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USE OF THORIUM FOR HIGH TEMPERATURE GAS-COOLED REACTORS

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ABSTRACT

The HTGR (High Temperature Gas-cooled Reactor) is a 4th generation nuclear reactor and is fuelled by a mixture of graphite and fuel-bearing microspheres. There are two competitive designs of this reactor type: The German “pebble bed” mode, which is a system that uses spherical fuel elements, containing a graphite-and-fuel mixture coated in a graphite shell; and the American version, whose fuel is loaded into precisely located graphite hexagonal prisms that interlock to create the core of the vessel. In both variants, the coolant consists of helium pressurised. The HTGR system operates most efficiently with the thorium fuel cycle, however, so relatively little development has been carried out in this country on that cycle for HTGRs. In the Nuclear Engineering Centre of IPEN (Instituto de Pesquisas Energéticas e Nucleares), a study group is being formed linked to thorium reactors, whose proposal is to investigate reactors using thorium for 233U production and rejects burning. The present work intends to show the use of thorium in HTGRs, their advantages and disadvantages and its feasibility.

1. INTRODUCTION

The continued research and development of the use of thorium as a fertile fuel material for nuclear reactors is endorsed by several major nuclear organisations on Earth, notably by the IAEA (International Atomic Energy Agency) and the ANS (American Nuclear Society).

Thorium is a potentially valuable energy source since it is about three to four times as abundant in the earth’s crust as uranium and is a widely distributed natural resource, which is readily accessible in many countries. The use of thorium as a fertile fuel material leads to the production of an alternative fissile uranium isotope, uranium-233; coproduction of a highly radioactive isotope, uranium-232, which provides a high radiation barrier to discourage theft and proliferation of spent fuel.

Since thorium is an abundant resource that can potentially be used as a fertile nuclear fuel, it is likely to be an important contributor to the future global nuclear enterprise in several countries. It is, therefore, paramount that the evolving global thorium fuel cycle (including fuel conditioning and recycling operations) incorporate the latest in safeguards and other proliferation-resistant design features so that the thorium fuel cycle complements the uranium fuel cycle and enhances the long-term global sustainability of nuclear energy.

The HTGR is a pretty fuel-versatile kind of reactor, which could be adapted to many fuel cycles, including the Thorium one. That characteristic, allied to thermal efficiency and safety, make the Thorium run HTGR concept an attractive idea.

2. HTGR'S FEATURES

The main features of HTGRs are enhanced safety, high thermal efficiency, economical competitiveness, and proliferation resistance, whose intensity is stronger when associated with the thorium fuel cycle.

2.1. Safety

About safety’s point of view, there are several advantages of HTGRs over conventional water cooled reactors. These water cooled carry a huge concern about the chemical interactions between fuel, moderator, and coolant, which, as wildly known, are potentially dangerous, once that water-zirconium reactions are exothermic at high temperatures and those reactions become autocatalytic. That risk is successfully avoided through the HTGR, due to the fact that Helium, a single phase and inert fluid, is used as coolant. Besides that, the large mass of the graphite moderator provides high heat capacity. Core materials are made of ceramic materials and usable at elevated temperatures.

The enhanced safety of the HTGR is also based on its coated fuel particle design consisting of minute uranium particles coated with layers of carbon and silicon carbide. Coated particles can withstand high internal gas pressure without releasing their fission products to the environment.

Fission product release rates are kept very low during normal operation and off-normal transients as long as the maximum fuel temperature is kept below 1600ºC. The HTGR cores are quite large in size; therefore, their core power density is appreciably low. With their low power density, the HTGR can accommodate decay heat removal passively from the reactor core by means of the large graphite volume without causing any radioactivity release. This is a critical issue in the case of off-normal transients such as the loss-of-coolant, or loss of flow, to keep the coated fuel particles intact by not exceeding their accident fuel temperature limit, typically 1600ºC for a short period of time. The highest normal operating fuel temperature should not be greater than 1250ºC. Fuel failure rates are extremely low below these temperatures and increases rapidly at much higher temperatures. However, accident fuel performance depends on temperature progressions, duration, burnup, fabrication quality, and must be demonstrated by specific fuel irradiation experiments, followed by out-of-reactor accident simulation testing.

The TRISO (tri-isotropic) particles have an overall diameter in the range of 500 to 1000 μm. Each particle contains a spherical fuel kernel (350 to 600 μm diameter) of fissile or fertile fuel materials, usually in the form of uranium dioxide (UO2), plutonium dioxide (PuO2), or an uranium oxycarbide (UCO) mixture. (Fertile thorium compounds, either alone or mixed with uranium or plutonium, can be used as fuel kernel material.) Typical fuel enrichments vary from 8 to 20%, as dictated by power rating and safety considerations. The fuel kernels are then coated with successive layers of pyrocarbon (PyC) and silicon carbide (SiC). First, a low-density PyC buffer coating is applied that provides void volume to accommodate fission gas and attenuates fission product recoils released from the fuel kernel. This layer is surrounded by successive coatings consisting of an inner PyC layer (IPyC), a silicon carbide (SiC) layer and an outer PyC layer (OPyC). The irradiation behaviour of the PyC coatings on either side of the SiC provides prestressing to assist in accommodating internal pressure. The SiC layer is the primary pressure vessel and is an effective barrier to fission product release. The coated particles are over coated with a resinated graphite powder to prevent particle-to-particle contact during either sphere making or compact formation.

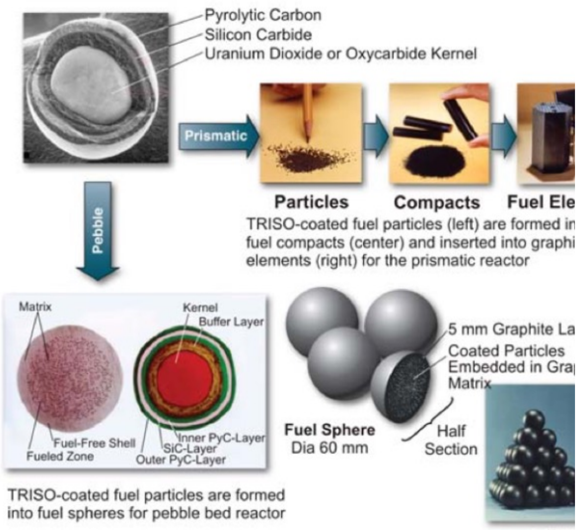

Figure 1: The TRISO particles at Pebble and Prismatic HTGR design

In the prismatic design, the over coated TRISO particles are imbedded within a graphite matrix to form cylindrical compacts. Approximately 3200 of these compacts are inserted into a hexagonal graphite fuel element. In the pebble bed design, over coated TRISO particles are also imbedded in a graphite matrix; however, in this case, in the form of a spherical element with hundreds of thousands of them making up the core.

2.2. Thermal efficiency and economical competitiveness

The HTGR generally presents gas outlet temperatures of 900 to 1000°C. So high temperature in the primary cycle provides the realisation of efficient thermal conversion cycles like the superheated steam cycle and the gas turbine cycle (as applied at the GT-MHR).

Net thermal efficiencies greater then 45% are within reach in some of the designs of High Temperature Gas-cooled Reactors. The high outlet gas temperatures may also be utilised as a thermal heat source in endothermic chemical processes. Examples are in coal chemistry and upgrading of hydrocarbons. Hydrogen production is another promising field for deployment of the HTGR.

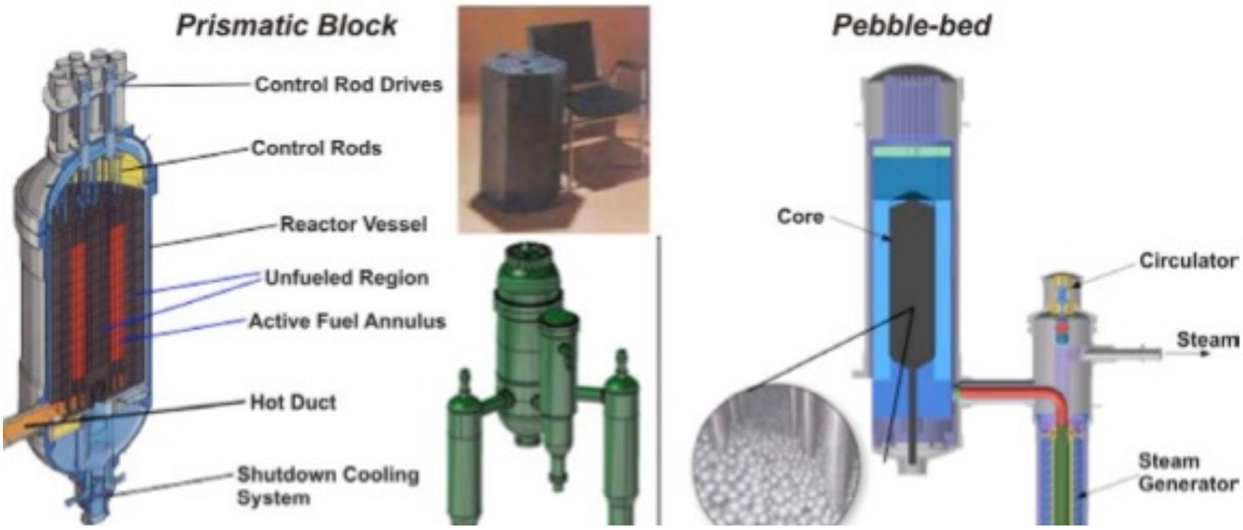

Figure 2: Prismatic and Pebble-bed HTGRs cores

An important difference of pebble bed reactors compared to the prismatic reactor is the capability to do on-line refuelling. Thus, the reactor can be run without having to be shut down for long period for refuelling. This may increase plant capacity factors. Another advantage of on-line refuelling is that the reactor can operate with very little excess reactivity and reduced enrichment.

2.3. HTGR's past achievements

Table 1: Data from some key HTGR around the globe

Experimental HTGRs					
	Peach Bottom (USA)	**Dragon** (UK)	**AVR** (Germany)	**HTTR** (Japan)	**HTR-10** (China)
Operational	1967-74	1968-75	1967-88	1998-xx	2000-xx
Status	safe encl.	safe encl.	Defueled	in operation	in operation
Thermal/electric power [MW_{th}/MW_{el}]	115/40	20/-	46/15	30/-	10/-
Fuel element type	pin	pin	Spherical	pin-in-block	Spherical
Power density [$MW_{th}.m^{-3}$]	8.3	14	2.6	2.5	2
He-inlet/ outlet temperature [°C]	377/750	350/750	270/950	385/850 and 950	250/350/ 700/900
Mean He pressure [MPa]	2.5	2	1	4	3
Enrichment	HEU	HEU/ LEU	HEU/ LEU	LEU	LEU
Fuel	Carbide	Oxide	Carbide/ Oxide	Oxide	Oxide
Coating	BISO	TRISO	BISO/ TRISO	TRISO	TRISO
Pressure vessel	steel	steel	Steel	steel	Steel

Prototype HTGRs		
	Fort St. Vrain (USA)	**THTR** (Germany)
Operational	1976-1989	1986-1989
Status	Decommissioned	safe enclosure
Thermal/electric power [MW_{th}/MW_{el}]	842/330	750/300
Fuel element type	Prismatic	Spherical
Power density [$MW_{th}.m^{-3}$]	6.3	6
He-inlet/-outlet temperature [°C/°C]	405/784	270/750
Mean He pressure [MPa]	4.5	3.9
Steam temperature [°C]	530	530
Electricity production [MWh]	5500	2890
Enrichment	HEU	HEU
Fuel	Carbide	Oxide
Coating	TRISO	BISO
Pressure vessel	PCRV	PCRV

Commercial HTGR Projects					
German designs	**PNP**	**HHT**	**HTR-500**	**HTR-Modul**	**HTR-100**
Thermal/electric power [MW_{th}/MW_{el}]	500/-	1240/500	1250/500	200/80	258/100
Fuel element type	spherical	block/ spherical	spherical	spherical	Spherical
Power density [$MW_{th}m^{-3}$]	4	5,5	7	3	3
He-inlet/-outlet temperature [°C/°C]	300/950	440/850	280/700	250/750	250/740
He pressure [MPa]	3.9	5.0	4.7	5.0	7.0
Steam temperature[°C]	850	-	530	530	530
Enrichment	LEU	LEU	LEU	LEU	LEU
Fuel	Oxide	Oxide	Oxide	Oxide	Oxide
Coating	TRISO	TRISO	TRISO	TRISO	TRISO
Pressure vessel	PCRV	PCRV	PCRV	Steel	Steel
International designs	**MHTGR** (USA)	**VGR-50** (Russia)	**VGM-400** (Russia)	**PBMR** (SA)	**GT/MHR** (USA/Russia)
Thermal/electric power [MW_{th}/MW_{el}]	350/140	136/50	1060/300	400/165	600/285

Fuel element type	prismatic	spherical	spherical	spherical	prismatic
Power density [$MW_{th}.m^{-3}$]	6	?	?	4.8	6.5
He-inlet/-outlet temperature [°C]	319/685	296/810	350/950	500/900	510/850
He pressure [MPa]	9	4	5	9	7
Enrichment	LEU	HEU	LEU	LEU	U/Pu
Fuel	UCO	Oxide	Oxide	Oxide	Oxide
Coating	TRISO	TRISO	TRISO	TRISO	TRISO
Pressure vessel	steel	steel	PCRV	steel	steel

	HTR-PM (China)	**HTR/VHTR [ANTARES]** (France)	**NGNP (VHTR)** (USA)	
Thermal/electric power [MW_{th}/MW_{el}]	2x250/ 200	600/-	600 (max)/-	500/200
Fuel element type	spherical	prismatic	prismatic	spherical
Power density [$MW_{th}.m^{-3}$]	3.215	?	undecided	6.0
He-inlet/-outlet temperature [°C/°C]	250/700	400/1000	-/ 850 to 950	350/ 850 to 950
He pressure [MPa]	7	5	undecided	9
Enrichment	LEU	LEU	LEU	LEU
Fuel	Oxide	UCO or UO_2	UCO	UO_2
Coating	TRISO	TRISO	TRISO	TRISO
Pressure Vessel	Steel	Steel	Steel	Steel

The US–led GIF (Generation IV International Forum) has identified very the HTGRs with helium coolant outlet conceptual temperature of 1000ºC as one of the candidate nuclear energy systems deployable by the year 2025. For this, the reference reactor concept has been a 600 MWt, helium cooled prismatic block fuel of the gas turbine modular helium reactor (GT–MHR) or the pebble fuel of pebble bed modular reactor (PBMR). Thorium based, ZrC coated fuel particles TRISO of oxide, mixed oxide, di–carbide or mixed di–carbides in graphite matrix are strong candidate fuels for this type of reactor.

The HTGRs have considerable adaptability to different fuel cycles without change of active core design and main plant components and offers attractive opportunities of thorium utilisation in combination with enriched uranium and plutonium. The studies of fuel loads on the base of thorium with weapon quality 235U and 233U–Th fuel and also experience of Fort St. Vrain reactor operation being the GT–MHR prototype showed a high effectiveness of these fuel compositions from the point of view of minimisation of fissile isotopes consumption. Thus the operational conditions (ratio of fuel reloading, time between fuel reloading, limitations on an available operative reactivity margin) met the design requirements.

3. THORIUM AS FUEL

3.1. Thorium's features

Natural thorium is not a fissile element, actually it is a fertile one. So, its use as a fuel component requires a breeder reactor. When Thorium-232 is hit by a neutron, it becomes Thorium-233, which, through β decay, becomes Protactinium-233, and, again through β decay, becomes Uranium-233, a very attractive fissile isotope.

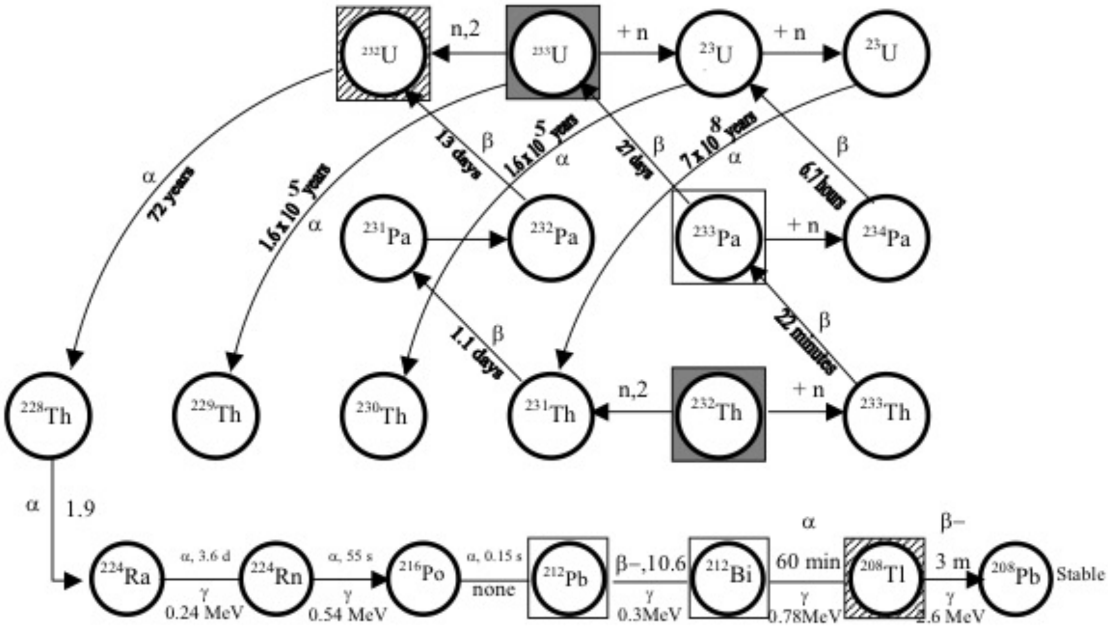

Figure 3: Illustrates the decay chain when a neutron hits the Th-232

Considering that the natural reserves of U-235 will no longer be enough to meet the global nuclear fuel demand at long term, the breeder reactors are the most feasible alternative. Among these reactors' technologies, it is remarkable that the Th-232 is a better fertile material than U-238 in thermal reactors because of the three times higher thermal neutron absorption cross-section of Th-232 (7.4 barns) as compared to U-238 (2.7 barns). Thus, conversion of Th-232 to U-233 is more efficient than that of U-238 to Pu-239 in thermal neutron spectrum though the resonance integral of Th-233 is one–third of that of U-238.For each successful U-233 nuclei fission, the number of neutrons liberated per neutron absorbed is about 2.7 over a wide range of thermal neutron spectrum, unlike U-235 and Pu-239. Therefore, if one liberated neutron reach other Th-232, and the other liberated neutron reach another U-233, the breeder condition for the reactor is achieved.

The advantages of the "Thorium-Uranium" cycle go beyond the Th-232 better "fertility".The U-233 features fissile characteristics are more attractive than the conventional ones due to the following reasons: the capture cross-section of U-233 is much smaller (46 barns) than the U-235 (101 barns) and Pu-239 (271 barns) for thermal neutrons, while the fission cross-section of all the three fissile isotopes is of the same order (525, 577 and 742 barns for U-233, U-235 and Pu-239 respectively). Thus, non-fissile absorption leading to higher isotopes (U-234, U-236 and Pu-240 respectively) with higher absorption cross-sections is much less probable. This makes recycling of U-233 less of a problem from reactivity point of view compared to plutonium burned in thorium systems.

Table 2: Data about some key isotopes

Atomic Number	90	90	92	92	92
Isotope	Th-232	Th-233	U-233	U-235	U-238
Abundance, atom %	100	0	0	0,72	99,27
Half-Life	1,41x 10^{10} years	22,1 months	1,62 x 10^{5} years	7,13 x 10^{5} years	4,51 x 10^{9} years
Total Cross Section	7,4 barns	1500 barns	573,1 barns	678,2 barns	2,73 barns
Fission Cross Section	X	15 barns	524,5 barns	577,1 barns	X
Radiative Capture Cross Section	7,4 barns	1485 barns	48,6 barns	101,1 barns	2,73 barns
Capture-to-Fission Ratio	X	99	0,093	0,175	X

Th–based fuels and fuel cycles have intrinsic proliferation-resistance due to the formation of U-232 via (n,2n) reactions with Th-232, Pa-233 and U-233. The half-life of U-232 is only 73.6 years and the daughter products have very short half-life and some like Bi-212 and Tl-208 emit strong gamma radiations. From the same consideration, U-232 could be utilised as an attractive carrier of highly enriched uranium (HEU) and weapons grade plutonium (WPu) to avoid their proliferation for non-peaceful purposes.

3.2 Data from reactors which used to run through thorium

In the past, thorium-based fuels have been successfully utilised in HTGRs in Germany, United States, Japan and The Russian Federation. The fuels were in the form of TRISO coated particles of ThO2, (Th,U)O2, ThC2 and (Th,U)C2 with a fuel kernel of diameter between 350 and 500 µm with multilayer carbon and silicon carbide coatings (nearly 100 µm buffer carbon layer on fuel kernel followed by inner and outer pyrolitic carbon coatings of nearly 40 µm with 35 µm SiC layer in between). In Germany, two Pebble Bed HTGRs, namely AVR 15 MW(e) and THTR 300 MW(e), successfully operated till the late 1980s after which they were terminated. Coated fuel particles of mixed uranium thorium oxide and di–

carbide, embedded in graphite, were also employed in the form of prismatic blocks in the helium-cooled HTGRs of USA, namely Peach Bottom (40 MW(e)) and Fort St. Vrain (330 MW(e)). The HTGR in UK, namely the Dragon reactor, has also used TRISO particular of mixed thorium uranium oxide and di–carbide in graphite matrix.

Table 3 : Past use of Thorium in nuclear reactors

Name and Country	Type	Power	Fuel	Operation period
AVR, Germany	HTGR Experimental (Pebble bed reactor)	15 MW(e)	Th+^{235}U Driver Fuel, Coated fuel particles Oxide & dicarbides	1967 – 1988
THTR, Germany	HTGR Power (Pebble Type)	300 MW(e)	Th+^{235}U, Driver Fuel, Coated fuel particles Oxide & dicarbides	1985 - 1989
Lingen, Germany	BWR Irradiation-testing	60 MW(e)	Test Fuel $(Th,Pu)O_2$ pellets	Terminated in 1973
Dragon, UK OECD-Euratom also Sweden, Norway & Switzerland	HTGR Experimental (Pin-in-Block Design)	20 MWt	Th+^{235}U Driver Fuel, Coated fuel particles Dicarbides	1966 - 1973
Peach Bottom, USA	HTGR Experimental (Prismatic Block)	40 MW(e)	Th+^{235}U Driver Fuel, Coated fuel particles Oxide & Dicarbides	1966 – 1972
Fort St Vrain, USA	HTGR Power (Prismatic Block)	330 MW(e)	Th+^{235}U Driver Fuel, Coated fuel particles Dicarbide	1976 - 1989
MSRE ORNL, USA	MSBR	7.5 MWt	^{233}U Molten Fluorides	1964 - 1969
Borax IV & Elk River Reactors, USA	BWRs (Pin Assemblies)	2.4 MW(e) 24 MW(e)	Th+^{235}U Driver Fuel Oxide Pellets	1963 - 1968
Shippingport & Indian Point, USA	LWBR PWR (Pin Assemblies)	100 MW(e) 285 MW(e)	Th+^{233}U Driver Fuel, Oxide Pellets	1977 – 1982 1962 - 1980
SUSPOP/KSTR KEMA, Netherlands	Aqueous Homogenous Suspension (Pin Assemblies)	1 MWt	Th+ HEU Oxide Pellets	1974 - 1977
NRU & NRX, Canada	MTR (Pin Assemblies)		Th+^{235}U Test Fuel	Irradiation–testing of few fuel elements
KAMINI, CIRUS, & DHRUVA, India	MTR Thermal	30 kWt 40 MWt 100 MWt	Al-^{233}U Driver Fuel ‘J’ rod of Th & ThO_2 ‘J’ rod of ThO_2	All three research reactors in operation
KAPS 1&2, KGS 1&2, RAPS 2,3&4, India	PHWR (Pin Assemblies)	220 MW(e)	ThO_2 Pellets For neutron flux flattening of initial core after start-up	Continuing in all new PHWRs
FBTR, India	LMFBR (Pin Assemblies)	40 MWt	ThO_2 blanket	In operation

4. CONCLUSION

It's wildly known that the thermonuclear energy sources are highly trustworthy and hold great power generation, whose discard would deliver failure to meet the world demand for energy. Therefore, it's important to assure a sustainable future to the nuclear energy. It includes to seek and improve fissile materials management strategies.

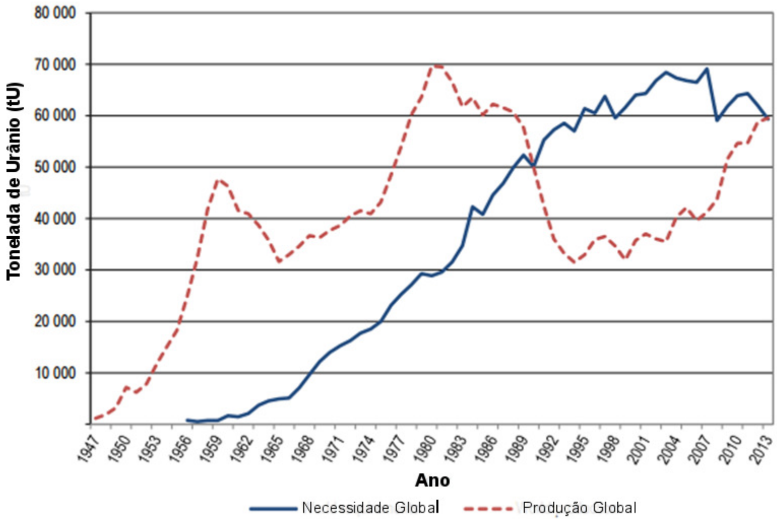

Figure 4: Features tonnes of uranium consumption (blue) and production (red) per annum

As a feasible answer to that quest, Thorium presents itself. Hence, prudence says that the study on Thorium fuel cycle, as well as reactors' design capable to run through Th fuel, must be endorsed.

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