2017 International Nuclear Atlantic Conference - INAC 2017
Belo Horizonte, MG, Brazil, October 22-27, 2017
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR – ABEN

Agência Brasileira do ISBN
ISBN 978-85-99141-07-6

PROSPECTIVE THORIUM FUELS FOR FUTURE NUCLEAR ENERGY GENERATION

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ABSTRACT

In the beginning of the Nuclear Era, many countries were interested on thorium, particularly during the 1950-1970 periods. Nevertheless, since its discovery almost two centuries ago, the use of thorium has been restricted to gas mantles employed in gas lighting. The future world energy needs will increase and, even if we assumed a conservative contribution of nuclear generation, it will be occur a significant increasing in the uranium prices, taking into account that uranium, as used in the present thermal reactors, is a finite resource. Nowadays, approximately the worldwide yearly requirement of uranium for about 435 nuclear reactors in operation is 65,000 metric t. Therefore, alternative solutions for future must be developed. Thorium is nearly three times more abundant than uranium in the Earth's crust. Despite thorium is not a fissile material, ^{232}Th can be converted to ^{233}U (fissile) more efficiently than ^{238}U to ^{239}Pu. Besides this, thorium is an environment-friendly alternative energy source and also inherently resistant to proliferation.. Many countries had initiated research on thorium in the past, Nevertheless, the interest evanesced due new uranium resources discoveries and availability of enriched uranium at low prices from obsolete weapons. Some papers evaluate the thorium resources in Brazil over 1.200.000 metric t. Then, the thorium alternative must be seriously considered in Brazil for strategic reasons. In this paper a brief history of thorium and its utilization are presented, besides a very short discussion about prospective thorium nuclear fuels for the next generation of nuclear reactors.

1. INTRODUCTION

The use of thorium in power reactors for nuclear energy generation has been considered since the 1950s. Nevertheless, despite some advantages over uranium for use in nuclear reactors, in the almost two centuries since its discovery, the use of thorium was restricted to use for gas mantles, especially in the early twentieth century. The great success of the Light Water Reactors, good availability of uranium and the high reliability of UO_2 fuels have provoked a decrease in the interest devoted to thorium fuel cycle. Considering nuclear energy as an option, uranium is a finite resource if used in a thermal reactor instead of a breeder. The worldwide uranium reserves could last for more than 50-80 years, depending on the amount of reactors in operation in the next future.

Unfortunately, thorium does not have a fissile isotope like the uranium-235. However, with thorium and breeding of ^{233}U in a reactor operating in the thermal spectrum this problem can be overcome. Therefore, although thorium does not present significant commercial value nowadays, in a not too distant future it will probably be an important commodity. Mainly, because the element thorium is considerably more abundant than uranium in the Earth's crust. The average abundance of thorium has been estimated three times as great as uranium. Besides this, the breeding of uranium-233 from thorium is more efficient than the breeding of plutonium from uranium-238. And last, but not least, the thorium fuel cycle generates a much lower quantity of long-lived actinides when compared to the uranium fuel cycle, resulting much less long-lived wastes, a very interesting characteristic from the environmental point of view.

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. Unfortunately, contrarily to what is happening in some countries in recent years, Brazil is paying little attention to the thorium, even less than in the past, despite its huge reserves. The reasonably assured Brazilian thorium reserves and the estimated additional resources can reach 1.3 million metric tons of ThO_2 [1]. It is important the discussion about the future use of thorium, particularly for Brazil, which has large mineral reserves of this strategic element, and the accumulated experience of the IPEN (Instituto de Pesquisas Energéticas e Nucleares) in the purification of thorium compounds.

The first reports on the exploitation of monazite in Brazil date back to 1886, when Englishman John Gordon began exporting to the ore from the municipality of Prado, State of Bahia to Europe, for use in incandescent gas lamps, before the advent of electricity from the 1920s [1]. Between the two World Wars, there was a gradual decline in the consumption of monazite. In 1951, the Brazilian government banned the export of concentrates of monazite. DNPM (National Department of Mineral Production) data [2] estimate that from 1886 to 1950, were exported from Brazil about 95,000 t of monazite concentrate.

Particularly, IPEN has a long tradition in the thorium technology and has accumulated since the 60's a wide experience especially in the purification of thorium compounds with properties suitable for nuclear use. The production and purification of thorium compounds was carried out at IPEN for about 18 years. The main product sold was the thorium nitrate with high purity (nuclear grade) having been produced over 170 metric tons of this material during this period, obtained through solvent extraction. The main raw material employed during the thorium nitrate production period was the thorium sulfate produced by Orquima (Indústrias Químicas Reunidas). This Company started the operations in Sao Paulo in 1949 for processing of monazite aiming at rare earth chlorides for export [3]. One of the byproducts was the thorium sulfate that was first transformed in thorium carbonate by addition of water, sodium carbonate and sodium hydroxide. Further, the carbonate was transformed in thorium nitrate by dissolution with nitric acid [4]. To obtain high purity thorium nitrate, the most satisfactory process is purification by solvent extraction. This material was produced by the unique thorium purification plant in south hemisphere. This facility has already been partially decommissioned [5].

2. RENEWED INTEREST IN THORIUM

During the last decades, the discoveries of new oil and gas reserves scattered around the world and new extraction technologies had an important impact in the reduction of the interest in the nuclear electric generation alternative. Before these discoveries and new technologies, the main world concern was the progressive depletion of fossil reserves and its economic impact. Nevertheless, the increasing of global concerns about the consequences of emission of greenhouse gases from burning of fossil fuels has created a renewed interest in nuclear energy, after some decades of uncertainties.

Mainly considering some serious constraints and concerns such as limited reserves of fossil fuels and their progressive depletion, global warming due the use of fossil fuels, increasing of global population, aspiration for better life conditions implicating in increasing of the power consumption, besides the high cost and inconstancy of some power generation alternatives, like aeolic and solar. Even in a pessimist scenario, a significant part of the electricity generation will be supplied by nuclear energy. There are about 435 nuclear reactors operating in the world nowadays and the electricity generation capacity is about 370 GW(e).The power reactor fleet needs more than 65.000 metric tons of uranium yearly. The nuclear energy generation capacity will reach in the next 30 or 40 years, in a pessimist scenario, 600 GW(e). In an optimistic scenario, 1,400 GW(e). In an intermediate scenario, there will be 1,000 GW(e) from nuclear in the world. Each gigawatt added means approximately 200 tU/year. Then, in an intermediate scenario, by 2050 the consumption of uranium will reach 190,000 tU/year. Therefore, it will be necessary the production of 125,000 tU/year in addition to the present production. So, advanced fuel cycles can increase the reserves of nuclear materials. For this reason they are very interesting. In this case it is particularly important the use of thorium to produce the fissile isotope ^{233}U.

Besides the fact that thorium is three to five times more abundant than uranium in the earth's crust, the mining of thorium is not necessary, since there are big quantities of thorium concentrates in the places where rare earths are explored. Additionally, thoria produces less radiotoxicity than the UO_2, because it produces fewer amounts of actinides. ThO_2 has higher corrosion resistance, besides being chemically stable; the burning of Pu in a reactor based in thorium also decreases the inventories of Pu from the current fuel cycles. There are some ongoing projects in the world, taking into consideration the proposed goals for Generation IV reactors, namely: sustainability, economics, safety and reliability, proliferation resistance and physical protection. Some developments on the use of thorium in reactors are underway, with the support of the IAEA (International Atomic Energy Agency) and some governs like molten salt reactor.

2.1. Reasons for Non Utilization of Thorium as Nuclear Fuel

Considering that ^{235}U is the only fissile isotope naturally existent, and the quantities are relatively very small (only about 0.72 wt.%), besides the fact that it is a finite resource, if used in the current thermal nuclear reactors, the present interest and use of thorium as a nuclear fuel alternative should be much larger.

Since the beginning of operation of the first nuclear power reactors, in mid of 1950 years, until the 1980 decade there was a relevant increasing of this kind of electric generation. Nevertheless, the nuclear accidents of Three Mile Island (USA, 1979) and Chernobyl (Ukraine, 1986) provoked an impact in the rate of growing of number of reactors and total of energy generated. Part of the public has developed an anti-nuclear attitude and the concern about safety of nuclear reactors and radioactive wastes generation has become predominant among many people. Of course, the public perception of the consequences of a nuclear accident has created an opposition to the expansion of this kind of energy. However, the positive consequence of those accidents was the implementation of more reliable safety practices and measures in nuclear industry and in nuclear reactors design and operational procedures. But, the general result was a reduction in the growing of nuclear energy generation and consequently in the uranium consumption.

The main material used for nuclear weapons is highly enriched uranium (HEU), containing about 90% ^{235}U. As a result of the obsolescence of some nuclear warheads and some agreements between USA, Russia and some former ex-USRR republics, part of this enriched uranium has been available for civilian use in commercial nuclear reactors. HEU can be blended with depleted uranium (0.2 to 0.3 wt. % U-235) or natural uranium (0.7% wt.% U-235) to produce fuel for power reactors, constituted by low-enriched uranium (LEU), with less than 5% U-235. Hundred tons of HEU were transformed in thousands of tons of LEU, available as fuel for commercial reactors. The agreement reduced the amount of nuclear weapons but significantly depressed uranium exploration activities and the uranium price. The availability of low price uranium (in some sense, artificially kept) reduced considerably the interest in alternative fuels, since the interest about thorium stems mainly from the uranium prices.

Another aspect that has provoked a decrease in the interest devoted to thorium fuel cycle was the great success of the Light Water Reactors and the high reliability of UO_2 fuels. Nevertheless, since this type of reactor is the most commercially utilized, mainly in a "once trough fuel cycle", many R&D programs had focused in the Th utilization in it [6].

3. PROSPECTIVE NUCLEAR FUELS WITH THORIUM

Presently, only the raw material uranium is used for commercial nuclear power production. Uranium ore resources appear to be assured for several decades but they are expected to become exhausted, at least for low cost uranium, in the middle (assuming high growth of nuclear energy) and towards the end of this century (assuming low growth of nuclear energy). Continued exploration efforts may further extend those uranium resources but probably at a higher cost. An attractive additional resource of natural fertile material will be the use of ores containing thorium. There is some reactor technology know-how available how to use thorium which was designed in order to convert ^{232}Th into ^{233}U [7].

There are many considerations and studies going on internationally to prepare options and plans for the further development of nuclear energy technology in the near (15–25 years) and in the more distant (>25 years) future. Most of them are aware that this development has to take care of and to synthesize demands of sustainability, i.e. of economics, safety and security, environment, waste, infrastructure and proliferation resistance. And most of them understand those efforts as part of necessary activities that include the whole nuclear fuel cycle. By far, the largest and strongest effort of this kind is the Generation IV and the GNEP initiative [7].

There are some ongoing projects in the world, taking into consideration the proposed goals for Generation IV reactors, an initiative of the DOE-USA, namely: sustainability, economics, safety and reliability, proliferation resistance and physical protection. Some developments on the use of thorium in reactors are underway, with the support of the IAEA (International Atomic Energy Agency) [8].

Another initiative is the International Project on Innovative Nuclear Reactors and Fuel Cycles (INPRO). INPRO was established in 2001 in response to a resolution by the IAEA General Conference. The INPRO´s objectives: to help to ensure that nuclear energy is available to contribute, in a sustainable manner, to meeting the energy needs of the 21st century; to bring together technology holders and users so that they can consider jointly the international and national actions required for achieving desired innovations in nuclear reactors and fuel cycles. INPRO provides an open international forum for studying nuclear energy options, the associated requirements and the potential deployment of innovative nuclear energy systems in IAEA Member States [7].

3.1. Thorium Fuel Perspectives

Since we are concerned about the necessity of a thorium program for Brazil, it would be important to prepare a review as comprehensive as possible. However, it is difficult to cover in one paper all types of thorium fuels for all types of commercial, evolutionary and innovative nuclear reactors. Then, it is necessary to categorize the several alternatives, in accordance with the reactor type or concept, composition of fuel and physical properties. Of course, not all concepts are applicable to the Country, at least in the near future. It is important emphasize that, in spite of the use of thorium-based fuels has been studied, it is necessary much effort to reach the level of information available for uranium fuels.

It is important to emphasize that the different future thorium-based fuels are directly related to the type of reactor in which they will be used. Despite GEN IV initiatives, none of the selected reactors [7] with potential for future use are really new concepts. They are in fact evolutionary concepts of prototypes already developed a few decades ago. The same can be said about the corresponding fuels.

Basically, four major fuel types can be viewed. The most common would be ceramic fuels, in the form of pellets, for use in heterogeneous reactors with liquid refrigerants and microspheres for gas-cooled reactors. In addition, liquid fuels, in the form of fluoride mixtures, for homogeneous reactors such as MSR are another important current but already known for a long time. As a more recent alternative, and where there is a need for more research effort, are the fuels based on uranium, thorium and possibly plutonium alloys.

3.1.1. Thoria and mixed oxides

The use of fuel constituted by pellets of thoria or mixed oxides is probably one of the most important applications due the prominence of the PWRs and VVERs in the world scene, besides the possibility of use in CANDUs, AHWRs (Advanced Heavy Water Reactors). In some cases, thoria pellets were used for power-flattening in the initial core, as occurred with the Indian PHWR [9]. In USA, some projects involving use of a thorium fuel cycle were performed in the past. Those projects were based on an once-through, proliferation resistant, high burnup and long refueling cycle use of thorium in light water reactors and they were directed mainly to the use of uranium-thorium dioxide fuels. The use of urania-thoria fuels also was object of interest by Brazilian and German institutions. A comprehensive report was prepared in the eight's years aiming the fabrication of these fuels for use in PWRs [10]. More recent works have availed the possibility and the viability of use of thorium and uranium mixed oxide fuel pellets in the more advanced PWRs, like AP-600 and AP-1000 [11].

Despite being a rather limited study, due to difficulties with obtaining resources, IPEN has started recently a study focused in the sintering of thoria pellets. Considering that thorium oxide is an important nuclear reactor material and since its ceramic fabrication process involves a very high sintering temperature, considering that thoria melting point is very high (3,650 K), an investigation about the use of niobium as a dopant for thoria sintering temperature reduction was initiated, taking into account the huge availability of thorium and niobium in the country. Cations of elements of the group VB (V, Nb and Ta) have a known effect in the reduction of thoria sintering temperature. Thoria used in the study was produced in the IPEN's pilot plant and different amounts of niobium oxide (Nb_2O_5) will be added to thoria by different routes [12].

3.1.2. Microspheres containing thorium

Germany, United States, Japan and Russian Federation have already used helium gas cooled reactors and combinations of thorium and highly enriched uranium, or thorium and plutonium, in the form of microspheres [13]. There are two basic high temperature reactors – HTR – designs. In Germany, It was adopted the pebble bed reactor using spherical elements, and in United States was adopted the prismatic fuel concept. Since the interest in the High Temperatures Gas Cooled Reactors has been renewed in the INPRO – Innovative Nuclear Reactors and Fuel Cycle Programme – lead by IAEA, and in the GIF - Generation IV International Forum, a program lead by the United States, and taking into account that this concept of reactor allows a wide choice of fuel designs/fuel cycle, a lot of development must be occur in the next years. Fuels capable of providing helium temperatures around 750°C are already available. Then, the research effort will occur mainly in providing fuels capable to reach around 1000°C of outlet temperatures.

3.1.3 Molten salt fuel containing thorium

Molten salt reactors (MSR) use graphite as moderator, molten fluoride salt of high boiling point (>1400°C) with dissolved 'fissile' and 'fertile' materials as fuel and primary coolant and operate in an epithermal neutron spectrum. The primary coolant, containing the fuel, constituted by the molten fluoride salt containing thorium uranium and plutonium, flows to a primary heat exchanger, where the heat is transferred to a secondary molten salt coolant and then flows back to the graphite channel of the reactor core [13].

The Experimental Molten Salt Reactor (MSRE), constructed in Oak Ridge National Laboratory (ORNL), USA in the 1960s was the first and the only thorium-based MSR in the world. The 8 MWt MSRE had a core volume of <2 m^3 and operated with a molten fuel / coolant salt of composition $7LiF/BeF_2/ThF_4/UF_4$ [13]. In spite of the MSRE was shutdown in December 1969, and the then proposed/designed MSBR–1000 was not constructed, MSR has been considered as a potential option for the GIF and INPRO. In recent years, several national and multinational collaborative programmes on thorium utilization in MSR have been initiated. These countries have identified MSR as a potential component of thorium-based 'closed' fuel cycle to efficiently burn actinides and reduce the long term radiotoxicity of nuclear wastes. In the case of MSR, it seems that the main effort related to the fuel in the next years will be done in the development of an online reprocessing unit for removal of fission products and for feeding of heavy nuclei (U, Th,etc.) to the MSR.

3.1.4. Thorium metallic fuels

In India, there has always been a strong incentive for development of thorium fuels and fuel cycles because of large availability of thorium in the country, associated with the search for the independence from overseas uranium sources. Besides several R&D activities on $(Th,U)O_2$ and $(Th,Pu)O_2$ fuels containing <5% uranium or plutonium oxide for use in water cooled reactors and $(Th, Pu)O_2$ containing from 20–80% PuO_2 for use in LMFBR, respectively, India has initiated a very important and interesting study about essential microstructural, mechanical and. thermophysical properties of thorium-uranium alloys [14].

Thorium-based metallic fuel is of great interest to LMFBRs with excellent safety features. The research of these properties in U-Th metallic fuels has been little explored and the study in question represents an important contribution to know the possibilities and limitations of these potential fuels. Th-U alloys present prospective possibility of using as nuclear reactor fuel in the future, since complementary studies prove they have favorable complementary properties, such as performance in operation, stability under radiation, corrosion, etc.

3.1.5. Innovative thorium fuels

The Nuclear Energy Research Initiative (NERI) project of the Department of Energy, USA has developed an innovative metal matrix dispersion, or cermet fuel consisting of $(Th,U)O_2$ microspheres (using LEU: <20% ^{235}U) of diameter ~50 micron in a zirconium matrix that can achieve high burnup in a 'once-through' cycle and disposed, as nuclear waste without processing. The metal matrix fuel has been manufactured by the 'powder–in–tube–drawing' technique, which consist of dry mixing or wet vibratory milling of zirconium powder with (Th, U)O_2 microspheres, loading the powder mixture in cladding tube and vibratory packing. The high thermal conductivity of zirconium matrix enhances heat removal and keeps the fuel center temperature significantly low [13].

Japan accomplished R&D activities on the innovative thorium-based hydride fuels for advanced Minor Actinides (MA) and plutonium burners with high safety characteristics. The U–Th–Zr–H fuel has high thermal conductivity and consists of U–metal, Th–Zr_2Hx and ZrHx phases [13].

4. CONCLUSIONS

In accordance with the premises of the GIF and INPRO, the future reactor types and fuel cycle options in different countries will depend on resource utilization, environmental impact, safety, public acceptance and energy security and sustainable supply.

Then, the discussion about the development of a thorium fuel cycle is of strategic importance for our country, on one hand, due to huge Brazilian reserves of thorium and the existence of an accumulated experience in thorium compounds purification and, on other hand, by the resurgence of the interest in the use of thorium in nuclear reactors, particularly for the advanced Generation IV 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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