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USE OF THORIUM IN THE GENERATION IV MOLTEN SALT REACTORS AND PERSPECTIVES FOR BRAZIL

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ABSTRACT

Interest in thorium stems mainly from the fact that it is expected a substantial increase in uranium prices over the next fifty years. The reactors currently in operation consume 65,500 tons of uranium per year. Each electrical gigawatt (GWe) additional need about 200 tU mined per year. So advanced fuel cycles, which increase the reserves of nuclear materials are interesting, particularly the use of thorium to produce the fissile isotope 233U. It is important to mention some thorium advantages. Thorium is three to five times more abundant than uranium in the earth's crust. Thorium has only one oxidation state. Additionally, thoria produces less radiotoxicity than the UO2 because it produces fewer amounts of actinides, reducing the radiotoxicity of long life nuclear waste. ThO2 has higher corrosion resistance than UO2, besides being chemically stable due to their low water solubility. The burning of Pu in a reactor based in thorium also decreases the inventories of Pu from the current fuel cycles, resulting in lower risks of material diversion for use in nuclear weapons. 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 and some governs. Can be highlighted some reactor concepts using thorium as fuel: CANDU; ADTR - Accelerator Driven Thorium Reactor; AHWR - Advanced Heavy Water Reactor proposed by India (light water cooled and moderated by heavy water) and the MSR - Molten Salt Reactor. The latter is based on a reactor concept that has already been successfully tested in the U.S. in the 50s, for use in aircrafts. In this paper, we discuss the future importance of thorium, particularly for Brazil, which has large mineral reserves of this strategic element, the characteristics of the molten salt reactor and the experience of the IPEN in the purification of thorium compounds.

1. INTRODUCTION

The world today faces many challenges and one of the most important is to maintain a sustainable society with a growing demand for products, increasing consume of energy from fossil fuels and coal. These may provide a positive feedback that amplifies the global warming and climate change [1]. Effort to decrease this positive feedback must be necessary to avoid economic and agricultural impacts in world. It is only be accepted by society if there is maintenance of their economic pattern. The energy from solar, wind and nuclear together will be the substitute. The approval for nuclear power after the Fukushima Daichii accident has declined. The change in this framework can only be effective when there is a perception, by the public opinion, that the new projects in nuclear energy are intrinsically safe. This characteristic can be covered by the concepts incorporated in the design of the Generation IV nuclear reactors.

The reactors currently in operation consume 65,500 tons of uranium per year. Each electrical gigawatt (GWe) additional need about 200 tU mined per year. So advanced fuel cycles, which increase the reserves of nuclear materials are interesting, particularly the use of thorium to produce the fissile isotope 233U. One of the technological alternatives proposed by the Generation IV nuclear reactors, that meets those needs, is the molten salt reactor (MSR) based on thorium. Projects under development by the United States, China and Europe indicate that in the coming decades this reactor type will have relevant developments.

In this paper, we discuss the future importance of thorium, particularly for Brazil, which has large mineral reserves of this strategic element, the characteristics of the molten salt reactor and the experience of the IPEN in the purification of thorium compounds.

2. GENERATION IV NUCLEAR REACTORS AND THE MOLTEN SALT REACTOR

The concepts adopted in the design of the Generation IV Nuclear Reactors are base in some premises: sustainability, economics, safety and reliability, proliferation resistance and physical protection. The Molten salt reactor (MSR) meets the specifications of the Generation IV Nuclear Reactors.

2.1. Molten Salt Reactor - MSR

With changing goals for advanced reactors and new technologies, there is currently a renewed interest in MSRs. The new technologies include: Brayton power cycles (rather than steam cycles) that eliminate many of the historical challenges in building MSRs and the conceptual development of several fast-spectrum MSRs that have large negative temperature and void coefficients, a unique safety characteristic not found in solid-fuel fast reactors.

Molten salt reactor (MSR) is a homogeneous reactor operating in thermal-neutron-spectrum. MSR systems use liquid salts as a coolant and a fuel together, since in a molten salt reactor the fuel is dissolved in a fluoride salt coolant. The MSR concept was first studied at the Oak Ridge National Laboratory (ORNL), with the Aircraft Reactor Experiment of a reactor for aircraft based on a liquid uranium fluoride fuel circulating in a BeO moderator. The concept of the MSR was developed in the 1950s and two small thermal-neutron-spectrum MSRs were successfully built in the 1960s. The first reactor was part of a program to build a nuclear-powered aircraft, whereas the second reactor was built to test the concept of a molten salt breeder reactor (MSBR). Between 1946 and 1961, the USAF sought to develop a long-range bomber based on nuclear power - the Aircraft Reactor Experiment (ARE). The programs ended in 1976 when the United States decided to concentrate on a single breeder reactor concept—the sodium-cooled fast reactor. Dr. Alvin Weinberg worked at Oak Ridge National Laboratory from 1955 to 1974 on the subject of fluid-fueled reactors, particularly those that used liquid-fluoride salts as a medium in which to sustain nuclear reactions [2-4].

Earlier MSRs were thermal-neutron-spectrum reactors. Compared with solid-fueled reactors, MSR systems have lower fissile inventories, no radiation damage constraint on attainable fuel burnup, no spent nuclear fuel, no requirement to fabricate and handle solid fuel, and a single isotopic composition of fuel in the reactor. With changing goals for advanced reactors and new technologies, there is currently a renewed interest in MSRs. The new technologies include (1) Brayton power cycles (rather than steam cycles) that eliminate many of the historical challenges in building MSRs and (2) the conceptual development of several fast-spectrum MSRs that have large negative temperature and void coefficients, a unique safety characteristic not found in solid-fuel fast reactors.

Liquid Fluoride Thorium Reactor or LFTR is a specific fission energy technology based on thorium rather than uranium as the energy source. The fuel is dissolved in a fluoride salt coolant constituted by a mixture of fluorides (LiF.BeF.ThF4.UF4). The nuclear reactor core is in a liquid form and has a completely passive safety system (i.e., no control rods). The MSR is based on the nuclear power generation by fission of U-233 obtained from the Th-232, using salt melted at 700°C. 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 inlet temperature of the coolant (e.g., fuel-salt mixture) is 565°C while the outlet temperature reaches 700°C. However, the outlet temperature of the fuel-salt mixture can even increase to 850°C when co-generation of hydrogen is considered as an option. The thermal efficiency of the plant can reach between 45 and 50%. In the Fig. 1 and 2 [2] are presented schematic drawings of the MSR illustrating the molten salt reactor concept.

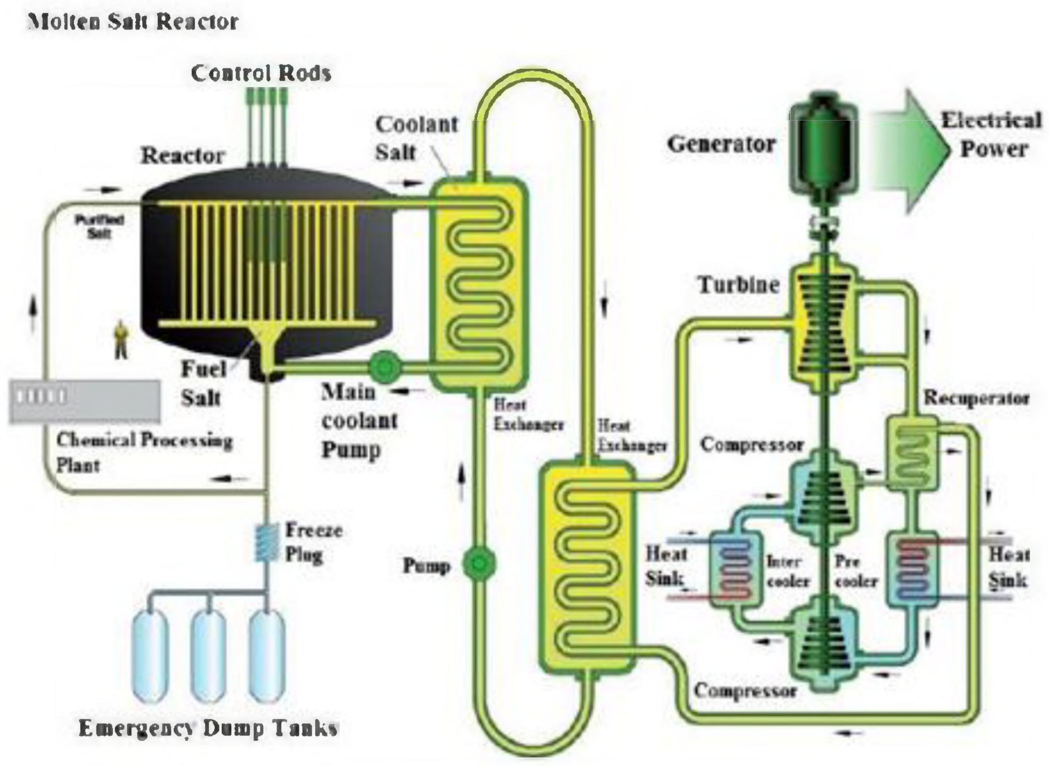

Figure 1. Scheme of Molten Salt Reactor (MSR) (US DOE, 2002).

The reactor has several applications, allowing its use in: power generation; radiopharmaceutical production, using the fission product yields Molybdenum Technetium; hydrogen production using the high reactor temperature 600°C; production of potable water by desalinization.

Fast-spectrum MSR concepts have been recently developed with unique capabilities in terms of actinide burning and fuel production. This is partly a consequence of a broader understanding of fluoride salt chemistry. The preferred salt is determined primarily by three factors: physical properties that determine its behavior as a coolant that must flow through the reactor core and heat exchangers, the neutronic and the chemistry. Different salts have different properties; thus, a viable molten salt for a thorium–^{233}U breeder MSR is different from the optimum salt for actinide burning. The development of fast-spectrum MSRs requires salts with higher solubility for fissile and fertile materials and less neutron moderation.

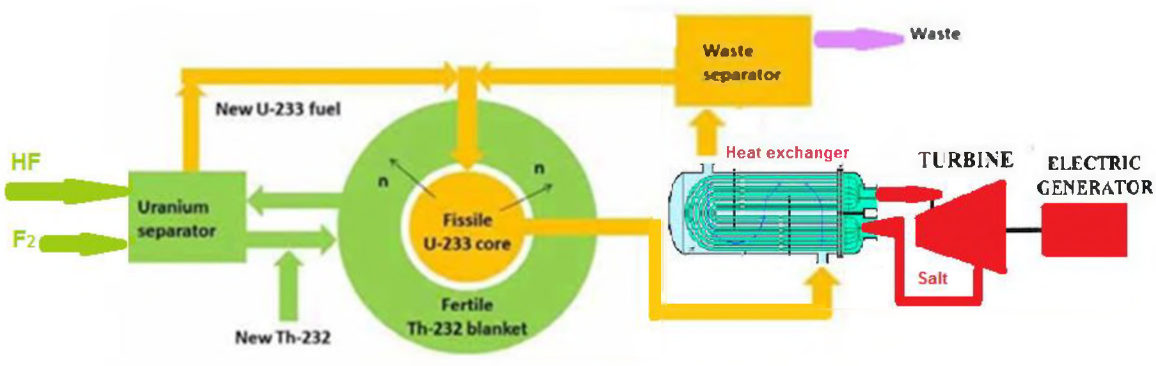

Figure 2. Schematic representation of the molten salt reactor.

2.1.1. Advantages and challenges

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. The MSR's liquid fuel allows addition of actinides such as plutonium and avoids the need for fuel elements fabrication. Actinides - and most fission products - form fluorides in the liquid coolant.

Another advantage is related the use of Brayton power cycles. Because of the melting points of molten salts (350 to 500°C), MSRs are intrinsically high-temperature reactors. When MSRs were first developed, steam cycles were the only power cycle options. Coupling steam cycles to MSRs was complicated because of the need to avoid freezing of the salt, diffusion of tritium through hot heat exchangers from the MSR into the steam, and other constraints. The development of closed helium and nitrogen Brayton power cycles has eliminated many of these technological challenges (salt freezing, tritium migration, etc.), significantly improved power plant efficiency, and reduced capital costs. Power cycles now exist that match the characteristics of MSRs.

Molten fluoride salts have excellent heat transfer characteristics and a very low vapor pressure, which reduce stresses on the vessel and piping [2]. It was not a light-water reactor, nor was it a fast-breeder reactor. It has a thermal (slowed-down) neutron spectrum which made it easier to control and vastly improved the amount of fissile fuel it needed to start. The MSR operates at atmospheric pressure rather than the high pressure of water-cooled reactors, obviating the need for a large containment dome and no danger of an explosion. A molten salt reactor cannot melt down because the normal operating state of the core is already molten.

The salts are solid at room temperature, so if a reactor vessel, pump, or pipe ruptured they would spill out and solidify. If the temperature rises, stability is intrinsic due to salt expansion. In an emergency an actively cooled solid plug of salt in a drain pipe melts and the fuel flows to a critically safe dump tank. The Oak Ridge MSR researchers turned the reactor off this way on weekends. The high heat capacity of molten salt exceeds that of the water in PWRs or liquid sodium in fast reactors, allowing compact geometries and heat transfer loops utilizing high-nickel metals. High temperatures enable 45% efficient thermal/electrical power conversion using a closed-cycle turbine, compared to 33% typical of existing power plants using traditional Rankine steam cycles. Cooling requirements are nearly halved, reducing costs and making air-cooled MSR's practical where water is scarce.

Major advantages of the LFTR include: significant reduction of nuclear waste (producing no transuranics and ~100% fuel burn up), 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.

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. The technical challenges and risks that must be addressed in a prototype development project include control of salt container corrosion, recovery of tritium from neutron irradiated lithium salt, management of structural graphite shrinking and swelling, closed cycle turbine power conversion, and maintainability of chemical processing units for U-233 separation and fission product removal.

One important point to be considered when thorium or uranium are compared for using in nuclear reactors is that, differently from the need of uranium mining, thorium is associated to rare earths. In other words, to obtain uranium, it is necessary to mine it, generating a considerable amount of wastes, since the U content is in the range of approximately 0.2-0.5% in mass. Nevertheless, in the case of thorium, the rare earths will be mined anyway and the tails containing thorium can be used without the generation of additional wastes.

Two fertile materials (^{232}Th and ^{238}U) can be converted to fissile materials and form the basis of a long-term sustainable closed fuel cycle. Thorium-232 plus a neutron yields fissile ^{233}U and ^{238}U plus a neutron yields fissile ^{239}Pu. The uranium–^{239}Pu fuel cycle generates large quantities of transuranic (TRU) actinides. The thorium–^{233}U fuel cycle generates almost no TRU actinides, because it takes many neutron captures to convert ^{233}U to a TRU isotope.

One problem associated to the reprocessing of thorium fuels is the presence of U-232, that is extremely radioactive, has a half-life around seventy years and it is dangerous even in small amounts. By one hand, the presence of U-232 makes the fabrication of thorium-uranium-233 mixed oxide fuel much more difficult but, by other hand, this problem is one of the reasons that makes thorium cycles more favorable from the point of view of proliferation risks. But, if the fabrication of oxide fuels is complex due the presence of U-232 in the form of particles that can be ingested or inhaled, when the use of thorium in a LFTR is compared with the use of thorium in a heterogeneous reactor, the first has a considerable advantage. LFTRs use fuel in the liquid state and there is no need of fuel elements fabrication steps employing powder metallurgy techniques. Then, the use of thorium is more advantageous when a homogeneous reactor with liquid fuel is considered, as it is the case of the LFTRs

In July 2010, an industry organization with members such as Toyota, Toshiba and Hitachi, IThEMS unveiled their plans to build the world's first commercial Thorium Molten-Salt Reactor (Th-MSR) power generator. The Fuji Molten salt reactor is a Japanese design that can run on thorium or a mix of thorium and uranium or plutonium. The first step on the path to commercially available Thorium Energy will be through their 10 MW mini FUJI (in 5 years). That will be followed by a larger capacity design called FUJI, delivering 200MW in ten years. In accordance with the organization, the Fuji Molten salt thorium reactor would generate power at a cost significantly lower than that of current Light Water Reactors (LWR) – at least 30% lower [5].

Commercialization of technology lowers costs as the number of units produced increases due to improvements in labor efficiency, materials, manufacturing technology and quality. Doubling the number of units produced reduces cost by a percentage termed the learning ratio, which is often about 20%. In The Economic Future of Nuclear Power, University of Chicago economists estimate it at 10% for nuclear power reactors. Reactors of 100 MW size could be factory-produced daily in the way that Boeing Aircraft produces one airplane per day. At a learning ratio of 10%, costs drop 65% in three years.

3. THORIUM AND URANIUM IN BRAZIL

3.1. Thorium

The mineral thorium was discovered by Berzelius in 1828. The natural thorium isotope is formed by the Th-232 that is a fertile element and can be transmuted in U-233 by bombardment with neutrons, as can be observed in the Fig. 3.

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 manufacture incandescent mantles.

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. The thorium mining in Brazil started in the 50s by Orquima company, in search of various minerals, especially rare earths, produced different kinds of RE and thorium concentrates obtained from monazite sands, a phosphate of rare earths and thorium.

Research and development related to thorium at IPEN, from 60's until the first years after 2000, included a semi-industrial unit for supply of thorium nitrate with purity above 99% for impregnation of mantles for incandescent gas lamps. The purification process used was solvent extraction in pulsed columns with TBP (trybutilphosphate) and Varsol as diluent. A picture of the purification pilot plant can be observed in the Fig. 4 [6]. It should also be recorded that there was at IPEN, from 1985 until 2002, the production in pilot-scale of over one hundred and seventy metric tons of thorium nitrate with high purity.

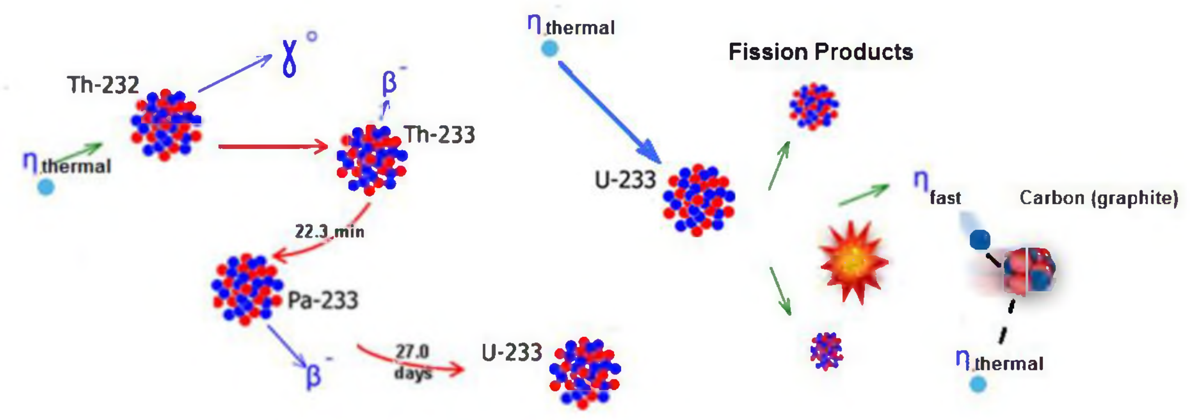

Figure 3. Nuclear reactions involved in the transmutation of ^{232}Th in ^{233}U

Figure 4: Pilot plant production of thorium nitrate.

3.2. Uranium

The uranium ore was discovered by Klaproth in 1789. The uranium exploration in Brazil began jointly with thorium and rare earths, having its biggest boost in the Brazilian nuclear program started in the 60's, looking for the expertise in obtaining nuclear enriched uranium needed to build nuclear reactors. At IPEN, the uranium purification was accomplished by solvent extraction using TBP and Varsol as diluent. It was possible the obtainment of uranyl nitrate with purity suitable for nuclear purposes. A view with the purification pilot plant can be observed in the Figure 5. The next step of the process at IPEN was the precipitation of the purified uranyl nitrate as ammonium diuranate (ADU) and its transformation into oxides by calcination. The UO_3 formed was converted first to UF_4. Two different routes were adopted at IPEN for the UF_4 obtainment: dry and wet routes. The next step was the conversion to UF_6 by reaction of UF_4 with F_2 produced by electrolysis. The technology of gas-solid reactions necessary for obtaining UF_4 was derived from a cooperation agreement involving IAEA and France [7].

The know-how of obtaining UF_6 until the fuel element was obtained by Brazilian scientists The main achievement in the nuclear fuel cycle technological domain at IPEN was the development of the U-235 isotopic enrichment using ultracentrifuges that was a result of a partnership between the Brazilian Navy and IPEN [8-10]. The enriched UF_6 was converted to UO_2 employing the AUC route (ammonium uranyl carbonate). Enriched UO_2 was transformed in green pellets and sintered. Pellets produced in the IPEN´s UO_2 Pilot Plant were used to the fabrication of the first core of the IPEN-MB 01, a research reactor built in IPEN also in cooperation with Brazilian Navy. The first criticality of this reactor, whose main purpose is the obtainment of nuclear parameters for the design of PWRs reactors, was reached in 1988. Reduction of UF_4 (natural enrichment) to metallic uranium by magnesium was also performed at IPEN in the nineties' [11]. In the 2000s, the reduction of UF_6 enriched to 20% was also held at IPEN for obtaining of U_3Si_2 used as fuel in the form of dispersions for the IEA-R1 research reactor.

Figure 5: Uranyl nitrate purification pilot plant.

4. CONCLUSIONS

The developments of uranium and thorium fuel cycles in Brazil involved several industries and Brazilian Research Institutes: Orquima, Nuclemon, INB, CNEN, CDTN, IRD and IPEN. Unfortunately, due changes in the Brazilian nuclear policy in the early 1990s, the continuity of these technological developments at IPEN were interrupted.

Brazil was part of the ten countries initially involved in the Generation IV Technology group. To advance nuclear energy to meet future energy needs, ten countries: Argentina, Brazil, Canada, France, Japan, the Republic of Korea, the Republic of South Africa, Switzerland, the United Kingdom, and the United States have agreed on a framework for international cooperation in research for a future generation of nuclear energy systems, known as Generation IV. Nowadays, Brazil and Argentina are not effective members of the group.

Nevertheless, Brazil should be involved in this research because energy resources from oil, coal or hydroelectric are for some decades, which can be considered a relatively short space of time, when you think about a nation. 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 50 – 80 years, depending on the amount of reactors in operation in the next future. With thorium and the breeder of U-233 in a reactor operating in the thermal spectrum this problem can be overcame. Besides this, Brazil has very important thorium resources and this should be considered to establish national priorities.

ACKNOWLEDGMENTS

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