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# THE IMPORTANCE OF THORIUM IN THE CONTEXT OF THE GENERATION IV ADVANCED REACTORS AND THE IPEN'S EXPERIENCE

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## ABSTRACT

Globally, the 80's and 90's years were characterized by a significant reduction in the rate of growth of nuclear energy. However, from the 2000's, there has been a significant change in the international arena, with the "renaissance" of interest in nuclear energy, even in countries that had abandoned nuclear power. To answer questions like security, reducing the generation of radioactive waste, control of proliferation risks and long-term sustainability, some initiatives have been adopted by some countries. In 2000, the Department of Energy - DOE - United States created the GIF - Generation IV International Forum for Nuclear Reactors. Six reactor concepts were selected based on criteria such as: reduction of radioactive wastes, safety and cost effective to meet the increasing energy demand on a sustainable basis, being resistant to diversion of materials for nuclear weapons proliferation and safer against terrorist attacks. In this context, it becomes important to use thorium as nuclear fuel for the Generation IV Advanced Reactors, with startup scheduled for 2030. Although the thorium does not present significant commercial value nowadays, in a not too distant future it will probably be an important commodity. Unfortunately, contrarily to what is happening in most developed countries in recent years, Brazil is paying little attention to the thorium, even less than in the past, despite its large reserves. Thorium is three to four times more abundant than uranium in the Earth's crust and, although not fissile, all thorium can be used to produce $^{233}U$, by absorption of neutrons and subsequent radioactive decay. This uranium isotope is an excellent fuel for use in almost all types of nuclear reactors. It is possible that the thorium constitutes the largest Brazilian energy reserve, supplanting much oil (despite the findings of the pre-salt) and uranium. Brazil has a long tradition in the thorium technology, from mining of monazite until the obtainment of high purity thorium compounds and IPEN accumulated since the 60's a wide experience in the purification of thorium, obtained primarily from the monazite processing. Studies were also conducted on obtaining nuclear fuel based on thorium, the reduction of $ThF_4$ to metallic thorium, neutronic studies and proposition of reactor concepts based on the element. It should also be recorded that there was at IPEN, during this period, the production in pilot-scale of over one hundred and seventy metric tons of thorium nitrate with high purity. In this paper, we present briefly the experience accumulated at IPEN-CNEN/SP-Brazil and the different areas that comprise the Thorium Fuel Cycle, and the possibilities and advantages of thorium use in the IV Generation Advanced Reactors.

## 1. INTRODUCTION

There is currently 370 GWe of nuclear power capacity in operation around the world, producing 15% of the world's electricity, the largest share provided by any non-greenhouse gas-emitting source. This reduces significantly the environmental impact of today's electricity generation and affords a greater diversity of electricity generation that enhances energy security. The importance of reducing greenhouse gas emissions is now universally recognized, and numerous strategies and scenarios are proposed in order to achieve more sustainable future energy supplies. In the majority of these, the prospects are good for nuclear energy's growth [1]. Many of the world's nations, both industrialized and developing, believe that a greater use of nuclear energy will be required if energy security is to be achieved. They are confident that nuclear energy can be used now and in the future to meet their growing demand for energy safely and economically, with certainty of long-term supply and without adverse environmental impacts. The nuclear power industry has been developing and improving reactor technology for more than five decades and is starting to build the next generation of nuclear power reactors to fill orders now materializing [2].

Several generations of reactors are commonly distinguished. Generation I reactors were developed in 1950-60s, and outside the UK none are still running today. Generation II reactors are typified by the present US fleet and most in operation elsewhere. Generation III (and III+) are the Advanced Reactors. The first are in operation in Japan and others are under construction or ready to be ordered. Generation IV designs are still on the drawing board and will not be operational before 2020 at the earliest [3].

However, challenges still exist to further large-scale use of nuclear energy: (1) nuclear energy must be sustainable from the standpoint of its utilization of nuclear fuel resources as well as the management and disposal of nuclear waste, (2) the units must operate reliably and be economically competitive, (3) safety must remain of paramount importance, (4) deployment must be undertaken in a manner that will reduce the risk nuclear weapons proliferation, (5) new technologies should help meet anticipated future needs for a broader range of energy products beyond electricity, and (6) governments need to support the revitalization of their nuclear R&D infrastructures. Challenging technology goals for Generation IV nuclear energy systems are defined in four areas: sustainability, economics, safety and reliability, and proliferation resistance and physical protection. By striving to meet the technology goals, new nuclear systems can achieve a number of long-term benefits that will help nuclear energy play an essential role worldwide [2]. To meet these challenges and develop future nuclear energy systems, the Generation IV International Forum (GIF) is undertaking necessary R&D to develop the next generation of innovative nuclear energy systems that can supplement today's nuclear plants and transition nuclear energy into the long term. Generation IV systems can be broadly divided into fast and thermal reactors that address the above challenges with differing emphasis and technology [1].

The Generation IV International Forum (GIF) was initiated in 2000 and formally chartered in mid 2001. It is an international collective representing governments of 13 countries where nuclear energy is significant now and also seen as vital for the future. Most are committed to joint development of the next generation of nuclear technology. Led by the USA, Argentina, Brazil, Canada, China, France, Japan, Russia, South Korea, South Africa, Switzerland, and the UK were charter members of the GIF, along with the EU (Euratom). Most of these are party to the Framework Agreement (FA) which formally commits them to participate in the development of one or more Generation IV systems selected by GIF for further R&D. Argentina and Brazil did not sign the FA, and the UK withdrew from it; accordingly, within the GIF, these three are designated as "inactive Members." Russia formalized its accession to the FA in August 2009 as its tenth member, with Rosatom as implementing agent. After some two years' deliberation, GIF (then representing ten countries) late in 2002 announced the selection of six reactor technologies which they believe represent the future shape of nuclear energy. These were selected on the basis of being clean, safe and cost-effective means of meeting increased energy demands on a sustainable basis, while being resistant to diversion of materials for weapons proliferation and secure from terrorist attacks. They are the subject of further development internationally, and expenditure so far is in line with the initial estimate of $6 billion over 15 years. About 80% of the cost is being met by the USA, Japan and France. In addition to selecting these six concepts for deployment between 2010 and 2030, the GIF recognized a number of International Near-Term Deployment advanced reactors available before 2015 [1].

Most of the six systems employ a closed fuel cycle to maximize the resource base and minimize high-level wastes to be sent to a repository. Three of the six are fast neutron reactors (FNR) and one can be built as a fast reactor, one is described as epithermal, and only two operate with slow neutrons like today's plants. Only one is cooled by light water, two are helium-cooled and the others have lead-bismuth, sodium or fluoride salt coolant. The latter three operate at low pressure, with significant safety advantage. The last has the uranium fuel dissolved in the circulating coolant. At least four of the systems have significant operating experience already in most respects of their design, which provides a good basis for further R&D and is likely to mean that they can be in commercial operation well before 2030 [3].

The effort today in Generation IV follows through on this basis, with the aim of developing and delivering viable, high-performance systems in a few decades. The six systems are outlined in Table 1. They are described below after a short introduction of the nuclear fuel cycle and followed by summaries regarding fuel cycles and overall sustainability, missions and economic outlook, the approach to safety and reliability, and proliferation resistance and physical protection.

**Table 1. Overview of the six Generation IV systems [1].**

| System | Neutron Spectrum | Coolant | Temperature °C | Fuel Cycle | Size (MWe) |
|---|---|---|---|---|---|
| **VHTR** (very-high temperature reactor) | Thermal | Helium | 900-1000 | Open | 250–300 |
| **SFR** (sodium-cooled fast reactor) | Fast | Sodium | 550 | Closed | 30–150, 300–500, 1000–2000 |
| **SCWR** (supercritical water-cooled reactor) | Thermal/fast | Water | 510–625 | Open/ closed | 300-700 1000–1500 |
| **GFR** (gas-cooled fast reactor) | Fast | Helium | 850 | Closed | 1200 |
| **LFR** (lead-cooled fast reactor) | Fast | Lead | 480–800 | Closed | 20–180 300–1200 600–1000 |
| **MSR** (molten salt reactor) | Fast/thermal | Fluoride salts | 700–800 | Closed | 1000 |

## 2. USE OF THORIUM IN NUCLEAR REACTORS

The utilization of the thorium fuel cycle has been considered attractive since the post World War II period, owing to the excellent neutron characteristics of uranium-233 and the availability of vast thorium resources. Starting around the end of the 50's, a great number of prototypes based on thorium were built. Since the fifties, the interest in thorium has been renewed for different applications. In the United States, one of the first commercial nuclear power plants – Indian Point I – was designed and started up using Th/U fuels, although they were only used in the first cycle of operation. However, the Shippingport reactor used Th/U fuels for several cycles. The reactor was full-power operated on thorium and $^{233}$U cycles from

1977 until 1982 at which time it was shut down. Besides this, the Molten Salt Breeder Reactor – MSBR – and the High Temperature Gas Cooled Reactor also demonstrated the feasibility of using thorium based fuels. In Germany, the development of thorium fueled reactors was connected with High-Temperature Reactor and the Heavy Water Moderated Thorium Breeder Reactor. In the eighties, a program of research and development on the Thorium Utilization in PWRs was performed in German-Brazilian cooperation. France, Japan, the former USSR and India also developed some efforts on thorium based fuels and reactor concepts. Nevertheless, the great success of the Light Water Reactors, the good availability of uranium and the reliability in the $UO_2$ fuels, lead to abandon in some extent the interest devoted to thorium cycle.

Thorium is 3 to 4 times more abundant than uranium and is widely distributed in nature as an easily exploitable resource in many countries. Unlike natural uranium, which contains ~0.7% 'fissile' $^{235}U$ isotope, natural thorium does not contain any 'fissile' material and is made up of the 'fertile' $^{232}Th$ isotope only. Although the cross section for fission at thermal energy is zero (non fissile material), and only fast fission would be possible by using thorium, given the high capture thermal cross section for the reaction, $^{232}Th(n,\gamma)^{232}Th \rightarrow ^{233}Pa \rightarrow ^{233}U$(fissile), makes that Th could be used to produce $^{233}U$(fertile), and used as fuel or in blankets( breeder) of fast reactors. Hence, thorium and thorium-based fuel as metal, oxide or carbide, has been utilized in combination with 'fissile' $^{235}U$ or $^{239}Pu$ in nuclear research and power reactors for conversion to 'fissile' $^{233}U$, thereby enlarging the 'fissile' material resources. During the pioneering years of nuclear energy, from the mid 1950s to mid 1970s, there was considerable interest worldwide to develop thorium fuels and fuel cycles in order to supplement uranium reserves. Thorium fuels and fuel cycles are particularly relevant to countries having large thorium deposits but very limited uranium reserves for their long term nuclear power programme. The feasibility of thorium utilization in high temperature gas cooled reactors (HTGR), light water reactors (LWR), pressurized heavy water reactors (PHWRs), liquid metal cooled fast breeder reactors (LMFBR) and molten salt breeder reactors (MSBR) were demonstrated [4].

The initial enthusiasm on thorium fuels and fuel cycles was not sustained among the developing countries later, due to new discovery of uranium deposits and their improved availability. However, in recent times, the need for proliferation-resistance, longer fuel cycles, higher burn up, improved waste form characteristics, reduction of plutonium inventories and in situ use of bred-in fissile material has led to renewed interest in thorium-based fuels and fuel cycles in several developed countries. The two main international projects, namely Innovative Nuclear Reactors and Fuel Cycles Programme (INPRO) initiated by the IAEA and the US-led Generation IV International Forum (GIF), are also considering thorium fuels and fuel cycles. Thorium fuels and fuel cycles have the following benefits [4]:

- Thorium is 3 to 4 times more abundant than uranium, widely distributed in nature as an easily exploitable resource in many countries and has not been exploited commercially so far. Thorium fuels, therefore, complement uranium fuels and ensure long term sustainability of nuclear power.
- Thorium fuel cycle is an attractive way to produce long term nuclear energy with low radiotoxicity waste. In addition, the transition to thorium could be done through the incineration of weapons grade plutonium (WPu) or civilian plutonium.
- The absorption cross-section for thermal neutrons of $^{232}Th$ (7.4 barns) is nearly three times that of $^{238}U$ (2.7 barns). Hence, a higher conversion (to $^{233}U$) is possible with $^{232}Th$

than with $^{238}U$ (to $^{239}Pu$). Thus, thorium is a better ‘fertile’ material than $^{238}U$ in thermal reactors but thorium is inferior to depleted uranium as a ‘fertile’ material in fast reactor.

- For the ‘fissile’ $^{233}U$ nuclei, the number of neutrons liberated per neutron absorbed is greater than 2.0 over a wide range of thermal neutron spectrum, unlike $^{235}U$ and $^{239}Pu$. Thus, contrary to $^{238}U$–$^{239}Pu$ cycle in which breeding can be obtained only with fast neutrons, the $^{232}Th$–$^{233}U$ fuel cycle can operate with fast, epithermal or thermal spectra.
- Thorium dioxide is chemically more stable and has higher radiation resistance than uranium dioxide. The fission product release rate for $ThO_2$–based fuels is one order of magnitude lower than that of $UO_2$. $ThO_2$ has favorable thermo physical properties because of the higher thermal conductivity and lower co-efficient of thermal expansion compared to $UO_2$. Thus, $ThO_2$–based fuels are expected to have better in–pile performance than that of $UO_2$ and $UO_2$–based mixed oxide.
- $ThO_2$ is relatively inert and does not oxidize unlike $UO_2$, which oxidizes easily to $U_3O_8$ and $UO_3$. Hence, long term interim storage and permanent disposal in repository of spent $ThO_2$–based fuel are simpler without the problem of oxidation.
- Th–based fuels and fuel cycles have intrinsic proliferation-resistance due to the formation of $^{232}U$ via (n, 2n) reactions with $^{232}Th$, $^{233}Pa$ and $^{233}U$. The half-life of $^{232}U$ is only 73.6 years and the daughter products have very short half-life and some like $^{212}Bi$ and $^{208}Tl$ emit strong gamma radiations. From the same consideration, $^{232}U$ could be utilized as an attractive carrier of highly enriched uranium (HEU) and weapons grade plutonium (WPu) to avoid their proliferation for non-peaceful purpose.
- For incineration of WPu or civilian Pu in ‘once-through’ cycle, (Th, Pu)$O_2$ fuel is more attractive, as compared to (U, Pu)$O_2$, since plutonium is not bred in the former and the $^{232}U$ formed after the ‘once-through’ cycle in spent fuel ensures proliferation resistance.
- In $^{232}Th$–$^{233}U$ fuel cycle, much lesser quantity of plutonium and long-lived Minor Actinides (MA: Np, Am and Cm) are formed as compared to the $^{238}U$–$^{239}Pu$ fuel cycle, thereby minimizing the radiotoxicity associated in spent fuel. However, in the back end of $^{232}Th$–$^{233}U$ fuel cycle, there are other radionuclides such as $^{231}Pa$, $^{229}Th$ and $^{230}U$, which may have long term radiological impact.

## 2.1 Molten Salt Reactor (MSR)

The MSR concept was first studied at the Oak Ridge National Laboratory (ORNL), with the Aircraft Reactor Experiment of a reactor for plane based on a liquid uranium fluoride fuel circulating in a BeO moderator. Between 1946 and 1961, the USAF sought to develop a long-range bomber based on nuclear power - the Aircraft Reactor Experiment (ARE). The Aircraft Nuclear Program had unique requirements, some very similar to a space reactor [6]:

- High temperature operation (>1500° F ~815°C), 3 times higher than sub reactors, critical for turbojet efficiency;
- Lightweight design, compact core for minimal shielding, low-pressure for minimal structure, minimal inventory and fuel additions;
- Ease of operability, inherent safety and control, easily removable, minimal reprocessing.

In order to test the liquid-fluoride reactor concept, a solid-core, sodium-cooled reactor was hastily converted into a proof-of-concept liquid-fluoride reactor. The Aircraft Reactor Experiment ran for 100 hours at the highest temperatures ever achieved by a nuclear reactor (1150 K) and produced 2.5 MW of thermal power. It operated from 11/03/54 to 11/12/54.

The molten salt circulated through beryllium reflector in Inconel tubes and it was constituted by $^{235}UF_4$ dissolved in $NaF$-$ZrF_4$. Gaseous fission products were removed naturally through pumping action with very stable operation due to high negative reactivity coefficient. It was demonstrated the load-following operation without control rods.

Studies were then oriented on a civilian application of this concept to electricity production. The Molten Salt Reactor Experiment (MSRE) managed from 1964 to 1969 the operation of a 8 MWth graphite-moderated MSR, with a liquid fuel made of lithium and beryllium fluorides. A third component of the salt was first enriched uranium, then $^{233}U$ and finally plutonium fluoride [7]. The main results of the MSRE are materials improvement against corrosion, control of reactivity and understanding of fuel behavior. Its success led to the Molten Salt Breeder Reactor (MSBR) project of a 1 GWe industrial reactor based on the thorium cycle. The MSR full system includes a graphite-moderated core with channels for $LiF$-$BeF_2$-$ThF_4$ fuel circulation and a pyrochemical reprocessing unit. The latter extracts most capturing Fission Products (FPs), which is essential for $^{233}U$ breeding with the MSBR thermal spectrum. Another feature is the temporary storage of the extracted protactinium, in order to let $^{233}Pa$ decay out of flux into $^{233}U$. The doubling time was evaluated to about 25 years, for a $^{233}U$ inventory of about one (metric) ton. The MSBR project was stopped at Oak Ridge in 1976.

In a Molten Salt Reactor (MSR), the fuel is dissolved in a fluoride salt coolant. Molten salt reactor systems use liquid salts as a coolant and a fuel together. The molten salt fuel flows through graphite core channels, producing an epithermal spectrum. The heat generated in the molten salt is transferred to a secondary coolant system through an intermediate heat exchanger, and then through a tertiary heat exchanger to the power conversion system. The reference plant has a power level of up to 1,000 MWe. The system has a coolant outlet temperature of 700°C, possibly ranging up to 800°C, affording improved thermal efficiency. The closed fuel cycle can be tailored to the efficient burn up of plutonium and minor actinides. The MSR's liquid fuel allows addition of actinides such as plutonium and avoids the need for fuel fabrication. Actinides - and most fission products - form fluorides in the liquid coolant. Molten fluoride salts have excellent heat transfer characteristics and a very low vapor pressure, which reduce stresses on the vessel and piping [5]. MSFR systems have been recognized as a long term alternative to solid-fuelled fast neutron systems with unique potential (negative feedback coefficients, smaller fissile inventory, easy in-service inspection, simplified fuel cycle, etc.).

The main benefits of the MSR system are that it offers an integrated fuel cycle, embodying a burner/breeder reactor concept whilst taking advantage of the excellent heat transport properties of molten salt. These properties imply that the building housing a MSR could be smaller than for other reactor concepts under development and that the thermal power output would be higher. A number of other promising applications for molten salts beyond the MSR itself have been identified. These use a variety of salt compositors that vary according to the envisioned application. These include: liquid fuel; primary or secondary coolant; and pyrochemistry solvent. Molten salts might also be used as a substitute for primary or secondary circuit working fluids in the SFR and VHTR. The molten salt chemistry and handling, with the resulting corrosion of reactor components, along with the development of materials and the fuel cycle, are the main challenges for the development of this system. In the Figure 1 is presented a diagram illustrating a schematic concept of the reactor system and does not represent the reference design [5].

Liquid Fluoride Thorium Reactor or LFTR is a specific fission energy technology based on thorium rather than uranium as the energy source. The nuclear reactor core is in a liquid form and has a completely passive safety system (i.e., no control rods). Major advantages include: significant reduction of nuclear waste (producing no transuranics and ~100% fuel burnup), inherent safety, weapon proliferation resistant and high power cycle efficiency. It is a type of nuclear reactor where the nuclear fuel is in a liquid state, suspended in a molten fluoride-based salt, and uses a separate fluid stream for the conversion of thorium to fissionable fuel to maintain the nuclear reaction. It is normally characterized by: operation at atmospheric pressure; high operating temperatures (>>600K); chemical extraction of protactinium-233 and reintroduction of its decay chain product, uranium-233; thermal spectrum run marginally above breakeven; Closed-Cycle Brayton power conversión.

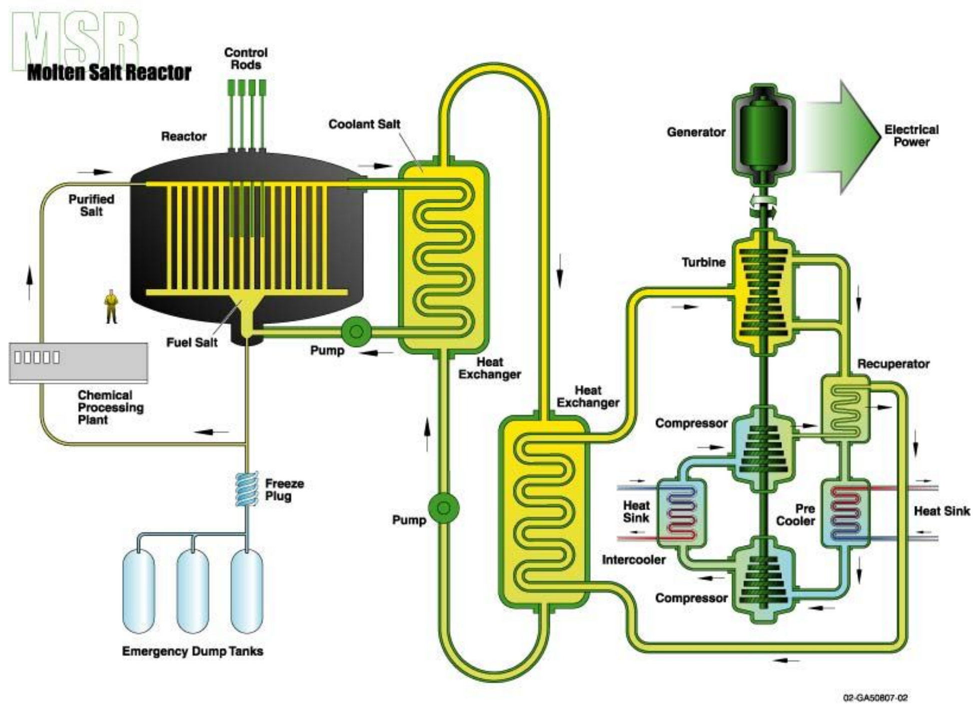


**Figure 1. Schematic concept of the Molten Salt Reactor – MSR [2, 5].**

## 3. BRAZILIAN INTEREST IN THORIUM AND IPEN'S EXPERIENCE

The Brazilian's interest in the nuclear utilization of thorium has started in the 50's as a consequence of the abundant occurrence of monazite sands. Brazil has one of the biggest world nuclear resources (uranium and thorium), being the sixth natural uranium resource in the world (309,000 t $U_3O_8$), one of the first world thorium natural resource. The reasonably assured reserves and the estimated additional resources can reach 1.3 million metric tons of $ThO_2$ as presented in the Table 2 [8]. Nevertheless, as the worldwide fuel industry and the reactor technology have been developed predominantly in the uranium field, the lack of interest in the thorium affected the prospecting and the reserve's evaluation, as well as the research and development in this matter in the country.

In 1960 the Brazilian Nuclear Energy Commission – CNEN – acquired the mining rights from the private companies that were exploiting the monazite in the country (SULBA and ORQUIMA). Nowadays the monazite mining is performed by the Brazilian Nuclear Industries – INB. The INB ore beds are the beach sands in the States of Rio de Janeiro, Espirito Santo and Bahia. Those resources are evaluated as 50.000 t of contained monazite. Unfortunately, reserves and resources of thorium have not been assessed so precisely. The lack of interest in the use of thorium in the industry and as fertile material in nuclear fuels has affected the prospecting. The figures would be much more precise if there were active prospecting programmes for the element. Therefore, it is only possible to have an estimated potentiality of some occurrences. The Brazilian thorium reserves are 73,500 t of $ThO_2$ and the estimated additional resources are 1,202,500 t of $ThO_2$.

**Table 2 . Thorium Potential Resources in Brazil [8]**

| Occurrence | Associated Mineral | Average Content (%) | Measured (t $ThO_2$) | Estimated (t $ThO_2$) |
|---|---|---|---|---|
| Coastal deposits | Monazite | 5 | 2,250 | - |
| Morro do Ferro (MG) | Thorite and others | 1 to 2 | 35,000 | - |
| Barreiro, Araxa (MG) | Pyrochlore | 0.09 | - | 1,200,000 |
| Area Zero, Araxa (MG) | Pyrochlore | 0.09 | 30,000 | - |
| Aluvial and Pegmatite | Monazite | 5 | 3,000 | 2,500 |
| Total | | | 73,500[a] | 1,202,500 |

[a] Including 3,500 t of Monazite sand of INB.

Note: The IAEA gives (1992) 606,000 t as indicated reserves and 700,000 t of inferred reserves.

Nevertheless, the most important source of thorium in Brazil nowadays is the concentrated obtained in the second cake (Torta II) of the sodic opening process of the monazite, for obtaining of rare earth salts. The Torta II is an impure hydroxide containing 20 % of thorium, 1 % of uranium and 6.5 % of rare earths. The amount of Torta II stored by INB can reache about 3,000 t of Th content and the company was interested in the development of a treatment of the Torta II for recovering of uranium and rare earths. The development of a treatment for the remaining waste and thorium recovery must be considered from both economical and environmental point of views. The main operations of the monazite opening process can be observed in the figure 2.

A review on thorium utilization in Brazil can be found in [9]. Since the beginning of Nuclear Energy Development in Brazil in the sixties, it was recognized the strategically of the thorium utilization for the country. In fact the first project was conducted by a research group from a Brazilian State, Minas Gerais, very rich in mineral resources, including thorium. This research group was called the "Thorium Group", and in the framework of a cooperation agreement with the French CEA aimed at the development of a thorium fueled PHWR with a concept of a pre stressed concrete reactor vessel. Other initiatives took place at IEAv (Institute for Advanced Studies), in São Jose dos Campos in the eighties, studying thorium-fuelled both gas-cooled and sodium fast reactors. Also energy scenarios for thorium-fueled in Molten Salt Reactors were studied at Federal University of Minas Gerais. Material studies, such as diffusion properties of ceramics/ $ThO_2$, $(Th,U)O_2$, and $(Th,Ce)O_2$ are studied at CDTN and Federal University of Ouro Preto [10]. Recently a proposal involving IPEN-CNEN/SP, CDTN-CNEN/MG and CTM-SP (Navy Center of Technology) to perform experiments in a water critical facility (IPEN-MB-01) with fuel rods constituted by thorium oxide and thorium-uranium mixed oxide was proposed [11].

### 3.1 Thorium Related Activities in IPEN

Since the sixties, IPEN has performed some activities and developments related to the thorium fuel cycle, mainly in solvent extraction purification process, thorium tetrafluoride preparation and its reduction to metallic thorium, studies of some properties of the $UO_2$-$ThO_2$ solid solutions [12-13]. Thorium was discovered in 1828 by J.J. Berzelius but had few uses until the invention of the Welsbach mantle in 1885. Thoria, $ThO_2$, is the major incandescent component in the fabrication of the Welsbach mantle employed in gas lighting. These mantles, consisting of thorium oxide with about 1 % of cerium oxide and other ingredients, glow with a brilliant white light when heated in a gas flame. Thorium is also used in magnesium alloys, imparting high strength and creep resistance at elevated temperatures and in tungsten filaments for electric lamps (to control the grain size of tungsten) and electronic tubes. Besides this, it is used in special highly refractive optical glass (high quality lenses for cameras and scientific instruments) and in catalysts for several industrially important chemical reactions as, for example, in the conversion of ammonia to nitric acid, in petroleum cracking and in producing sulfuric acid. The oxide is also employed for high-temperature laboratory crucibles, since $ThO_2$ has the highest melting point - 3300 $^oC$ - of all oxides.

The production and purification of thorium compounds was carried out at IPEN for about 18 years. The raw materials used were some thorium concentrates obtained from the industrialization of monazite sands, a process carried out in S. Paulo between 1948 and 1994 on an industrial scale by the company ORQUIMA, later NUCLEMON. During that period, the main product sold was the thorium nitrate with high purity (nuclear grade), having been produced over 170 metric tons of this material in the period, obtained through solvent extraction [14-15]. The thorium nitrate was supplied to the domestic industry and particularly used for gas portable lamps (Welsbach mantle). During production, the process went through a series of changes and a new pilot plant was designed. The construction of the new facility would allow the recovery of production capacity, streamlining and simplifying the process, besides reducing operational costs. An alternative method using a thorium concentrate known as raw thorium hydroxide (HTBR) was used as the precursor for the preparation of pure thorium nitrate at IPEN. Constituted mainly by 60.1% thorium oxide ($ThO_2$), 18.6% rare earth oxides ($TR_2O_3$), and common impurities like silicium, iron, titanium, lead and sodium, this material was produced industrially from the monazite processing in Brazil. To obtain a thorium nitrate as a mantle grade quality, the crude thorium hydroxide was treated with hot nitric acid and after the digestion and addition of flocculants it was filtered for the separation of the insoluble fraction. Using this nitrate solution, the thorium peroxide was precipitated by controlled addition of hydrogen peroxide. Nevertheless, it was decided to suspend the production in 2002-03 and the pilot plant was partly decommissioned in 2003-04. Although the thorium compounds produced have not been employed in the nuclear area, several studies were conducted with a view to conversion of nitrate nuclear-grade thorium oxide suitable for the manufacture of fuel pellets, manufacture of mixed oxide pellets $(U,Th)O_2$, obtaining of thorium tetrafluoride and its reduction to metallic thorium.

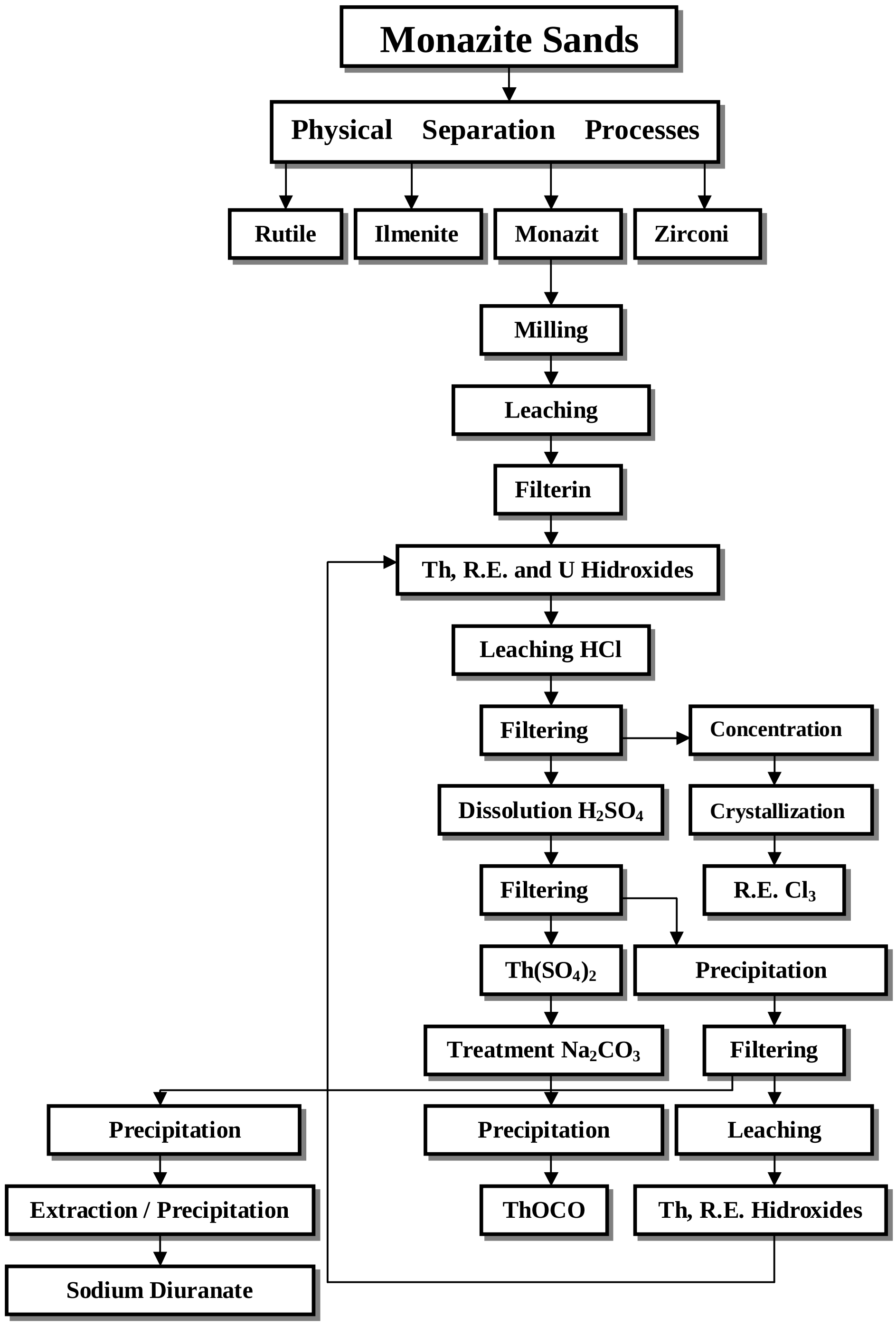


**Figure 2. Main operations of the monazite opening process.**

## 4. CONCLUSIONS

Novel reactor fuel-cycle concepts based on the utilization of thorium are now being considered. The thorium fuel cycle presents some advantages, such as: good characteristics of the U-233, from a neutronic point of view; the thermal stability of $ThO_2$ (melting point around 3300$^{o}$C) that permits high-burn-ups and high temperatures; the ecological argument of much lower quantity of long-lived actinides generated from fission with the thorium cycle, resulting much less long-lived wastes; the average abundance of thorium in the earth's crust that has been estimated three times as great as uranium. Therefore, the accomplishment of those activities and the accumulated experience are of strategic importance for the country, on the one hand due to huge Brazilian reserves of thorium, on the other hand by the resurgence of the interest in the use of thorium in nuclear reactors, particularly for the Generation IV Advanced Reactors.

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