power plant. The full-length fuel rod shall be irradiated for altogether 4 operational cycles and finally be examined in the spent fuel pond of the power plant. Intermediate examinations are planned to be performed during each refueling shutdown if the time schedule for the refueling allows.

## 7. Investigation on the back end of the fuel cycle

For the most interesting case - the use of LWR recycling Pu as fissile material in Th fuel - worth mentioning savings are already achieved in the once through-put away cycle. Thus no reprocessing is needed for this fuel. But the long term storage of spent thoria fuel has to be still considered. Preferentially dry storage techniques will be used. Based on the excellent experience on the dry storage of spent $UO_2$ containing LWR-fuel / 17 /, no major problems should arise in this field. Only the different decay heats from $(Th,Pu)O_2$, in comparison to the $UO_2$ fuel, require an assessment at which periods after shut down dry storage may start in both versions: unconsolidated or consolidated manner. The problem areas of final disposal have not yet been addressed.

Using HEU as fissile material reprocessing is needed to achieve resource conservation (U- ore savings) and savings in the fuel

Table 4 : Overview on the pathfinder irradiation to be performed in ANGRA I (Beginning with 3. reload 1988)

| **Test rod identification** | **Test rod insertion period** | | | |
|---|---|---|---|---|
| | **host fuel assembly No. 1** | | | **host fuel assembly No. 2** |
| | **1 cycle** | **2 cycle** | **3 cycle** | **4 cycle** |
| Segmented fuel rod No. 1 | X<br>target burnup<br>11 GWd/tHM | | | |
| Segmented fuel rod No. 2 | | X<br>target burnup<br>27 GWd/tHM | | |
| Segmented fuel rod No. 3 | | | X<br>target burnup<br>33 GWd/tHM | |
| Segmented fuel rod No. 4 | | | | X<br>target burnup<br>45 GWd/tHM |
| Full length fuel rod | | | | X<br>target burnup<br>45 Gwd/tHM |

cycle cost. A scoping study on reprocessing showed the following results:

Cold dissolution experiments have shown that in the dissolution of (Th,U)$O_2$ fuels by Thorex solution, the Zircaloy clad is also dissolved to some extent and, besides this the dissolution time is 30 % increased if Zircaloy cladding is present. Nevertheless, final statements about dissolution behaviour of fuel with high burn up can only be made after hot experiments / 6 /. Those experiments are in progress.

For an optimization of the solvent extraction processes, distribution data covering the whole range of interest have been evaluated. Interpolations and extrapolations are possible by a computer program. Considerations about the readioactivity of reprocessed U-233 fuel from power reactors on the one side and the experience gained so far with the THOREX process variants on the other side lead to the proposal of a single-cycle THOREX process / 6 /.

## 8. Conclusions

The major results of the program work, confirmed in all related working areas, can be summarized so far:

- The standard PWR may use (Th,U)$O_2$-fuel and even (Th,Pu)$O_2$ fuel without any changes within the reactor system, within the core or in the fuel assembly design.
- Nearly the same performance of the reactor can be expected using (Th,U)$O_2$-fuel or (Th/Pu)$O_2$ as for of $UO_2$-fuel in a standard PWR nearly
- (Th,U)$O_2$-fuel can be easily manufactured within the same specification limits used for $UO_2$-fuel. A slightly modified ex-gel process from the HTR-technology as a chemical conversion is combined with the standard pelletizing, sintering and grinding process which is in commercial use for LWR-fuel fabrication. The development of (Th,Pu)$O_2$ fuel by means of Ce as simulator material is in good progress.

- Thermal and mechanical design of fuel rods containing thoria-based fuel can be done as well as with standard $UO_2$ fuel pins. The performance predictions made for the irradiation tests have been validated so far by the irradiation results.
- A favourable strategy to avoid the need of early reprocessing and to strive for worthmentioning savings in U-ore might be the use of $(Th,Pu)O_2$ fuel in a "once through-put away cycle" with extended burnups. Problems encountered with long term storage of spent Thoria-based fuel are very similar to that known from urania-fuel. It should not pose any extra problems.
- Thoria fuel may in the HEU-cycle require reprocessing to realize substantial savings in fuel cycle costs. Reprocessing of spent LWR-fuel with $(Th,U)O_2$-fuel is principally feasible using reprocessing schemes derived form experience gained in the context with HTR work and in combination with a chop-leach technique using the Thorex solvent.

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# THORIUM CYCLE IN UNMODIFIED PWRs

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## Abstract

For more than ten years the Commissariat à l'Energie Atomique and the Electricité de France have jointly carried out experimental and design studies in different fields : HTRs, MSRs, PWRs.

With regard to the PWRs, the studies first concerned the use of Plutonium with Thorium to start the cycle. A satisfactory solution has been obtained by recycling the Plutonium with Thorium as from the first core loading. The solution proposed consists of loading the whole reactor with Th-Pu assemblies from the first core and no longer using the standard checkerboard loading ; the assemblies are loaded in rings. Two positive points appear : burnable poisons are no longer necessary and the assemblies are no longer divided into several zones (as with Plutonium recycle with Uranium in reloadings). There are two rather negative characteristics : the second generation Plutonium may be used only in FBRs and the conversion factor is rather small. Nevertheless the proposed solution gives acceptable pin power peak, temperature coefficients, etc...

Satisfactory solutions have also been obtained for cores loaded with Th-U233 assemblies. The main problem is, in this case, the moderator temperature coefficient. Different solutions may be found to solve this problem : we have chosen the one which gives simultaneously an acceptable moderator coefficient and a conversation factor improvement (the use of spectrum shift control).

## INTRODUCTION

For some years now Electricité de France (EDF) has been carrying out studies concerning the Thorium cycle. These studies, begun in 1969 in relationship to High Temperature Reactors (HTR), have been performed jointly with the Commissariat à l'Energie Atomique (CEA) /1/ : the Nuclear Studies Center at Saclay for theoretical studies concerning designs and codes developments ; the Nuclear Center at Cadarache for the integral experiments made necessary by the search for improvements in the accuracy of the basic nuclear data. These studies were then completed in the field of Molten Salt Reactors (MSR) (from 1975 on) and more recently (from 1979 on) in the field of Light Water Reactors (LWR) /2/ /3/.

## PLUTONIUM RECYCLE WITH THORIUM IN UNMODIFIED PWRs

To start the Th-U233 cycle it is necessary to begin with a Thorium-Fissile Material couple from which the fissile element may be easily obtained. Two possibilities may be considered :

The High Enriched Uranium/Thorium/U233 cycle
The Plutonium/Thorium/U233 cycle.

The first case seems to be inconsistent with the present development of PWRs using Low Enriched Uranium. Furthermore, since important Plutonium quantities will be available it is obvious to try to use this material to start the Thorium cycle ; moreover the Plutonium recycle with Thorium appears as the logical continuation of the studies concerning the Plutonium recycle /4/ /5/ and the uranium recycle /6/ /7/.

The use of this PWR Plutonium, containing rather important even isotopes proportions, is of course particularly favourable in Fast Breeder Reactors (FBRs). The best way to use the Plutonium is thus clearly defined : it must be used in the LMFBRs. Nevertheless, taking into account the important production of Plutonium (250 kg/GWe/year in a PWR) and the rather slow development of LMFBRs, it seems natural to try to find uses for this element in thermal reactors with Uranium or with Thorium /8/. In addition this recycling may be considered as an attractive storage before use in LMFBRs /9/ ; the quality of the Plutonium, with regard to the FBRs is in fact only very little affected by the recycling /10/.

## Reactor characteristics

The PWR considered for our studies is the 920 MWe Bugey 2 reactor. In order to introduce no other problems than the use of Thorium, we have not modified the assemblies characteristics.

The reactor contains 157 17 x 17 fuel rod assemblies. The initial enrichments for the three zones are respectively : 2.1% ; 2.6% ; 3.1%.

## Physical characteristics of the $PuO_2$ - $ThO_2$ lattice

The moderator ratio optimum for a $ThO_2$ - $PuO_2$ lattice, with a $PuO_2$/($PuO_2$ + $ThO_2$) ratio equal to 7.27% (this concentration allows, at equilibrium, the same cycle length to be obtained as with the standard Uranium cycle), is approximately equal to 8 (Figure 1) instead of 4 for the Uranium fuel. The spectrum is particularly hard (Figure 2).

The fact of keeping the 1.92 standard moderator ratio leads to an important Plutonium concentration.

The changes in heavy nuclides concentrations as a function of the burn-up is given on figure 3.

## Use of $ThO_2$ - $Pu_2$ assemblies in the reloadings of a Uranium core at the equilibrium

If we try to put $ThO_2$ - $PuO_2$ assemblies in the reloadings of a reactor which is at equilibrium (in the standard Uranium cycle), as with the conventional Plutonium recycle for instance, important difficulties appear.

An acceptable pin power peak has not been obtained during the transitory cycles between the Uranium loaded core and Pu - Th loaded core. It is due to strong absorption differences beetween $UO_2$ assemblies and $ThO_2$ - $PuO_2$ assemblies. The use of assemblies divided into three zones (4% of Pu in the outer zone, 6% of Pu in the intermediate zone and 9% of Pu in the central zone) is not sufficient to obtain an acceptable value for the pin power peak.

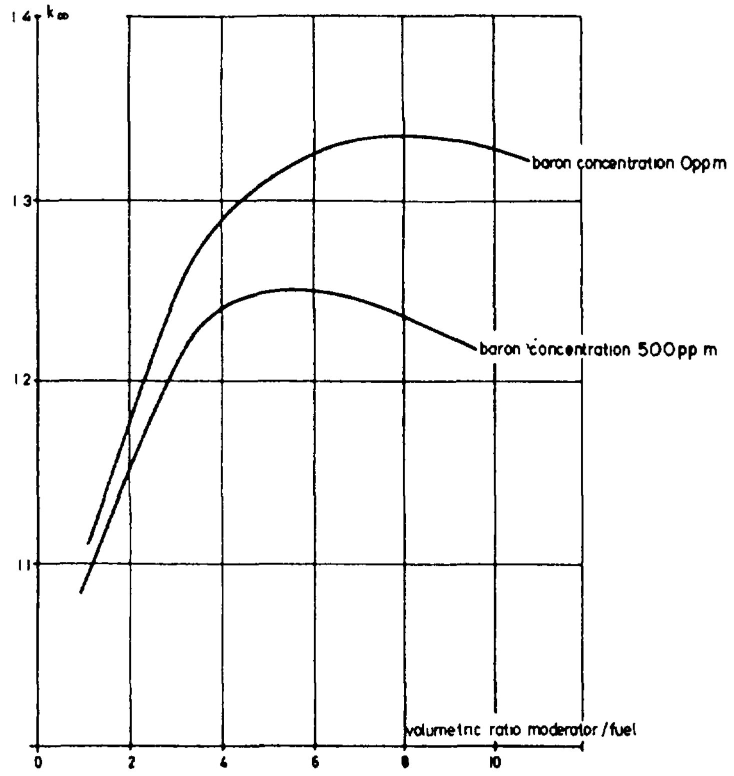

Fig 1 Multiplication factor k∞ , of the $ThO_2$ - $PuO_2$(7.27% Pu) lattice as a function of volumetric moderator/fuel ratio

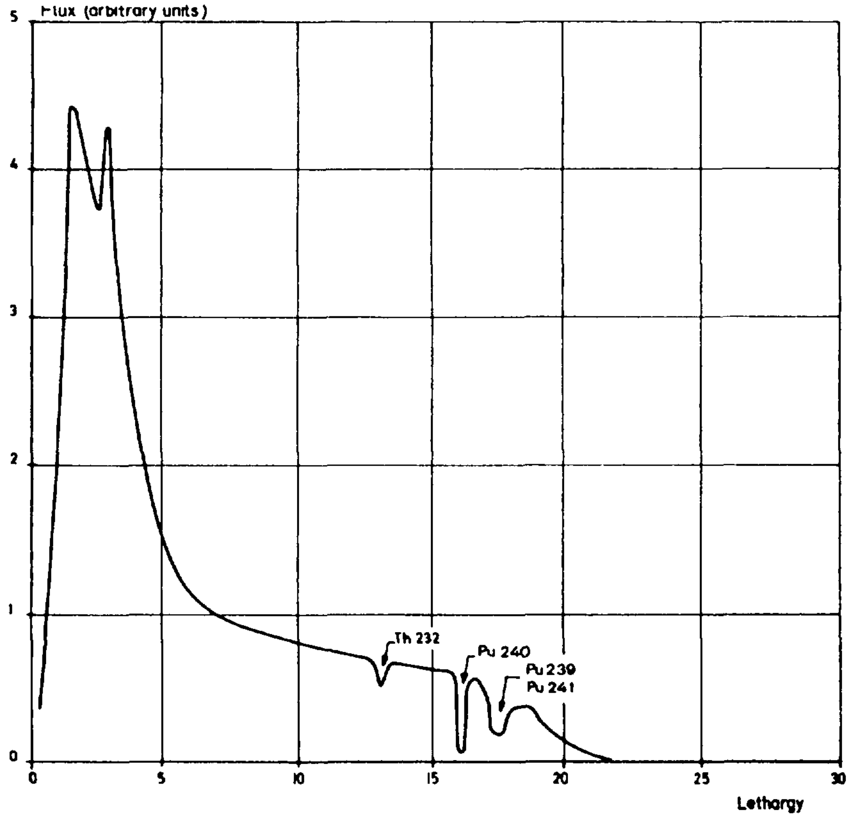

Fig 2 Energy distribution of neutrons in the $ThO_2$ - $PuO_2$ lattice

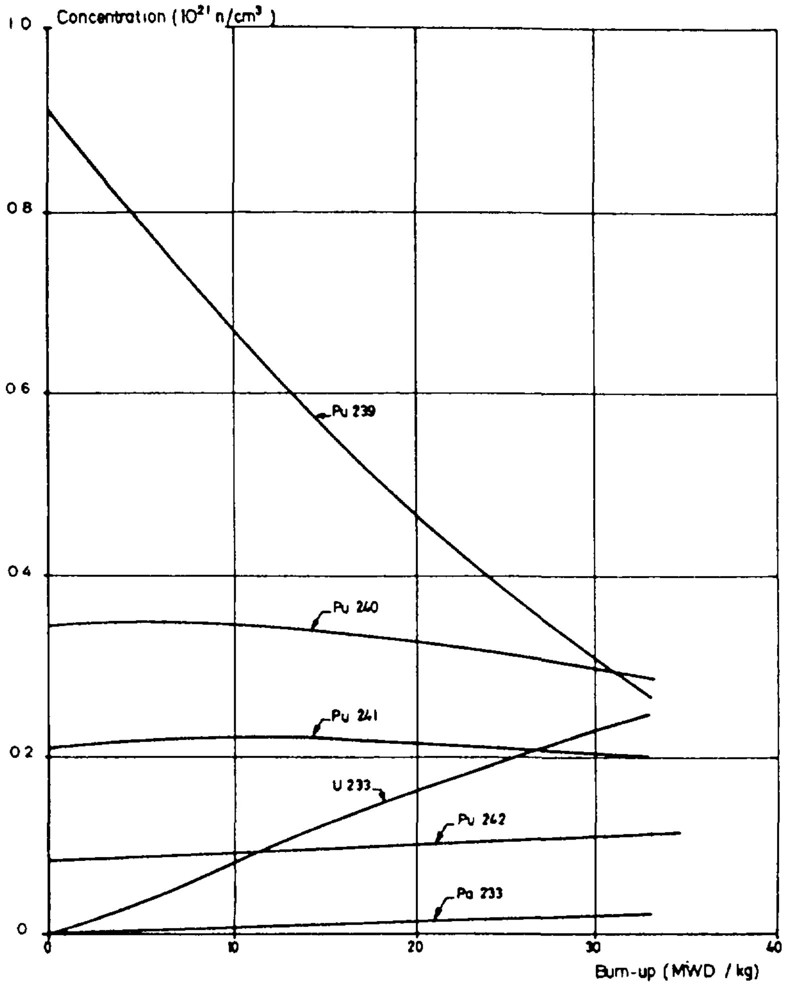

Fig 3 Changes of Pu, U, Pa concentrations in a $ThO_2$-$PuO_2$ lattice

Other solutions might be studied, for instance :

- smaller Pu concentrations giving sthorter cycle lengths (but we want in this study to keep the standard 10.6 GWD/T value)

- use of burnable poisons

- etc...

We have prefered to consider an absolutely different solution : the $ThO_2$ - $PuO_2$ assemblies will be introduced into the whole reactor from the first loading.

## Use of $ThO_2$ - $PuO_2$ assemblies from the first core loading

When the reactor is entirely loaded with $ThO_2$ - $PuO_2$ assemblies, absorption transitory (therefore neutron spectrum transitory) at the bottom of assemblies are of course eliminated.

In these conditions it has been possible to find a non-standard core loading map, with rings configuration (Figure 4), with only very simple assemblies (they are not divided into several zones). Furthermore, it has been shown, in the detailled study, that the use of burnable poisons (as with Uranium cycle) was not necessary.

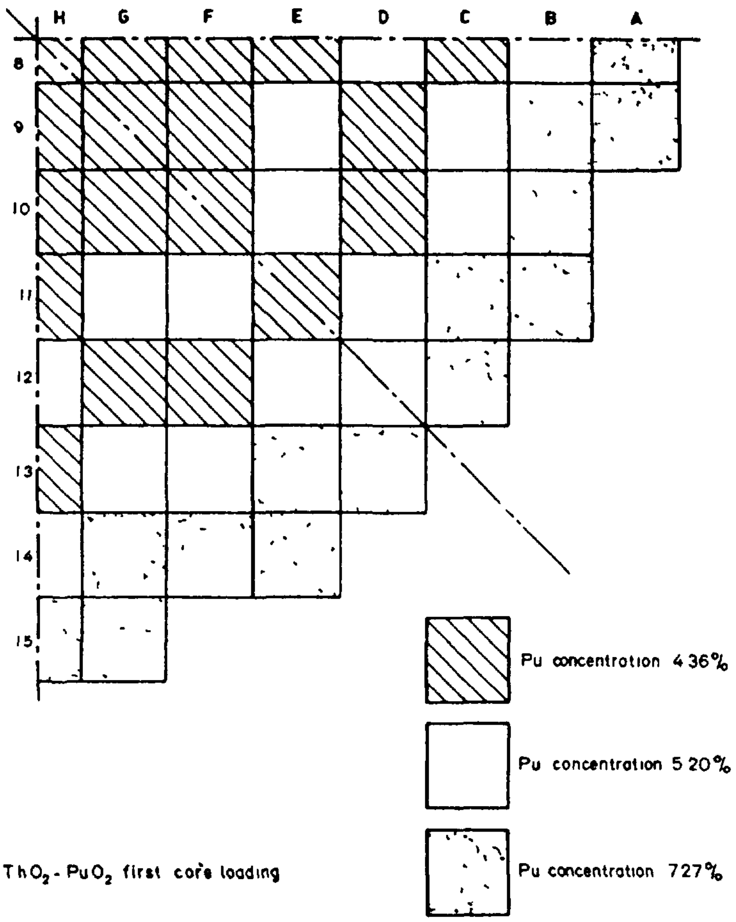

Fig 4 $ThO_2$-$PuO_2$ first core loading

On figure 5 are represented the three assembly types multiplication factor changes as a function of burn-up and the equilibrium core loading map is given on figure 6.

For the equilibrium core :

The Boron efficiency is equal to - 3.13 pcm/ppm (BOL) and - 3.70 pcm/ppm (EOL).

The Doppler coefficient is equal to - 4.45 pcm/°C (BOL) and - 3.54 pcm/°C (EOL).

The moderator temperature coefficient is equal to - 36 pcm/°C (BOL) and - 53 pcm/°C (EOL).

The delayed neutron proportion is equal to 330 pcm.

The prompt neutron generation time is equal to 0.44 $10^{-5}$ s.

The 48 control rods are sufficient to assure the antireactivity margins (2580 pcm at the beginning of cycle and 2850 pcm at the end).

The different heavy nuclide masses for the first core and for the core at the equilibrium are given respectively in tables 1 and 2.

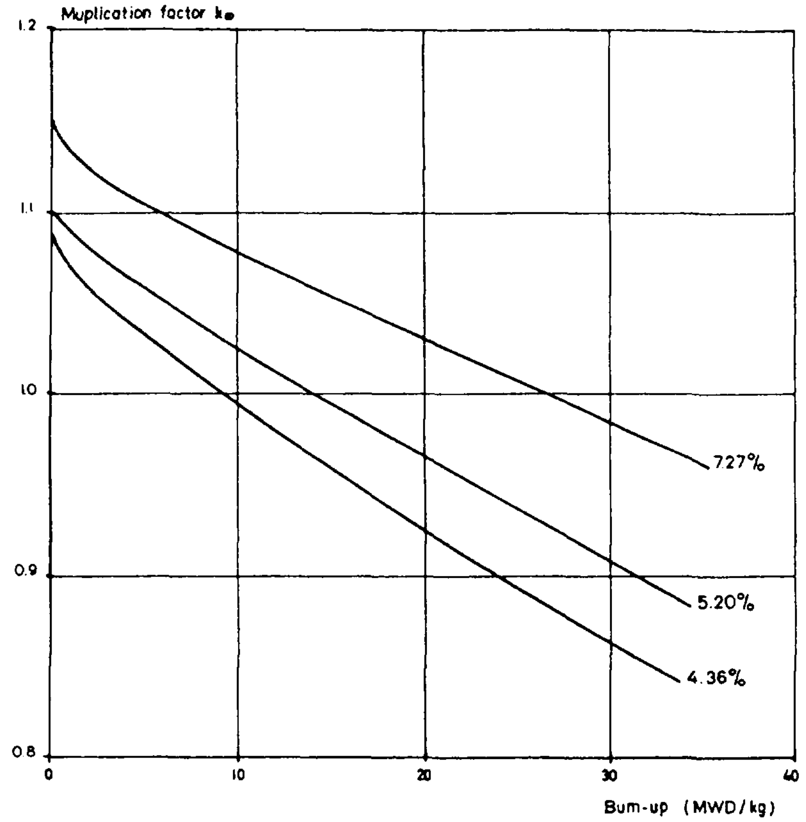

Fig 5 Infinite multiplication factor of $ThO_2$-$PuO_2$ lattices (4.36% ; 5.20% ; 7.27 %) as a function of burn-up

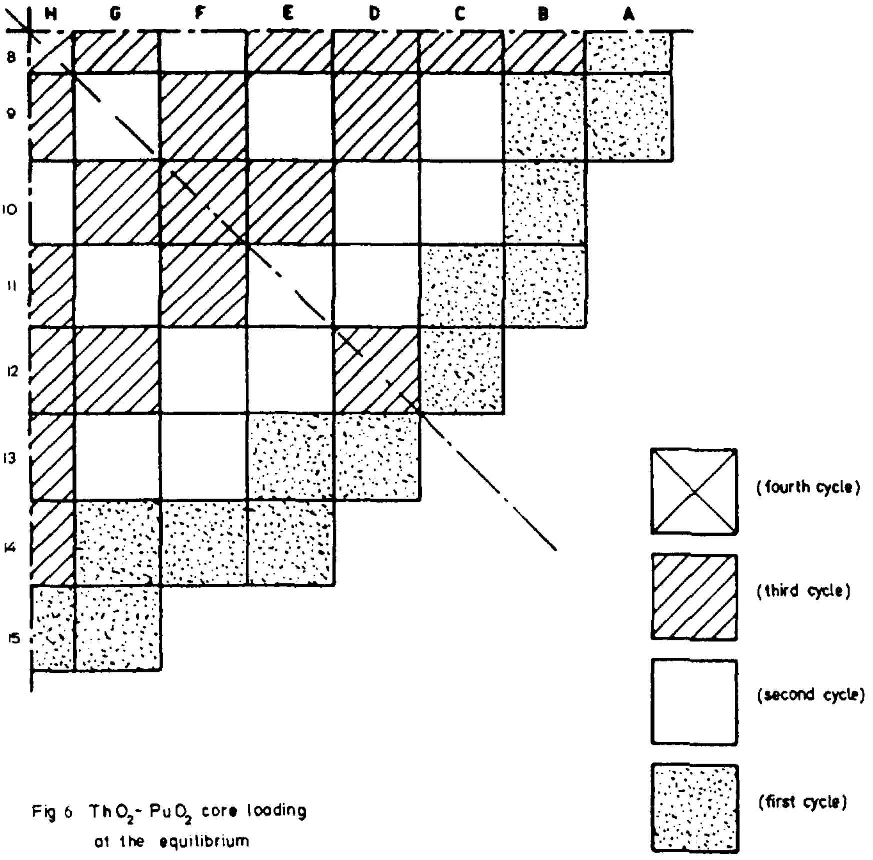

Fig 6 $ThO_2$- $PuO_2$ core loading at the equilibrium

**TABLE 1 : FIRST $PuO_2$ - $ThO_2$ CORE INVENTORY (kg)**

| ISOTOPES | Zone 1<br>4.36%<br>53 ass. | Zone 2<br>5.2%<br>52 ass. | Zone 3<br>7.27%<br>52 ass. | TOTAL |
|---|---|---|---|---|
| Th 232 | 21,963 | 21,098 | 20,640 | 63,431 |
| Pu 238 | 10 | 11 | 16 | 37 |
| Pu 239 | 571 | 669 | 935 | 2,175 |
| Pu 240 | 218 | 255 | 357 | 830 |
| Pu 241 | 136 | 159 | 223 | 518 |
| Pu 242 | 53 | 63 | 87 | 203 |
| Fissile Pu | 707 | 828 | 1,158 | 2,693 |
| Total Pu | 988 | 1,157 | 1,618 | 3,763 |
| TOTAL | 22,681 | 22,255 | 22,258 | 67,194 |

## $THO_2$ - $U233O_2$ ASSEMBLIES LOADED CORE

The $ThO_2$ - $PuO_2$ reactors, described in the precedent paragraph, produce, at the equilibrium, about 300 kg of Uranium by GWe and by year. The Uranium composition is :

| | |
|---|---|
| U233 | : 92.14% |
| U234 | : 6.50% |
| U235 | : 1.06% |
| Other isotopes | : 0.30%. |

This type of Uranium will be represented by $U^T$. $U^TO_2$ - $ThO_2$ assemblies are now loaded in the surrounding zone of a Bugey-2 type reactor, supposed to be at the equilibrium in the conventional Uranium cycle. It is important to know how would such standard reactors would operate with $U^TO2$ - $ThO_2$ fuels because they might be used during the transitory period before starting specially adapted Thorium cycle reactors. It would be the same for the $ThO_2$ - $PuO_2$ fuel reactors : a better conversion would be probably found by moderator ratio modifications.

**TABLE 2 : $ThO_2$ - $PuO_2$ CORE INVENTORY (kg) AT EQUILIBRIUM**

| ISOTOPES | LOADED ASSEMBLIES BOL | % | DISCHARGED ASSEMBLIES EOL | % | BALANCE |
|---|---|---|---|---|---|
| Th 232 | 20,640 | 92.73 | 20,260 | 94.10 | - 380 |
| Pa 233 | - |  | 20 | 0.09 | + 20 |
| U 233 | - |  | 238 | 1.11 | + 238 |
| Pa 233 + U 233 | - |  | 257 | 1.20 | + 257 |
| U 232 | - |  | 1.3 | 60 ppm | + 1.3 |
| U 234 | - |  | 19 | 0.09 | + 19 |
| U 235 | - |  | 3 | 0.01 | + 3 |
| Total U | - | - | 280 | 1.30 | + 280 |
| Pu 238 | 16 |  | 16 | 0.07 | - |
| Pu 239 | 935 |  | 284 | 1.32 | - 651 |
| Pu 240 | 357 |  | 300 | 1.39 | - 57 |
| Pu 241 | 223 |  | 209 | 0.97 | - 14 |
| Pu 242 | 87 |  | 119 | 0.55 | + 32 |
| Fissile Pu | 1,158 | 5.20 | 493 | 2.29 | - 665 |
| Total Pu | 1,618 | 7.27 | 928 | 4.30 | - 690 |
| Am 241 | - |  | 12 | 0.3 | + 60 |
| Cm 242 | - |  | 3 |  |  |
| Am 243 | - |  | 34 |  |  |
| Cm 244 | - |  | 11 |  |  |
| TOTAL | 22,258 | 100 | 21,528 | 100 | - 730 |

## $ThO_2$ - $U^TO_2$ cell physical characteristics

On figure 7 are represented the changes, as a function of the moderator ratio of the 3.45% $U^T$ multiplication factor (this 3.45% proportion allows the same cycle length to be obtained at equilibrium, as for the standard Uranium cycle).

On figure 8 is represented, as a function of irradiation, the 3.45% $U^T$ multiplication factor, in comparision with 3.1% U235 cell and 7.27% Pu $PuO_2$ - $ThO_2$ cell ; on figure 9 are given the Uranium isotope concentration changes.

The equilibrium core loading map, which is very similar to the loading map of a standard reactor, is given on figure 10.

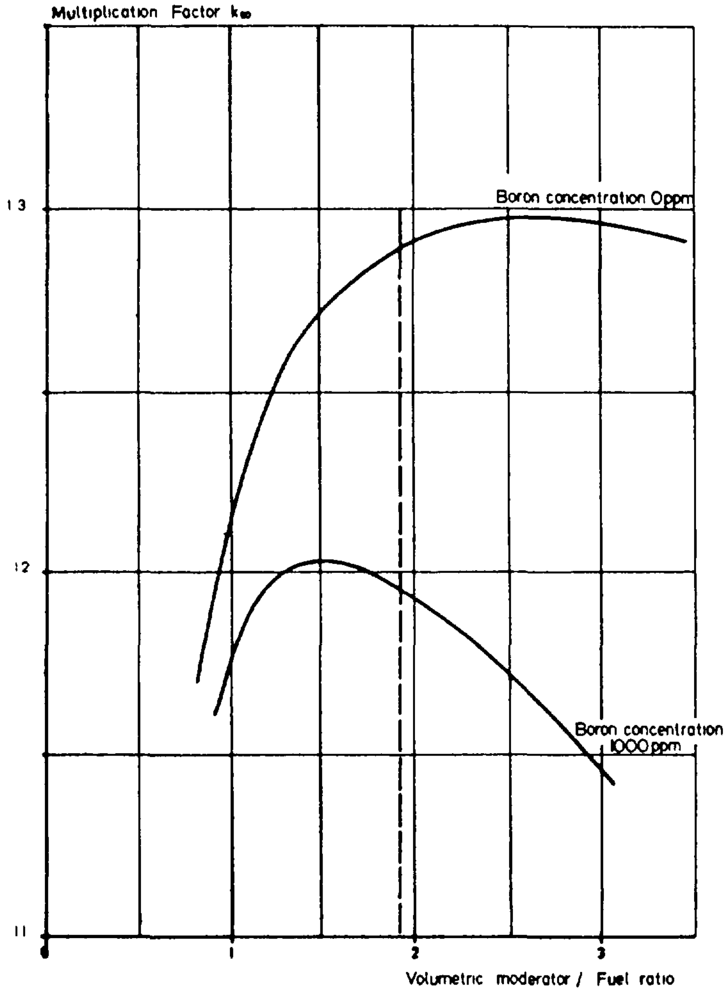

Fig 7 Multiplication factor of the $ThO_2$-$U^TO_2$ lattice (3.45%) as a function of moderator / fuel ratio

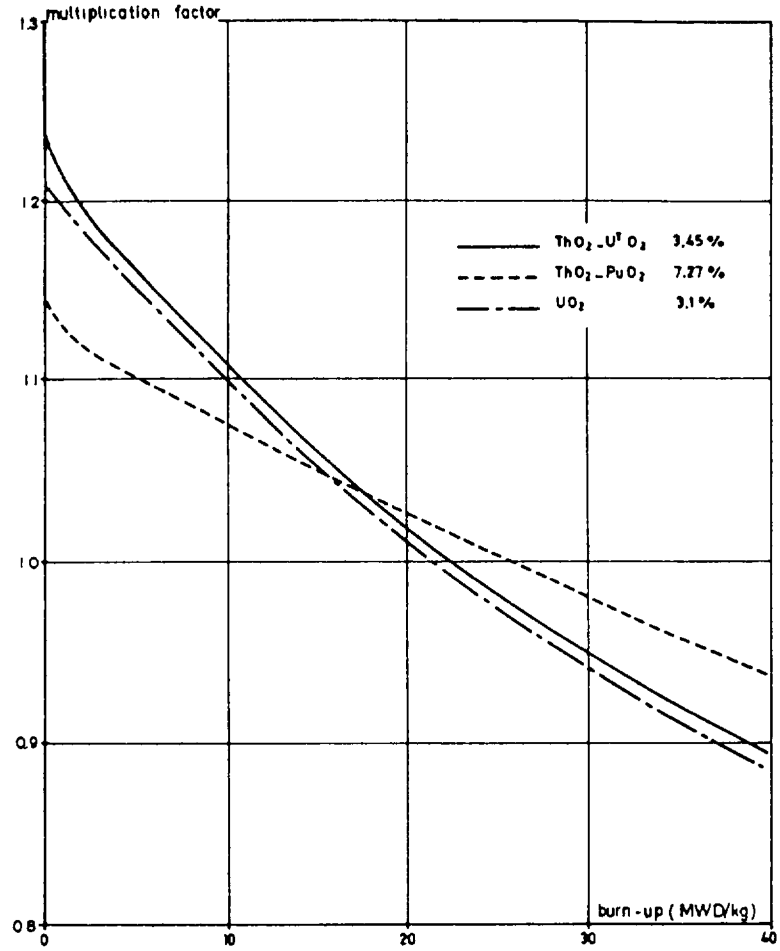

Fig 8 Multiplication factor of ThO$_2$-U$^T$O$_2$, ThO$_2$-PuO$_2$, UO$_2$ lattices as a function of burn-up

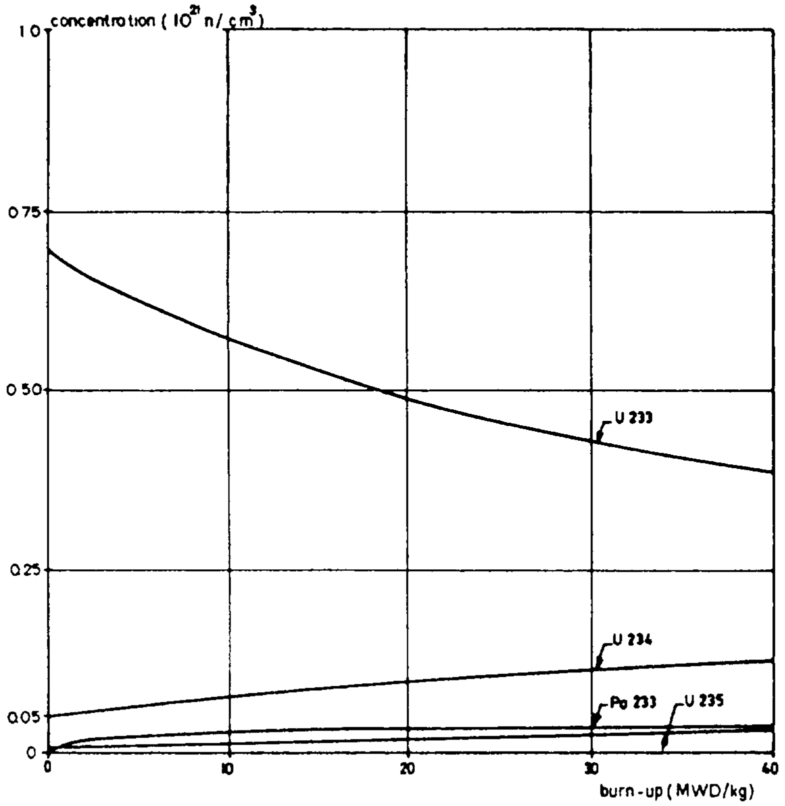

Fig 9 Changes of U, Pa concentrations in the ThO$_2$-U$^T$O$_2$ lattice

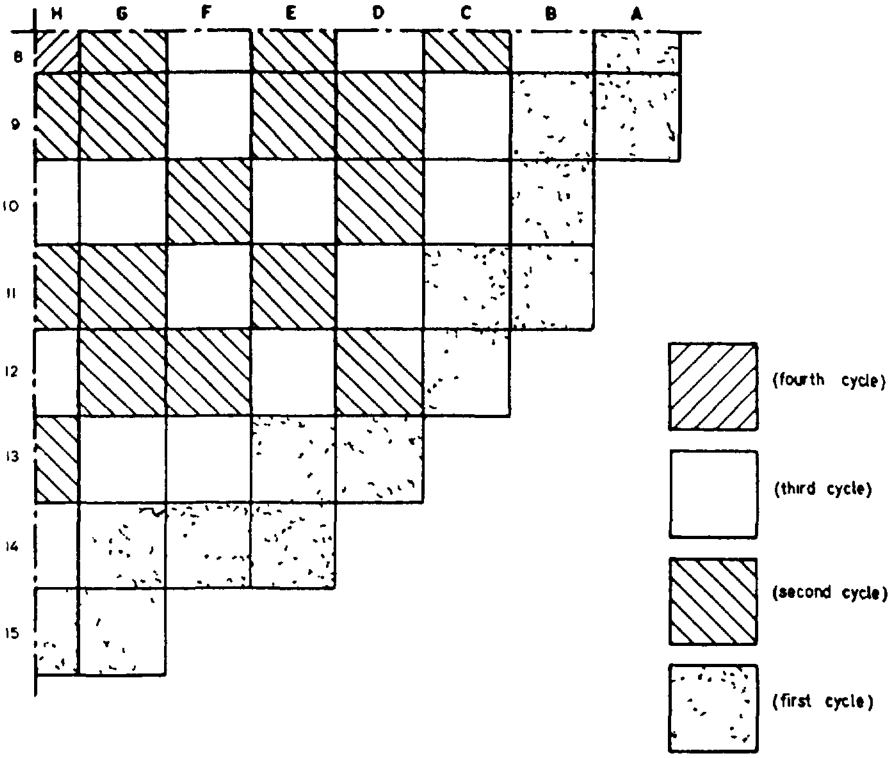

Fig 10 $ThO_2$- $U^TO_2$ core loading at the equilibrium

For the core, at equilibrium :

The Boron efficiency is equal to - 7.53 pcm/ppm (BOL) and - 9.76 pcm/ppm (EOL).

The Doppler coefficient is equal to - 2.9 pcm/°C (BOL) and - 2.63 pcm/°C (EOL).

The moderator temperature coefficient is equal to + 15 pcm/°C (BOL) and - 8 pcm/°C (EOL).

The delayed neutrons proportion is equal to 315 pcm.

The prompt neutron generation time is equal to 1.5 $10^{-5}$ s.

The 48 control rods are sufficient to assure the antireactivity margins (6580 pcm at the begining of cycle and 6900 pcm at the end).

**Reactor inventory**

The core inventory is given in table 3.

A very important conversion improvement may be obtained by using the Spectrum Shift Technique. In this case, the positive moderator temperature coefficient at the beginning of the cycle is eliminated and the mean conversion ratio reaches 0.87 instead of 0.64 with a light water moderator (figure 11).

## TABLE 3 : $U^TO_2$ - $ThO_2$ CORE INNVENTORY (kg), AT EQUILIBRIUM

| ISOTOPES | LOADED ASSEMBLIES BOL | % | DISCHARGED ASSEMBLIES EOL | % | BALANCE |
|---|---|---|---|---|---|
| Th 232 | 21,486 | 96.55 | 20.890 | 97.23 | - 596 |
| Pa 233 | - | - | 31 | 2.08 | + 31 |
| U 233 | 706 | 3.18 | 415 | | - 291 |
| Pa 233 + U 233 | 706 | 3.18 | 446 | 2.08 | - 260 |
| U 232 | 3.1 | 0.014 | 1.8 | $84\ 10^{-6}$ | - 1.3 |
| U 234 | 51.5 | 0.24 | 116 | 0.54 | + 64.5 |
| U 235 | 7 | 0.3 | 25 | 0.12 | + 18 |
| U236 | 0.3 | | 4.5 | 0.02 | + 4.2 |
| Total U | 768 | 3.45 | 593 | 2.76 | - 175 |
| TOTAL | 22,254 | 100 | 21,482 | 100 | - 768 |

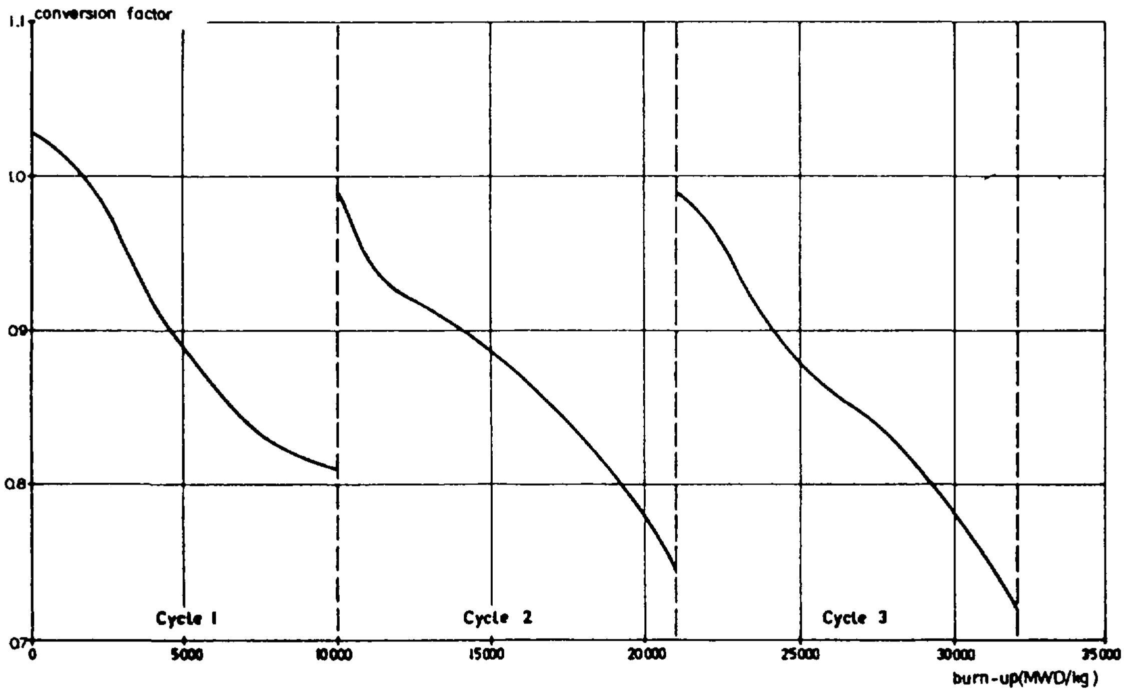

Fig 11 $ThO_2$-$U^TO_2$ (SSCR) conversion factor as a function of burn-up for the first three cycles

## CONCLUSION

The solution proposed to start the th-U233 cycle in unmodified PWRs consists of loading the whole reactor with Th-Pu assemblies from the first core and no longer using the standard cherkerboard loading ; the three assembly types (3 Pu concentrations) are loaded in rings.

The U233 being obtained, its best use is in Spectrum Shift Control Reactor (SSCR). This system gives simultaneously an acceptable moderator coefficient and a good conversion factor.

The Th-U233 in unmodified PWRs does not give large improvements with regard to uranium consumption (comparated to conventionnal plutonium and uranium recycle). Its main objective is to open the way of the thorium cycle in reactors ; the "royal way" being probatly the Molten Salt Reactors.

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# THORIUM UTILIZATION IN SOLVING THE NUCLEAR POWER FUEL PROBLEM

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**Abstract**

The future large nuclear energy appears to be a many-branched industry. Creation of such an industry and dimensions of its beneficial influence on the development of industrial production depends to a great extent on the capability to generate relatively low-priced fuel on the basis of commercial nuclear breeding. So far no universal approach has been available for solving the nuclear fuel problem, though some realistic and effective ways to case its ocuteness have been developed. The most radical means for solving the fuel problem is application of fast breeder reactors in combination with improved fuel utilization characteristics of the thermal reactors. Among the ways realizing the improved fuel balance in the thermal reactors emphasis should be placed on use of the Uranium-233 - Thorium cycle. The available estimates of fuel utilization by the thermal reactors on U/Th cycle are indicative of its attractiveness. The paper considers a method for Uranium buildup in the blankets of current fast reactors, required for transition to the Uranium-233 - Thorium cycle in the thermal systems. It is shown that thorium utilization contributes noticeably to the nuclear fuel resources. Combination of the Uranium-Plutonium and Thorium cycles ensures long-term fuel supply, makes the nuclear energy production more flexible and, at realistic requirements to the level of fast reactor breeding, enables the self-provision regime to be realized in future.

Successful development of the world nuclear power production (NP) and, primarily, nuclear electrical energy production, is probing its capability to contribute essentially to the overall energy production system. The developed nuclear energy production opens a wide possibility of using the nuclear energy in various technological processes, in heat production for industrial and district heating.

The future large NP is thought as a many-branched industry and its creation depends on the capabilities of providing it with relatively inexpensive fuel first with Uranium-235 and then with recycled fuel, plutonium and Uranium-233 on the basis of commercial breeding. Taking into account the scale of energy requirement, the organization of fuel breeding is an indispensable condition, though at the current stage of NP development when the Uranium-235 resources are still large and the overall energy production by existing nuclear power stations is relatively low, the role of breeding is unjustifably underestimated.

It may be expected that deficiency of natural uranium resources would become noticeable in the NP economy by the end of this and beginning of the next century. By that time the measures for sharp reduction of natural uranium utilization will have to be developed and their realization begun.

By present some real methods for easing the NP fuel problem have been worked out. One of them is connected with improvement of the fuel cycle of the thermal reactors and reprocessing of spent fuel enabling the natural uranium requirement to be reduced by 20-30% (see Table 1). An additional and very essential benefit may be got from the measures directed to increase the breeding ratio in the thermal reactors by use of dense fuel (e.g. uranium silicides), dense fuel lattices (APWR-type reactors) etc. However, the radical solution of the NP fuel prob-

Table I. Specific natural uranium utilization in loading feeding and separative works E for the VVER-1000 reactors with uranium-233 and 235 (direct replacement)

| | | U-235 | | |
|---|---|---|---|---|
| | | VVER-1000<br>open cycle | VVER-1000<br>closed cycle | VVER-1000<br>U-233-Th |
| g | kg/MW(e) | 460 | 250 | 110 |
| q | kg/MW | 230 | 140 | 60 |
| $E_g$ | kg/MW | 250 | 180 | 100 |
| $E_q$ | kg/MW.year | 140 | 100 | 50 |

lem lies in breeding, the fast reactors operating on the uranium-plutonium cycle being the most available breeding scheme. The requirements to be imposed on the breeder reactors will change in the course of the NP development. At present, when the nuclear fuel is still relatively inexpensive, the main efforts must be directed to restrain the rate of the energy production cost. Today the problem of maximum excess fuel production is not still acute, but of great importance are the non-fuel cost components such as investment (capital) costs. In the future the efforst will inevitably shift to improvement of the breeding gain.

The recent studies of the methods for improving the fast sodium reactor breeding, including development of the heterogeneous oxide metalic core concept, have revealed a real possibility for increasing the breeding ratio up to 1.55-1.6 /2/. However even at so high characteristics the self-provision fuel cycle may be only realized when the fast reactor fraction in NP were significant, no lower than 60%. For the many-component energy production structure the requirement for such a high fast reactor fraction may prove to be too stringent.

The fuel problem might be helped by use of the uranium-plutonium cycle in combination with the uranium-thorium mode, which allows a significant reduction in fuel requirement for the thermal reactors feeding. Here important is not utilization of thorium as fuel but the better nuclear and physical characteristics of uranium-233 in the thermal reactor spectrum. As a result the net production of secondary fuel determined by the breeding gain (BG) increases from - 0.5-0.6 to - 0.2 - 0.3 in the light water reactors with the oxide uranium-thorium fuel at high burnups, though the fast neutron fission fraction reduces sharply because of the low thorium fission cross section.

Transition to dense fuel (e.g., metallic thorium) is less effective than in the case of the uranium fuel. However at relatively low burnups this transition may permit the fuel requirement for fuel feeding to be practically eliminated (Table 2).

Table 2. Characteristics of thermal reactors
with U-235-Th cycle (fuel delay time-3 years)

| | Natural uranium requirement | | Separative work requirement | |
|---|---|---|---|---|
| | loading t /GW(e) $10^{-3}$ | feeding t/GW(e).$10^{-3}$ | loading | feeding |
| VVER | 0.6 | 0.25 | 0.6 | 0.25 |
| RBMK | 1.2 | 0.35 | 1.2 | 0.36 |
| HTGR | 0.4 | 0.17 | 0.35 | 0.13 |

Even greater effect is reached when the thorium fuel cycle is used in the high-temperature thermal reactors (HTGR) primarily designed for technological purposes /3/. For example, in the linear development of two-component energy production structure involving advanced fast sodium reactors and HTGRs with uranium-233 and with the capabilities increased at a rate of 20-30 GW(e) per year, the fuel self-provision is reached after the amount of uranium-235 contained in 2-2.5 mln tons of natural uranium and about 0.2 mln tons of thorium at a maximum thorium output not exceeding $10^4$ tons per year have been spent.

Introduction of the uranium-thorium fuel cycle for the thermal reactors is associated with a number of complicated problems which are to be solved:

- the thorium-uranium reactors use a highly-enriched uranium for startup and increased amounts of uranium and large quantity of separative work at the first operation stage while the rejection of the reactor feeding leads to reduction of fuel burnup and to growth of fuel reprocessing.

If a part of the thermal reactors are used in the uranium_ 233 production mode for the rest of the reactors, then in the exponential growth of the NP capabilities at doubling times of 20 and 15 years the natural uranium requirement over 30 years will not exeeed 1-2 mln tons and 2-3 mln tons, respectively. Thus NP development on the basis of the thermal reactors operating on $^{235}$U- Th and $^{233}$U-Th cycles leads to noticeable reduction of the natural uranium requirement as compared to the open uranium cycle though these requirements remain still very high. However use of this cycle makes it impossible to organize the self-provision mode and this is its major disadvantage,

- commercialization of remote reprocessing of the uranium-thorium fuel at a sufficient burnup is required.

The current state of the reactor and fuel cycle facility development as well as the fuel problem timing suggest the following logics in the development and combining of the uranium-plutonium and uranium-thorium cycles for NP.

The NP development began with the thermal uranium-fuelled reactors. Transition to the uranium-plutonium closed cycle, development of spent fuel regeneration, plutonium accumulation paves the way for rapid inroduction of fast reactors which are to be the basis of future nuclear fuel breeding. The acuteness of the NP fuel problem can be then eased additionally by transition of the thermal reactors to utilization of the uranium-233 accumulated in the fast breeder reactor blankets. This way of uranium-233 production is beneficial for the following reasons:

- the above mentioned problem of enhanced uranium-235 expanses for initial buildup of uranium-233 in the thermal reactors is eliminated;

- uranium-233 losses because of parasitic radiation capture by protactinium-233 are minimized;

- remote technology of reprocessing and fabrication of the uranium-thorium fuel, developed for uranium-plutonium cycle can be extensively used. Investigations shows general similarity of technological processes for both types of fuel.

The current assessments of uranium-233 buildup in the thorium blankets of advanced fast sodium reactors with improved breeding (heterogeneous reactor core concept) indicate that replacement of uranium-233 with thorium in the sodium breeder reactor blankets enables uranium-233 to be produced in amounts slightly (by 5%) lesser than the plutonium output in the uranium blankets. Replacement of the uranium blankets with thorium ones slightly affects also the core physical characteristics: the inner breeding ratio decreases by 1-2%, and the specific fuel load increases by about 1-2%.

Use of thorium in the blanket, the region of lower neutron flux, permits hazardous effect of protactinium on the neutron balance and reactor control to be reduced. The effect of the protactinium poisoning in the fast reactor blankets is by an order of magnitude lower than in the thermal reactor ones.

From the joint of view of effective introduction of thorium to NP, of interest are also fast reactors with mixed fuel cycle. The main fissile nuclides in this cycle are uranium-233, uranium-235 and in the blankets-thorium. The sodium fast reactors with mixed fuel have the following advantages:

- smaller initial loading in comparison to the plutonium reactor and breeder reactor on U-233-Th;
- integrated breeding ratio close to the plutonium one and noticeably higher than that for the uranium-233-thorium fuel;
- better sodium void reactivity effect with the uranium-233 fuel located in the central part of the core and plutonium - in the periphery, the power distribution profile noticeably stabilized, the specific fuel load and the doubling time reduce.

The analysis of the NP fuel cycles reveals that use of thorium contributes appreciably to the NP fuel resources. Combination of the uranium-plutonium and uranium-thorium cycles ensures long-term fuel supply with moderate amount of thorium mining, makes NP more flexible, eases the fuel problem significantly and for long time, with real requirements to breeding in the fast reactors and balanced structure of NP, enables the fuel self-provision mode to be realized. Introduction of thorium to nuclear energy production is most beneficial through uranium-233 production in the breeder-reactor blankets after technology of the closed uranium-plutonium fuel cycle is mastered.

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# BASIC RESEARCH ON UTILIZATION OF THORIUM-BASED NUCLEAR FUEL

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**Abstract**

The dependences of build-up of U-233, U-232, Pa-233 and fission products on integral neutron flux for $ThO_2$ samples irradiated in high flux ETR at constant neutron flux and steady spectrum have been investigated. From these experiments, we have accumulated experiences on further research for the behaviours of the irradited $ThO_2$ under different irradiation conditions in reactor.

## 1)Introduction

It has been well known that thorium is not a nuclear fission fuel, but it could be transformed into U-233, an actual fission fuel with good nuclear properties, by capturing a slow neutron. Before thorium-based nuclear fuel could be used in reator, a full understanding of the build-up of related nuclides in reactor (mainly U-233, U-232, Pa-233 and fission products) is very important. In order to clarify the characteristics of the build-up of these nuclides in reator, we have initiated a program to study the accumulations of U-233, U-232, Pa-233 and fission products in $ThO_2$ in different irradiation conditions, including different integral thermal neutron fluxs and diffenrent neutron spectra in reactor. In this work, the dependences of build-up of U-233, U-232, Pa-233 and fission products on integral thermal neutron fluxs for $ThO_2$ samples in high flux ETR at constant neutron spectrum have been investigated.

## 2) Experimental

### Nuclear reactor

Irradiation was carried out in high flux ETR in Southwest China. The reactor has thermal power of 125 MW. Maximum thermal neutron flux is $6.2{:}10^{14}n/cm^{2}.s$ and fast neutron flux is $5.7.10^{14}n/cm^{2}.s$. It was built in 1981.

### Target preparations and irradiation conditions

$ThO_2$(50mg) was pressed into discal tablet of 4 mm.diameter and loaded it into a container. Then all containers were put into irradiation tube.(figure1) Six samples were placed in two irradiation tubes.(figure2) Two tubes were put in Be-reflecting layer. Mean thermal neutron flux was $2x10^{14}$ $n/cm^{2}.s$. in this area. Six samples could receive different thermal neutron flux, because thermal neutron flux distribution from the bottom to the top were not uniform as we could see in fig.3.(maximum non-homogenuous distribution coefficient was 1.4) $Q_{13}$ and $Q_{14}$ were irradiated by one and three runs respectively. We supposed that fast/thermal neutron flux ratio was 1.03 for two irradiation tubes. Integral thermal neutron fluxs were $2x10^{20}$ to $2x10^{21}n/cm^{2}$.

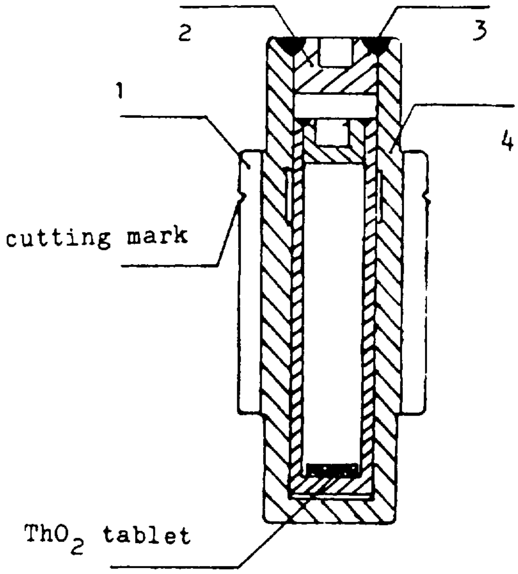

Fig.1. Irradiation container

1. External cylinder. 2. cap of external cylinder.
3. Cap of internal cylinder. 4. INternal cylinder.

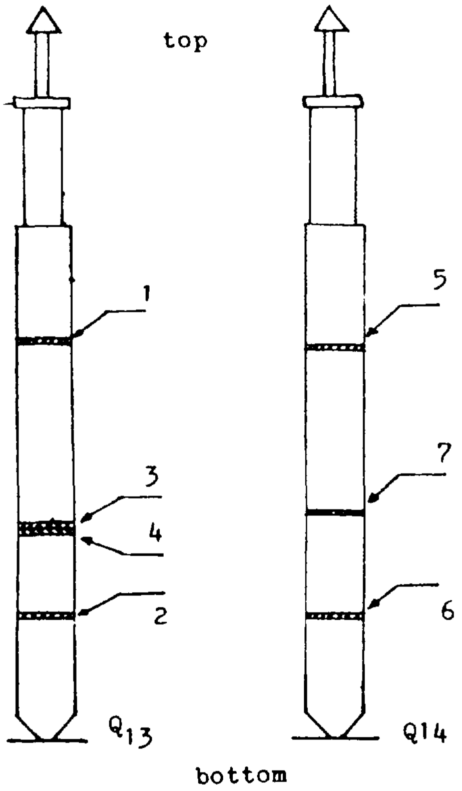

Fig.2. Irradiation positions of samples in reator.

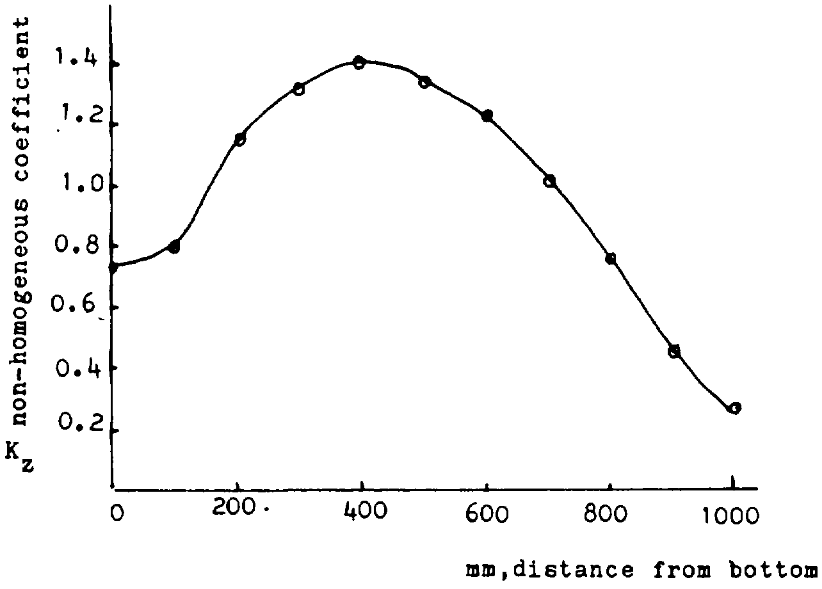

Fig.3 Axial distribution of neutron flux in reator.

### Radiochemical separation and determinations

After irradiation and "cooling", $ThO_2$ samples were dissoled into HCl solutions, from which yields of Pa-233 and fission products could be determind by Ge(Li) $\gamma$ detector. Then the solution was adjusted into 8N HCl and feeded on an anion exchanger ZerolitFF(Cl form,50-60 mesh). After eluting out Th, Pa-233 and fission products from the column, U was eluted by 0.05 $HNO_3$. U-232/U-233 ratio was determined by $\alpha$-spectrometry and total U amount was determined by Asenazo -3 photospectrometric method in U solution.

### 3) Results and discussions

THe results could be seen in table 1and also by fig.4,5, and 6. From fig.4 it shows that U-233 yields increase with increases of integral thermal neutron fluxs. U-233 in thorium could reach 1.5% when integral thermal neutron flux reachs $2\times10^{21}$ n/cm$^2$. According to theoretical calcalution, U-233 could reach a saturation value at about $2\times10^{21}$ n/cm$^2$. So we are planing to work at higher integral thermal neutron fluxs to see if U-233 reach a saturation.

Table 1. Correlations between U-233, U-232/U-233, Pa-233 Cs-137, Zr-95 and integral thermal neutron fluxs.

| sample | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|
| $^{232}Th$ $10^{20}$ atoms | 1.500 | 1.476 | 1.499 | 1.491 | 1.486 | 1.481 |
| $\Sigma\phi_{Th}$ $10^{20}$neutron/ cm$^2$ | 3.499 | 4.543 | 5.040 | 12.35 | 15.91 | 17.59 |
| $^{233}U$ $10^{17}$ atoms | 3.489 | 4.771 | 6.156 | 15.81 | 17.83 | 21.99 |
| $^{233}U/^{232}Th$ $10^{-3}$ | 2.33 | 3.23 | 4.10 | 10.6 | 12.0 | 14.8 |
| $^{232}U/^{233}U$ $10^{-5}$ | 2.0 | 1.7 | 2.2 | 8.6 | 7.5 | 10.0 |
| $^{233}Pa$ $10^{17}$ atoms | 2.450 | 3.284 | 4.215 | 5.878 | 6.998 | 7.872 |
| $^{137}Cs$ $10^{14}$ atoms | 4.066 | 6.522 | 11.77 | 173.9 | 295.6 | 416.4 |
| $^{95}Zr$ $10^{15}$ atoms | 0.3367 | 0.5316 | 0.9554 | 11.27 | 17.82 | 25.27 |

* All data were extrapolated to the end of irradiation

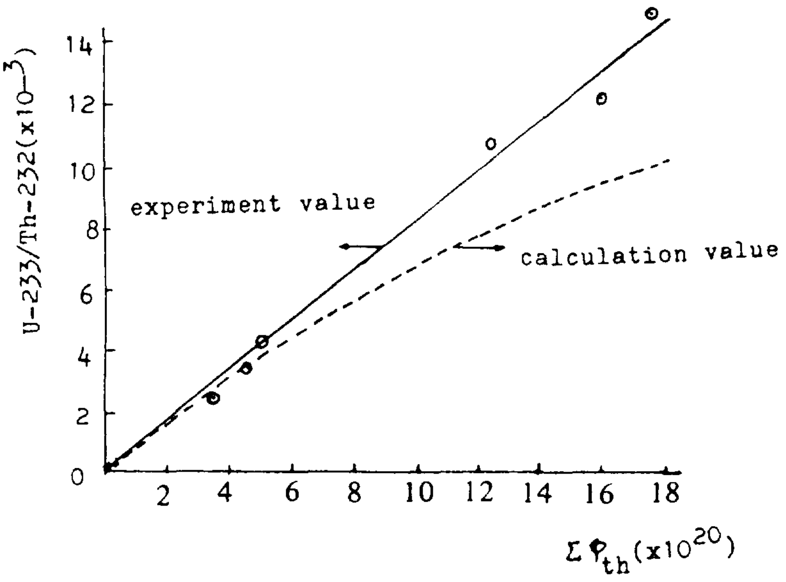

Fig.4. Correlations between U-232/Th-232 and $\Sigma\Phi_{th}$

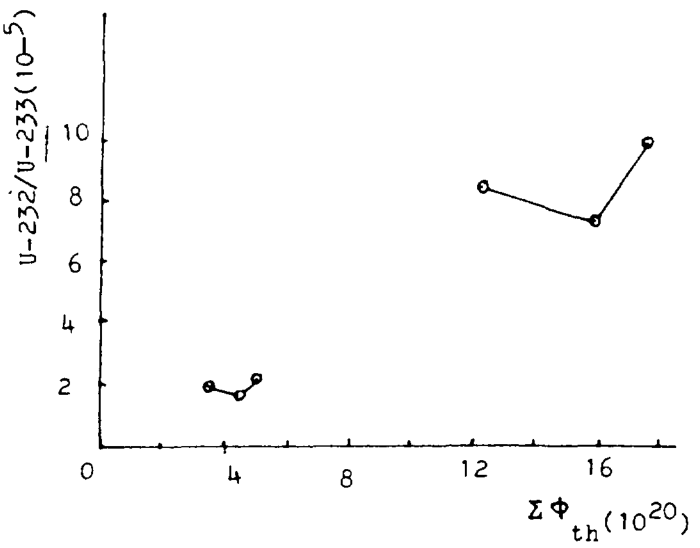

Fig.5. Correlations between U-232/U-233 and $\Sigma\Phi_{th}$

From fig.5, U-232/U-233 ratios increase with increases of integral thermal neutron flux as we anticipited, but linear correlation was not so good. We thought it is because of comlex neutron spectra in reactor. For one tube, fast/thermal neutron flux ratio was not the same from the bottom to the top as we supposed. Generally speaking, U-232/U-233 ratios increase when fast/thermal neutron ratios increase. Research on correlation between U-233,U-232/U-233 and different fast/thermal neutron ratios is in progress.

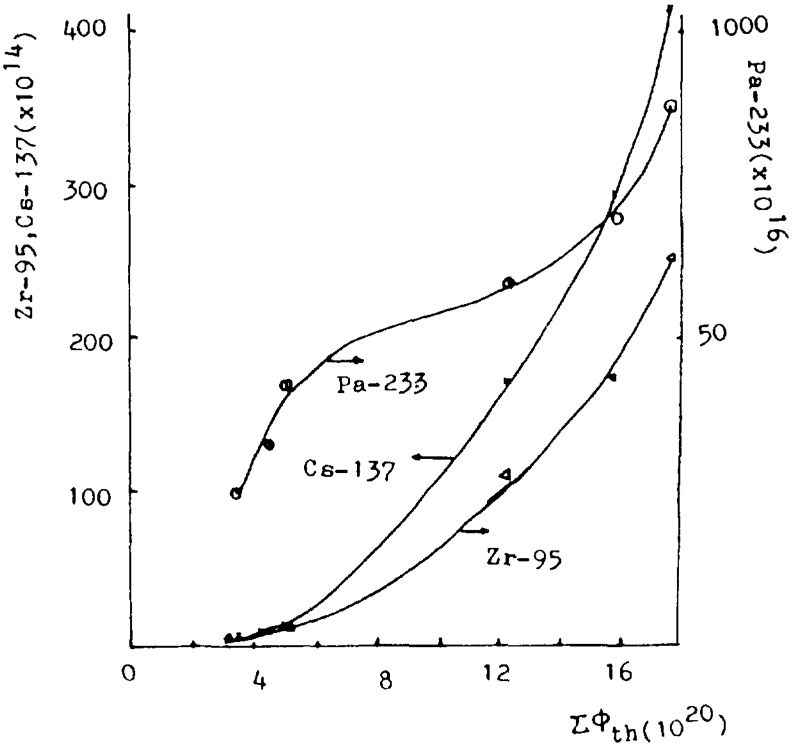

Fig.6. Correlations between Pa-233, Zr-95, Cs-137 and $\Sigma\Phi_{th}$

It is necessary to do some tests in small reactor before we use thorium-based nuclear fuel in large reactor. So a zero power reactor(3% enriched $UO_2$ as fuel, light water as moderator) in our institute will be partly loaded with $ThO_2$ fuel rods instead of $UO_2$ fuel rods to observe the influence on reactor performances.

There are still a lot of work on basic research about the utilization of thorium-based fuel left to be done. We should be continuing our work in this area and accumulate necessary experimental data. Thereby, thorium will put into large scale use in near future and play an appropriate role in nuclear power development.

# THORIUM OXIDE BLANKET FABRICATION FOR INDIAN FAST BREEDER TEST REACTOR

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## Abstract

*For an effective utilisation of Thorium in FBRs, use of fertile blanket of thorium oxide/thorium must be considered. Development of a reliable and economic fabrication technique of this fertile material, thus is an important step in the ultimate goal of utilization of thorium in FBRs.*

*Fabrication and supply of blanket assemblies containing thorium oxide pellets to the requisite quality, for full core of Indian Fast Breeder Test Reactor, is one of the important tasks of Nuclear Fuel Complex at Hyderabad, a unit of the Department of Atomic Energy.*

*Starting with* $ThO_2$*-powder obtained by oxalate precipitation and calcination, green pellets are produced through powder metallurgical route. In order to achieve sintered densities more than 94% T.D., addition of Mg in small quantities, during oxalate precipitation stage, have been found quite effective.*

*The effect of compacting pressure, green density and sintering temperature on final sintered density of pellets and percentage of shrinkage have been studied to arrive at the optimum fabrication parameters, to yield high density in pellets requiring minimum amount of grinding.*

*The compactability and sinterability characteristics seem to be affected by long storage of powder. Ball milling of powder before pre-compaction and granulation almost restore the original characteristics of the powder.*

*The paper deals with the Indian experience on fabrication of thoria pellets for the Fast Breeder Test Reactor, with some*

*emphasis on the work carried out for standardization of the parameters, quality characteristics of the pellets and on blanket assemblies made.*

*Performance test of these assemblies under actual operating conditions of FBTR, followed by reprocessing studies, will be of great value for the Indian programme of utilization of thorium in the envisaged FBR's.*

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## *1. INTRODUCTION*

*With continued increasing demand on uranium and its known limited reserves, there is a growing realization for the need to utilise thorium to broaden the resource base of supply of fissionable materials.*

*Various concepts including LWRs, Canadian PHWRs, FBRs, and symbiotic reactor systems like HTR-FBR have been studied [1] [2] [3] for utilisation of thorium.*

*For India which has rather limited known reserves of uranium and a vast reserve of thorium, its special interest in utilization of thorium in its reactors is obvious.*

*Studies elsewhere [4] as well as our own [5] have shown that thorium in radial blanket region of FBRs has only marginal neutron economic penalty compared to complete uranium cycles. With this in view, it was decided to fabricate assemblies containing high density thoria pellets for the entire radial blanket region of Fast Breeder Test Reactor (FBTR), which has been commissioned in October, 1985.*

*The initial development work on preparation and characterization of thoria powder to yield high sintered density was carried out at Bhabha Atomic Research Centre (BARC) [6]. Emphasis, however, was laid on optimization of fabrication parameters only during fabrication of radial blanket assemblies, at Nuclear Fuel Complex.*

*This paper presents the experiences gained in thoria powder and pellet production, optimization of certain fabrication parameters, assembly fabrication and quality control aspects.*

## *2.0 Radial Blanket Assembly:*

*FBTR has a thick blanket and has 342 positions in its core for radial blanket assemblies. The assembly consists of a 'foot' section, a cluster of seven elements containing thoria pellets with gas plenum at the top, a hexagonal tube enveloping the element cluster, and a lifting head. The elements are wrapped with spacer wire and the ends are closed with end-plugs. The 'foot' section and the lifting 'head' are welded to the hexagonal tube. Fig.1 shows an element and the finished assembly.*

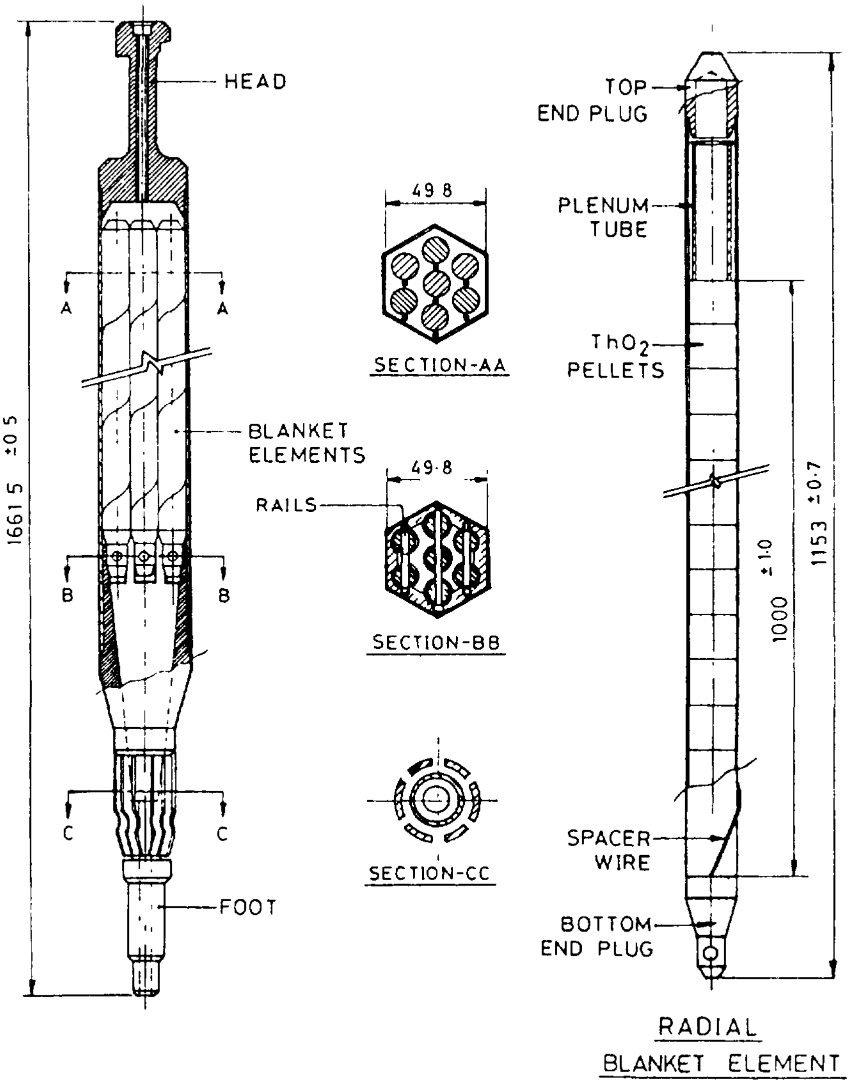

*Fig.1 - Blanket Assembly.*

The salient features of a radial blanket assembly are summarised below:

Overall length of assembly - 1661.5 mm
No. of elements/assembly - 7
Approx.weight of fertile material/assembly - 12.5 Kg.
Density of thoria pellets - 94% T.D.
Dia of spacer wire - 0.7 mm
A/F dim.of SS Hex.tube - 49.8 mm
OD & ID of SS clad tube - 16.5 & 15.5 mm

## 3.0 Fabrication of High Density Thoria Pellets:

3.1 The Process: Fig. 2 shows schematically the thoria pellet fabrication process.

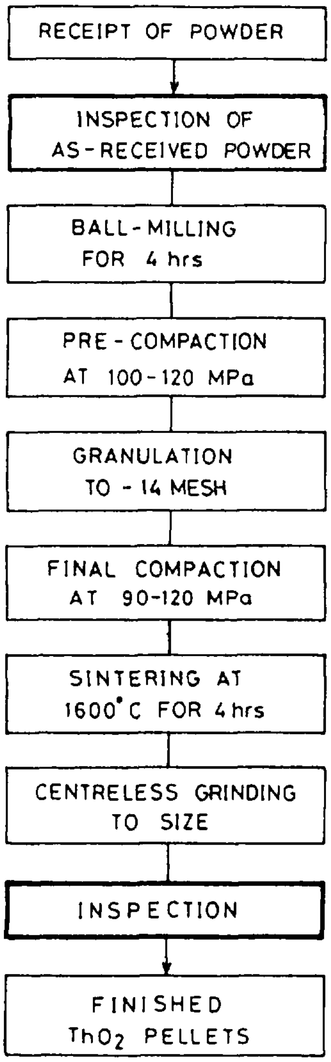

Fig.2 - $ThO_2$ pellet fabrication process flow-sheet.

3.1.1 Starting Powder: Thoria powder is supplied by M/s. Indian Rare Earths Ltd, a Public Sector Undertaking in Department of Atomic Energy. The powder is obtained by calcination of thorium oxalate, precipitated from pure thorium nitrate solution. The bulk and tap densities of the starting powder are respectively 0.9 to 1.2 g/cc and 1.5 to 1.9 g/cc. The particle size is in the range of 0.8 to 1.5 microns.

A typical chemical analysis of thoria powder being supplied, is given in Table I. The powders are accepted on the basis of a sinterability test.

Table I - Typical analysis: Values of elemental impurities in Thorium Oxide.

| ELEMENTAL IMPURITIES | p.p.m. (less than) | ELEMENTAL IMPURITIES | p.p.m. (less than) |
|---|---|---|---|
| Al | 5 | V | 0.2 |
| B | 0.25 | U | 1.0 |
| Be | 0.1 | Zr | 30 |
| Cd | 0.1 | Ce | 4 |
| Cr. | 5 | Dy | 0.2 |
| Cu. | 5 | Eu | 0.08 |
| Co. | 1 | Er | 0.2 |
| Fe. | 50 | Sm | 0.2 |
| Mn | 1 | S | 100 |
| Pb | 20 | P | 50 |
| Sb | 1 | Ca | 30 |
| Sn | 1 | Mg | 300 |
| Mo | 5 | Gd. | 0.08 |

### 3.1.2 Ball Milling:

Light ball milling the powder was introduced, when it was observed that pellets produced from as received powders, revealed unacceptable surface pits after grinding. This was attributed to be due to formation of hard agglomerates in the powder which had been stored for a long time. Ball milling the powder for 4 hours, resulted in considerable improvement in acceptability of the sintered and ground pellets.

### 3.1.3 Pre-compaction & granulation:

*The ball milled powder is then compacted into slugs of 60 mm dia and 60 mm height, at a pressure of about 100 MPa using a hydraulic press.*

*The slugs are granulated to minus 14 mesh size granules which show considerable improvement in flowability of the powder. Bulk and tap densities of granules are respectively in the range of 3.5 to 3.6 and 4 to 4.4 gms/cc.*

### 3.1.4 Final compaction:

*The final compaction of flat end pellets is done using a two hole die in a hydraulic press. A green density of 6.2 to 6.3 g/cc and a L/D ratio of 1.2 to 1.4 are maintained in the green pellets. No binder is used during compaction. The die wall and plungers are however lubricated with a saturated solution of stearic acid in acetone, during each compacting cycle.*

### 3.1.5 Sintering:

*Sintering of green pellets is carried out in a batch type resistance furnace, under hydrogen atmosphere. The temperature of the furnace is raised gradually to 1600°C and the pellets are soaked at that temperature for 4 hours. The charge is cooled under hydrogen atmosphere to a temperature of about 150°C, before the pellets are discharged from the furnace.*

### 3.1.6 Grinding:

*In order to obtasin the finished size and perfect cylindrical shape, the pellets are centreless ground using silicon carbide grinding wheel.*

*The sintered and ground thoria pellets are normally light grey to pure white in colour. However, at times, black patches on the surface appear. Chemical analysis and lattice parameter measurements carried out on samples containing these coloured patches have confirmed it to be thorium dioxide.*

## 3.2 Quality Control & Inspection:

Quality control on as received powder and pellets fabricated, include assuring chemical purity of starting powder, powder morphology, dimensional and density requirements as per specifications and visual examination of finished pellets. As regards cracks, pits, chips or other surface defects, the same acceptance criteria as used for PHWR fuel pellets, have been used for the present compaign of $ThO_2$ pellet fabrication. This may be probably more stringent than necessary, keeping in view the service conditions of blanket assemblies. Perhaps, this could be relaxed with consequent improvement in recovery, as the experience and confidence builds up after use.

## 3.3 Optimisation of certain process parameters:

### 3.3.1 Choice of Powder Production Process:

The very early development work carried out at BARC [7] on thoria powders produced by different routes and their sinterability, had shown that powders produced from (a) direct ignition of thorium nitrate (b) decomposition of thorium hydroxide and (c) decomposition of thorium carbonate, had coarse particle sizes, poor compactibility and sintered to low densities. On the other hand, powder obtained by calcination of oxalate had fine particle size and sintered to comparatively higher densities.

During later development work, [6] it was found that addition of small quantities of magnesium during oxalate precipitation, further improved the sinterability of thoria powder.

Thus the optimized method of powder preparation, presently used, consists in

- addition of magnesium sulphate in requisite quantity * to a batch of oxalic acid.

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*The residual magnesium in powder produced is in the range of 300-400 ppm.

- *Continuous addition of pure thorium nitrate solution to this oxalic acid, to co-precipitate thorium-magnesium oxalate.*
- *calcination of oxalate at 900°C in air, to obtain sinterable thoria powder.*

### *3.3.2 Choice of Pre-compacting Pressure:*

*The powder produced by oxalate route has poor flowability. Attempts to compact pellets directly from the powder resulted in laminated compacts, apart from non-uniform die fill. Hence pre-compaction and granulation of starting powder was considered essential.*

*The pre-compaction pressure was observed to have significant effect on integrity of green pellets at final compaction stage. Slugs compacted at a pressure of 90-120 MPa and granulated, yielded integral green pellets. Granules from slugs compacted at lower pressures resulted in broken or laminated pellets.*

### *3.3.3 Choice of final compacting pressure:*

*The final compacting pressure was optimised with following objectives.*

- *to have sufficient green strength in the pellets to withstand handling without chipping or breaking.*
- *to obtain sintered density more than 94% theoretical.*
- *to minimise variation in shrinkage after sintering which would eliminate or minimise grinding of sintered pellets to get the final dimensions.*

*With above requirements in view, pellets were compacted at seven different pressures and were sintered at 1600°C for 4 hours under hydrogen atmosphere. The powder used was from a single lot. The pellets were evaluated for green and sintered densities, shrinkages and green strength. The results are presented in Figs 3 & 4. As may be seen from the figures*

- *green as well as sintered densities increased with increase in compacting pressure.*
- *variation of green density decreased as the pressure increased.*

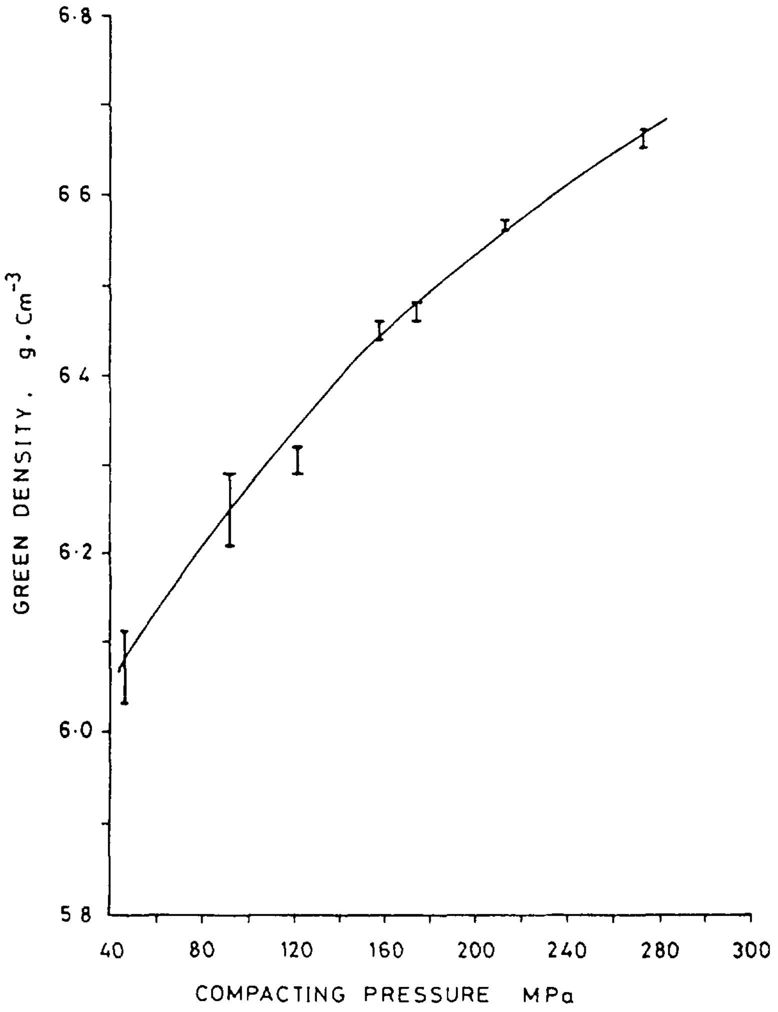

*Fig.3 - Effect of compacting pressure on green density.*

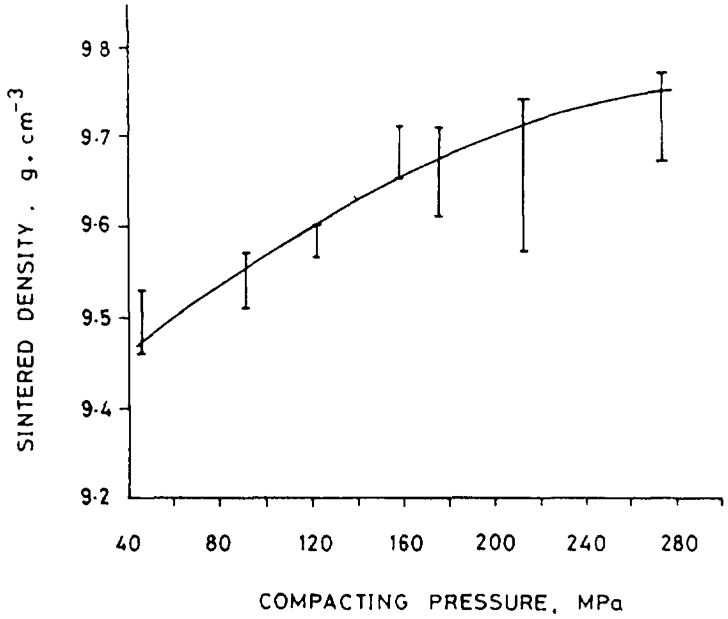

*Fig.4 - Effect of compacting pressure on sintered density.*

- *variation in sintered density (hence shrinkage) is comparatively more at low or high pressures of compacting.*

*The same trends were observed, when pellets were compacted using powders from randomly chosen different lots, which can be considered to represent more closely the actual conditions during production. Again the pellets were compacted at seven different pressures and sintered at 1600°C for 4 hours under hydrogen atmosphere. Results based on four different lots are presented in Fig.5. Pellets compacted in the pressure range of 90-120 MPa yielded the most suitable end-product. Comparatively larger variations in both green and sintered densities at lower pressures and in sintered densities at higher pressures are evident. Thus the above pressure range which normally results in a green density of 6.2 to 6.3 g/cc is used for production of thoria pellets.*

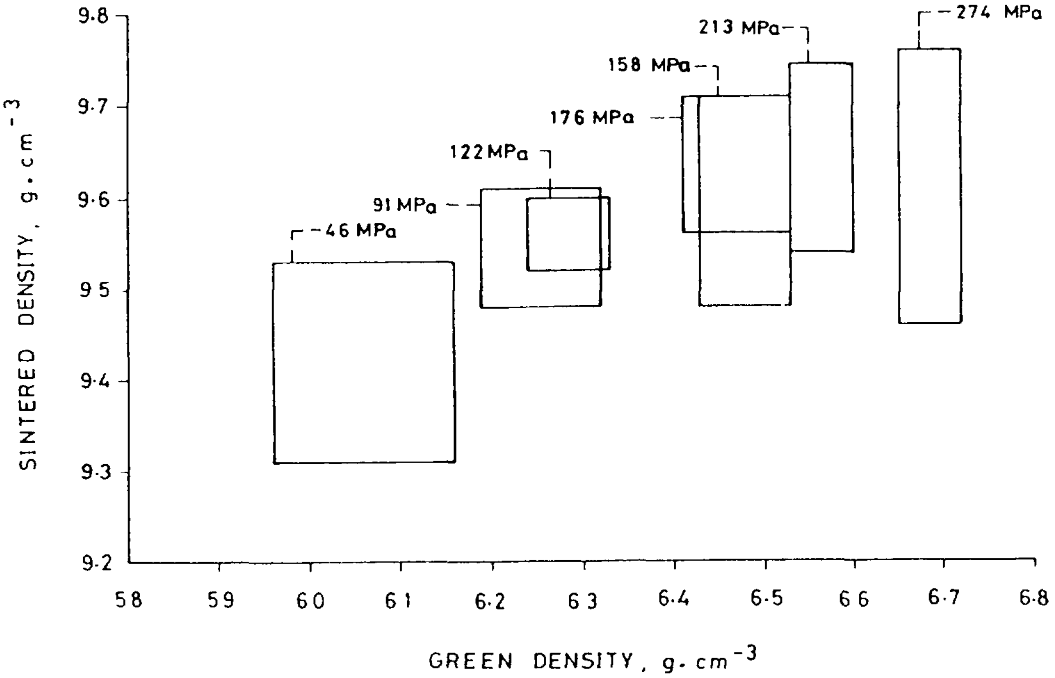

*Fig.5 - Variation in green and sintered densities at constant pressures based on different lots.*

### 3.3.4 Absorption of moisture:

*Thoria powder is known to absorb moisture and carbon-di-oxide from atmosphere. Same trend was observed in the case of sintered pellets. Pellets which were dried at 350°C for 2 hrs. and analysed for moisture content within 2 hrs. were found to contain moisture less than 1 cc at STP per 100 gm of thoria. This moisture content increased with increasing time of exposure to atmosphere.*

*The method followed for moisture analysis consisted in heating the pellets upto 800°C under vacuum and estimating the moisture in the pellet.*

## 4.0 Blanket Element and Assembly Fabrication:

*The blanket element and assembly fabrication processes are shown schematically in Figs. 6 & 7.*

### 4.1 Fabrication of Elements:

### 4.1.1 Washing, Drying & Loading of Pellets:

*Thoria pellets, ground to size and accepted for loading, are washed with hot demineralised water and dried in a drying oven for 2 hours at 350°C.*

*The dried pellets are stacked in lengths of 1000 ± 1 mm and loaded into clad tubes, one end of which is closed by welding bottom end plug to the tube. A gas plenum tube is kept at the head of each stack before closing the top end by welding top end-plug.*

*In order to prevent absorption of moisture from atmosphere by sintered pellets, the second end of clad tubes is necessarily closed within 2 hrs. of drying.*

### 4.1.2 Second-end plug welding:

*The second end of the clad tube of 316 M Stainless Steel is closed by welding the top plug to its open-end. While the first end-plug welding is carried out by TIG process with argon gas, the top plug is welded in an enclosed chamber*

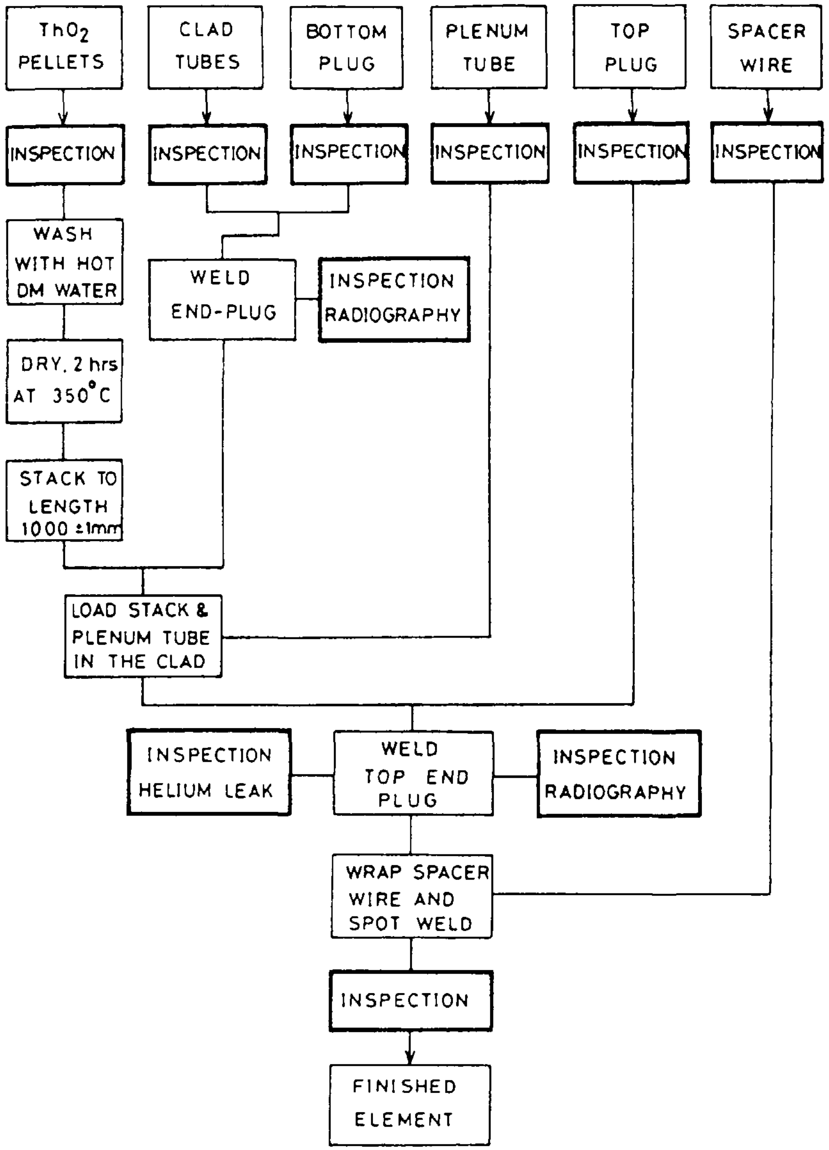

*Fig.6 - Blanket element fabrication process flow-sheet.*

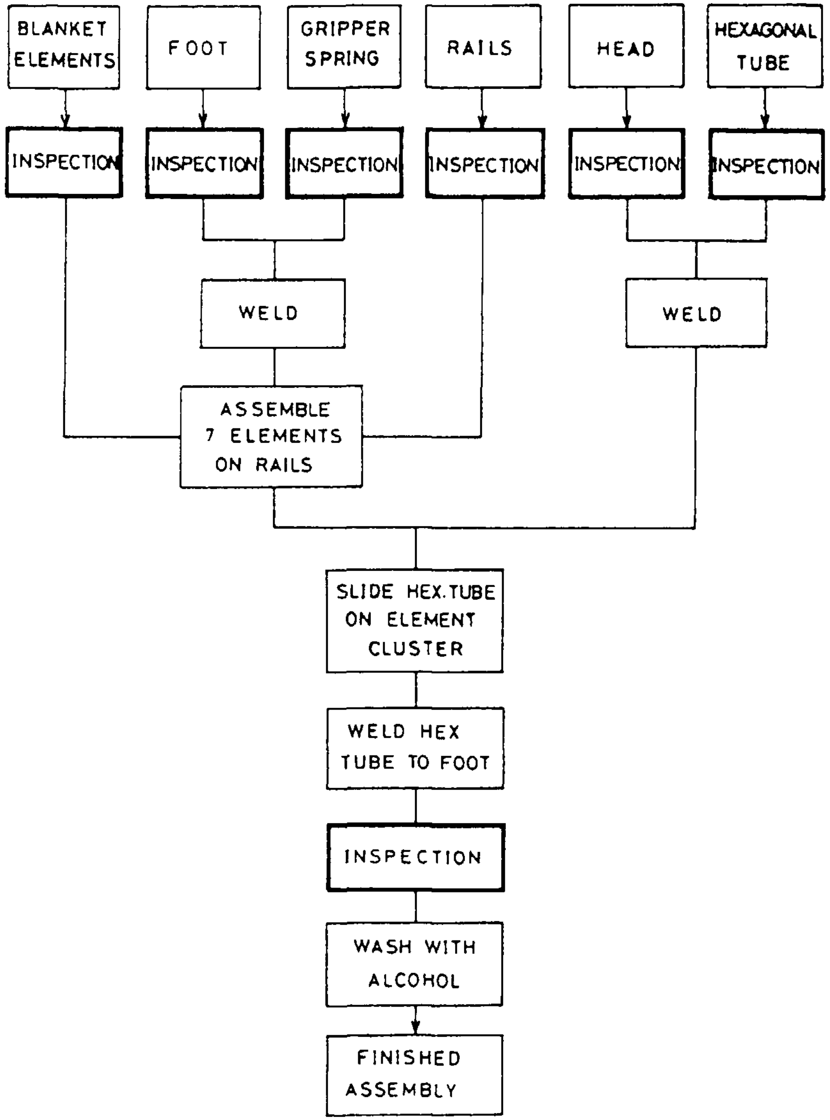

*Fig.7 - Blanket assembly fabrication process flow-sheet.*

Table II - End-plug welding parameter.

| PARAMETER | BOTTOM PLUG | TOP PLUG |
|---|---|---|
| WELD CURRENT | 30A | 25A |
| R.P.M. | 4 | 6 |
| TIME AT WELD CURRENT | 35 Secs. | 12 Secs. |
| DOWN SLOPE TIME | 15 Secs. | 12 Secs. |
| ELECTRODE SIZE | 1 mm | 1 mm |
| ELECTRODE GAP | 0.4 mm | 0.4 mm |
| COVER GAS | Argon | Helium |
| GAS FLOW | 7 l.p.m. | - |

*filled with helium for helium bonding the pellets to the clad tube. The end-plug welding is carried out with an automatic welding cycle having provision for up-slope, down-slope controls of welding current. No filler wire is used during welding of either top or bottom plugs. The welding parameters used for top & bottom plug welding is given in Table II.*

### 4.1.3 Spacer Wire Wrapping:

*Clad tubes, filled with thoria pellets and both ends closed by end-plugs, are then wrapped with a 0.7 mm dia S.S. wire on a helical pitch and spot welded to the clad tube at every quarter turn. Spacer wire wrapping and spot welding is done using a special fixture fabricated for this purpose.*

### 4.2 Assembly of Elements:

*In a blanket assembly, the elements are held to the 'foot' by pin like rails which pass through the holes provided in the foot and the bottom end-plug of an element.*

*For assembly, the 'foot' section is held vertically in a fixture. Seven elements are assembled one by one over the 'foot' with the help of rails. The elements, when assembled, assume a hexagonal geometry having across flat dimension equal to the inside across flat dimension of a hexagonal tube which envelopes the cluster of element assembly. To this hexagonal tube is already welded the 'head', the upper section of the assembly, normally used for handling* and lifting the assemblies in the core. Holding the elements vertically in the fixture, the hexagonal tube with 'head' welded to it, is slid over the elements and over the 'foot,' till the step in the 'foot', meant for welding hexagonal tube to 'foot.'

### 4.3 Welding of Hexagonal tube to 'Foot':

In order to meet the stringent requirements of straightness of finished assemblies, the welding is carried out in another fixture. In this special purpose fixture, the two sections to be welded could be precisely aligned and held tight to prevent distortion after welding. The bed of the fixture, which holds the assembly can be swivelled in horizontal as well as vertical position.

The fiuxture bed is swivelled to vertical position and tac welding of hexagonal tube to 'foot' is done. Next the bed is swivelled to horizontal position and full face welding is completed. No filler wire is used for welding. A similar fixture and process is used for welding 'head' to hexagonal tube.

Straightness of the finished assembly is measured at the top end of 'head' at middle of the assembly and at the lower end of the hexagonal tube, just above the welding joint.

Occasionally, balance welding has to be done at the 'foot' side of the weld to improve straightness.

### 4.4 Alcohol Washing:

Washing the finished assembly with alcohol is the final step in fabrication of radial blanket assembly. After the components have been degreased, all subsequent handling is done under clean-room condition and with lint-free hand gloves on. However, before the assemblies are packed and despatched for reactor use, they are washed with alcohol.

Alcohol washing system is a closed circuit system. It consists of a double walled chamber, a pump, a filter and an alcohol storage tank. The assembly is lowered in *the chamber and the lid is closed. Alcohol, when pumped, enters the assembly through its 'foot', leaves through its 'head', flows over its outer surface and then to the annulus of the double walled chamber. The level of alcohol rises in the annulus and the chamber till the outlet which is provided at a level little over the level of 'head' in the chamber. The outlet from washing chamber is connected to alcohol storage tank. The assembly is thus washed with circulating alcohol for about ten minutes before it is removed and air-dried.*

## 5.0 *Quality Control:*

*Strict quality control is exercised at every stage of fabrication of assemblies. Starting with the clad tubes, which are produced in one of the plants of the Nuclear Fuel Complex, other components and semi-finished products like elements are inspected to assure compliance with the drawings and specifications.*

*The clad tube, besides meeting the mechanical properties and metallurgical conditions as per requirement, are tested by ultrasonic testing method for internal defects. The components, which make the assemblies are intricate in shape and involve precision machining and inspection. Weld integrity of welds besides qualification set-up welds and process weld evaluation.*

## 6.0 *Conclusions:*

*Based on rather limited experience so far, certain conclusions could be drawn as follows:*

- *Well characterised thoria powders, produced via oxalate route, are amenable to normal powder metallurgical methods of fabrication of shapes for reactor use.*
- *Defect free pellets with high green densities are obtained at comparatively lower pressures, which probably is indicative of good compactibility of thoria powders.*
- *Sinterable grade thoria powder is strongly sensitive to the route by which it is obtained.*
- *An economic and viable recycling method for rejected sintered pellets as well as powder needs to be developed.*

- *A more realistic specification for acceptance/rejection of pellets has to be evolved, taking into account the actual service conditions in FBRs. This of course probably takes some time till experience builds-up in actual use of this material in FBR conditions.*

## 7.0 Acknowledgement:

*Authors gratefully acknowledge the help and assistance given by Atomic Fuels Division, BARC; M/s. Indian Rare Earths Ltd., and Design Group, Reactor Research Centre in connection with the work reporeted in this paper.*

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# REPROCESSING AND REFABRICATION OF THORIUM-BASED FUEL

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## Abstract

In the last decade extensive R&D has been carried out at our institute on the thorium fuel cycle, particularilly on reprocessing and refabrication of High Temperature Reactor (HTR) fuel.

For reprocessing thorium-based spent fuel, the dual cycle THOREX process has been thoroughly investigated. The study demonstrates that there are no significant differences between the two extraction modes with either acid or acid deficient feed solution. The investigations have also shown that the feed solution of the second extraction cycle need not be acid deficient since the history of zirconium does not play a role for its decontamination. For re-extraction, the original flow-sheet has been changed such that the Th/U partition is already achieved in the first cycle. The formation of a crud produced by a $Th(DBP)_4$ precipitate can thus be avoided.

For refabrication of U-233 bearing HTR fuel kernels, the EGT sol-gel process has been developed. The process starts with a solution containing $Th(NO_3)_4$ and $UO_2(NO_3)_2$. The kernel production is completed in a few steps and besides ammonia and water no other chemicals are necessary. In conjunction with the chemical process, equipment for continuous and remotely controlled operation was developed, e.g. a particle transport system, a belt drier, a sintering furnace, etc.

On the basis of the EGT process a gel pelletization route for the production of $(Th,U)O_2$ pellets has been developed. Two important changes were necessary for the sol-gel process: the sol must contain carbon black as well as $Ca(NO_3)_2$. The pores remaining after burning off the carbon black improve the compactability and provide for closed porosity in the pellets whereas calcium improves the sinterability. At a pressing load of 350 MPa and a sintering temperature of 1773 K, pellets with a density of ≥ 96 % TD and an excellent microstructure can be produced.

## 1. Introduction

Uranium-233, bred in the thorium fuel cycle, exhibits the highest neutron yield of all fissile materials when used in reactors with a thermal neutron spectrum. Because of this reason, considerable work was conducted on this cycle in addition to that on the uranium-plutonium cycle during the fifties and sixties, when the technology was developed to technical maturity. Significant efforts were made in many countries to utilize thorium as a fertile material. In contrast, the present interest in the thorium fuel cycle is quite limited. This change is attributed to several causes.

The use of thorium as a fertile material offers the greatest advantages in heavy water moderated reactors, where very high conversion ratios are reached and even genuine breeding might be expected. However, it is the light water reactor, not the heavy water reactor, which has been commercially established in the last two decades. In this system the thorium cycle is much less attractive. Instead, this reactor system uses the uranium-plutonium fuel cycle, as does the sodium-cooled fast reactor.

The use of thorium is also promising in a second reactor system; that is, in the graphite-moderated high-temperature reactor. Using the thorium-uranium fuel cycle instead of the uranium-plutonium cycle, reductions in uranium consumption of 60 % are possible. To date, however, the expectations of commercially introducing this reactor type have not been fulfilled. This too has contributed to decreased interest in the thorium fuel cycle. Development work on the thorium fuel cycle was greatly impaired, moreover, by the INFCE results. The decision to abandon the use of highly enriched uranium for nuclear energy applications virtually rules out the thorium fuel cycle, since economic utilization of thorium as a fertile material requires the use of highly enriched material. The resulting unavailability of highly enriched uranium for the foreseeable future has led to the decision in the Federal Republic of Germany to abandon the use of thorium in future HTR's and to rely on the uranium-plutonium fuel cycle instead.

Of course, this decision also greatly affected experimental work on reprocessing and refabrication of thorium based fuel at the Institute of Chemical Technology of Kernforschungsanlage Jülich.

All plans for the construction and operation of a hot pilot plant for reprocessing thorium based HTR fuel were deferred and only cold development of the THOREX process was continued for several years. On the other hand, R&D on refabrication of thorium based HTR fuel was completely abandoned. However, on the basis of our thoria sol-gel process for HTR fuel production, a procedure for production of pellet fuel for water cooled reactors was developed. This gel-pelletization process should be particularly suitable for recycling of thorium based fuel since it is amenable to continuous and remote operation and does not produce radioactive dusts.

## 2. Cold reprocessing studies

### 2.1 Distribution data

The commonly used solvent extraction procedure for reprocessing spent thorium based nuclear fuel is the THOREX process with its different variants. Like the PUREX process, the THOREX process works in a nitrate medium and uses a 30 % solution of tributyl phosphate in an organic diluent such as dodecane for extraction of the heavy metal nitrates into the organic phase.

So far, distribution data for the system $Th(NO_3)_4$:$HNO_3$/30 % TBP in a paraffinic diluent have only been evaluated by SIDDALL who used ultrasene, a mixture of paraffins [1]. However, these data do not give all the information that is necessary, e.g. for flowsheet development. Also computer calculations of distribution data yield unsatisfactory results, at least in certain concentration ranges [2].

Therefore, we have evaluated distribution data for the systems $Th(NO_3)_4$:$HNO_3$/30 % TBP in dodecane and $Th(NO_3)_4$:$UO_2(NO_3)_2$:$HNO_3$/30 % TBP in dodecane. By means of these data a calculational model has been derived which makes it possible to determine data in the investigated range. A method is also described for the evaluation of distribution data at very low thorium concentrations [3,4].

### 2.2 Studies on the dual cycle THOREX process

#### 2.2.1 General remarks

The first THOREX process, which uses only $HNO_3$ as a salting agent, was developed by ORNL [5]. The process uses a feed solution of 1.1 M $Th(NO_3)_4$, a special feature being an acid deficiency of about - 0.15 M. The scrub solution is 1 M $HNO_3$. The acid deficiency is adjusted by steam stripping of the feed solution. Fission product decontamination (particularly with respect to zirconium) is reported to be improved due to the presence of hydrolyzed species in the feed solution.

On the other hand, undesirable precipitates can be encountered during feed adjustment from power reactors. For this reason, a dual cycle THOREX process was developed by Farbwerke Hoechst [6] as part of the German project.

This process (FIG. 1) uses a 1 M $HNO_3$ feed solution in the first extraction cycle and a 0.1 M $HNO_3$ scrub, thus avoiding hydrolytic precipitates during feed adjustment. After having separated the main volume of fission products in the first cycle, an acid deficient feed solution is used in the second cycle to reach the desired decontamination factors. In this cycle the scrub is 1 M $HNO_3$.

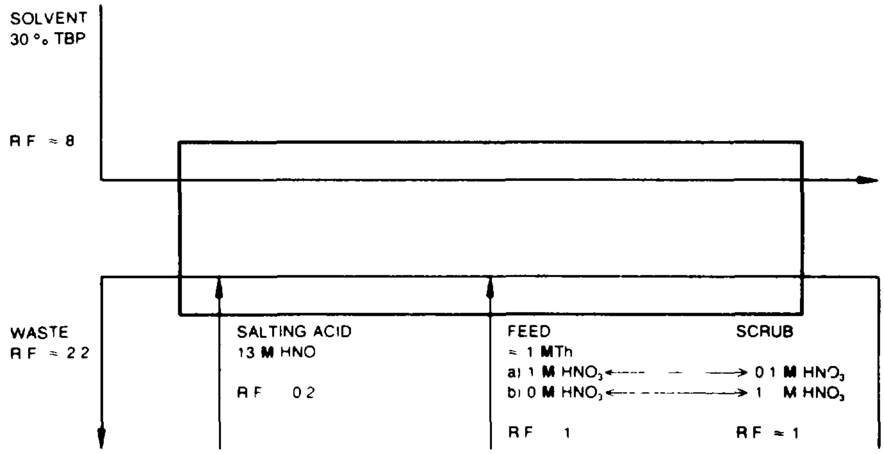

FIG 1: General flow-sheet of the dual cycle THOREX process

Re-extraction of thorium and uranium is carried-out as co-stripping in the first cycle. In the second cycle, separation of thorium and uranium is accomplished by subsequent re-extraction.

### 2.2.2 Investigations of the extraction modes

As can be seen in FIG 1, the two extraction procedures of the dual cycle process differ very much in their $HNO_3$ supply via feed solution and scrub. The low $HNO_3$ concentration of the incoming scrub solution in the THOREX process with acid feed solution suggests that there is a substantially lower $HNO_3$ concentration in the scrub section. The question arises whether both flow sheets can be stable under such different process configurations.

An analysis of the $HNO_3$ profile in mixer-settlers and pulsed columns has provided the answer (FIG. 2). The acidity is almost the same for both process variants, only in some parts of the scrub and

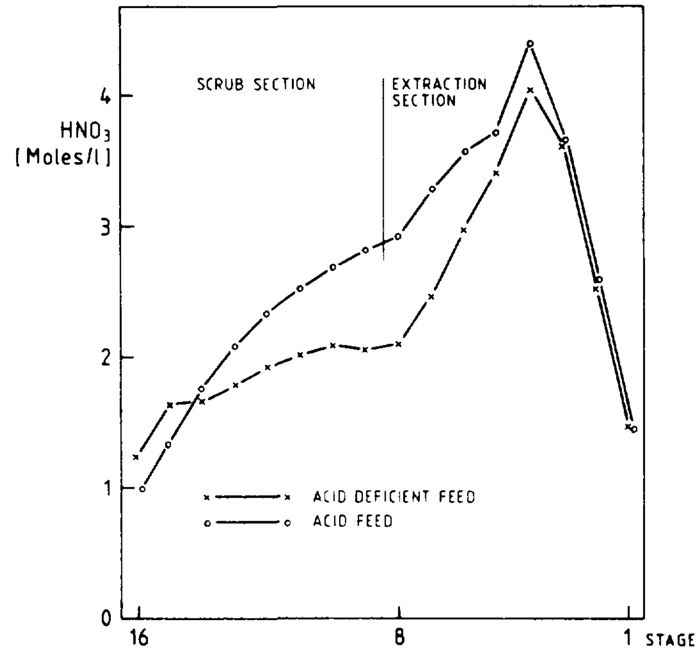

FIG 2: Acidity profiles in the dual cycle THOREX process

extraction sections the $HNO_3$ concentration of the variant with acid feed solution is somewhat higher. Obviously, $HNO_3$ is transported into the scrub section via the organic phase.

Cold extraction runs with zirconium and Zr-95 as a tracer have revealed a slightly better zirconium decontamination for the acid deficient variant (DF = 80 instead of 40). This difference is only due to the lower acidity in the scrub section, since batchwise zirconium extraction experiments (again with Zr-95 as a tracer) have demonstrated that the history of the zirconium does not play a role. Hydrolyzed Zr species produced during adjustment of the acid deficiency are obviously converted within minutes to the species present in a highly acidic medium ($\geq 2$ $\underline{M}$ $HNO_3$) as found under extraction conditions.

### 2.2.3 Re-extraction studies

The original dual cycle THOREX process provides a co-stripping in the first cycle.

Our investigations have shown that during co-stripping a process-perturbing crud formation occurs in both mixer-settlers and pulse columns. Initially the crud is formed in the vicinity of the inlet of the aqueous phase by interaction with the unloaded organic phase, i.e. in those places where the acid concentration is lowest.

In-depth investigations have shown that a precipitate of thorium with dibutyl phosphate (DBP) is produced. The precipitate has exactly the formula $Th(DBP)_4$. DBP is generated in the extraction step by hydrolysis and radiolysis of TBP and then transported to re-extraction via the organic phase. At the low acidity necessary for co-stripping, DBP is also re-extracted and precipitates with thorium.

In cold experiments, the DBP concentration formed by hydrolysis during the extraction process was sufficient for the formation of the $Th(DBP)_4$ crud. The disturbances due to the crud could be avoided in mixer-settlers by substantially reducing the flows (while maintaining the flow ratios). This leads to sufficiently long residence times in the settler chambers for the emulsion to separate again. Hot tests will have to show whether this measure will suffice for high-burn-up fuel when much higher DBP concentrations are to be expected.

The formation of a crud can be avoided if the strip solution has an acidity of $\geq 0.5$ $\underline{M}$ $HNO_3$ because $Th(DBP)_4$ does not precipitate at $\geq 0.7$ $\underline{M}$ $HNO_3$. Since partitioning but not co-stripping is possible at this nitric acid concentration, Th and U should be already separated after the first extraction. A modified partitioning using 0.5 $\underline{M}$ $HNO_3$ as stripping solution yields a good Th/U separation and avoids crud problems.

### 2.2.4 Recommended flow-sheet

For the reprocessing of high burn-up fuel from power reactors an acid feed solution must always be used in the first cycle. Our experiments have demonstrated that an acid deficient feed solution is not necessary for the second cycle. A modified "acid" process with reduced $HNO_3$ concentration in the scrub section could provide the same decontamination factors as a process with acid deficient feed solution.

However, when looking at the entire thorium fuel cycle the question arises whether a dual cycle process with high decontamination factors is required at all. It is known that bred U-233 always contains up to a few hundred ppm U-232, which has some very intensive $\gamma$-emitting daughter radionuclides. During refabrication, U-233 can only be handled applying heavy γ-shielding, so that a gross fission product decontamination factor of better than $10^3$ is not necessary. The same applies to reprocessed thorium which contains considerably more Th-228 than its equilibrium value. Thorium, if recycled immediately, must thus be processed in hot cell facilities just as U-233.

These considerations lead to the proposal to return to a single cycle THOREX process. An optimized process with acid feed solution ($\leq 1$ $\underline{M}$ $HNO_3$) should provide the required decontamination factors of up to about $10^3$ for both uranium and thorium.

Co-stripping should be avoided as a re-extraction procedure since $Th(DBP)_4$ precipitates and produces a crud at the low acidity ($\leq 0.1$ $\underline{M}$ $HNO_3$) necessary for this step. Partitioning of uranium and thorium by a separate re-extraction of thorium can be carried out at an acidity ($\approx 0.7$ $\underline{M}$ $HNO_3$) which reliably avoids crud formation. Further decontamination of thorium and uranium can then be accomplished in separate steps.

## 3. Process for fabrication of $ThO_2$ and $(Th,U)O_2$ fuel kernels

### 3.1 Scope of the process development

Dense ceramic $ThO_2$ and $(Th,U)O_2$ microspheres of $\approx 0.5$ mm dia. are used in the fuel elements of High Temperature Reactors (HTR). These particles are produced by aqueous chemical processes, viz. sol-gel processes [7,8]. The EGT ( External Gelation of Thorium) process of KFA has been especially developed for the fabrication of fuel kernels from bred U-233 and irradiated thorium. It has already been mentioned above that U-233 always contains a few hundred ppm of U-232 and its gamma-emitting daughters. The irradiated thorium has an elevated Th-228 concentration. Therefore, production amounts of these materials must be handled in hot cells, even after decontamination of the heavy metals by reprocessing. Thus, special requirements have to be fulfilled for the refabrication in the thorium fuel cycle.

For kernel preparation the development must be directed to simple processes yielding little waste. Because of the necessity of remote operation and maintenance, the number and complexity of process steps should be minimized and equipment should be kept simple. In view of these requirements, external gelation processes seem to be the most suitable. Common advantages of all external gelation processes in use are the comparatively easy preparation and stability of the broths, a fast gelation reaction (which directly influences the production rate and yield), and finally, the small amount of waste produced.

## 3.2 Chemistry of the EGT process

The block diagram of the so-called EGT process is shown in FIG. 3. The chemistry of thorium allows the use of a sol-gel technique for kernel preparation. The method of sol preparation is in principle similar to that described for the SNAM process [9]. But in contrast with the SNAM-process concentration of 0.5 - 0.7 mole/l, a heavy metal concentration of 2 moles/l is used here. In addition, the degree of pre-neutralization is not 70 - 80 % but 87.5 ± 0.5 %. This means that 87.5 ± 0.5 % of the ammonia stoichiometrically necessary for the complete precipitation of $Th(OH)_4$ is added. Since the resulting sols have a viscosity of about $3 \cdot 10^{-2}$ Pa · s the use of a thickener can be avoided if a suitable drop

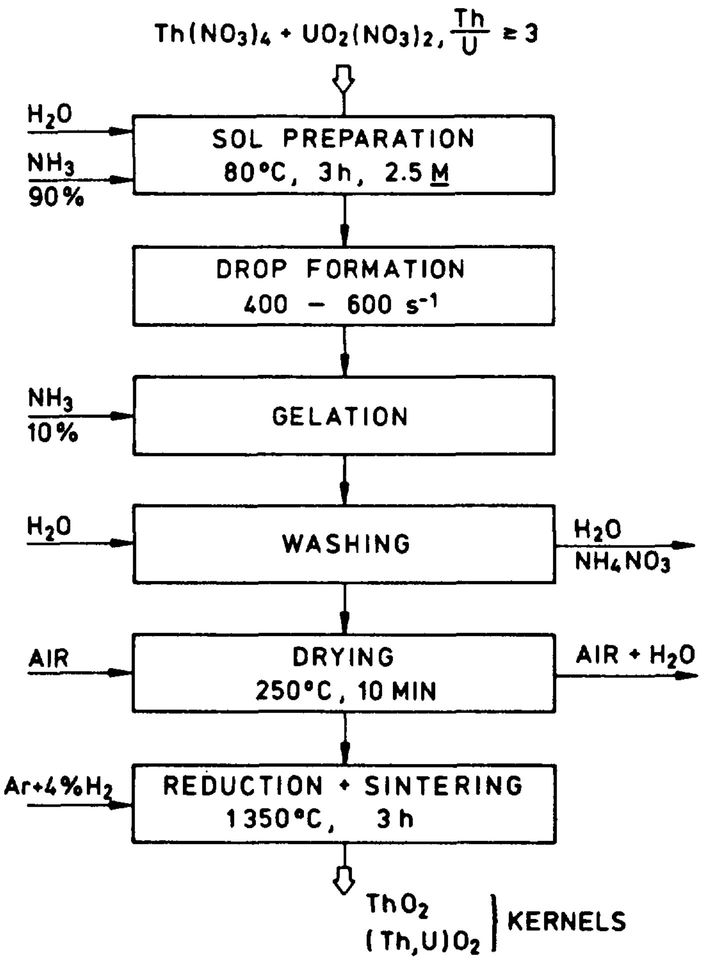

FIG. 3: Chemical flow-sheet of the EGT process

formation device is used. It should be emphasized that apart from ammonia no other chemicals are needed for the process.

Prior to gelation, the sol is dispersed into uniform droplets.

Gelation is a very fast reaction since it takes place by the action of ammonia on sols which are stabilized by hydrogen ion adsorption. The gelation has to start before impact on the surface. As in the SNAM process, the droplets must, therefore, travel through an $NH_3$-gas atmosphere from the time they leave the nozzle until they enter the aqueous ammonia bath. A skin is then formed stabilizing the spherical shape of the droplets. Gelation is completed in the ammonia bath. The ammonia concentration must be of the order of 1 w/o.

Immediately after gelation, washing can take place. This step is necessary for the removal of $NH_4NO_3$ and can be carried out with water in the EGT process. It has been found that the relatively dense, water washed gel kernels can be dried without cracking if the drying takes place in a wet atmosphere. In too dry an environment the outer shells of the kernels quickly become impermeable to water vapor so that the pressure inside the kernels increases and cracks them. The use of super-heated steams is well known for drying kernels but it was found by us that drying can also be done in sufficiently wet air at a temperature between 200° and 400°C.

Sintering of $ThO_2$ kernels can be carried out without any particular temperature program. The kernels reach a density of better than 99 % of the theoretical density after 10 minutes at 1000°C. This is due to the small crystallite size of the sol/gel system. However, to obtain crystallite growth and a heat resistant ceramic material, it is necessary to sinter the kernels at 1250 - 1300°C for about 3 hours. The hexavalent uranium in the mixed kernels must be reduced between 600 and 800°C with $H_2$ or $Ar/H_2$ to obtain mixed oxides, $(Th,U)O_2$. Beyond this temperature, the same heat treatment as for pure $ThO_2$ kernels can be applied.

## 3.3 Description of the process equipment

FIG. 4 shows an equipment flow-sheet of the EGT process. The process starts with the preparation of the sol. Gaseous ammonia is fed into a solution containing $Th(NO_3)_4$ and, if necessary, $UO_2(NO_3)_2$. The gas flows through the hollow shaft of a mechanical

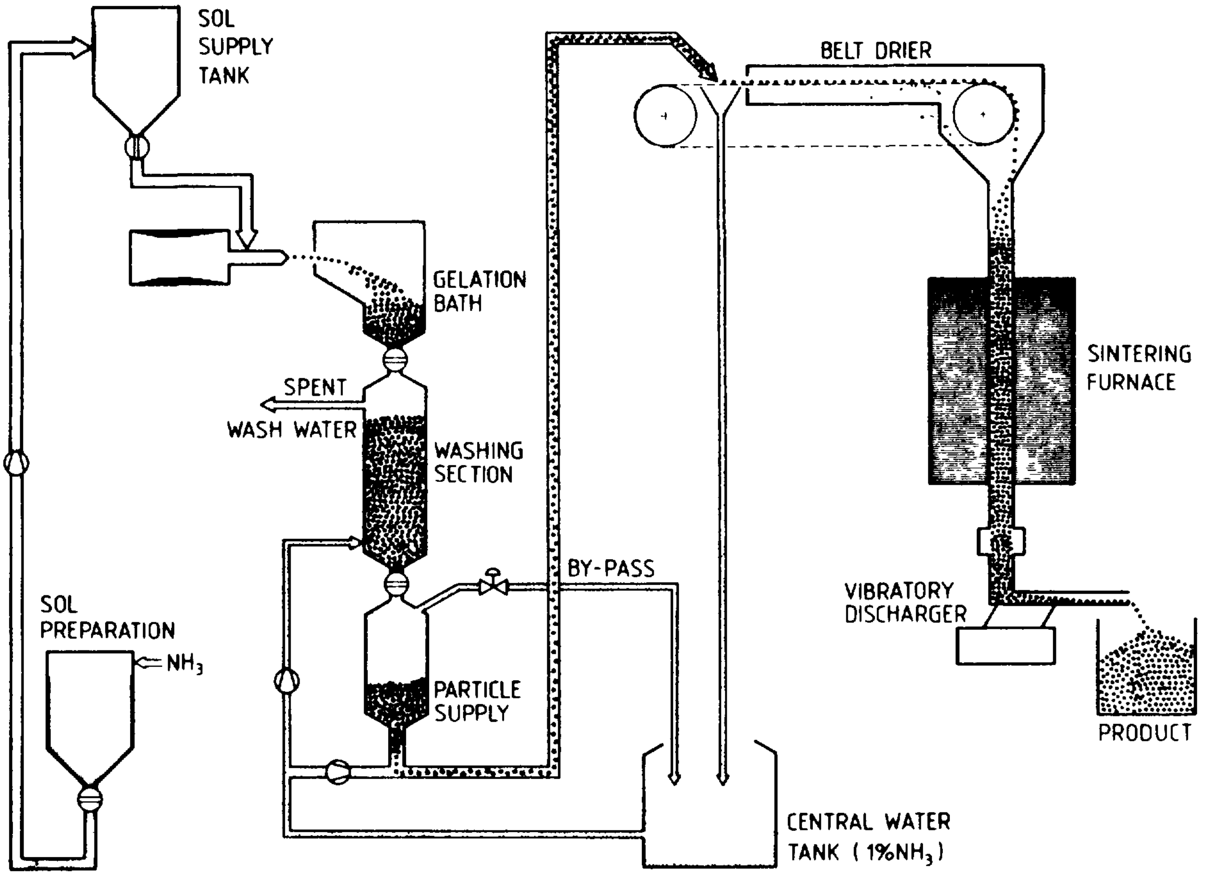

FIG. 4: Equipment flow-sheet of the EGT process

disperser and exits at the circumference of the rotor. This way the gas outlet does not become blocked. The state of the sol is controlled automatically by pH measurement and viscosity control.

In the next step the sol is dispersed into uniform droplets. This is done by feeding a steady laminar flow through a nozzle which vibrates with a constant frequency (typically 500 Hz). Special equipment has been developed to fulfil these requirements [10].

After dispersion of the sol, the droplets enter the gelation box where they first pass an ammonia atmosphere prior to falling into the aqueous ammonia bath for final gelation.

The gel particles are transported from the gelation box to the washing column by gravity where washing is done batchwise. The overall continuous flow is, however, not interrupted since washing is carried out in a few minutes and sufficient storage capacities are available in the gelation box and supply tank beneath the column.

The dried particles are fired and, if necessary, reduced in a continuous operation in a vertical sintering furnace. The microspheres move in a packed bed through the alumina tube of the furnace. The transport of the particles is achieved by gravity and a vibratory discharger. The reducing gas (argon with 4 % $H_2$) enters the furnace at the bottom.

## 4. Sol-gel microsphere pelletization of $ThO_2$ and $ThO_2$-$UO_2$

Thorium is mostly used as a fertile material in the form of $ThO_2$ blankets in both thermal and fast reactors [11,12]. Several countries [13-15], are also considering $ThO_2$-$U^{233}O_2$ ($\leq$ 5 w/o) as an advanced fuel for PHWRs and PWRs on the basis of balanced utilization of uranium and thorium resources and favourable neutron physics characteristics of $Th^{232}$-$U^{233}$ in a thermal spectrum. Both the blanket and fuel materials should have a high density ($\geq$ 96 % T.D.) for enhancing breeding and a microstructure consisting of uniformly distributed 2 - 5 micron pores for satisfactory in-pile performance.

A process flowsheet (FIG. 5) based on "sol-gel microsphere pelletization" (SGMP) has been developed for fabrication of high density $ThO_2$ and $ThO_2$-2 % $UO_2$ pellets at relatively low compaction pressures ($\approx$ 350 MPa) and low sintering temperatures ($\approx$ 1773 K).

The SGMP is a hybrid of the conventional "sol-gel" and "pellet" processes and use sol-gel derived calcined microspheres rather than fine powder derived granules as feed material for pellet pressing. The

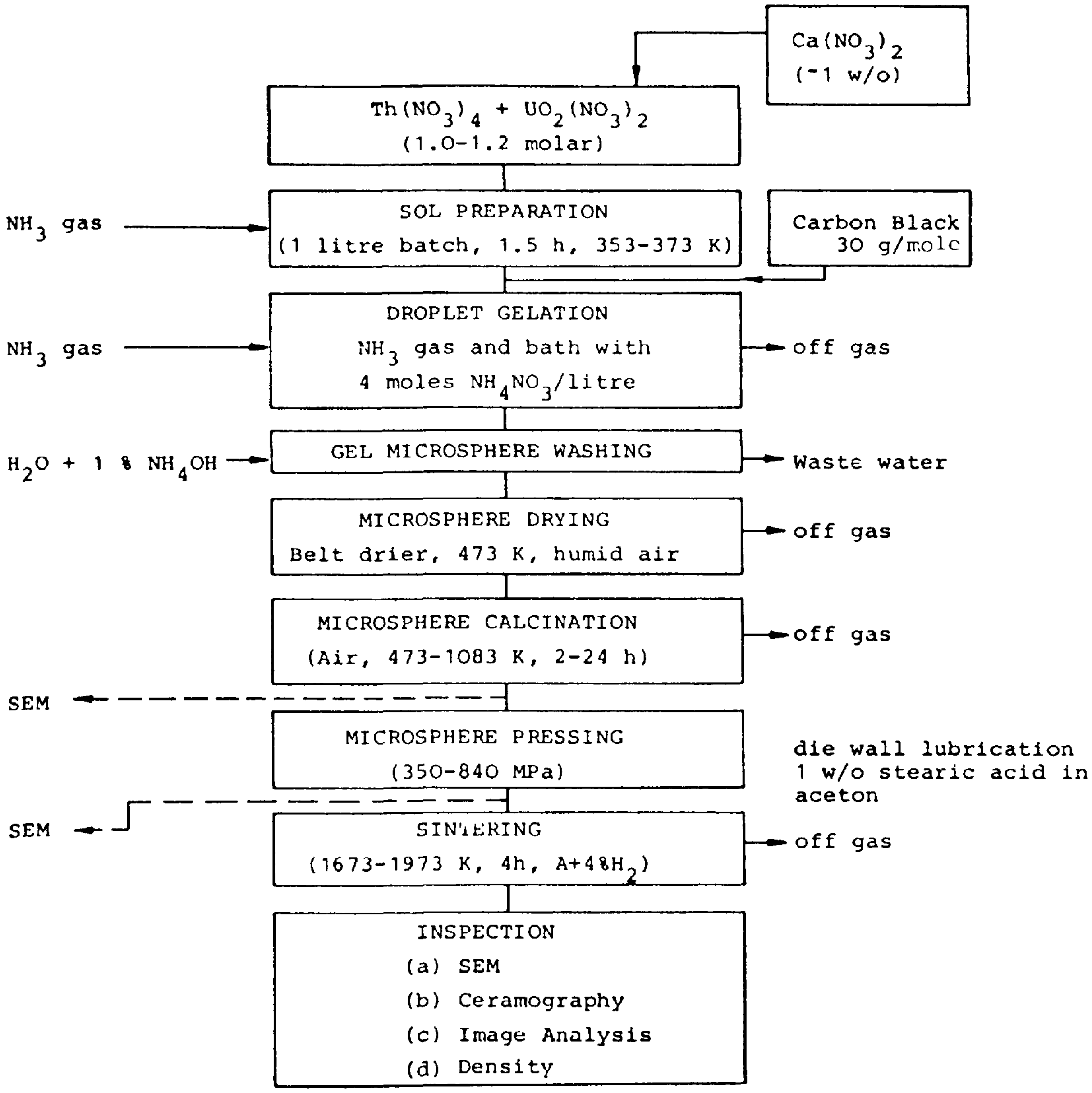

FIG 5: Sol gel microsphere pelletization process for fabrication of $ThO_2$ or $ThO_2$-$UO_2$ fuel, adapting the KFA-EGT process

advantages are as follows:

- radiotoxic aerosols are not produced
- dust free and freeflowing nature of microspheres make remote fabrication easier
- for mixed oxide uniform dispersion of uranium in fuel is ensured
- fabrication of high density pellets of desirable microstructure is possible.

The KFA-EGT facility (FIG. 4) was used for preparation of gel microspheres. However, 3 major modifications were made in the reference EGT process (8) in order to produce gel microspheres which could be calcined to microspheres suitable for pelletization and sintering. First, heavy metal nitrate feed solutions of lower molarity (1 - 1.2) were found to be more suitable. Secondly, around 1 w/o calcium nitrate was added to the feed solution in order to have around 0.4 w/o CaO "sintering aid" in the subsequent calcined microspheres. The $Ca^{2+}$ substitutes for $Th^{4+}$, resulting in the formation of oxygen ion vacancies which enhance the volume diffusion of thorium ion and in turn improvesthe densification process during sintering. Thirdly, 30 g carbon black per mole heavy metal were added to the sol prior to gelation. The pores formed in the sol-gel microspheres after burning off the carbon black particles reduce the crushing strength of the microspheres and facilitate pelletization.

The optimum process parameters developed for fabricating sol-gel derived microspheres are as follows:

(a) concentration of thorium nitrate or mixed thorium uranyl nitrate feed solution: 1 to 1.2 molar
(b) sintering aid: ≈ 0.4 % CaO

| | | |
|---|---|---|
| (c) | pore forming additive: | 30 g carbon black per mole of heavy metal's |
| (d) | drying of sol-gel microspheres: | On a continous belt drier at 473 K for 10-15 minutes in humid air |
| (e) | calcination of microspheres and simultaneous removal of carbon black particles: | 973 K, 24 hours, air |

The CaO doped porous $ThO_2$ and $ThO_2$-2 % $UO_2$ microspheres, thus prepared, have a low crushing strength ($\approx$ 2.5 N/microsphere), an optimum specific surface area (40 - 50 $m^2/g$) and an excellent sinterability. These microspheres could be pelletized, without difficulty, to a green density between 52 and 54 % T.D. at 350 MPa, and later sintered to a high density ($\geq$ 96 % T.D.) at a relatively low sintering temperature (1773 K).

The pellet density versus sintering temperature plots of $ThO_2$-2 % $UO_2$ pellets prepared from 4 different categories of microspheres are shown in FIG. 6. Porous microspheres without CaO sintering aid could be easily pelletized at 350 - 540 MPa to green densities in the range of 52 to 54 % T.D. but needed a higher sintering temperature ($\geq$ 1973 K) in order to achieve high sintered pellet densities.

The as-polished microstructures of sintered pellets prepared from different categories of microspheres are shown in FIG. 7. Pellets prepared from microspheres without any additives had a typical blackberry structure, showing the boundaries of the microspheres prominently (FIG. 7a). With CaO addition,

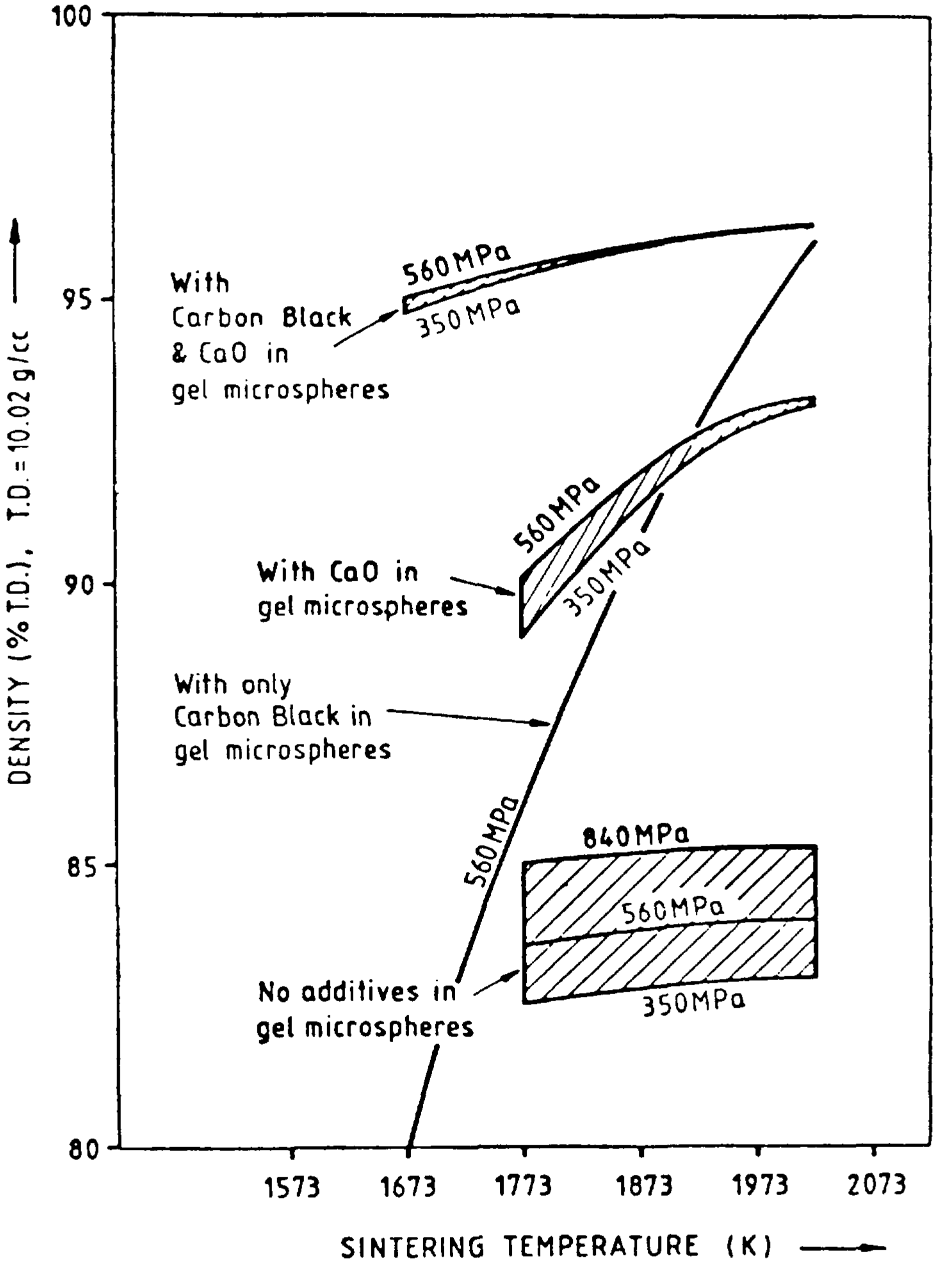

FIG 6: Density versus sintering temperature of pellets prepared from different categories of sol-gel microspheres

the microsphere boundaries were only partly dissolved (FIG. 7b). Only with porous microspheres, the sintered pellets had practically no microsphere boundaries (FIG. 7c). In fact, the porous microspheres,

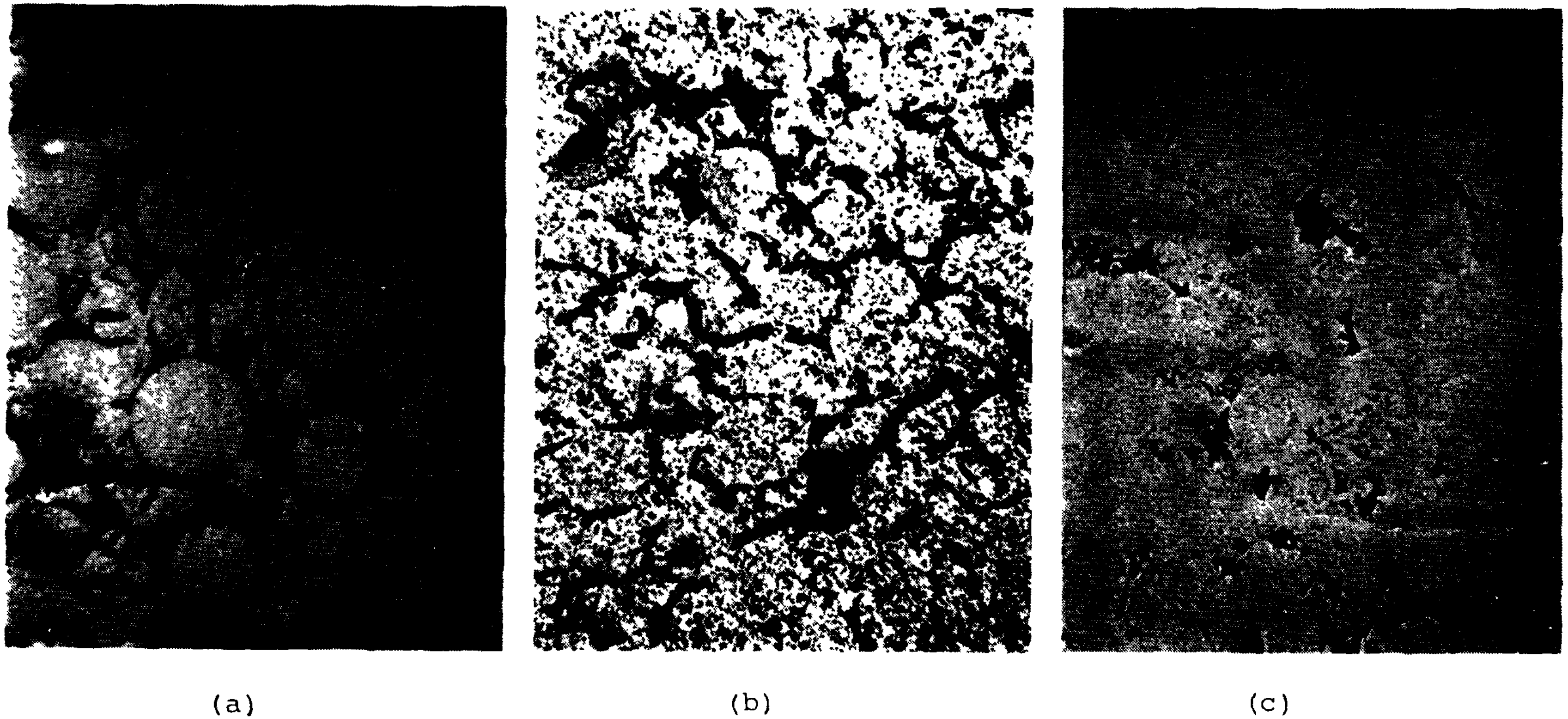

FIG 7: As polished microstructure of sol-gel microsphere pelletized $ThO_2$-2 % $UO_2$ pellets (X50)
(a) without any additive
(b) with 0.4 w/o CaO
(c) with 0.4 w/o CaO and carbon black pore former

because of their low crushing strength, completely disintegrated during the pellet pressing step itself and lost their individual identity.

The SEM picture (FIG. 8) of a fractured surface of a sintered pellet prepared from porous microspheres show uniformly distributed pores and absence of any microsphere boundary. The etched microstructure (FIG. 9) of the same pellets show single phase equiaxed grains in the size range of 15 - 20 micron. The pore size and spatial distributions were determined by the "individual method" of KfK, Karlsruhe with the Leitz TAS analyzer [16]. More than 85 % of the pores in the pellets, prepared from porous microspheres and sintered at 1773 K (with CaO) and 1973 K (without CaO), were in the size range of 2 and 3 micron. A typical plot of pore size and distribution is shown in FIG. 10.

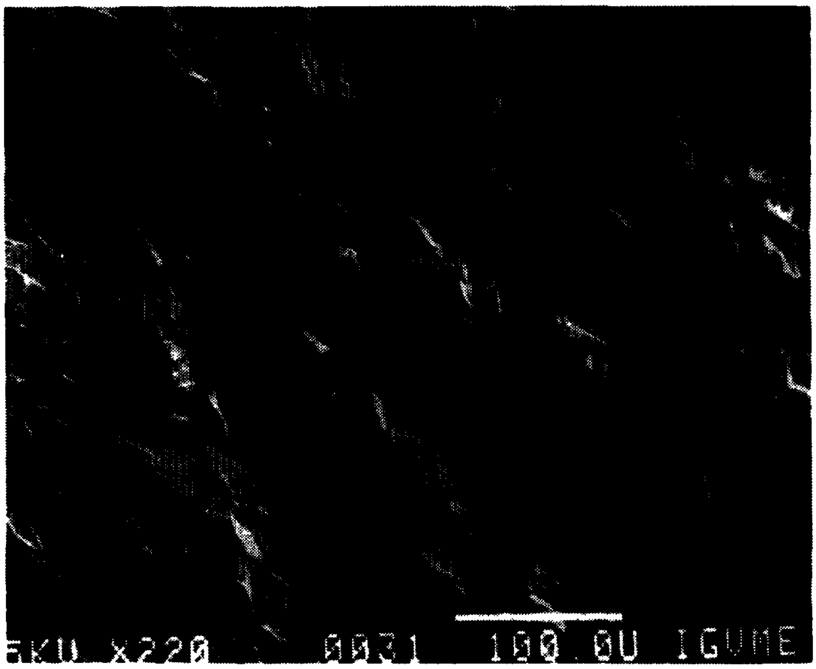

FIG 8: SEM picture of a fractured surface of a $ThO_2$-2 % $UO_2$ sintered pellet prepared by SGMP process (X220)

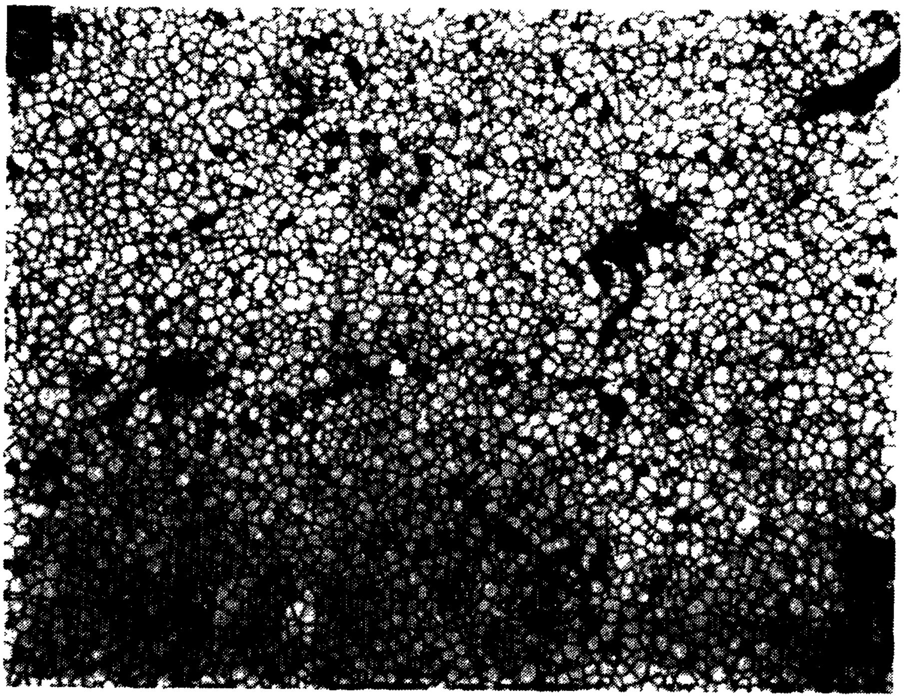

FIG 9: Photomicrostructure of an etched $ThO_2$-2 % $UO_2$ pellet prepared by the SGMP process (X100)

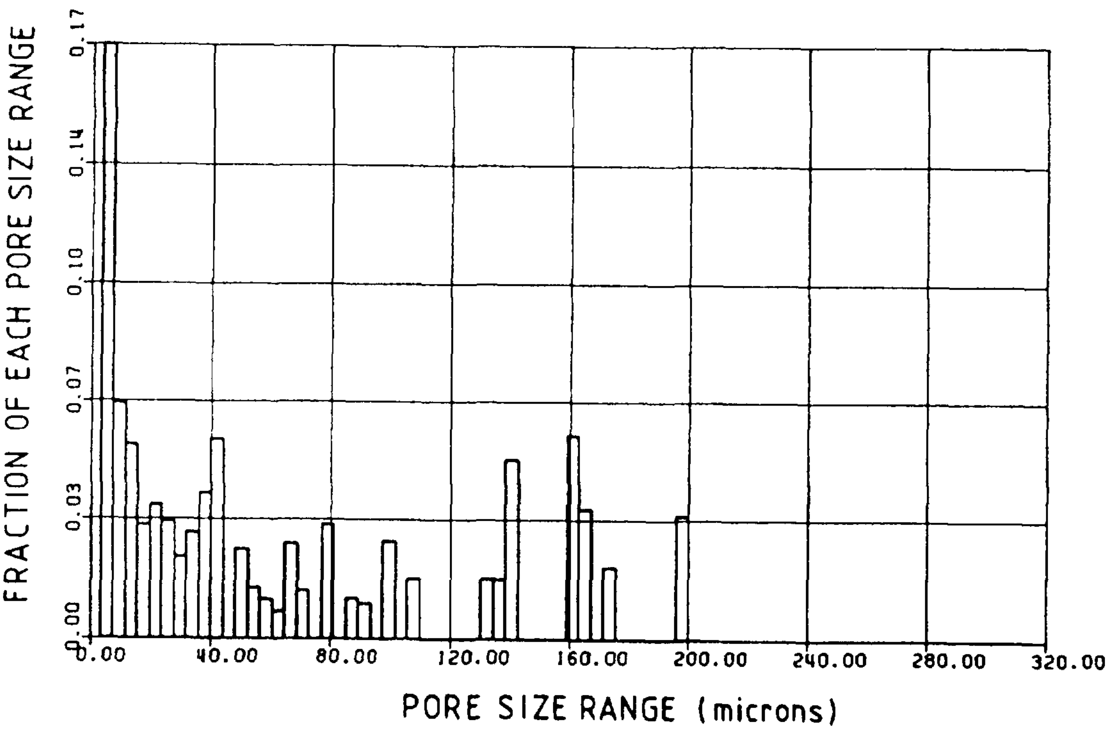

FIG 10: Pore size and distribution of a high density $ThO_2$-2 % $UO_2$ sintered pellet prepared by SGMP process (X122)

Thus, the SGMP process not only avoids dust generation and is easy to remotize but also produces high density pellets of desired microstructure at compaction pressures and sintering temperatures, which are even lower than what are reported [17] for the conventional powder route.

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# OPERATIVE EXPERIENCE IN FIRST CAMPAIGN FOR REPROCESSING OF URANIUM-THORIUM ELK-RIVER FUEL ELEMENTS

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## Abstract

OPERATIVE EXPERIENCE IN FIRST CAMPAIGN FOR REPROCESSING OF URANIUM-THORIUM ELK-RIVER FUEL ELEMENTS.

The main characteristics which differentiates the ITREC pilot plant from the other reprocessing plants are summarized. The report describes: a brief history of the Uranium-Thorium Cycle Program developed in Italy since 1960; the results obtained during the cold tests in the Remote Refabrication Cell until 1974, year in which these activities have been stopped; the operating experience in the Campaign for reprocessing of 20 Uranium-Thorium Elk-River irradiated fuel elements. In particular are described the results of the following operations: dismantling fuel elements and pool water treatment, chopping, dissolution, extraction in a Thorex acid-deficient flow-sheet. Some works of maintenance and modification during hot operations (dismantling machine, replacement of the high level waste evaporators) are also presented. The planned utilization of the ITREC plant is indicated.

## 1. HISTORY OF THE URANIUM-THORIUM PROGRAM DEVELOPED IN ITALY

. The program was carried out in cooperation with the USAEC. In 1960 a joint Italian-American team, started the preliminary design work on the reprocessing and remote refabrication plant.

. The purpose of the research work to be done within the framework of the U-Th Cycle Program was to develop:

- knowledge of the technical and economic feasibility of the application of the Th-U cycle to power reactor, as an alternative to the U-Pu cycle;
- processes and technologies in the field of the processing of irradiated fuels and that of the remote refabrication of nuclear fuels.

. The basic facility for the execution of this research work is the fuel Element Processing and Refabrication Plant (ITREC) built at the TRISAIA Center.

. For the hot experiments in ITREC plant have been utilized the fuel elements of the Elk River Reactor first core.

. In 1974 the Italian Committee of Ministers for Economic Planning (CIPE) decided to stop the U-Th Fuel Cycle Program.
In accordance with that resolution and as regard the ITREC Plant it was decided to stop all activities concerning the fuel remote refabrication and was planned to use ITREC as a pilot reprocessing plant for the performance of hot and cold testing on advanced components (centrifugal contactors, continuous dissolver etc.).

## 2. GENERAL CHARACTERISTICS OF THE ITREC PLANT

The ITREC (Reprocessing and Refabrication pilot plant of fuel elements of the ceramic oxide) differs from most reprocessing plants in the following main aspects:

- the plant was originally conceived and built as an integrated unit for the reprocessing and for the remote refabrication **(Fig. 1)**;

- among the various possible methods for the processing of fuel elements containing uranium and thorium, that used in the ITREC does not involve the separation of thorium and uranium and therefore, after extraction of the fission products, the processed material contains Th-228, a strong gamma emitter which makes it impossible to handle the material without adequate protection. The refabrication must therefore be carried out in a cell fitted with adequate shielding, using remote-operated equipment and techniques;

- all equipments of the main chemical processing are installed in modular units (the Racks Removal System), an arrangement which provides for the remote-controlled removal (after appropriate decontamination) of the individual mobile unit (rack) for maintenance work and modification of components and equipments.

Thanks to this arrangement it is also possible, without particular difficulties and at limited cost, to replace entirely complete sections of the plant to adapt it to new research program. For example, the experiment with centrifugal contactors in the extraction section will be possible by simply replacing the equipment installed on a single movable rack.

This feature, as well as the various alternatives envisaged in the original design, makes the plant extremely flexible. Thus, for instance, the plant can carry out the Th-U co-decontamination flow-sheet as well as the Th-U partition flow-sheet. Also, in the case of adaptation to the reprocessing of fuel from different reactors, the plant is equipped for both the reception and pool storage of the new type of elements and for an increase in dissolving capacity (from 15 to 30 Kg/d).

## 3. REMOTE REFABRICATION CELL: COLD TESTS

In the remote refabrication cell (warm cell) is carried out the last phase of the chemical process and the last cycle of the operations connected with the mechanical refabrication of the fuel elements.

From 1968 until 1974, year in which these activities have been stopped, the refabrication equipment has been tested with U and Th not irradiated, in order to optimize the various process conditions and to improve the remote operation procedures.

All these operations are indicated in the **Fig. 2.**
The coprecipitated Th-$U^{4+}$ oxalates are converted into oxide by calcining the oxalate cake at 920°C.
Green pellets are obtained by cutting the extruded rods into pieces after drying.
To carry on the ceramic fabrication in remote handling conditions a considerable amount of work was devoted to development of special tools.
**Table I** summarizes the characteristics of the sintered pellets obtained since the process was set up in the main lines. With exception of two lots, the number of the pellets meeting the specific density is 90%.
After loading the accepted pellets into the tube the second end plug welding is remotely performed.
Metallographic checks demonstrate that the quality of remotely performed welds is up to the standard of the welds made by direct operation.
All the welds are routinely Xrayed in an apparatus which was developed for the purpose.
The pins are assembled into an Halden type element still inside the refabrication cell.

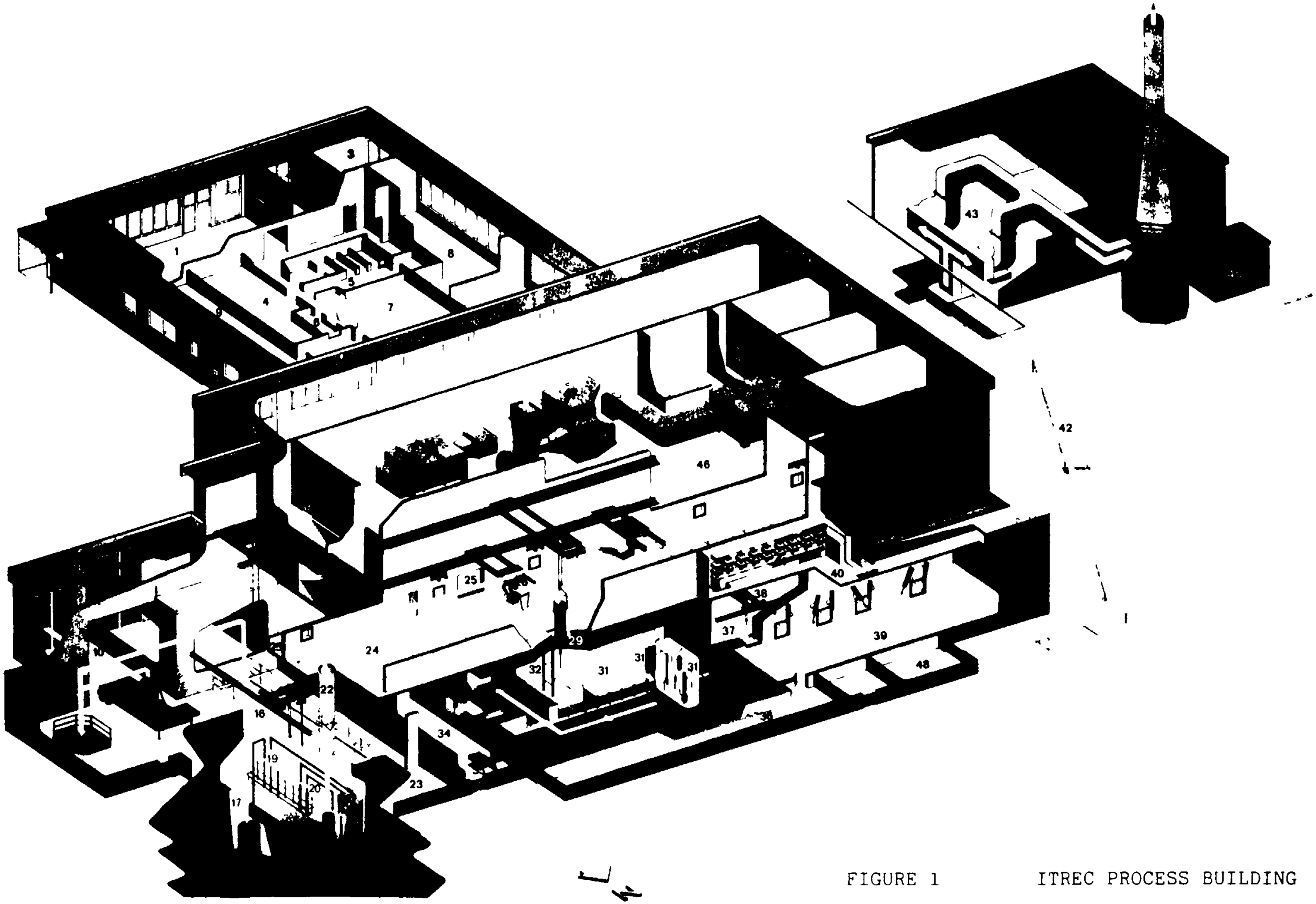
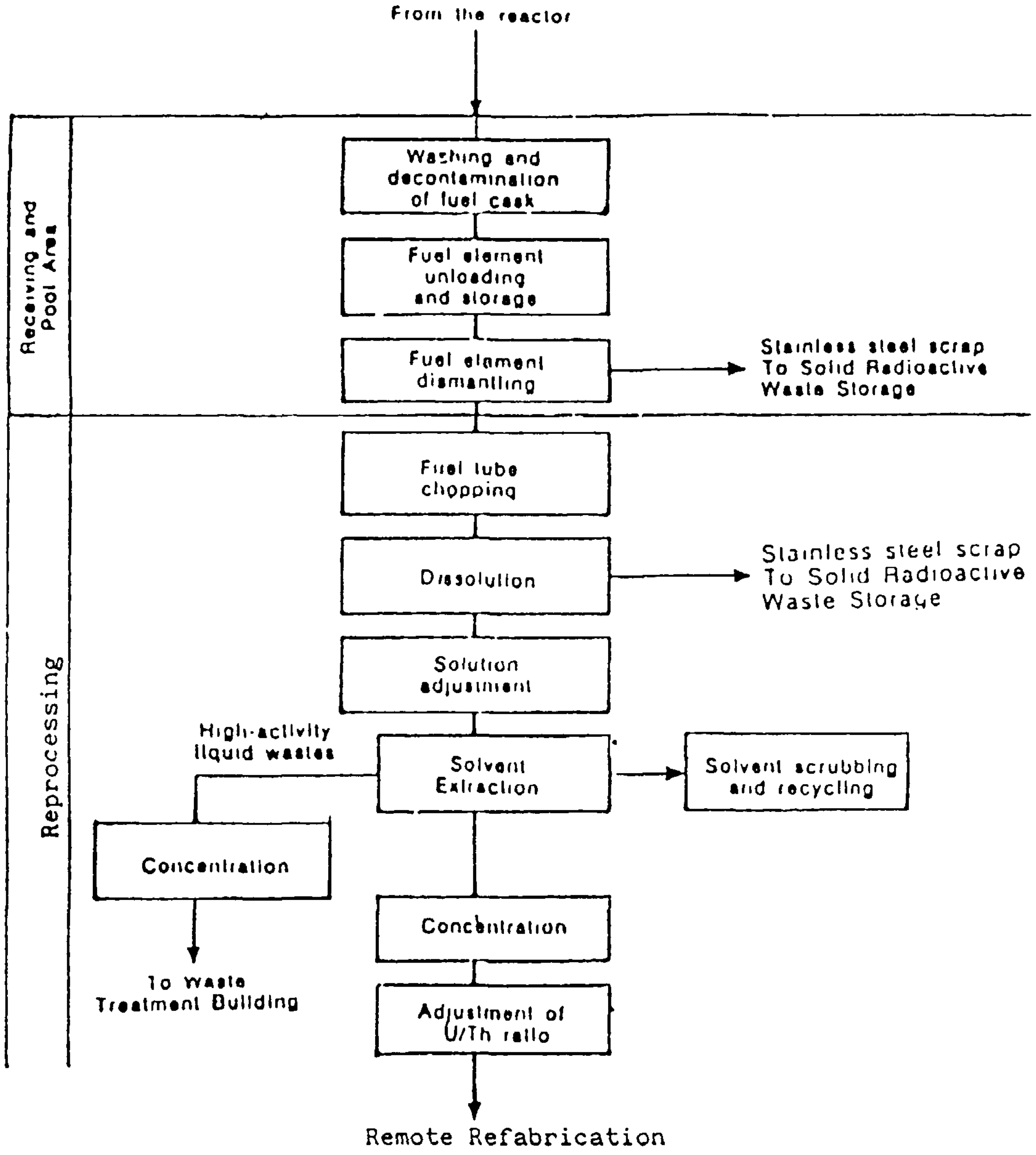


FIGURE 1 ITREC PROCESS BUILDING

# FIGURE 1 - ITREC PROCESS BUILDING

Legenda

| | |
|---|---|
| 1 | Entrance - Display Room |
| 2 | Waiting Room |
| 3 | Relax Room |
| 4 | Dressing Room (cold) |
| 5 | Services |
| 6 | Guard Room |
| 7 | Dressing Room (hot) |
| 8 | Quality Control Laboratory |
| 9 | Laundry |
| 10 | Receiving Area |
| 11 | Cask |
| 12 | Cask Washing Area |
| 13 | Cask Carrier |
| 14 | Crane 50 ton. |
| 15 | Crane 10 ton. |
| 16 | Pool Fuel Storage |
| 17 | Well Fuel Discharge |
| 18 | Fuel Dismantling Machine |
| 19 | Fuel Element |
| 20 | Disassembled Element |
| 21 | Pins container |
| 22 | Air-lock |
| 23 | Pool Water Treatment |
| 24 | Corridor Hot Cell |
| 25 | Operating Area Corridor |
| 26 | Electronic Manipulator MASCOT-1 type |
| 27 | Crane 10 ton. |
| 28 | Master Slave Manipulator |
| 29 | Fuel Chopping Machine |
| 30 | Hot Cell |
| 31 | Rack |
| 32 | Dissolver |
| 33 | Decontamination Cell |
| 34 | Operating Area of Decontamination Cell |
| 35 | Rack Removal System |
| 36 | Fire-Fighting System |
| 37 | Refabrication Cell |
| 38 | Crane 3 ton. |
| 39 | Operating Area Refabrication Cell |
| 40 | Plenum Air Filtration |
| 41 | Hot Filters Battery |
| 42 | Off-Gas Header |
| 43 | Ventilating Fan |
| 44 | Stack |
| 45 | Off-Gas Monitoring |
| 46 | Air Conditioning Area |
| 47 | Warm Filters Battery |
| 48 | Intermediate Storage Low Leve Solid Wastes |


From the reactor
Receiving and Pool Area
Washing and decontamination of fuel cask
Fuel element unloading and storage
Fuel element dismantling
Stainless steel scrap To Solid Radioactive Waste Storage
Reprocessing
Fuel tube chopping
Dissolution
Stainless steel scrap To Solid Radioactive Waste Storage
Solution adjustment
High-activity liquid wastes
Solvent Extraction
Solvent scrubbing and recycling
Concentration
To Waste Treatment Building
Concentration
Adjustment of U/Th ratio
Remote Refabrication

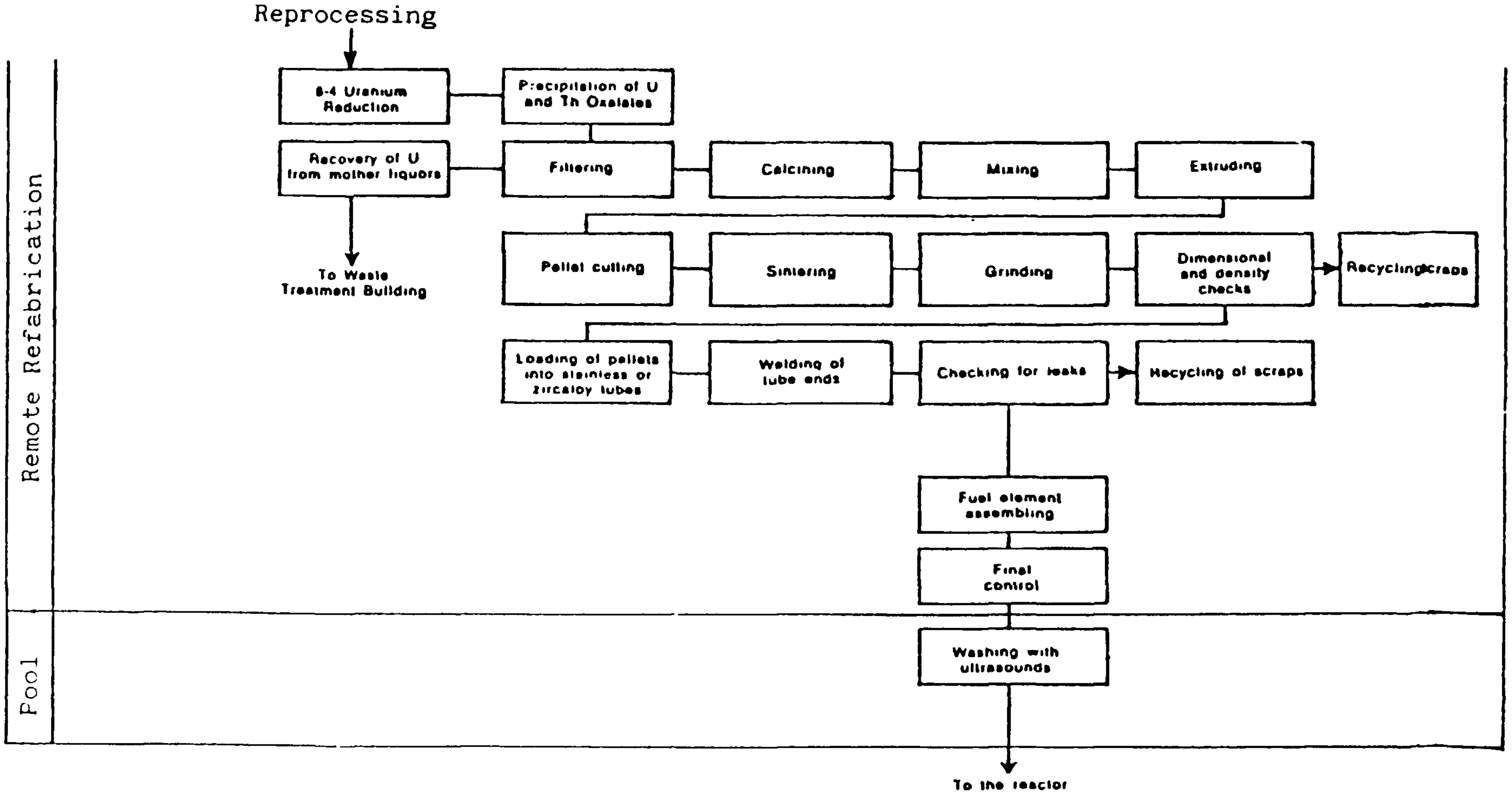


FIGURE 2 BLOCK CHART - ITREC PROCESS BUILDING

TABLE I CHARACTERISTICS OF A SAMPLE OF SINTERED PELLETS OBTAINED DURING PRENUCLEAR TESTS IN REFABRICATION CELL

| LOT No. | Pellets produced | DENSITY | AVERAGE SHRINKAGE % | | | | BEND | | |
|---|---|---|---|---|---|---|---|---|---|
| | | % of pellets meeting spec. | Ø | Length | Volume | Weight | Average Value mm | Standard Deviation | $\frac{n}{n_t}$ (****) |
| 11 | 61 | 90 | 20.52 | 21.11 | 50.16 | 2.49 | 0.36 | 0.15 | 24.5 |
| 12 | 49 | 53 | 19.02 | 19.25 | 47.05 | 1.70 | 0.48 | 0.17 | 18.4 |
| 13 | 72 | 100 | 22.96 | 20.17 | 52.63 | 1.87 | 0.34 | 0.11 | 38.9 |
| 14 | 83 | 97.6 | 23.18 | 20.02 | 52.36 | 1,81 | 0.34 | 0.12 | 41.0 |
| 15 | 60 | 88 | 21.96 | 19.07 | 50.70 | 1.84 | 0.35 | 0.12 | 28.1 |
| 16 | 55 | 100 | 23 | 20.53 | 52.6 | 4.45 | 0.35 | 0.1 | 14.5 |
| 17 (*) | 68 | 20 | 22.81 | 21.07 | 53.17 | 2.16 | 0.28 | 0.14 | 20.6 |
| 18 | 54 | 80 (***) | 20.91 | 19.19 | 49.16 | 1.65 | 0.30 | 0.20 | 20.4 |
| 19 | 37 | 90 | 21.13 | 19.45 | 49.85 | 1.49 | 0.30 | 0.09 | 18.9 |
| 20 (**) | 57 | 0 | 23.27 | 20.80 | 53.36 | 1.84 | 0.31 | 0.10 | 26.3 |
| 21 | 56 | 92.7 | 22.41 | 19.73 | 51.69 | 1.81 | 0.19 | 0.07 | 22.2 |

NOTES:

(*) - Lot excessively heated in mixer-muller; quantity of PAA doubled and clay re-worked in mixer-muller

(**) - Extrusion pressure excessively low (0.15 ton/cm$^2$ instead of the regular value > 0.3 ton/cm$^2$)

(***) - Pellets out of spec. only for density higher than 96% theor. Very high extrusion pressure applied

(****) - n = number of pellets within $\bar{x} \pm \frac{\sigma t}{\sqrt{n_t}}$ ($\bar{x}$ = average bend, $\sigma$ = standard deviation, t = Student's factor, Probability 0.05, $n_t$ = number of pellets produced).

A number of modifications, suggested by work experience have been made on the various unit operations to optimize the ceramic process and the equipment functionality (for example the sol-gel process instead of oxalate precipitation and pelletization instead of extrusion).

## 4. ERR FUEL CAMPAIGNS RUN IN ITREC PLANT

Following the decision to phase-out the remote fabrication activity, only the chemical process section of the plant, together with the mechanical head-end (fuel assembly, dismantling and individual pin cutting) has been put into active operation.

The hot operation has been started in July 1975, with the reprocessing of 20 ERR Th-U spent fuel elements, irradiated in the Elk-River (USA) reactor. For this first campaign the maximum cooled and less irradiated elements have been chosen, whicn characteristics are reported in **Table II.**

The experience acquired in the hot operations can be considered as complete and regards the following activities·

- Mechanical operations in the receiving, dismantling and chopping of fuel elements.

- Chemical processes in the unit operations as dissolution, feed adjustment, solvent extraction, solvent recovery, wastes and pool water treatment.

### 4.1. Receiving and Dismantling Area

In case of the receiving area, hot work experience regards the cooling, washing and unloading of the 3 Elk River casks.
The dismantling operations involved 36 of the 84 Elk River fuel elements stored in the ITREC pool.
The dismantling operations showed that the machine was not functionally perfect because of its complexity.

The principal troubles were experienced in the.

. deformation of the element holder along the vertical axis

. block of the knife extractors probably due to seizure

. oxidation and deformation of the rotable finger due
to error in the choice of materials.

TABLE II ISOTOPIC COMPOSITION AND ACTIVITY

| ISOTOPIC COMPISITION | 20 ELK-RIVER ELEMENTS REPROCESSED | EVALUATED DATA FOR OTHER 64 ELK-RIVER ELEMENTS |
|---|---|---|
| Th-232 Kg | 495.570 | 1565.100 |
| U-Tot " | 20.490 | 64.710 |
| U-235 " | 16.845 | 48.290 |
| U-233 " | 1.465 | 7.810 |
| U-238 "<br>34-36 | 2.170 | 8.610 |
| ACTIVITY | | |
| Gross α Ci/KgU | 3.750 | 14.000 |
| Gross β " | 236.540 | 2.600 |
| Gross γ " | 84.230 | 1.100 |

Therefore, a series of complexed operations (decontamination, extraction from the pool etc.) were made for a radical maintenance of the dismantling machine.
The pool water treatment takes place on a mixed resin bed after filtration.

### 4.2. Fuel Chopping and Dissolution

After the dismantling of the fuel element assembly, each pin was cut into 2 cm. pieces, which fall into the dissolver.

A Thorex "acid-deficient" flow-sheet was used for the codecontamination of U-Th in a single cycle 30% TBP solvent extraction.

In case of the chopping and dissolution section hot operation has confirmed the good results of the prenuclear tests.

The dissolution yields on the 4 batches are as theoretically foreseen, whereas on the first 3 batches (input solution) they are 98-99% for Th and 99.5-99.8% for U.
Visual examination of the hulls and the spectrum of a sample of hulls analyzed prove both the perfect chopping operation and the complete dissolution of the nuclear materials.
The losses of $HNO_3$ in the OFF-GAS dissolver give approximately 10 - 12% when, during chopping, the dissolver solution temperature reaches 100°C and drop to 6 - 7% when the attack solution is at room temperature.

During hot operation the complete chopping group was replaced in order to check the remote intervention system.

### 4.3 Extraction system

For the first campaign the flow-sheet selected, from a range of possibilities due to the extreme versatility of the plant, was U-Th co-decontamination **(Fig. 3/4)**.

The U-Th product is stored as concentrated solutions of Uranyl and Thorium nitrate with the possibility of recycling in the input tank for the subsequent Th-U partition campaign or the subsequent sol-gel process conversion. **(Tab. III).**
Theoretical FP's decontamination factor is about $5 \cdot 10^4$ for beta-emitters and $5 \cdot 10^3$ for gamma-emitters. Actual decontamination factors in normal operating conditions were about 1000 for gross-beta and about 1000 for gross-gamma.

The hot run was stopped twice: firstly because of a microloss appeared in HLW evaporator (which concentrates the acqueous raffinates 1 AW flow from first co-decontamination extraction battery) and secondly for the obstruction in the extraction mixer-settlers battery. The second maloperation has caused the bad decontamination of end product and therefore the stored product is a mixture of the end products obtained from the normal and abnormal operations of mixer-settlers battery. In **Figg. 5/6** is reported the behaviour of gross-beta and gross-gamma Decontamination's Factors as well as for some particular gamma-emitters, during the normal plant operation.

In the 1AW current the losses of Th are very near to the theoretical values of approximately $1^\circ/_{oo}$ while it is practically impossible to determine the losses of U ($< 1^\circ/_{oo}$; the concentration found is at the sensitivity limit of the test method).

In the re-extraction section theoretical and actual losses also proved extremely low.

In conclusion, the process may be considered operatively safe and satisfactory.

### 4.4. Remote Maintenance

In general the maintenance philosophy of the hot facilities in the plant is based on remote removal and transfer of parts to the decontamination cell. In the ITREC Plant two different approaches have been chosen to solve the practical problems connected with this philosophy:

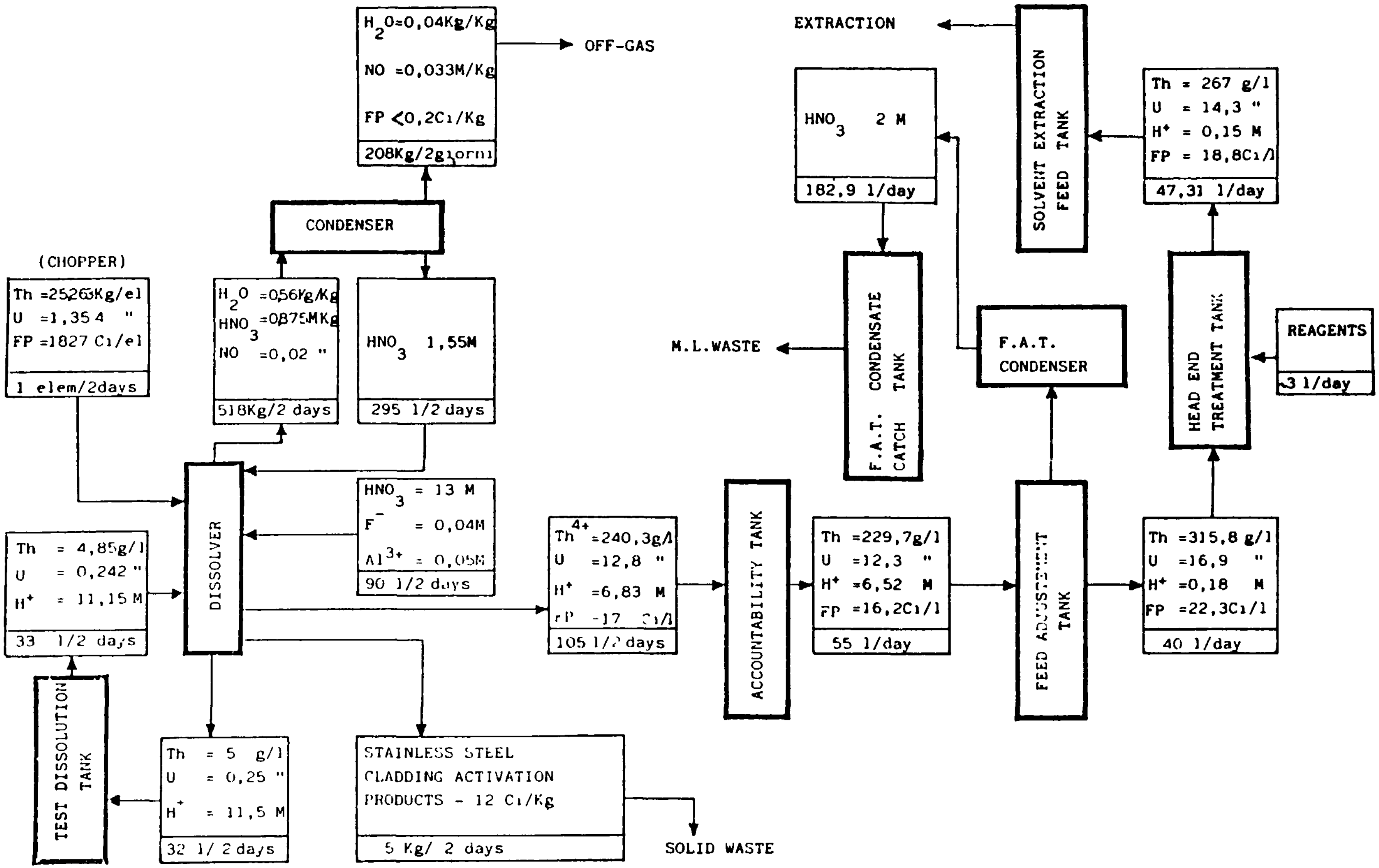


FIGURE 3 HEAD-END REFERENCE FLOW-SHEET

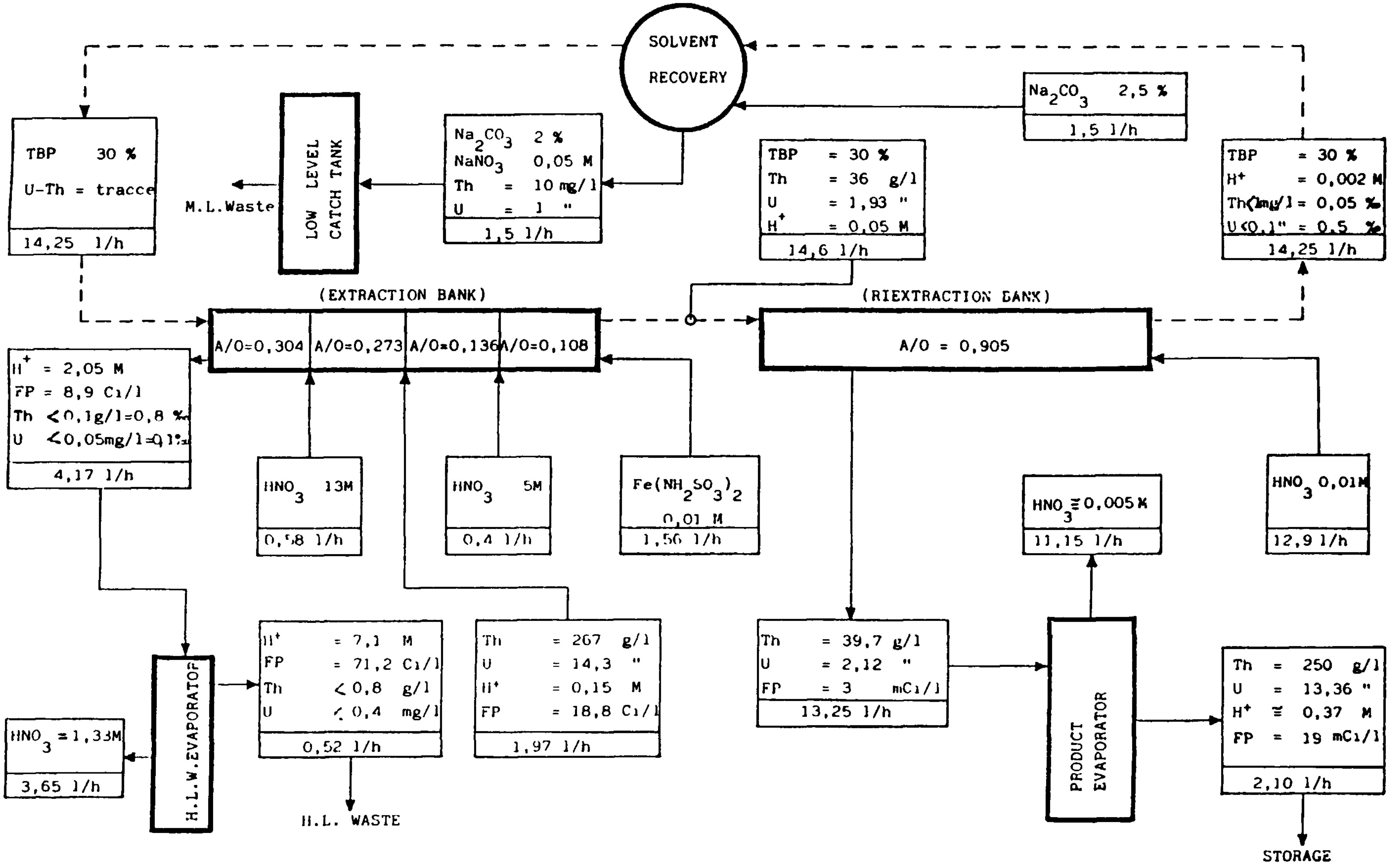


FIGURE 4 U-TH COEXTRACTION REFERENCE FLOW-SHEET

TABLE III CHARACTERISTICS OF ELK RIVER REACTOR ELEMENTS REPROCESSED IN ITREC PLANT - MATERIAL BALANCE

| | CHARGE IN REACTOR gr/elem. | DISCHARGE FROM REACTOR (A) gr.elem. | EXPERIMENTAL DATA IN INPUT TANK (B) gr./elem. | A-B S.R.D % | OUTPUT (C) gr./elem. | B-C M.U.F. % |
|---|---|---|---|---|---|---|
| Th. | 25572 | 25465 | 26410 | -2.6 | 25527 | 3.3 |
| U-TOT. | 1238 | 1149 | 1035 | 9.9 | 1044 | -0.8 |
| U-235 | 1153 | 939 | 851 | 9.4 | | |
| U-233 | -- | 88 | 74 | 15.9 | | |

BURNUP Min. MWD/HT= 3640

BURNUP Max. " =10000

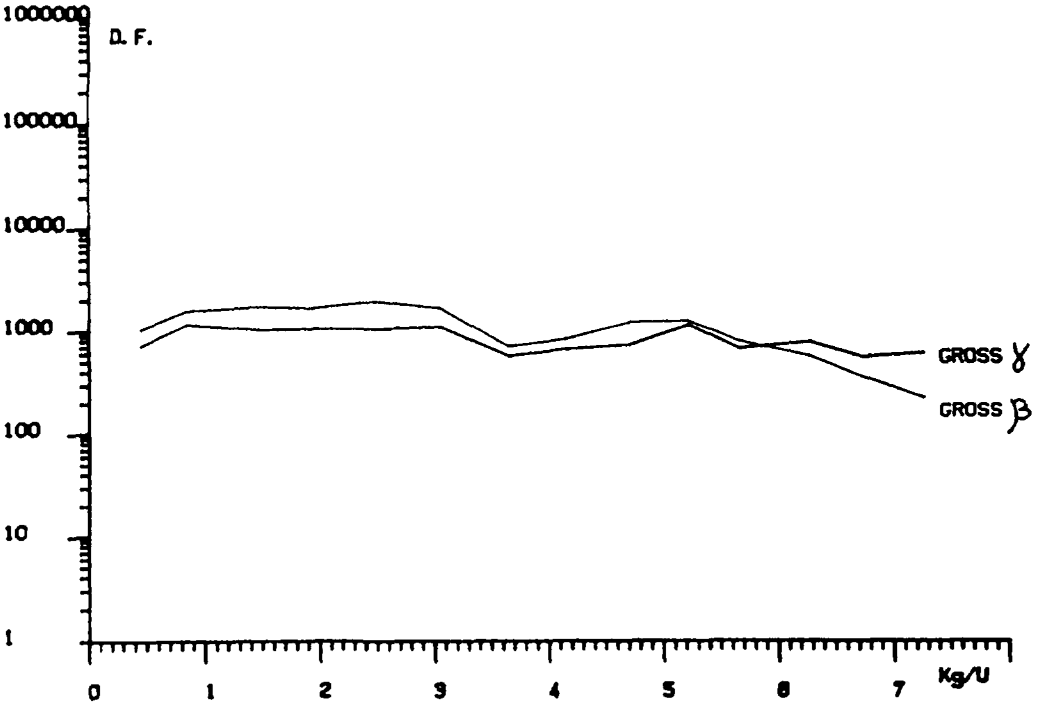


FIGURE 5 DECONTAMINATION FACTOR

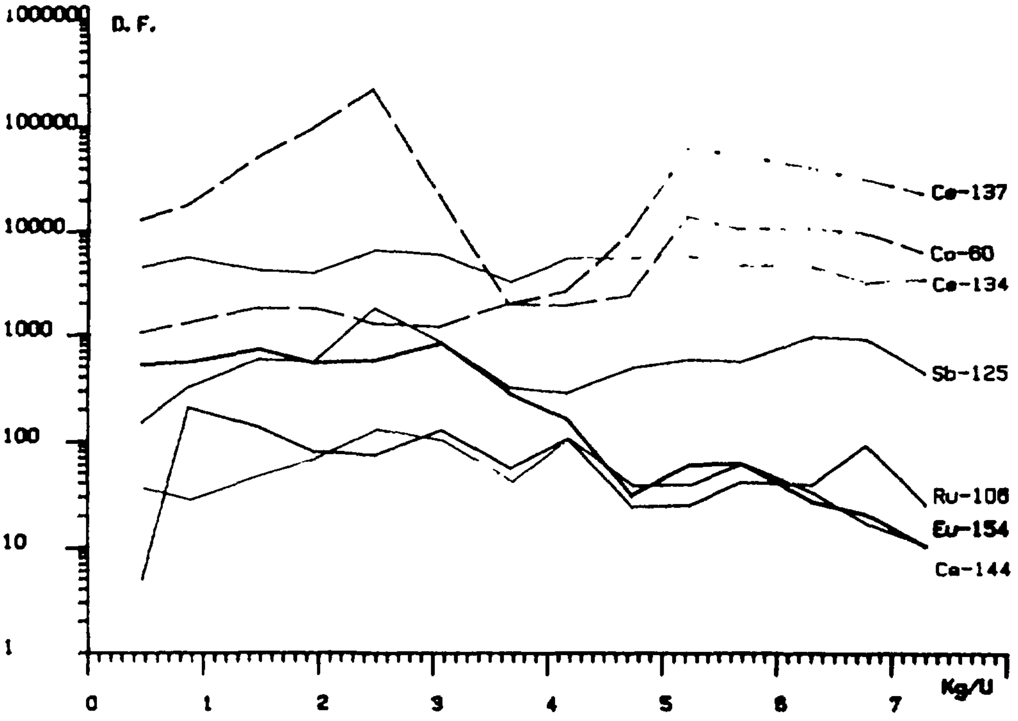


FIGURE 6 DECONTAMINATION FACTOR

1) first approach is installation of all apparatus on removable racks that can be transferred to the Decontamination Cell.

2) second approach is adaptation of every major item by itself as suitable for handling with regular master-slave manipulators.

The first approach (Rack Removal System) has been adopted in the reprocessing section, where the removal of each rack is controlled through positive mechanical devices in so strict a sequence that no provision has been made to even see the operation (no shielding window or camera).
After long operation the system has not induced any suspicion of developing faults and is currently considered as proven.

The second approach inspired the installation of the refabrication equipment. Every item was custom made to fit independently into a general arrangement of the cell, that provides supports and connections preset accordingly to a modular disposition. This accomodation required a considerable amount of engineering work but resulted in more compact and flexible arrangement.

The transfer of equipments to and from the decontamination cell, and of irradiated fuel from pool to process is done through the "corridor" which is an area of restricted access.
An electronic manipulator has been added. The manipulator is of the "Mascot" type, developed by ENEA. It has proved to be most useful as a substitute for man in fine adjustment jobs.

### 4.5. Operational Troubles

The number and variety of incidents which occurred made it possible to gain wide experience in the hot maintenance operations and at the same time had no consequences for operators or for the environment.
Most of these maintenance operations have concerned:

- clogging in the sampling system
- obstruction of ejectors
- blockage of remote head pumps in the extraction process
- chopping machine

The most serious problem concerned the leak on the steam piping of the H.L.W. evaporator. **(Fig. 7).**

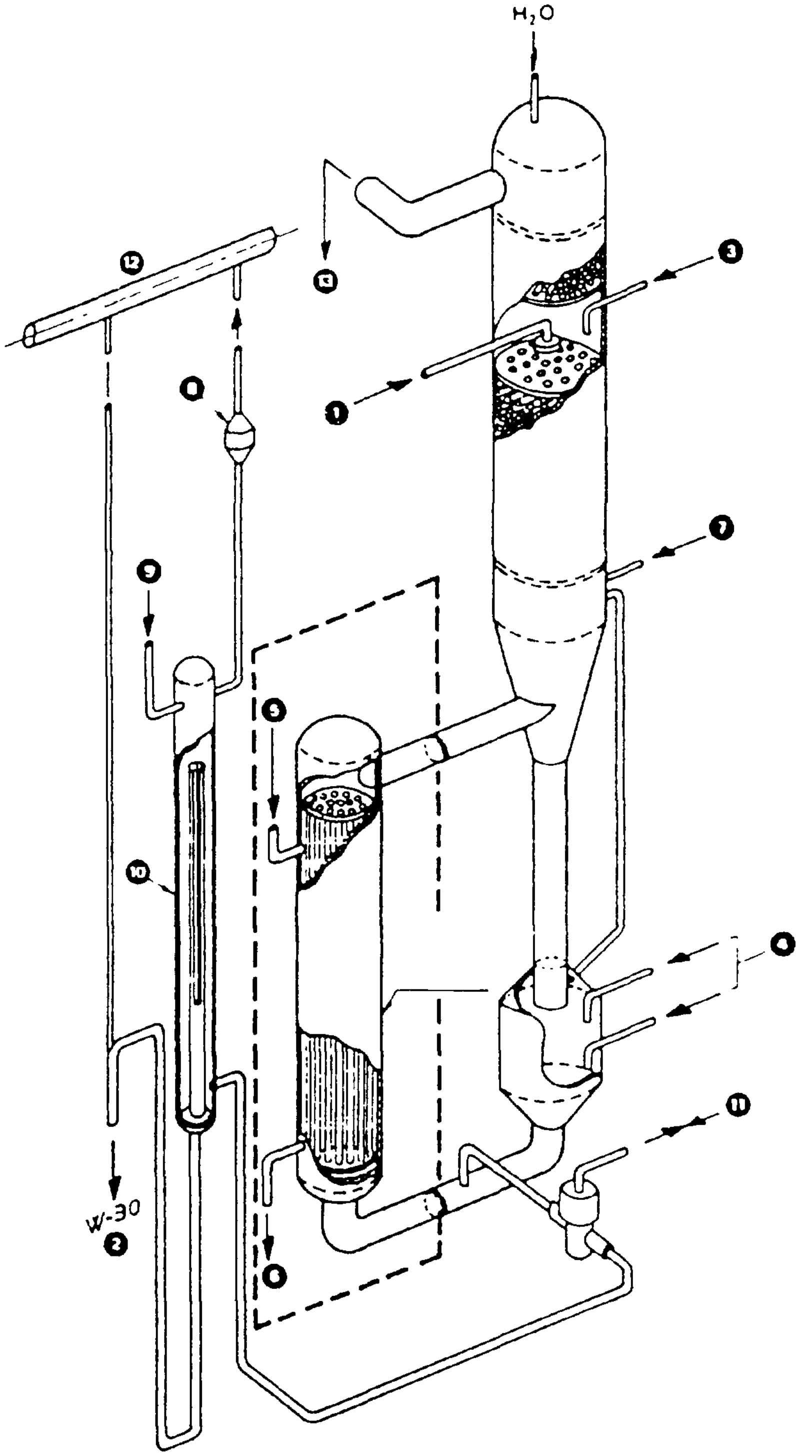


FIGURE 7 LAYOUT OF W-20 EVAPORATOR

*(1) feed inlet; (2) concentrate outlet, (3) level probe; (4) density probe, (5) steam inlet; (6) to monitoring system, (7) level reference, (8) calibrated orifice; (9) air line for pressurized system; (10) air pressurized discharge system, (11) on/off discharge valve; (12) dissolver off-gas; (13) to condenser.*

In 1976, during the first nuclear campaign when the reprocessing of 20 Elk River elements was started a microloss appeared in the HLW evaporator, which concentrates the aqueous raffinates-1AW solution. The Rack No. 6, containing the evaporator, was removed from the hot cell to the decontamination cell. It is the section more difficult to decontaminate as it includes the first mixer-settler extraction battery and the evaporator of the 1AW flow.

The decontaminating solutions used are stocked in the HLW and/or LLW tanks of the plant. The need to ensure compatibility of a long storage of these solutions with plant materials (AISI-304L), has brought to the exclusion of all commercial decontamination agents, because highly corrosive, whose efficiency is based essentially on the combined action of high oxidant substances with high complexing power.

Decontamination of the mixer-settler battery and of rack No. 6 was completed in the decontamination cell by successive washing with: $Na_2CO_3$ at 20%; $COO(NH_4)_2 = 0.3M$ + EDTA = 0.18M; $HNO_3 = 6M$, using for every washing 100 lt. of solution introduced in the first stage of the battery and subsequently in other equipment.
The removed exchanger suitably packed and enclosed in a special container, will be examined internally to verify causes and degree of damage to the steam piping.
After decontamination the intensity of exposure in contact with the shell reaches the maximum values of 300-500 mR/h.
During the intervention on the evaporator 3 ejectors have been replaced because of the frequent obstructions during the hot operation, especially due to solid particles contained in the steam.

The total dose for the whole intervention in the decontamination cell was 2.4 rem, distributed among 19 operators. The maximum individual dose was 433 rem, absorbed during 14 interventions in a 60-day period.
The duration of the hot intervention was 34 hours and 13 minutes, representing 113 man hours.

After the intervention the rack No.6 was replaced in the hot cell and the hot plant operations were restarted to complete the reprocessing of the remaining 13 Elk River fuel elements planned for the first nuclear campaign.

## 5. NEXT PROGRAM OF ITREC PLANT UTILIZATION

In accordance with the strategic destination of the ITREC plant the next program foresees the following main activities:

- 2nd Hot Campaign (1986-1987) with the reprocessing of 16 U-Th Elk River fuel elements for hot tests of a special type centrifugal contactors in the extraction section.

- 3rd Campaign for U-Th partition, recycling in the plant the uranyl and Th nitrate solutions produced in the First and Second Campaigns and subsequent Uranyl nitrate conversion by sol-gel process.

- Experience on the decommissioning technology within the framework of the C.E.C. Program.

- Experience on the storage of spent fuel elements utilizing one lot of the Elk River fuel elements.

# SUMMARY OF PAPERS

Nine papers were presented at the meeting, dealing with the following:

- evaluation of world thorium resources and incentives for further exploration;
- basic research of physical, chemical and nuclear properties of thorium;
- reactor core and blanket concepts regarding utilization of Th-based fuel;
- advanced thorium fuel fabrication technology;
- reprocessing of Th-based fuel.

## 1. Evaluation of world thorium resources and incentives for further exploration

Thorium in association with uranium and Rare Earth Elements (REE) occurs, as shown in the paper presented by Mr. Jayaram, India, in diverse rock types: as veins of thorite, thorianite, uranothorite and as monazite in granites, syenites, pygmatites and other acid intrusions. It also occurres as an associated element with REE bearing bastnaesite in carbonites. Monazite also occurs in quartz-pebble conglomerates, sandstones and in fluviatile and beach placers.

The current knowledge of thorium resources in the world is small because of the relatively low-key exploration efforts arising out of insignificant demand. Resources recoverable at less than $80/kg Th in the Reasonably Assured Resources (RAR) and Estimated Additional Resources-Group (EAR-I) categories are estimated at around 1.4 million tonnes and one million tonnes respectively. And this is due only to the effort made by several countries to identify either uranium or REE. India has about 31% of them. Argentina, Australia, Brazil, Burma, Canada, China, Republic of Korea, Indonesia, Malaysia and South Africa have also considerable resources of thorium. The possibility of discovering new resources, in addition to increasing the known resources contained in the deposits already known, is good.

In spite of today's apparent unfavourability for the thorium utilization situation in the uranium market (low prices for uranium and oversupply) it seems that the perspective of thorium utilization as a nuclear fuel is realistic in the near future. This could be justified by the forecast for uranium utilization in both once-through or closed fuel cycles. Also countries which have limited resources of uranium may have to use thorium to ensure continuous growth rate of electricity generation. Taking this into account the IAEA will contribute further to the exchange of information and co-ordination of national efforts aimed at maintaining and improving knowledge on thorium resources and its utilization as a nuclear fuel.

## 2. Basic research of physical, chemical and nuclear properties of thorium

In general fundamental investigations are concerned with preparing the base for studying the physical properties of nuclear reactors, working with either a mixed fuel cycle or Th-based fuel.

The dependences of U-233, U-232, Pa-233 and fission products buildup upon integral thermal neutron fluxes in high flux ETR-reactor (China) have been studied, including detailed correlation between the above-mentioned values and fast to thermal flux ratios in wide range. These results may be helpful in assessing breeding potential of Th-based fuel in different type of reactors. French work (Attachment IV) on revising and reevaluations of Th-232, U-233 data revealed the following:

The value of Th-232 absoption in thermal range is apparently well-known, though absorption cross sections seem to decrease more rapidly than $^{1}/_{v}$ law. Accurate experimental values should be desirable to improve temperature coefficients forecast. The most desirable further work is in high energy range. The knowledge of U-233 cross sections now is not as good as that of Th-232. Further work is still needed, particularly in the thermal range of energy. This work should include a new accurate measurement of $\nu$, $\sigma_f$, fission spectrum, as well as, benchmark "clean" critical experiments, which can be easily interpretated.

## 3. Reactor Core and Blanket Concepts Regarding to Th-based Fuel Reactors

Strategy investigation, reactor core and blanket design are still important for successful development of Th-based fuel reactors.

The current state of the reactor and fuel cycle facility development suggest the following logics (USSR) in combination with the uranium-plutonium and uranium-thorium cycles for nuclear power. Transition to the U-Pu closed cycle, development of spent fuel regeneration, plutonium accumulation paves the way for rapid introduction of fast breeder reactors which are to be the basis of future nuclear fuel breeding. The accuteness of fuel problem can be then eased by transition of the thermal reactors to utilization of the U-233 accumulated in the fast breeder reactor blankets. That idea is supported by the Indian plans for the behaviour Th-232 investigations in blankets of fast test reactors.

There is, among others, the French idea to use Pu-Th cycle in unmodified PWRs. The main goal of this work is to find a real way for opening the Th-cycle in up-date reactors. The solution proposed consists of initial loading the whole reactor PWR-type with Th-Pu assemblies. Besides, it is an attractive means of storage plutonium in case of rather slow development of LMFBRs. U-233 can be perfectly used in Spectrum Shift Control Reactors. Regarding optimization of neutronics, calculations were being made, including pin power peak, moderator temperature coefficient etc.

Other optimistic results were described in cooperative Brazilian-German paper on Th-utilization in unmodified PWR type reactors. It can be established, that standard PWR may use $(Th, U)O_2$ fuel and even $(Th, Pu)O_2$ fuel without any changes within the reactor system. A favourable strategy to avoid the need of early reprocessing and to strive for savings in uranium might be the use of $(Th, Pu)O_2$ fuel in the once-through cycle with extended burnups.

## 4. Advanced thorium fuel fabrication technology

For refabrication of U-233 bearing HTR fuel kernels the EGT (External Gelation of Thorium) sol-gel process has been developed (paper, presented by Mr. Zimmer, FRG). On the basis of this technology developed for HTR fuel production, a procedure for production of fuel pellets for water cooled reactors has also been developed (paper, presented by Mr. Peehs, FRG).

Three major modifications were made in the referred EGT process in order to produce gel microsphere which could be calcined to microspheres suitable for pelletization and sintering. First, heavy metal nitrate feed solutions of lower molarity were found to be more suitable. Secondly, calcium nitrate was added to the feed solution in order to have around 0.4 w/o CaO "sintering aid" in the subequent calcined microspheres. Thirdly, carbon black were added to the sol prior to gelation. The pores formed in the sol-gel microspheres after burning off the carbon black particles reduce the crushing strength of the microspheres and facilitate pelletization.

The Sol-Gel Microsphere Pelletization (SGMP) process not only avoids dust generation and is easy to remotize but also produces high density pellets of desired microstructure at compaction pressures and sintering temperatures, which may be even lower than for the conventional powder route.

HTR fuel kernels are easily produced by the EGT process which needs no chemicals apart from $NH_3$ necessary for precipitation of heavy metals in all fuel production processes.

In the paper regarding Indian experience in fabrication of $ThO_2$ pellets emphasize was made on optimization pre-compacting and final compacting parameters which will lead to the reliable and economic fabrication technique of this fertile material.

The goal of this work is fabrication and supply of blanket assemblies containing thorium oxide pellets to the requisite quality, for full core of Indian Fast Breeder Reactor at Kalpakkam.

Starting with $ThO_2$ powder obtained by oxalate precipitation and calcination, green pellets are produced through powder metallurgical route. In order to achieve sintered densities more than 94% addition of Mg in small quantities, during oxalate precipitation stage.

An economic and viable recycling method for sintered pellets as well as powder needs to be developed.

## 5. Reprocessing of Th-based fuel

KFA Jülich GmbH experience in the utilization of dual cycle THOREX process for extraction of Th and U was reported (by Mr. Zimmer), as well as, the results of cold tests.

The THOREX solvent extraction process is simple to carry out since separation of Th and U is accomplished by the different extraction behaviour of the elements in their stable valency state, rather than in the PUREX process where the Pu has to be reduced to and stabilized in the trivalent state for separation.

Reprocessing of the 20 ELk-River reactor (USA) Th-U spent fuel elements was done in the ITREC plant, Italy (paper by Mr. Candelieri). The ERR used fuel had a burnup in the range of 3,650 -10,000 MWd/t. A THOREX "acid-deficient" flowsheet was used for code-contamination of U-Th in a single cycle 30% TBP solvent extraction. Unfortunately, the project terminated in 1974 and the facility is presently used for testing some of the components used in a reprocessing plant. No pellet refabrication has been done in this facility.

# SUMMARY OF THE PANEL DISCUSSION

In spite of some leveling in the demand for nuclear energy and in some decrease in U-costs a number of IAEA Member States are still interested in the utilization of Th as nuclear fuel. This is in order to evaluateed long-term fuel supply and to prevent for shortages in enriched U. RAR and EAR-I resources of Th which could be recovered at costs less than US $80/kg are estimated at around 2,4 million tonnes.

Conventional nuclear power plants can be designed for operation either in the U/Pu cycle, in Th-HEU cycle or in Th-Pu cycle. The main advantage of the Th-based fuel cycles is that is has a higher neutron yield of U-233, in comparison with the neutron yield of Pu 239 in the U/Pu cycle. Those benefits may be equalized by commercial disadvantage, due to the requirement of HEU and the problems related to reprocessing and refabrication in a closed fuel cycle since full remotization is needed. The once-through cycle by the use of Th/Pu fuel offers the possibility when applicated to LWR-systems to avoid those problems and having considerable savings in U-ore and enrichment work units.

In fundamental and reactor conception problems the papers concentrated mainly on the following questions: are there enough neutronics data and how to choose the optimal way for starting the Th-based fuel cycle in nuclear power, to prove the neutronic feasibility of thorium cycle in unmodified PWR's For other types of reactors there are no real plans involving a Th-based cycle. FRG has abandoned the utilization of Th in the future HTRs in favour to low enriched uranium. The reason is the current inavailability of HEU.

A crucial item in the nuclear core design is the uncertainties in the cross-section data for Th-232 and U-233. In France, CEA recommended data is in use which is updated from time to time. The work in this area is summarized in the enclosure, presented by the French delegate, Mr. Greneche from CEA, Saclay. In general, the data for Th-232 are more reliable than those for U-233. In the FRG data are used which are elaborated by detailed studies based on the available experimental data base and from irradiation experiments. China feels necessary to elaborate those data more in detail, specifically for LWR application.

In India an Expert Group is working on data for thermal and fast flux conditions. USSR is working with ABBN data (Russian origin) but also data from Japan and France are used to improve the data base.

For fuel manufacturing at least four different procedures are in practice:

- Powder production ex-oxalates and cold processing, sintering in $H_2$/Ar atmosphere;
- Powder production ex-oxalates and extrusion, sintering in $H_2$/Ar atmosphere;
- Vibrocompaction of sol/gel kernels;
- Pelletizing ex sol/gel kernels and sintering in $H_2$/Ar atmosphere.

It is acknowledged that the microstructure optimization is needed for high burnup fuel. This can at the moment only be reached by pelletizing ex sol/gel kernels, improved in its pressing performance by the addition of carbon black (Brazil, FRG). Fuel fabrication for blanket element do not need a micro-structure optimization due to its low burnup. Here the powder production ex oxalates and cold processing shows sufficient performance (India). To lower the sintering temperature sintering aids like Ca or Mg are successfully in use (FRG, India). It was also emphasized, that the time for fuel dissolution in the reprocessing is influenced by its microstructure (FRG, Brazil). Also the addition of Ca or Mg seems to influence positively the dissolution behaviour (India, FRG) of irradiated fuel.

- The thermal design of pin-type fuel is not very much advanced. It seems that in the different countries different material data are used and different computer codes. Some countries are doing own measurement (Brazil, France, FRG, India) and some are using data from literature.

- The reprocessing and refabrication is heavily influenced by the radioactive by-products requiring heavily shielded devices. Italy has some experience in this area by the work with spent Elk River fuel from the USA. Generally, basic experiences are available but no work is going on on refabrication for the time being, even if some national strategies need reprocessing and refabrication (France, India).

- All work presented in the meeting was addressed to oxide fuel. The question on the potential of metallic Th-fuel could not be answered in the Panel. However, it seems that work is going on in the USA at ANL to investigate this fuel type as a FBR-fuel. Alloys with Th-20%U - 4%Zr and Th-20%U - 10% Pu are under investigation. Irradiation testing up to 100 GWd/t seems to be available. The fuel pins have SS-clad and Na-bonding.

After termination of the Panel Discussion a list of major activities worldwide in the area of Th-based nuclear fuels was compiled by the delegates from the different countries. No information was available in the Meeting from Canada, Japan, Romania and USA. IAEA staff members, Messrs. F. O'Hara, M. Ugajin and Mr. P. Bull of the Permanent Mission of Australia, prepared the reports on the status of the thorium-based fuel research and utilization programmes in the USA, Japan and Australia, correspondingly (attached). The report on "Status of Spent Fuel Management in Canada" presented at the IAEA Advisory Group on Spent Fuel Management in 1984 and reports by the Institute for Nuclear Power Reactors, Pitesti, Romania, presented to the IAEA, in the course of a research contract on "Data base for a PHWR operating on a once-through, low enriched uranium-thorium cycle" were used for filling the above mentioned list.

LIST OF MAJOR ACTIVITIES WORLDWIDE IN THE AREA OF TH-BASED NUCLEAR FUELS

| Country | Programme | | Directed towards | | | | | Areas of R and D | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | terminated | running | HTR | HWR | LWR | FBR | OTHER | Basic research | Strategy | Core design | Technology | Irradiation | Reprocessing | Refabrication |
| ARGENTINA | | X | | X | | | | X | X | | X | | | |
| AUSTRIA | | X | X | X | X | X | | Dynamic System Analysis<br>Fuel Cycles (by literature) | X | | | | | |
| AUSTRALIA | 1980 | X | | | | | | X see (attach-<br>X ment III) | | | | | | |
| BRAZIL | | X | | | X | | | | X | X | X | X | X | |
| CANADA | | X | | X | | | | | X | | | | | |
| CHINA | 1971 | X | X | | X | | | X<br>X | X | | X | X | X | |
| FRANCE | 1979/82 | X | X | | X | X | X | X | X<br>X | X<br>X | X | X | X | |
| FRG | 1985 | | X | X | | | | | | | X | | X | X |
| FRG/BRAZIL Jointly | | X | | | X | | | | | X | X | X | X | |
| INDIA | | X | | X | | X | | X | X | | X | X | X(planned) | |
| ITALY | 1974 | | | | X | | | X | | | | | X | X |
| JAPAN | | X | X | | | | X | X | X | X | X | | X | |
| PAKISTAN | 1983 | | | | | | | X | | | X | X | | |
| ROMANIA | | X | | X | | | X | X | X | X | | | | |
| USA | 1982 | | X | | X | | X | X | X | X | X | X | X | X |
| USSR | | X | X | | X | X | | X | X | X | | | | |

## Recommendations

- The capture cross-sections of Th is rather well-known in the thermal region. But in the fast energy range capture fission cross-sections and n-2n cross-sections must be known with a better accuracy. This could be achieved by differential thermal measurements. For U-233 better knowledge of cross-sections is needed in the whole energy range. For this aim it could be very helpful to have results available from clear integral experiments (bench-marking). It would be beneficial if this work could be done in cooperation.* Further information is needed in cross-sections for Pa-233 and U-234.*

- It appears that there may be discrepancies in the results obtained by different countries concerning core calculations for Th cycles (e.g. pin power peak calculations for PWR). It should be worthwhile to promote bench-marking studies on this problem.*

- There is a lack of information on the treatment of used Th for closed Th/U fuel cycles. Work on the reuse of Th would be emphasized.

- The powder technology for pelletizing should be investigated to assess its capability to optimize the microstructure of the fuel similarly to the pelletizing ex sol/gel kernels.

- The Member States working in the area of metallic fuel are invited to inform about their experience in order to assess its potential as on nuclear fuel. The IAEA is asked to distribute a reference list from the available reports in this area.

- It is recommended to hold in approximately 5 years a meeting on the progress in Th utilization. Possible objectives in this area meeting could be the evaluation of world-wide Th resources, basic

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* The Scientific Secretary was asked in cooperation with the other Divisions (Nuclear Data Base Section and Advanced Nuclear Power Technology Section) to endorse in the Agency the activities on collection and analyses of data on Th and U-233 cross-sections and on active core designs regarding utilization of Th fuels.

research of nuclear, physical and chemical properties of Th, strategies, reactor core and blanket concepts for different reactor types; fuel technology; reprocessing and refabrication in closed Th fuel cycles.

- Upcoming results on the different areas of activity in the Th investigation may be presented at the IAEA meeting.

# Annex I

# STATUS OF US PROJECTS RELATED TO THORIUM UTILIZATION

Two major thorium-fuelled reactor programmes have been actualized in the US. These are the Molten Salt Breeder Reactor and the Light Water Breeder Reactor. A third, the High Temperature Gas Reactor, was envisioned to utilize thorium, but this had not been accomplished. Instead this reactor utilized enriched uranium.

The study and development of molten-salt reactors was begun in the US at the Oak Ridge National Laboratory in 1947. The potential of MSBR for civilion power production was recognized and a development programme was established in 1956. A Molten Salt Reactor Experiment was operated from 1969. It was fuelled with a $U^{235}$-$U^{238}$ mixture during the initial two years of operation and with $U^{233}$ during the remaining 1.5 years of operation. The successful operation of MSRE and the favorable projected system characteristics attracted significant US industrial and utility interest. The development of molten-salt reactors was interruped in 1973 when the programme was terminated. The programme was resumed briefly in 1974 but finally terminated in mid 1976.

The second major reactor concept utilizing thorium was the Light Water Breeder Reactor. The concept, then called seed-blanket, was originally introduced in 1951 as a means of minimizing the separative work required for the fuel of a light water reactor. The seed-blanket concept was employed in the design of the first commercial PWR plant at Shippingport, Penn.

It was long thought to be impractical to breed with light water; however, since the value of $\eta$ for $U^{233}$ is only slightly lower in the epithermal region, while that of $U^{235}$ and $Pu^{239}$ are greatly reduced, the thorium cycle appeared to be most attractive for a thermal breeder. After preliminary work in the early 1960s indicated the feasibility of breeding in a light water seed-blanket core on the thorium cycle, the US AEC authorized a demonstration in the Shippingport Plant. This is the Light Water Breeder Reactor Project. Full-power operation of the demonstration began in December 1977.

The reactor has operated on thorium and $U^{233}$ cycles until 1982 at which time it was shut down. It is understood that the spent fuel is presently being reprocessed to provide an accurate check against physics calculations. Many reports on the project should be forthcoming.

# Annex II

# CURRENT STATUS OF THE THORIUM FUEL RESEARCH PROGRAMME IN JAPAN

No thorium fuel reactor is currently planned to be built in Japan, but basic R & D works are under way to pursue the possible diversification of nuclear fuel resources through thorium utilization in the future.

## 1. R & D in Japan Atomic Energy Research Institute

The Japan Atomic Energy Research Institute started in 1975 basic R&D works on thorium and thorium-uranium mixed oxides to develop laboratory-scale fabrication methods, examine irradiation behaviour and to measure physico-chemical properties of these fuels.

Microsphere and pellet-type fuels have been investigated taking into account potential applications of the acquired data for the fabrication and performance analyses of both types of nuclear fuels. The main accomplishments are the following:

- development of new sol-gel process to prepare crack-free microsphere fuel with better sphericity aiming at HTGR applications and to prepare starting material for making high-density pellet with varying Th/U ratios;
- measurement of fission-gas release/irradiation-induced damage and data analyses to predict irradiation stability and densification mechanism;
- measurement of new data on equilibrium oxygen potential/stoichiometry and its effect on chemical behaviour of burn-up simulated $(Th,U)O_2$.

## 2. R & D in Universities

The Research Programme on Thorium Fuel has been performed since 1980 on the university basis, under the support of Grant-in-Aid for Energy Research of the Ministry of Eduction, Science and Culture of the Japanese Government. The main results have been published in the English-written report "Research on Thorium Fuel (SPEY-9,1984)".

It covers nuclear data evaluation and measurement; reactor physics experiment and analysis; fuel fabrication, irradiation and property measurement; actinides production/separation and down-stream process development; biological effect and molten salt reactor engineering.

# Annex III

# THORIUM RESOURCE STUDIES IN AUSTRALIA

Australia's thorium resources are those contained in monazite, itself a by-product of mineral sands mining. Australia has some 12780 tonnes of reasonably assured resources of thorium (recoverable at less than $80/kg TH- however, if monazite was mined solely as a source of thorium, this resource would not be recoverable at less than $80/kg TH).

There are no specific exploration programs for thorium in Australia. Exploration effort for mineral sands (containing thorium-bearing monazite) has responded generally to changes in the price of titanium and zirconium ores.

Thorium is not produced in Australia. All monazite production is exported. Latest production figures of monazite concentrates and contained thorium are:

Production (tonnes)

| | 1983 | 1984 (prelim) |
|---|---|---|
| Monazite concentrates | 15141 | 16707 (prelim) |
| - thorium content | 893 | 969 |

Summary of work on the utilization of Th-based fuel in Australia (provided by the Australian Atomic Energy Commission).

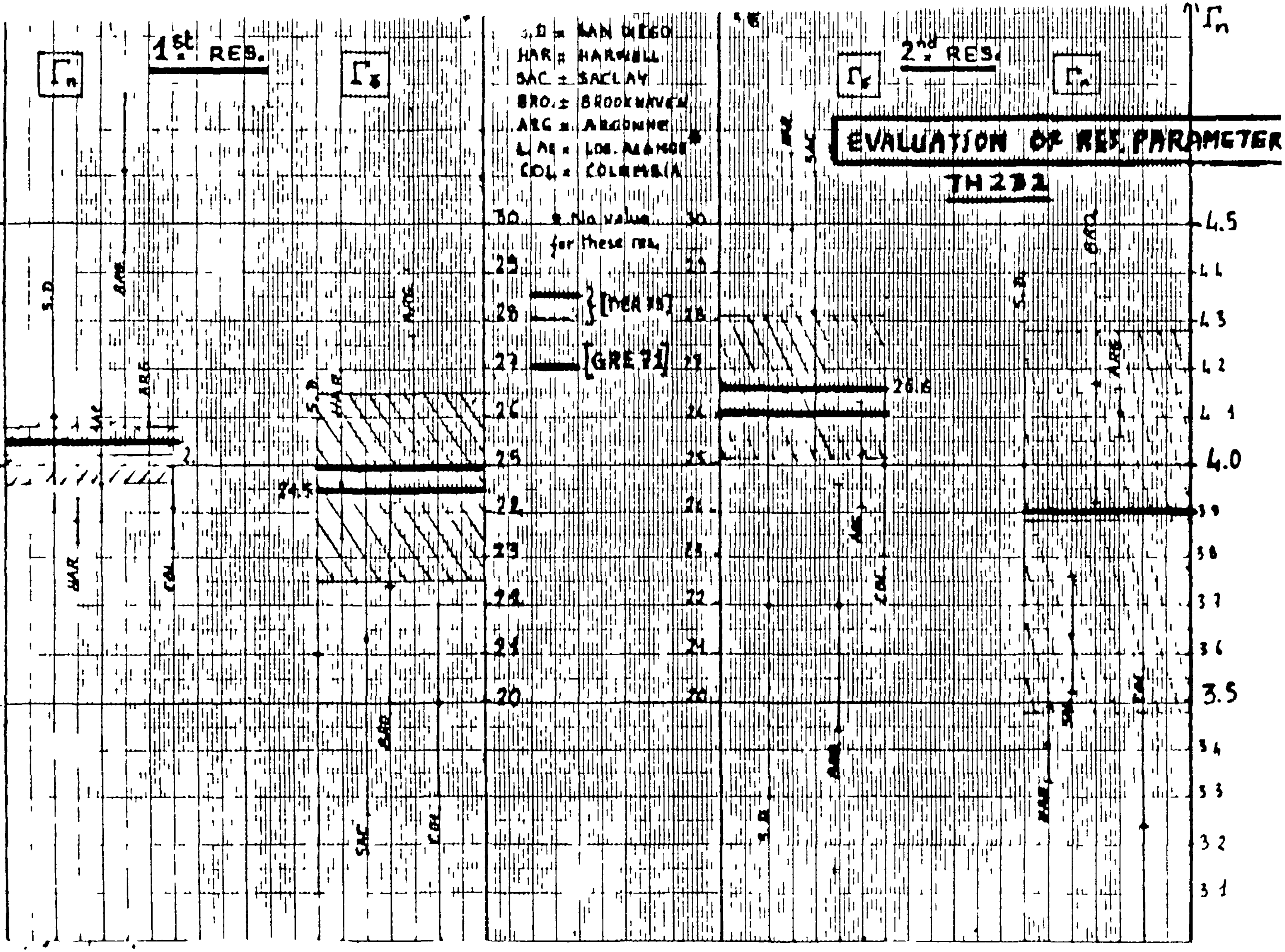
"The AAEC has carried out four experiments in the area of nuclear data on thorium isotopes: fission fragment angular distriubtions for thorium 232; the average number of prompt neutrons emitted in the fission of thorium 232; fission fragment angular distributions for thorium 230, and the fission cross-section of thorium 230. The first three experiments (concluded by 1980) aimed to examine the existence of a triple-humped fission barrier in the fission of thorium. The data from each experiment were in good agreement with such a barrier shape. The fourth experiment (still running) measured the sub-threshhold fission resonance in thorium 230 near 715 keV to confirm the energy resolution and energy accuracy of the first three experiments."

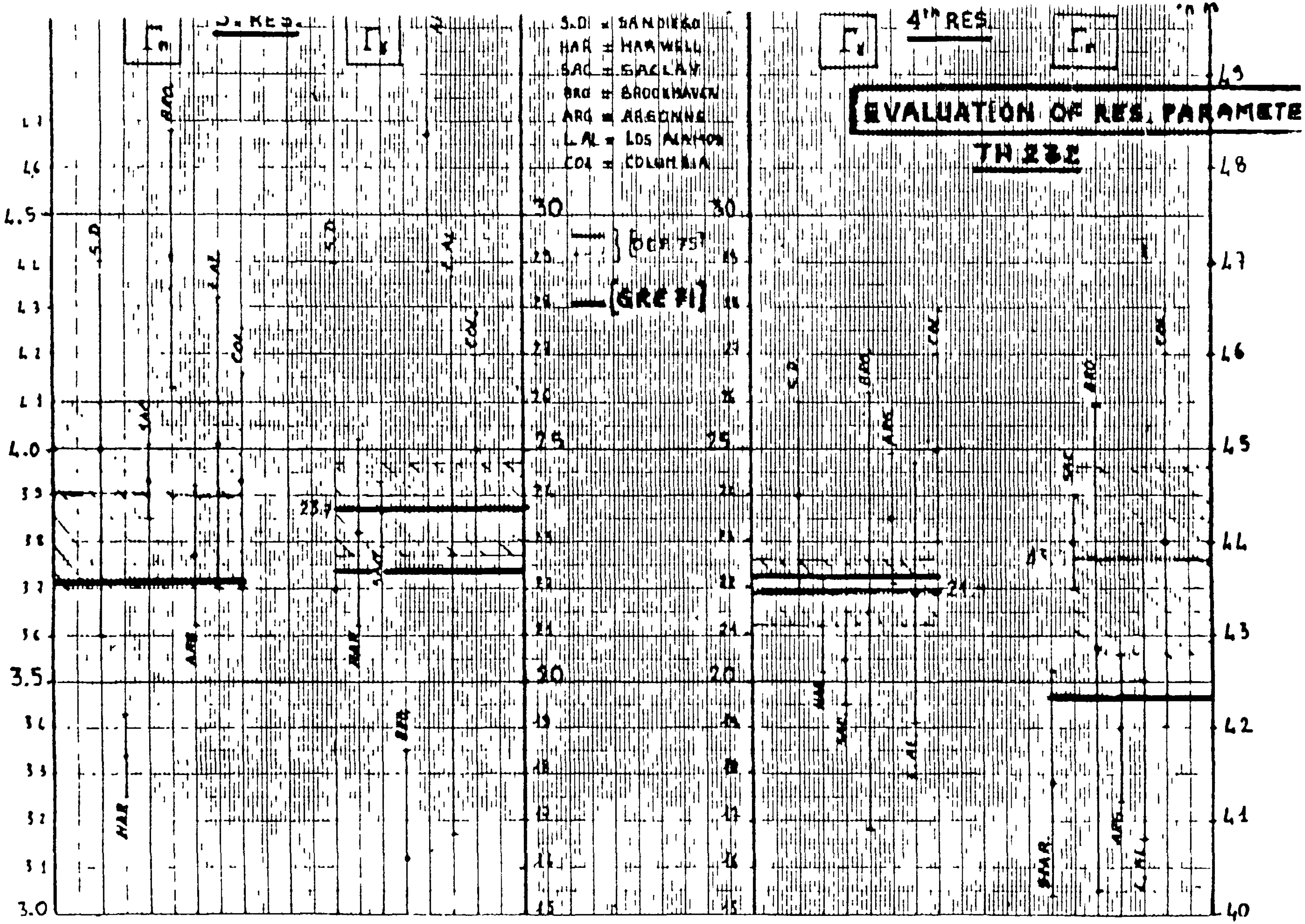
# Annex IV

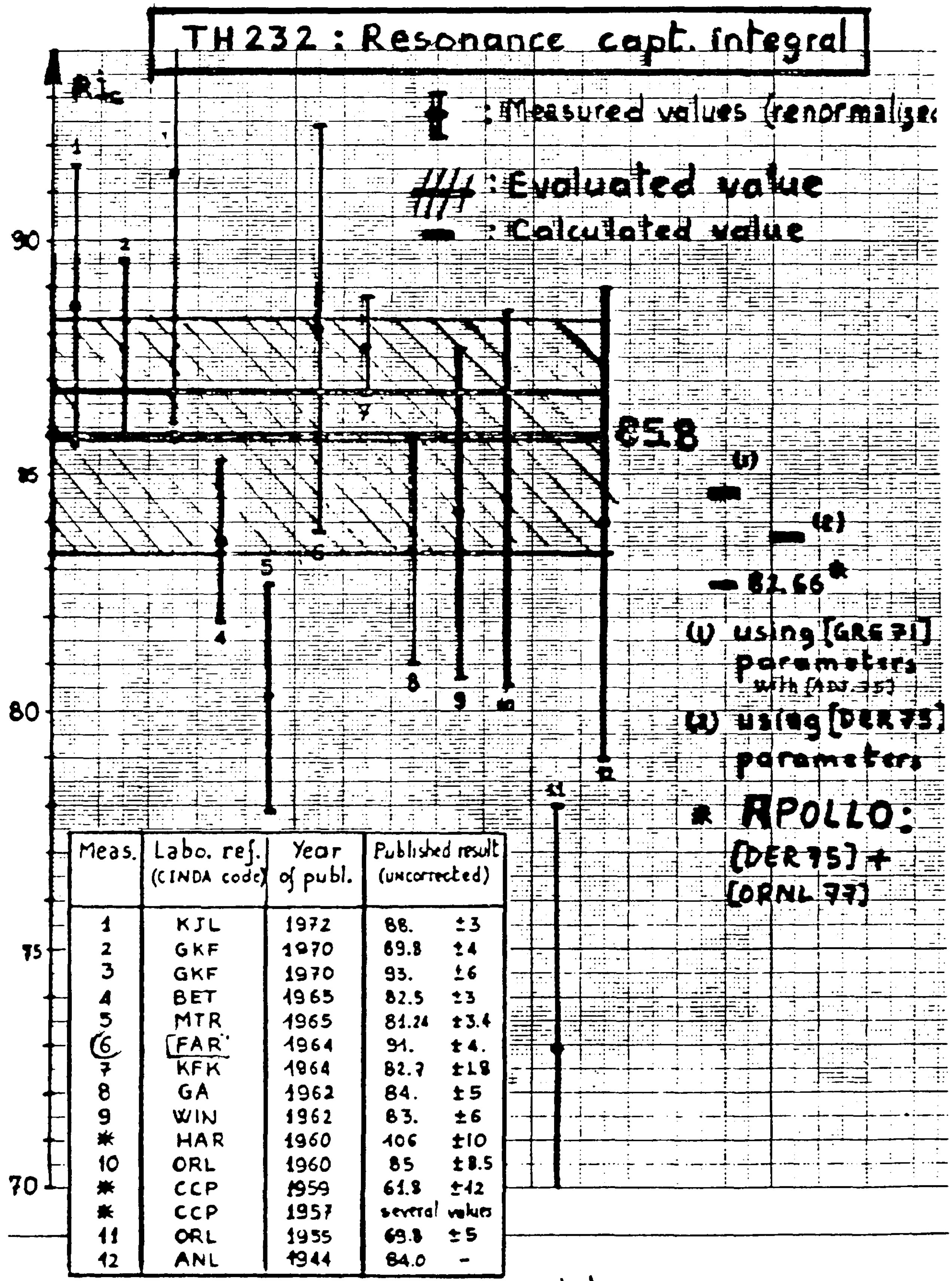
# NUCLEAR DATA FOR Th-232 AND U-233

## 1.1 Thorium 232

| | Thermal range<br>$E \lesssim 1$ ev | Region of resolved resonances<br>$1 \text{ ev} < E <$ few kev | High energy<br>$E >$ few kev |
|---|---|---|---|
| 1968<br>↓<br>1971 | *Cross sections derived from UKAEA data library (1968 version)*<br>[UKAEA 68] | | |
| 1971<br>↓<br>1975 | For $E_0 = 0.0253$ ev<br>$\sigma_a^0 = 7.4$ b<br>(BNL 325b)<br>$\sigma_a(E) = \sigma_a^0 \sqrt{\frac{E_0}{E}}$ | Evaluation of res. parameters from all available exper. data<br>(142 res.)<br>[GRE 71] | [UKAEA 68] |
| 1975<br>↓<br>1978 | id. | id. | Adjustment of UKAEA data:<br>• Up to 500 Kev: DERRIEN evaluation (calculation from average statist. param)<br>• E>500 Kev: Library of the GA spectrum code MICROX<br>[ADJ 75] |
| 1978<br>↓<br>NOW | $\sigma_a^0 = 7.4 \pm 0.08$ b<br>$\sigma_a(E) = \frac{a - b\,E}{\sqrt{E}}$<br>$a = 1.189$<br>$b = 0.45 \pm 0.1$ | New evaluation of reson. parameters by DERRIEN in 1975, from selected exper. data compiled in [GRE 71] | Recommandation of DE SAUSSURE and MAKLIN in 1977<br>(ORNL/TM 6161) |
| | [DER 75] | | [ORNL 77] |

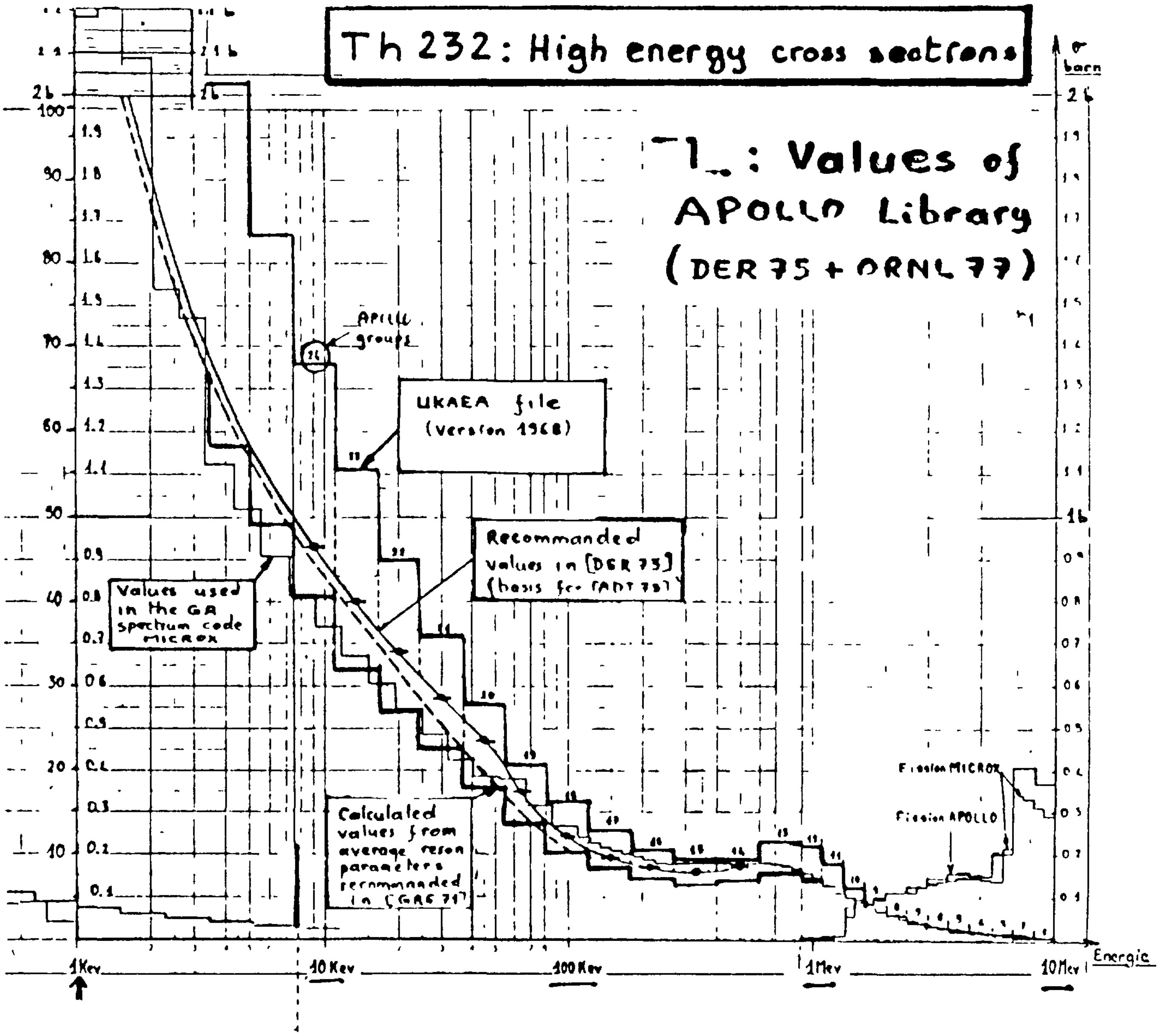

EVALUATION OF RES. PARAMETER
TH 232
1st RES.
2nd RES.
Γn
Γγ
Γγ
Γn
S.D = SAN DIEGO
HAR = HARWELL
SAC = SACLAY
BRO = BROOKHAVEN
ARG = ARGONNE
L.AL = LOS ALAMOS
COL = COLUMBIA
* No value for these res.
[MER 75]
[GRE 71]
2.5
2.0
1.5
30
25
20
4.5
4.0
3.5
24.5
26.6
S.D
HAR
SAC
ARG
COL
BRO


EVALUATION OF RES. PARAMETE
TH 232
3. RES.
4th RES.
Γn
Γγ
S.D = SAN DIEGO
HAR = HARWELL
SAC = SACLAY
BRO = BROOKHAVEN
ARG = ARGONNE
L.AL = LOS ALAMOS
COL = COLUMBIA
[OCF 75]
[GRE 71]
S.D
HAR
SAC
BRO
ARG
L.AL
COL
23.7
24
3.0
3.5
4.0
4.5
15
20
25
30
4.0
4.1
4.2
4.3
4.4
4.5
4.6
4.7
4.8
4.9
m



| Meas. | Labo. ref. (CINDA code) | Year of publ. | Published result (uncorrected) |
|---|---|---|---|
| 1 | KJL | 1972 | 88. ±3 |
| 2 | GKF | 1970 | 89.8 ±4 |
| 3 | GKF | 1970 | 93. ±6 |
| 4 | BET | 1965 | 82.5 ±3 |
| 5 | MTR | 1965 | 81.24 ±3.4 |
| 6 | FAR | 1964 | 91. ±4. |
| 7 | KFK | 1964 | 82.7 ±1.8 |
| 8 | GA | 1962 | 84. ±5 |
| 9 | WIN | 1962 | 83. ±6 |
| * | HAR | 1960 | 106 ±10 |
| 10 | ORL | 1960 | 85 ±8.5 |
| * | CCP | 1959 | 61.8 ±12 |
| * | CCP | 1957 | several values |
| 11 | ORL | 1955 | 69.8 ±5 |
| 12 | ANL | 1944 | 84.0 - |

* These measurements have been disregarded in evaluation


Th 232: High energy cross sections
σ barn
1: Values of APOLLO Library (DER 75 + ORNL 77)
APOLLO groups
UKAEA file (Version 1968)
Recommanded values in [DER 75] (basis for [ADT 79])
Values used in the GA spectrum code MICROX
Calculated values from average reson parameters recommanded in [GRE 71]
Fission MICROX
Fission APOLLO
2b
1b
100
90
80
70
60
50
40
30
20
10
1.9
1.8
1.7
1.6
1.5
1.4
1.3
1.2
1.1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
1Kev
10 Kev
100 Kev
1Mev
10Mev
Energie

# Th 232 : CONCLUSIONS

## 1 - Thermal range

- The value of $\sigma_a^0$ (7.4 b) is apparently well known
  (there are no large discrepancies between experimental results)
- $\sigma_a(E)$ seems to decrease more rapidely than an "$\frac{1}{\sqrt{E}}$" low
  (negative resonance)
  Accurate experimental values should be desirable to a aquire a better knowledge of the real shape of $\sigma_a(E)$ in this region
  (temperature coef. calculation)

## 2 - Resonance region

- Resonance parameters are determined within few percent.
  While this is a fairly good accuracy it must be improved again to be quite satisfactory
  (a detailed investigation is particularly needed for the 3$^{d}$ resonance ($\Gamma_n$))

## 3 - High energy range

- It is in this range that further work is the most desirable.

## 4 - Resonance integral

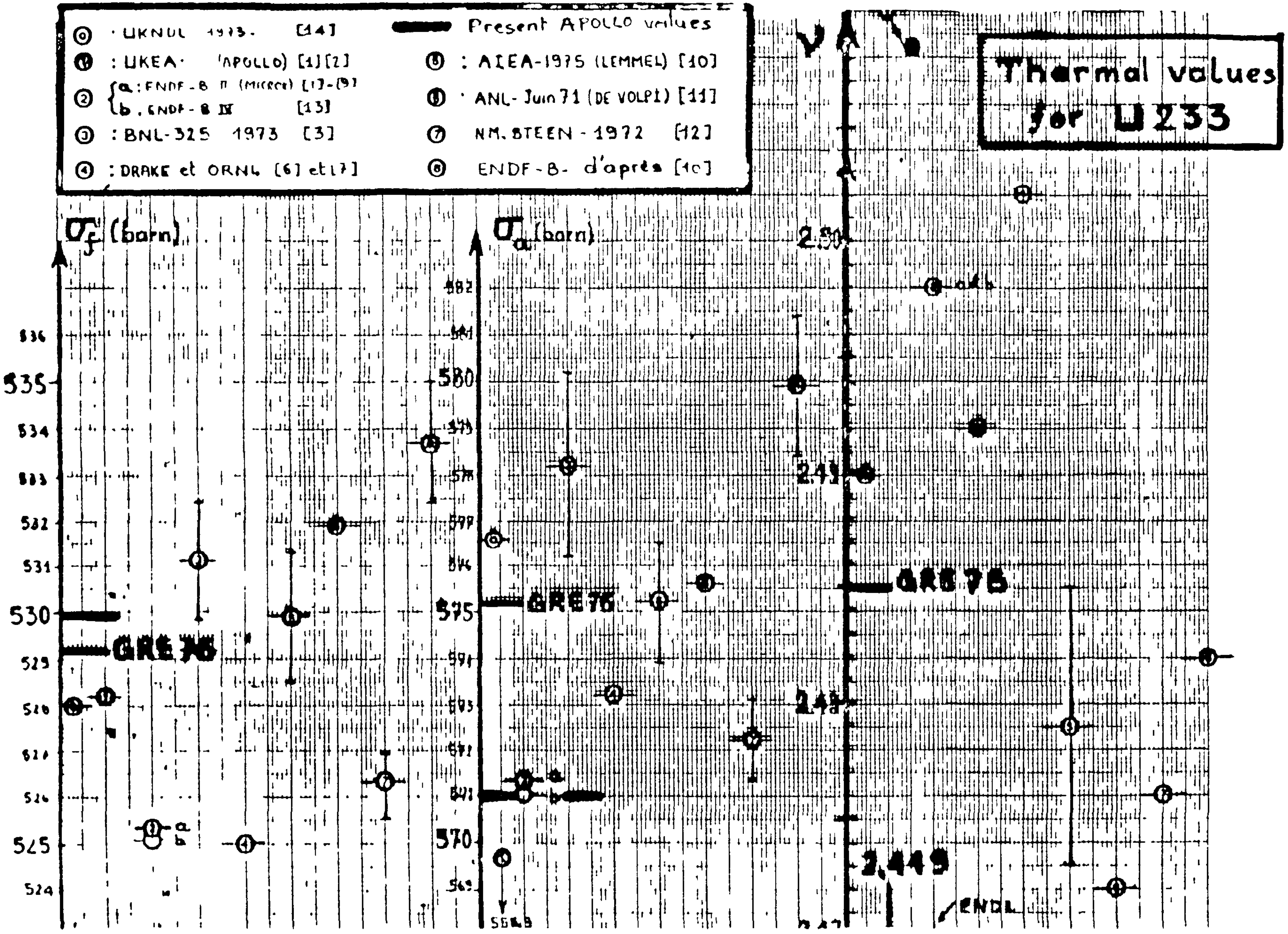
A good agreement is now achieved between evaluated values from experimental results and calculated values from resonance parameters

# 2.2 : U233 nuclear data

| Dates | ORIGIN OF THE DATA | Thermal values (E=0.0253 eV) | | | |
|---|---|---|---|---|---|
| | | $\sigma_a^0$ | $\sigma_f^0$ | $\nu$ | $\eta$ |
| 1968<br>↓<br>1975 | . UKAEA data Library [UKAEA 68]<br>Remark: no data available to take into account the self shielding effect. | 557.9<br>very low | 528.3 | 2.510 | 2.377 |
| 1975<br>↓<br>1976 | . Overall adjustment of UKAEA data:<br>$\sigma_a(E) \longrightarrow \sigma_a(E) \times \underline{1.0343}$<br>$\sigma_f(E) \longrightarrow \sigma_f(E) \times \underline{1.0023}$<br>("Tendancies" from analysis of some homogeneous critical experiments performed in France and USA) | 577.0 | 529.5 | id. | 2.303 |
| 1976<br>↓<br>1978 | . detailed adjustment of UKAEA data based upon a general review of available data and evaluations<br>Remark: same as the first one<br>[GRE 76] | 575.2 | $\boxed{525.2}$<br>(AIEA) | 2.483 | 2.286 |
| 1978<br>↓<br>1979 | . ENDF-B/IV data<br>. Tabulation allowing to take into account self shielding effect.<br>[ENDF/B-IV. | 571.0 | 525.1 | 2.498 | 2.297 |
| 1979<br>↓<br>NOW | . Few adjustments of ENDF/B IV data in the thermal range<br>("tendancies" derived from an analysis of a series of critical experiments: work published in ref [6] of the paper)<br>[ADJ.79] | id. | $\boxed{529.9}$<br>AIEA | $\underline{2.449}$* | 2.272 |

* In the analysis the "best" overall agreement with exper. results is obtained when $\nu\sigma_f$ is decreased by -1.1%. Then, it is proposed to:
- . adopt the IAEA value for $\sigma_f$ which is 0.9% higher than ENDF B/4 value
- . Compensate by a significant reduction of the $\nu$ value: -1.96%

This change on $\nu$ is not consistent with all available evaluations.


Thermal values for U 233
⓪ · UKNDL 1973. [14]
① : UKEA · (APOLLO) [1] [2]
② a : ENDF-B II (MICRO) [7]-[9]
b . ENDF-B IV [13]
③ : BNL-325 1973 [3]
④ : DRAKE et ORNL [6] et [7]
Present APOLLO values
⑤ : AIEA-1975 (LEMMEL) [10]
⑥ · ANL-Juin 71 (DE VOLPI) [11]
⑦ NM. STEEN - 1972 [12]
⑧ ENDF-B- d'après [10]
$\sigma_f$ (barn)
536
535
534
533
532
531
530
529
528
527
526
525
524
GRE 76
a
b
$\sigma_a$ (barn)
582
581
580
579
578
577
576
575
574
573
572
571
570
569
GRE 76
a
b
568.8
ν
2.50
2.49
2.48
2.449
GRE 76
a & b
ENDL

# References for U-233 cross sections

[1] "APOLLO, Code multigroupe de résolution de l'équation du transport pour les neutrons thermiques et rapides" - A. HOFFMANN et Col. Rapport SERMA "T" N°1193.

[2] "The aldermaston Nuclear Data Library as at May 1963" K. PARKER AWRE Report-O-79/63 - Septembre 1963

[3] BNL-325 : "Neutron cross section" 3ème édition, Volume I - Juin 1973 S.F. MUGHABGHAB and C.I GARBER

[4] "MICROX, a two-region flux spectrum code for the efficient calculation of group cross sections" - by P. Wälti and P. Koch - Rapport Gulf-GA-A10827 - Avril 1972

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[6] "Neutron cross section for U-233" USAEC Report GA-7076 - General Dynamics, General Atomic Division, Septembre 1966 - DRAKE

[7] RPI preliminary ENDF/B data March 1967, Weston L.W. (ORNL-TM)

[8] "Interprétation d'expériences critiques homogènes" M. MIKHAIL, P. REUSS, H. TELLIER, J. VANUXEEM - Rapport SERMA N°248 "T" - Octobre 1975

[9] ENDF-B/III : "Evaluated Nuclear Data File" - Version III

[10] Conférence de Washington de mars 1975. "Neutron cross sections " p. 286 3ème évaluation de H.D. LEMMEL (AIEA - Vienne)

[11] "Discrepancies and possible adjustements in the 2 200 m/s Fission parameters" - Rapport ANL-7830 - Juin 1971 - DE VOLPI

[12] "Revision of the thermal parameters of U-233" Rapport WAPD-TM-1052 (Sept. 1972) - N.M. STEEN

[13] ENDF-B/IV : "Evaluated Nuclear Data File" - Version IV

[14] UKNDL (U.K. Nuclear Data Library) - 1973

# U233 cross sections :

## CONCLUSION

- The knowledge of U233 c.s. is not as good as that of Th 232 c.s. (see for example the problem of $\nu$)

- Then: Further work is still needed particularly in the thermal range.

- This work should include
  - New accurate measurement of $\nu$, $\sigma_f$ and fissible spectrum
  - New "clean" critical experiments which can be easily interpretable

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