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REVIEW OF BRAZILIAN ACTIVITIES RELATED TO THE THORIUM FUEL CYCLE AND PRODUCTION OF THORIUM COMPOUNDS AT IPEN-CNEN/SP

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ABSTRACT

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. Since the sixties, IPEN-CNEN/SP has performed some developments related to the thorium fuel cycle. The production and purification of thorium compounds was carried out at IPEN for about 18 years and the main product was the thorium nitrate with high purity, having been produced over 170 metric tons of this material in the period, obtained through solvent extraction. The thorium nitrate was supplied to the domestic industry and used for gas portable lamps (Welsbach mantle). Although the thorium compounds produced have not been employed in the nuclear area, several studies were conducted. Therefore, those activities and the accumulated experience are of strategic importance, on one hand due to huge Brazilian thorium reserves, on the other hand by the resurgence of the interest of thorium for the Generation IV Advanced Reactors. This paper presents a review of the Brazilian research and development activities related to thorium technology.

1. INTRODUCTION

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. 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 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, from the absorption of neutrons and subsequent radioactive decay. This uranium isotope is an excellent fuel for use in practically all nuclear reactors types. Before the advent of atomic energy and the appearance of thorium as a source producing secondary fuel (uranium-233), its main application was in the incandescent mantles manufacture. Globally, the 80 and 90 decades were characterized by a significant reduction in the nuclear power growth rate. However, from the year 2000, there was a significant change in the international arena, with the "renaissance" of interest in nuclear power, even in countries that had abandoned nuclear power. In this context, it becomes important to use thorium as nuclear fuel of the Advanced Generation IV reactors, with start-up scheduled for 2030. Unfortunately, contrary to what is happening in most developed countries in recent years, Brazil is paying little attention to thorium, even less than in the past, despite its large reserves. Brazil has one of the biggest world nuclear resources (uranium and thorium), being the sixth natural uranium resource in the world, one of the biggest worldwide thorium resources and only one third of its territorial area has been prospected until now. It is possible that thorium constitutes the major Brazilian energy reserve, supplanting much oil (despite the findings of the pre-salt) and uranium. However, the lack of incentives prevented a greater knowledge of the mineral occurrences and their dimensions in Brazilian territory.

2. HISTORICAL BACKGROUND OF THE BRAZILIAN INTEREST IN THORIUM

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 1 [1]. 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.

Brazilian systematic investigations on the use of thorium fuel cycles in nuclear power reactors started in 1965. During the 60's and early 70's the work was mainly concentrated on the thorium utilization in heavy water reactors (HWRs). This work was performed in the framework of a cooperation agreement with the French Commissariat à l'Energie Atomique - CEA., by the so-called Thorium Group. It was motivated by the results of a long term study of fuel requirements, 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 fuelled pressurized heavy water reactor concept with prestressed concrete reactor vessel. In the frame of the International Nuclear Fuel Cycle program, Brazil started in 1979 an R&D program on the thorium utilization in pressurized water reactors (PWRs), within the scope of a Brazil-Germany cooperation agreement. This program lasted for almost ten years. The activities of both programs were performed at the CDTN - Nuclear Technology Development Center, in Belo Horizonte city, State of Minas Gerais [2, 3].

In the 60's and 70's, also started in São Paulo and Minas Gerais, continuing through the following decades, theoretical and experimental studies with a view not only to the use of thorium reactors, but also to the physical and chemical processes for extraction, purification of compounds, manufacturing and deployment of fuel methods of characterization, recognizing the strategic importance that this element would have for Brazil. However, there was never a national program lasting for a period sufficient to produce practical results of effective use of thorium in the Brazilian reactors. Worse, the specific research activities on different topics of the thorium cycle have declined over time, due to the lack of interest and support. An autonomous nuclear program was carried out in the country in the 80's. However, major political changes occurred in the program in the early 90's. These changes contributed to the further reduction of interest in alternative nuclear fuel cycles.

Table 1. Thorium Potential Resources in Brazil [1]

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	-
Alluvial 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.

3. THORIUM RELATED ACTIVITIES AT IPEN-CNEN/SP

Since the sixties, IPEN has performed some activities and developments related to the thorium fuel cycle, mainly related to neutronic of thorium in nuclear reactors [4-10], thorium compounds chemistry and purification process [11-33], metallic thorium and thorium powder metallurgy [34-44], physical-chemical characterization of thorium compounds [45-59] and radiological protection [60-63].

3.1 Thorium chemistry and processing of monazite sands in Brazil

Brazil has a long tradition of thorium technology, from mining of monazite to obtain thorium with purity suitable for nuclear use. The first reports on the exploitation of monazite in Brazil date back to 1886, when Englishman John Gordon began exporting to Europe the ore mined in the municipality of Prado, Bahia State, for use in lighting (incandescent gas lamps), before the advent electricity from the 20’s, when there was a decline in the consumption of monazite [1]. Interest in monazite resurfaced during World War II, due to its content of thorium. A typical Brazilian monazite contains 39% of cerium oxide, 5% of yttrium, 6% of thorium oxide and 0.3% of U_3O_8. In Brazil, monazite occurs mainly on the beaches of the States of Bahia, Espirito Santo and Rio de Janeiro [14]. In the late of nineteenth and early of twentieth centuries, the interest in monazite increased owing to the use of thorium nitrate by gas mantle industries. Later, the use of lanthanide elements turned monazite into a much more important commodity than it was in pre-war years [64, 65].

In the 40's was started in Brazil in the processing of monazite sand, with a view to exports of rare earths, uranium and thorium, with the work of Brill, Krumholz and colleagues [11]. Orquima S/A started processing of monazite in São Paulo - SP aiming at taking advantage of the rare earths produced, sodium diuranate and rare earth basic carbonate, purchased by the Federal Government and rare earth chlorides for export. In 1951, the Brazilian government banned the export of concentrates of monazite. DNPM data [66] estimate that from 1886 to 1950, were exported from Brazil about 95,000 tons of monazite concentrate. In 1949, the chemical processing of monazite, to produce lanthanide chlorides and tri-sodium phosphate, was started at the Santo Amaro mill (USAM - Orquima S/A), located in São Paulo, the largest Brazilian city. The first phase of the monazite processing consists of the extraction, washing and drying of monazite bearing sands taken from beaches. Then physical separation processes separate the four minerals: ilmenite, rutile, monazite and zircon. Owing to public pressure, economic and radiological problems, the chemical processing of monazite stopped in 1992 [64].

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 most important source of thorium in Brazil nowadays is the concentrated obtained in the second cake (Torta II) of the soda 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 reach more than 3,000 t of Th content.

3.2 Thorium compounds purification at IPEN-CNEN/SP

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. In the course of the industrial treatment of monazite sands in São Paulo, Brazil, several concentrates containing thorium and rare earth elements were produced During the period 1985 - 2003, the main product sold was the thorium nitrate with high purity (mantle grade), having been produced over 170 metric tons of this material in the period, obtained through solvent extraction [14, 23]. The thorium nitrate was supplied to the domestic industry and particularly used for gas portable lamps (Welsbach mantle). Although the thorium compounds produced have not been employed in the nuclear area, several studies were conducted with a view to conversion of nitrate to 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. The main raw material employed during the thorium nitrate production period was the thorium sulfate produced in ORQUIMA. The crystallized thorium sulfate was first transformed in thorium oxocarbonate by addition of water, sodium carbonate and sodium hydroxide. Further, the oxocarbonate was transformed in thorium nitrate by dissolution with nitric acid. To obtain high purity thorium nitrate, the most satisfactory process is purification by solvent extraction. During the period of production, it was employed the solvent extraction in pulsed columns as shown in Figure 1.

Different alternative methods also were studied. A thorium nitrate solution is submitted to an extraction using tributyl phosphate (TBP) diluted in varsol. Thorium nitrate dissolves in tributyl phosphate to form $Th(NO_3)_4$. 2TBP. It is, therefore, extracted from aqueous solution as an unionized compound water free. The solubility of thorium nitrate in TBP is explored to purify the salt in a continuous counter-current mixer-settler unit. At the end of the process, it is obtained a thorium nitrate with purity higher than 99.6%, besides a rich concentrate of rare earths, which will be purified later [26]. Having in mind an eventual substitution of the normally used thorium sulfate as the raw material for the production of pure thorium nitrate, the use of a thorium brute concentrate - HTBR - was also investigated. In the experiments using the HTBR for the separation of thorium from the rare earth elements, some alternative processes were performed: fractional hydroxides precipitation, ion exchange chromatography and precipitation with hydrogen peroxide. The experimental investigations allowed concluding that it is possible to separate thorium from the great majority of the rare earth elements using those techniques [20, 22, 24, 27, 28]. Some experiments were carried out at laboratory scale to evaluate the thorium oxide pellets dissolution and partial denitration of the thorium nitrate solutions obtained. The study corresponded to the head-end operations of the Acid THOREX process and the best conditions of thorium oxide pellets dissolution were determined [15,16]. Nevertheless, radical changes of the Brazilian nuclear policy, in the beginning of 90's, determined the interruption of most R&D fuel cycle activities and the facilities shutdown in the IPEN. It was decided to suspend the thorium nitrate production in 2002-03 and the pilot plant was partly decommissioned in 2003-04 [67, 68].

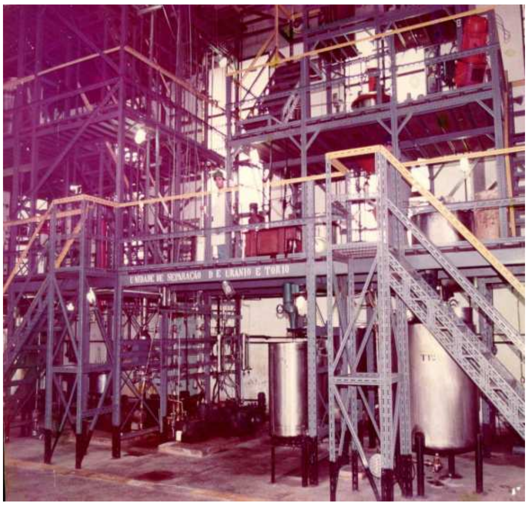

Fig 1. Pilot plant for thorium nitrate purification using pulsed columns.

3.3 Thorium metallurgy and thoria-urania mixed oxide preparation

In the 60's several developments were performed in the laboratory of the Division of Nuclear Metallurgy, Institute of Atomic Energy, regarding the production of metallic thorium and processing by powder metallurgy of the thorium sponge obtained. The studies encompassed the production of thorium achieved by reduction of thorium oxide by calcium, in special reduction bombs, in the presence of calcium chloride and iodine and processing by powder metallurgy of the thorium obtained [34-37]. Some evaluations of different ways of obtaining $(Th\text{-}U)0_2$ mixed oxides were accomplished. Co- precipitation, mechanical mixing of uranium and thorium oxide powders and the sol-gel technique were studied in order to get knowledge of the different processes performance.[25, 39 - 41].

4. CONCLUSION

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 3200-3300^{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. Since the beginning of Nuclear Energy Development in Brazil in the fifties, it was recognized the strategic importance of the thorium utilization for the country. Brazil has a long tradition in the thorium technology, from mining of monazite to the obtainment of thorium with purity suitable for nuclear use. The processing of monazite sand was started in Brazil in the end of 40's, and IPEN-CNEN/SP produced more than 170 t of thorium nitrate of high purity. The accumulated experience is of strategic importance for the country, on 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. Nevertheless, the lack of a Brazilian Thorium Program and the quick aging / retirements of the personnel involved are important factors determining the loss of the acquired knowledge.

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