CLE INVESTIGATIONS

BRAZILIAN EXPERIENCE ON THORIUM FUEL CYCLE INVESTIGATIONS

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1. INTRODUCTION

The major incentives for thorium use in power reactors in Brazil were and still are:

- the estimated large thorium resources of the country (see Appendix A), and
- the improved fissile fuel utilization of the thorium fuel cycle in thermal reactors (due to the higher η value of the ^{233}U as compared to that of ^{239}Pu), which results in a better utilization of the uranium reserves as well as in a reduction of the uranium enrichment requirements.

Today, the non-proliferation characteristics of the thorium fuel cycle, for instance:

- the contamination of the ^{233}U by the ^{232}U and its daughter products, some of them being hard gamma emitters;
- the capability of the ^{233}U to be denatured with ^{238}U;. and
- the reduction of plutonium buildup, if considered a future mix of thermal reactors operating in the U/Pu cycle and of Th-fueled reactors (and possibly even the burning of plutonium).

add a new incentive.

2. PAST EXPERIENCE ON THORIUM FUEL UTILIZATION

The most important activities in Brazil aiming at the introduction of thorium-fueled power reactors in the long term were developed in two different occasions by different institutions, in the framework of two projects in close cooperation with international partners, as shown in Table I.

Table I. Brazilian Projects on Thorium Fueled Power Reactors

Period	Institutions*)	Project	Country partner
From mid 1965 to mid 1970	CNEN & UFMG	"Instinto/Toruna"	France
From mid 1979 to mid 1988	NUCLEBRÁS	"Thorium Utilization in PWRs"	Germany

*) CNEN - Comissão Nacional de Energia Nuclear (Brazilian National Nuclear Energy Commission);
UFMG - Universidade Federal de Minas Gerais (Federal University of Minas Gerais);
NUCLEBRÁS - Empresas Nucleares Brasileiras S.A.

The Instituto de Pesquisas Radioativas - IPR (Institute for Radioactive Research), in Belo Horizonte, later renamed Centro de Desenvolvimento da Tecnologia Nuclear - CDTN (Nuclear Technology Development Center), was in charge of both projects.

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2.1 "Instinto/Toruna" Project

The first project was developed by the so-called Thorium Group, in the framework of a cooperation agreement with the French Commissariat à l'Energie Atomique - CEA. It was motivated by the results of a long term study of fuel requirements [1], one of the tasks of the Study's Committee for the first Brazilian Power Reactor, created by the Presidency of the Republic in 1965 and coordinated by the Brazilian National Nuclear Energy Commission CNEN. This project was ambitious and aimed at the development of an indigenous thorium-fueled pressurized heavy water reactor concept with prestressed concrete reactor vessel. It was scheduled in the three phases shown in Table II.

Table II. "Instinto/Toruna" Project Phases and Objectives

Phase	Main Objective
From 1966 to 1967	Evaluation of a thorium-fueled pressurized heavy water reactor - PHWR concept ("Instinto Project")
From 1968 to 1971	Research and Development centered on a natural uranium fueled PHWR concept ("Toruna Project")
From 1971 on	Development of a PHWR prototype (eventually)

The studies of the first phase were completed with the release of a Final Report with positive conclusions and recommendations which led to the continuation of the project [2].

The second phase was oriented towards the conceptual design of a natural uranium fueled PHWR, required for the production of fissile material (plutonium) for the startup of the thorium fuel cycle. This work was concluded also with a Final Report [3, 4]. This phase contemplated also a strong effort on research and development in different areas, such as fuel technology, core physics design, reactor and plant thermal-hydraulics design, reactor vessel design, materials and components testing, and fuel cycle economics. Formation and training of personnel had a major incentive. Several facilities and laboratories were designed and implemented at the IPR institute: the heavy-water subcritical facility ("Capitu"), the experimental thermal-hydraulic loop ("CT1"), the Fuel and Materials Laboratory, and the Components Testing Laboratory.

The continuation of this project in its third phase, which included a greater R&D effort (design and construction of a critical facility and a high power thermal-hydraulic loop) aiming at the design and (eventually) the construction of a PHWR prototype, was not implemented, due to the decision of adopting the pressurized light water reactor - PWR for the Brazilian first nuclear power plant (NPP). The reasons for the choice of the PWR for Angra 1 and the following NPPs, as well as a short description of the "Instinto/Toruna" project, which may be considered as an important contributor to the genesis of the Brazilian NPP program, are given in reference [5].

The main results of this project may be summarized as:

- the assessment and own development of know-how related to the technology and economics of water-cooled reactors, which made it possible to give a strong support to the Brazilian NPP implementation program, based on light water reactors; and, perhaps the most fruitful result,
- the formation of a staff highly qualified in different aspects of the analysis and design of nuclear power plants, later requested by the different organizations involved in the

implementation of the nuclear program (Angra 1 and Angra 2/3 NPPs; formation and training of personnel; and research and development).

2.2 R&D Program on the Thorium Utilization in PWRs

The second project was developed in the framework of a cooperation agreement between Brazil and Germany. The motivation was directed towards the improvement of knowledge in the field of thorium utilization in power reactors aimed at fulfilling the "Governmental Agreement on Cooperation in the Field of Science and Technology" (1969) and the "Memorandum of Understanding between the Kernforschungszentrum Jülich, GmbH - KFA and the Empresas Nucleares do Brasil S.A - NUCLEBRÁS" (1978). The "International Nuclear Fuel Cycle Evaluation - INFCE", organized at the same occasion (1977), was an incentive to the development of the joint CDTN, KFA Jülich, Siemens A.G./Group KWU and Nukem GmbH R&D program.

The general objectives of the program were:

(a) to analyze and prove the thorium utilization in PWRs,
(b) to design the PWR fuel element and core for the different thorium fuel cycles,
(c) to manufacture, test and qualify Th/U and Th/Pu fuel elements under operating conditions, and
(d) to study the closing of thorium fuel cycles by reprocessing of spent thorium-containing PWR fuel elements.

Technology transfer by joint work on the different tasks presented another major objective of the program. Three phases were foreseen at the start of the program (Table III):

Table III. Phases and Objectives of the "R&D Program on Thorium Utilization in PWRs"

Phase	Main Objective
From 1979 to 1982	Adaptation of the existing methods and technologies to PWR thoria fuels and irradiation testing of $(Th,U)O_2$ fuel rods in a test reactor
From 1982 to 1986	Research and development effort concentrated on the demonstration of the behavior of $(Th,U)O_2$ fuel in a power reactor
From 1986 on	Demonstration of $(Th,Pu)O_2$ fuel

The first phase was concluded in 1983 and the results were published in a Final Report [6].

The program was terminated in the second phase, by mid 1988, after nine years of successful cooperation before entering the pathfinder demonstration phase with a $(Th,U)O_2$ fuel bearing test assembly in a commercial PWR (Angra 1) [7].

The results of the program, discussed deeply in the program Final Report [8], confirmed in detail that the developed thoria based fuels, produced by merging of the standard light water reactor pelletizing process with the chemical ex-gel process developed for the high temperature reactor fuel, can be used in present PWRs. No changes in the fuel assembly and in the core design are needed. This holds both for $(Th,U)O_2$ and $(Th,Pu)O_2$ fuels in 3- and 4-batch operation. The latter shows high burnup potential beyond the four-cycle scheme. In this case, the inserted fissile plutonium is strongly depleted and the once-through put-away cycle becomes very attractive. As far as the technology development and transfer for the Th/U fuel are concerned, the program objectives were accomplished. However, large scale demonstration of Th/U fuel in a power reactor, fabrication and qualification of Th/Pu fuel as well as closing the fuel cycle would require substantially more effort.

The main conclusions of this Brazilian-German joint program were:

(a) The utilization of thorium in PWRs presents a long term option providing in some respects interesting results. The most attractive application of Th-based fuels is the use of recycle plutonium in an extended burnup once-through fuel cycle.

(b) From the point of view of cooperation and technology transfer, the program experience showed the importance of using hardware oriented goals, clear definitions of required outputs and sufficient communication including joint work on interacting tasks.

2.3. Other Activities

Other activities on thorium utilization took place also in other Brazilian institutions. For instance, from mid 60's to end of the 70's, the Instituto de Pesquisas Energéticas e Nucleares - IPEN (Institute for Energy and Nuclear Research), in São Paulo, has developed experimental activities on thorium-fuel technology [9, 10]. In the 70's, the IPEN spent a great deal of effort in a series of studies of the High Temperature Reactor concept [e.g. 11, 12]. It has, in addition, developed some activities (M.Sc. thesis work) on thorium utilization in PWRs and in gas cooled fast breeder reactors (feasibility study of a subcritical assembly and evaluation of thorium metal blankets) [13, 14].

The Instituto de Estudos Avançados - IEAv (Institute of Advanced Studies) of the Centro Tecnológico Aeroespacial - CTA (Aerospace Technology Center), in São José dos Campos, spent some effort in the 70's and 80's in studying thorium-fueled either gas-cooled or sodium-cooled fast breeder reactor concepts [15, 16, 17].

3. RECENT ACTIVITIES AND PLANS

Several activities involving thorium utilization in power reactors are either under way or being discussed.

3.1 Determination of Fuel Diffusion Properties

The transport properties of nuclear materials are very important to better understand the different phenomena influenced or controlled by them, such as oxidation and reduction, sintering, mechanical deformation at high temperature (fluence), grain growth, and densification. For the fuel designer it is necessary the knowledge of the autodiffusion and the heterodiffusion coefficients, in order to simulate properly, by using complex computer models, the fuel behavior under power reactor conditions.

In addition to that, the published values of the transport properties present, in general, a high dispersion, which makes difficult the establishment of a criteria to select the diffusion coefficients to use in the fuel design calculations.

This was the motivation for the CDTN research center and the Department of Physics of the Universidade Federal de Ouro Preto - UFOP (Federal University of Ouro Preto) to join their efforts, since 1992, to develop a research project aiming at the determination of the diffusion properties of nuclear materials manufactured in Brazil. In a first step, the anionic and cationic diffusion properties of the following ceramic materials are being studied:

- uranium dioxide fuel: UO_2,
- uranium/gadolinium mixed oxide $(U,Gd)O_2$, a burnable poison,
- uranium/cerium mixed oxide $(U,Ce)O_2$, a simulator of uranium/plutonium mixed oxide $(U,Pu)O_2$,

This step, from which first results are already available [e.g. 18 and 19], will be followed by the study of the diffusion properties of

- thorium/uranium and thorium/cerium mixed oxides: $(Th,U)O_2$ and $(Th,Ce)O_2$

Due to the multidisciplinary aspects and the complexity of the techniques involved in the determination of these properties (e.g. making use of the secondary ions mass spectroscopy technique - SIMS), this project has gained the support of other laboratories, in particular of the French laboratories: Laboratoire des Composés Non-Stoechimétriques, Université Paris XI, Orsay, and of the Laboratoire de Physique des Solides, Centre National de Recherche Scientifique - CNRS, Belleville-Meudon.

3.2 Energy Scenarios and Thorium-Fueled MSBRs

Investigations of scenarios for long term energy requirements and the introduction of breeder reactors in Brazil are being studied at the School of Engineering's Nuclear Engineering Department (DEN) of the Federal University of Minas Gerais - UFMG, in Belo Horizonte [20].

Considering the fact that the primary energy consumption per capita is a good index of the overall development of a country, since it reflects all desirable components of the development (e.g., the social, economical and technological components), the study made assumptions for three long term global energy demand scenarios (Table IV):

Table IV. World Energy Demand Scenarios

Average Energy Demand	1990	2015	2060		
			I	II	III
kW per capita	2.55	2.48	3.0	4.0	5.0

The following assumptions were made for Brazil:

(1) The population growth projection in the period 1990-2100 is summarized in Table V; it was assumed that the population becomes stabilized in about 280 million inhabitants by the end of the next century.

(2) In order to achieve a minimum reasonable development of the country in the long term, the primary energy consumption per capita projection in 2060 was assumed to be *at least* equal to the global average energy consumption per capita of the conservative Scenario I (Table IV).

With these assumptions the total long term energy requirements were derived, as shown in Table V.

The most important Brazilian energy resources are hydro, nuclear (uranium and thorium) and biomass (with a great production yield due to the very favorable location in the tropics). Fossile resources, oil in particular, are rather limited.

Table V. Brazil - Population Growth and Energy Consumption Scenarios

Year	Population, 10^6 inhabitants	Energy Consumption, kW/cap	Yearly Total Energy Requirements,		Total Cumulative Energy Requirements, Q	
			TW/yr	Q/yr *)	Period	Total
1989	147	1.84	0.27	8.062	-	-
2025	226	2.48	0.56	16.72	427	427
2060	264	3	0.79	23.64	768	1,195
2100	280	3	0.84	25.08	974	2,169

*) Q - Quad: 1 Q $=10^{15}$ Btu.

The study considered the indigenous energy resources and reserves presented in Table VI:

Table VI. Brazil - Indigenous Energy Resources

Resources	10^6 tEP	Q	TW
Depletable (Fossile) - Total *) 1)	***4,894***	***209.7***	***7.08***
Sustainable - Nuclear (Fission) *):			
– Uranium in light water reactors (LWRs)	1,342	57.5	
– Uranium in LWRs with recycling (+ 35%)	1,813	77.6	
– Uranium in breeder reactors 2)	107,360	4,600	
– Thorium in breeder reactors 3)	214,720	9,200	
– ***Nuclear (fission) - Total***		***14,012***	***473,08***
Renewables:			
– Hydro 4)	271.0/yr	11.4/yr	0.38/yr
– Biomass - excluding liquid fuels	200.0/yr	8.4/yr	0.28/yr
Renewables - Total 5)		***63.0/yr***	***2.13/yr***

*) Source: BEN-1990 [21].
Remark: Other energy sources, sustainable and renewables, were not considered due to its present low potential

1) Oil , natural gas, coal "in situ", shale and tar (assuming 100% resource recovery).
2) Uranium reserves taken from BEN, assuming 70% recovery and 20%. recycling losses. The uranium reserves may be higher, since only part of the country has been surveyed (aerogeographical survey: 60%; terrestrial survey: 40%).
3) Assuming 20% recycling losses. Thorium reserves were assumed to be at least twice those of uranium. As it is known, Brazil is a country with high thorium resources, but no systematic survey has been done until now.
4) Assuming that all hydro potential will be used.
5) Including ethanol (without co-products or sugar production), vegetable oil, hydrocarbons (from Euphorbias in semi-arid regions), and assuming, for each case considered, a land use equivalent to 10% of the national territory. Energy products from biomass may use land not useful for agriculture.

From the results showed in the above tables the following conclusions are derived:

(1) The nuclear energy that could be produced by uranium and thorium, *if totally used*, is by far the largest non-renewable energy resource (not only in Brazil but worldwide also);

(2) The long term energy requirements cannot be supplied by hydro power and by indigenous fissile fuels only, if reactors of poor fuel utilization (like the LWRs) are considered*).

(3) Use of biomass and of uranium and thorium *in breeder reactors* seem to be essential in order to fulfill the forecasted energy demand.

Following these conclusions, the DEN started to make a comparison between different breeder reactor concepts as possible candidates for the long term electricity supply. Part of this work is being done as a series of M.Sc. thesis work, which includes also exergy analysis of the nuclear fuel cycles.

Although the development of the sodium-cooled fast neutron breeder reactor (FBR) has had a strong support in several countries, culminating with the operation of several prototypes, demonstration reactors, and even of a large nuclear power plant, the full commercialization of this reactor concept is not expected to occur earlier than the second quarter of the next century. Taking this into account, the DEN considered that the molten salt-cooled thermal breeder reactor concept (MSBR), originally developed in the U.S.A. [22] and now being pursued in Japan [23], could be a possible breeder reactor candidate for the long term energy supply.

A comparison between the FBR and the MSBR is now under way, considering the different characteristics of both concepts, including:

- core physics (conversion and breeding factors, doubling time, initial fissile inventory),
- thermodynamic efficiency,
- fuel cycle flexibility,
- safety,
- economics, and
- nonproliferation aspects.

Preliminary results of this comparison have recommended to follow up the MSBR development abroad, as well as to start more detailed investigations of this reactor concept as a possible alternative to the FBR.

3.2 Proposal of a Sodium-Cooled Research Reactor

Recently, as a continuation of the studies mentioned in 2.3 above, the IEAv made a proposal to study a low power sodium-cooled research reactor with a moderated neutron spectrum. This would be a first step of using sodium in a reactor system in Brazil, looking ahead towards the long term penetration of fast reactors, possible using thorium [24].

3.3 Proposal of Thorium-Fueled PWR Lattice Experiments

The IPEN institute and the Centro Tecnológico da Marinha (Navy Technology Center) in São Paulo - CTM-SP made a joint proposal to CDTN, now under discussion, of continuing the investigations of the thorium fuel cycle, with emphasis on fuel development and on lattice experiments with ThO_2 and/or $(Th,U)O_2$ fuel in their critical facility IPEN/MB-01 [25]. This critical facility was designed for experiments with typical PWR lattices, but it is quite flexible to accept different geometries and different fuel/absorber materials.

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*) The available Brazilian uranium reserves amount to 301,490 tU_3O_8. About 2/3 are measured and indicated reserves and 1/3 are inferred ones.

Preliminary reactivity calculations were performed for lattices containing ThO_2 and $(Th,U)O_2$ rods, with the uranium enriched at 5 w/o. Three square arrays with rods containing thorium inserted in the central part of the UO_2 lattice were investigated (Table VII).

Table VII. Thorium Rod Arrays for Lattice Experiments

Material	Rod Array	Approx. number of rods*)
ThO_2	8 x 8	about 64
(Th, 25w/oU)O_2	10 x 10	about 100
(Th, 50w/oU)O_2	12 x 12	about 144

*) Some positions of the central part of the lattice are occupied by control and safety rods.

The cross sections of the fuel cells were generated with the HAMMER-TECHNION code [26] and the effective multiplication factors (k_{eff}) were calculated with CITATION [27]. These number of rods correspond to the maximum allowable, since an increase in the thorium containing region in any of the three arrays would make the core subcritical. These calculations were performed in order to estimate the materials requirements.

The general objective of the proposed lattice experiments is to get acquainted with mixed Th/U cores, by means of measuring, e.g.:

- neutron spatial flux distributions in mixed Th/U cores;
- temperature reactivity coefficients;
- reactivities of ThO_2 and of $(Th,U)O_2$ rods;
- reaction rates inside these rods,

To perform the proposed lattice experiments a great effort should be spent in the production of Th and Th/U fuel pellets and of the fuel rods. A cost/benefit analysis of this proposal shall be made before a final decision.

4. BARRIERS FOR THE INTRODUCTION OF THORIUM FUEL CYCLE

The major barrier to the introduction of the thorium fuel cycle in Brazil is the fact the startup of the thorium fuel cycle will require an initial inventory of fissile material. This means that, either an enrichment facility with the capability for the production of highly enriched uranium or the closing of the U/Pu fuel cycle with the implementation of a reprocessing plant for obtaining plutonium, is required. Both alternatives face a series of problems, which can be summarized as:

- technology development,
- high capital investment, and
- non-proliferation policy.

In this respect it is worth to mention that the nuclear power policy in Brazil is following the recommendations of the Commission, established in 1986 by a Decree of the President of the Republic, for the Evaluation of the Brazilian Nuclear Program. The Final Report with the evaluation and the recommendations of the Commission was published by the Brazilian Academy of Sciences in 1990. One of the recommendations states:

"To postpone the implementation of the spent fuel reprocessing project as it was originally planned, in view of its high costs and because it is not required in the medium term."

As a consequence of this recommendation, the NUCLEBRÁS' Reprocessing Project was interrupted and the staff was dispersed [28].

In practice, this means that the Brazilian government does not include the closing of the nuclear fuel cycle as a medium term option. This of course will change in the long term, particularly if the need of using thorium as an energy source becomes a requirement.

6. CONCLUSIONS

From the effort already spent in Brazil on investigating the thorium use in power reactors and from the gained experience, it can be concluded that:

- The utilization of thorium in power reactors presents an important long term option.

- The most attractive application of thoria-based fuels in pressurized water reactors is the use of recycled plutonium in an extended burnup once-through fuel cycle. However, the large scale demonstration of thorium containing fuel in a power reactor, the fabrication and qualification of thorium/plutonium fuel as well as the closing of the fuel cycle would require substantially effort of R&D and high investment costs.

- The benefits of working in partnership with the share of experience and costs and the importance of interacting tasks and good communication have been demonstrated.

- The adoption of well defined hardware oriented goals, clear definitions of the outputs and continuous effort are important factors for the succes of the projects.

- The systematic follow up of the worldwide developments oriented towards the thorium utilization in power reactors is important in order to subsidize future decisions. This also apllies to the demonstration and developments of breeder reactors.

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APPENDIX A

Thorium Reserves in Brazil

A recent survey was made for the "Diagnosis Project" of the Brazilian Association of Metallurgy and Materials - AMB [29]. Reference [25] gives the following data for thorium reserves in Brazil adopted in this survey:

Table A-I. Thorium Reserves in Brazil

Reserves, in t ThO_2	Indicated		Inferred	
	1972	**1992**	**1972**	**1992**
Brazil	10,000	606,000	20,000	700,000

Source: IAEA

The Brazilian thorium reserves are not easily comparable to other international categories, whose extraction and processing costs range from US$10 to US$20 per kg of ThO_2. In Brazil, thorium resources are considered those deposits of detritic monazite along the coast (monazite sands) which can be explored economically for the production of monazite and its associates. Rare earth salts (as main product) and thorium oxide (at a cost lower than US$20/kg ThO_2) are obtained from the monazite.

This last reference presents also the Table A-II for the natural potential resources of thorium in Brazil:

Table A-II. Thorium Potential Resources in Brazil

Occurrence	Associated Mineral	Average Content, %	Resource, t ThO_2	
			Measured	**Estimated**
Coastal deposits	Monazite	5	2,250	-
Morro do Ferro (State of MG)	Thorite and others	1 to 2	35,000	-
Barreiro, Araxá (MG)	Pyrochlore	0.09		1,200,000
Área Zero, Araxá (MG)	Pyrochlore	0.09	30,000	-
Alluvial and pegmatite deposits	Monazite	5	3,000	2,500
Total			73,750*)	1,202,500

*) Including 3,500 t of monazite sand of Indústrias Nucleares do Brasil S.A.- INB (in the States of RJ, ES and BA).