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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 to have a substantial increase in uranium prices. So, advanced fuel cycles which increase the reserves of nuclear materials are interesting, particularly, the use of thorium is to produce the fissile isotope ^{233}U. Thorium is three to five times more abundant than uranium in the earth's crust. 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, and 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 govern like molten salt reactor. 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 (Instituto de Pesquisas Energéticas e Nucleares) in the purification of thorium compounds.

Key words: Molten salt reactor, Generation IV reactors, thorium, uranium-233.

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 and 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 is necessary to avoid economic and agricultural impacts in world. It is accepted by society only if there is maintenance of their economic pattern.

The energy from solar, wind and nuclear will be the substitute. The approval for nuclear power has declined after the Fukushima Daichii accident. 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 t U mined per year. So advanced fuel cycles, which increase the reserves of nuclear materials are interesting, particularly, the use of thorium is to produce the fissile isotope ^{233}U. One of the technological alternatives proposed by the Generation IV nuclear reactors, that meets those needs, is the MSR (molten salt reactor) 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

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2. Generation IV Nuclear Reactors and the Molten Salt Reactor

The concepts adopted in the design of the Generation IV nuclear reactors are based on some premises: sustainability, economics, safety and reliability, proliferation resistance and physical protection. The MSR meets the specifications of the Generation IV nuclear reactors.

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 is a homogeneous reactor operating in thermal-neutron-spectrum. MSR systems use liquid salts as a coolant and fuel at the same time, since in a molten salt reactor, the fuel is dissolved in a fluoride salt coolant. The MSR concept was first studied at the ORNL (Oak Ridge National Laboratory), 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 MSBR (molten salt breeder reactor). Between 1946 and 1961, USAF (the United State Air Force) sought to develop a long-range bomber based on nuclear power—the ARE (aircraft reactor experiment). 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 Brayton power cycles [2] (rather than steam cycles) that eliminate many of the historical challenges in building MSRs and the conceptual development of several fast-spectrum MSRs [3] that have large negative temperature and void coefficients, a unique safety characteristic not found in solid-fuel fast reactors.

LFTR (liquid fluoride thorium reactor) 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, ThF_4, UF_4). 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 ^{233}U obtained from the ^{232}Th, 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 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%, and has a power level of up to 1,000 Mwe. Figs. 1 and 2 presented schematic drawings of the MSR illustrating the molten salt reactor concept [2].

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 ^{232}Th-^{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.

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 assembly fabrication. Actinides and most fission products form fluorides in the liquid coolant.

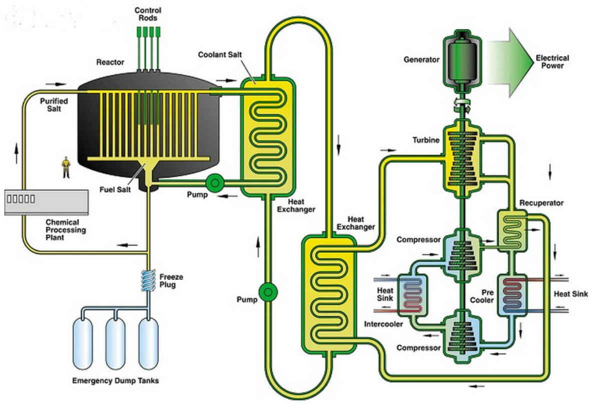

Fig. 1 Scheme of MSR (US DOE, 2002).

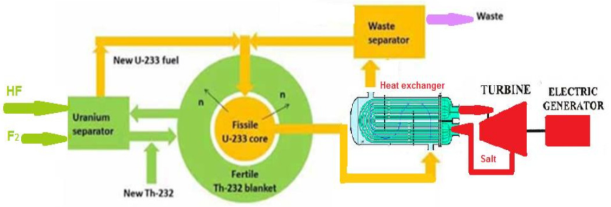

Fig. 2 Schematic representation of the molten salt reactor.

Another advantage is related to the use of Brayton power cycles. Because of the melting points of molten salts (350 °C 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 that 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 (pressurized water reactors) 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 approximately 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 (> 326 ^{o}C); chemical extraction of ^{233}Pa and reintroduction of its decay chain product, ^{233}U.

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 ^{233}U 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. ^{232}Th plus a neutron yields fissile ^{233}U and ^{238}U plus a neutron yields fissile ^{239}Pu. The ^{238}U-^{239}Pu fuel cycle generates large quantities of transuranic actinides. The ^{232}Th-^{233}U fuel cycle generates almost no TRU actinides, because it takes many neutron captures to convert ^{233}U to a transuranic isotope.

One problem associated to the reprocessing of thorium fuels is the presence of ^{232}U, that is extremely radioactive, has a half-life around seventy years and it is dangerous even in small amounts. In one hand, the presence of ^{232}U makes the fabrication of ^{232}Th-^{233}U mixed oxide fuel much more difficult, but, in the other hand, this problem is one of the reasons that make thorium cycles more favorable from the point of view of proliferation risks. But, if the fabrication of oxide fuels is complex due to the presence of ^{232}U 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 assembly fabrication steps employing powder metallurgy techniques. Then, the use of thorium is more advantageous when a homogeneous reactor with liquid fuel is considered, as is the case of the LFTRs.

In July 2010, an industry organization with members such as Toyota, Toshiba and Hitachi, IThEMS (International Thorium & Molten-Salt Technology Inc.)

unveiled their plans to build the world's first commercial Th-MSR (thorium molten-salt reactor) power generator [5]. 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 is that commercially available Thorium Energy will be through their 10 MW mini FUJI (in five years). That will be followed by a larger capacity design called FUJI, delivering 200 MW 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 LWR (light water reactors)—at least 30% lower [6].

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 [7]. 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

3.1 Thorium

The mineral thorium was discovered by Berzelius in 1828. The natural thorium isotope is formed by the ^{232}Th that is a fertile element and can be transmuted in ^{233}U by bombardment with neutrons, as can be observed in 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 ^{233}U 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 LWR, 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 (^{233}U), 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 has 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 50's by Orquima company, in search of various minerals, especially RE (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 99.5% for impregnation of mantles for incandescent gas lamps. The purification process used was solvent extraction in pulsed columns with TBP (try butyl phosphate) and Varsol as diluent. A picture of the purification pilot plant can be observed in Fig. 4 [8]. 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.

3.2 Uranium

The uranium ore was discovered by Klaproth in 1789. The uranium exploration in Brazil began jointly

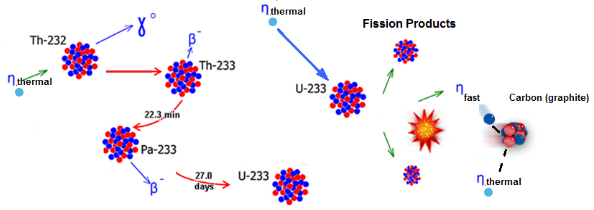

Fig. 3 Nuclear reactions involved in the transmutation of ^{232}Th in ^{233}U.

Fig. 4 Pilot plant production of thorium nitrate.

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 that the obtainment of uranyl nitrate with purity is suitable for nuclear purposes. A view with the purification pilot plant can be observed in Fig. 5. The next step of the process at IPEN was the precipitation of the purified uranyl nitrate as ADU (ammonium diuranate) and its transformation into oxides by calcination. The UO_3 formed was first converted 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 (International Atomic Energy Agency) and France [9].

The know-how of obtaining UF_6 until the fuel assembly was obtained by Brazilian scientists. The main achievement in the nuclear fuel cycle technology 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 [10-12]. The enriched UF_6 was converted to UO_2 employing the AUC (ammonium uranyl carbonate) route. 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' [13]. 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.

Fig. 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, Industrias Nucleares Brasileiras, Comissão Nacional de Energia Nuclear, and IPEN. Unfortunately, due to the changes in the Brazilian nuclear policy in the early 90's, the continuity of these technological developments at IPEN was 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 an effective member of the group.

Nevertheless, Brazil has a long tradition in the thorium technology, from mining of monazite to the obtainment of thorium with purity suitable for nuclear use. Therefore, 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. Besides this, Brazil has very important thorium resources and this should be considered to establish national priorities.

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. With thorium and the breeder of ^{233}U in a reactor operating in the thermal spectrum this problem can be overcame.

The liquid-fluoride thorium reactor design presents several advantages relating to the benefits of thorium as a nuclear fuel. Some benefits that could be pointed are: benefits with regards to nuclear proliferation, since there are not information available about nuclear weapon fabricated from ^{233}U obtained from thorium; LFTRs can be used to consume nuclear weapons material; the use of LFTRs would make possible the reduction in nuclear waste that would be produced in comparison with other reactors design; LFTRs present much better safety features; LFTRs operate at low-pressure, and a strong vessel is not necessary, like in PWRs, for instance; since LFTRs are homogeneous reactors, the cost of fuel elements production is considerately reduced. Some countries have already identified the strategic importance of LFTRs for the future, like China, for instance.

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