In Memoriam to: Prof. Kazuo Furukawa "Champion for the Th-MSR" who passed away 14 Dec. 2012.

An Alternative Source for Venezuelan Nuclear Energy Production: The Thorium Molten Salt Reactor

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Abstract

The Thorium Molten Salt Reactor (MSR), as the FUJI reactor project by Furukawa and collaborators, is considered as one of the generation IV six innovative concepts for alternative nuclear power plants. The thorium cycle for its several advantages should be the future energy source for developing countries. Recently Venezuela signed an agreement with the Russian Federation to install in the near future a PWR-type Nuclear Power Plant, since is the more used and standard technology. However, in the near future the Country should orient his nuclear program toward the thorium cycle MSR due to simplicity, safety and non proliferation properties. This technology is suitable for Venezuela having proved thorium resources and experience with molten salt at industrial scale. We report theoretical calculations for a sub-critical assembly dealing with energy transfer and the effect on the neutron balance of delayed neutrons.

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1 Introduction

The energy consumption is a direct evidence of any country productivity; the GDP *per capita* is often assumed as an indicator of a country's living standard. Since the tendency is to improve it, necessarily the energy demand will increase and may double in the next 30 years. The development and introduction of new technologies to tap wind and solar renewable energy is currently on the agenda. However, they are low in energy density, irregular in output and so far it seems to be uneconomical and impractical for large industrial power plants. There is evidence that the nuclear option is still a strong candidate to be employed and increasingly technological interest arises in this field in spite of the opposition of some from the general public. Today 436 Nuclear Power Plant (NPP) are in operation and already 63 new installations are in construction in spite of all the policy makers declarations that in few leading countries the nuclear program halted. To mention an example Rosatom (Russian Federation) is planning this year to connect to the electric grid a 1.2GW Novovoronezh-2-1 type VVER-NPP and further 16 units are planned to enter operation by 2020; among them, some of a more advanced GenerationIV⁺ (GEN-IV⁺) reactors are considered.

So far all commercial NNP are based on the uranium cycle, however it is not the only option. Already several initiatives have started toward new concepts e.g. employing the thorium fuel cycle. Initiatives toward development of nuclear plants employing this cycle rose during the past years in several countries motivated for its inherent technical advantages¹. For instance its superior performance over PWR or BWR (most often commercially employed NPP) was demonstrated during the Thorium Molten Salt Reactor Experiment (MSRE) constructed and successfully operated at Oak

Ridge National Laboratory (ORNL), USA during the decade 1960's ². The 8-MW(t)-MSR had a ⁷LiF-BeF₂ salt nd graphite moderator; this is a material that is compatible with the corrosive fluoride salt having a stable performance during neutron irradiation³. The mentioned research reactor prototype was a good demonstration assembly toward safety operation, neutron balance due to a removal system of xenon and krypton from the fluid fuel, and the employment of different fissile materials such as ²³⁵U, ²³³U, and plutonium. This unit operated successfully for 13,000 equivalent full-power hours between 1965 and 1968. Later on the ORNL-MSR design was further development e.g. the FUJI reactor project by Furukawa and collaborators (1980s – 2000s)⁴. A concept scheme of a MSR-NPP is given in figure 1.



Fig.1: Concept scheme of a MSR-NPP adapted from Forsberg 5. Main features are described in the text.

The main features of the MSR, scheme of Figure 1, consist in having a graphite reactor core crossed by the molten salt fluid followed by an off-gas system at the core out-let and a heat exchanger; after the primary salt pump, an actively cooled freeze plug being a safety valve connecting the primary circuit to dump tanks. The secondary circuit is a conventional one, in which hot molten salt and multy-reheat helium Brayton cycle is considered for its inherent thermodynamical advantages. In fact, since it employs helium (or nitrogen) gas, this cycle efficiently convert high-temperature heat to mechanical energy; in opposition to the steam cycle where the maximum value set on temperature is ~550°C that in turns imposes a limiting mechanical conversion capability. The development of the technology related to high-temperature Brayton cycles, allows higher conversion efficiency and therefore provide higher electrical energy output compared to any other thermodynamical cycle found in commercial NPP.

Other conceptual NPP schemes are under review and several "generation IV" concepts already have been proposed. For instance, Mazzini et al.⁶, started recently the EU-PUMA project based on Th-Pu fuel cycles working in pebble-bed High Temperature Reactor (HTR) concept. The outcome has pointed out that the thorium breeder, due to its liquid fuel characteristics, offer a better and less risky reprocessing operation mainly due to the fact that actinides could be returned to the reactor core. In this way fission fragments are modified and radio-toxicity reduced accordingly. Therefore these highly radioactive waste products should not be removed from the reactor by costly processes and transported

to reprocessing centers; consequently, further advantages exist from the resulting reduction of risk and accidents.

A few others design concepts are worth mentioning briefly: one is the TMSR-NM (nonmoderated thorium molten salt reactor) being so far the GEN-IV reactor with the highest energy density. This could be considered as an extreme industrial NPP concept being a combination of the thorium-based fuel cycle with accelerator driven systems (ADS-MSR known as the Rubbia proposal⁷). The other method is the hybrid fusion driven system concept proposed by H. Bethe (1970)⁸ related to two step power generation i.e. nuclear fusion to produce the neutron field and fission that will take advantage of it⁹. The last one to which we refer is the modified Liquid Metal Fast Reactor, known as the traveling-wave reactor proposed in the 1950s; that could breed its own fuel inside the reactor core, consume wastes from LWRs and run on depleted Uranium as a fuel¹⁰. In table 1, we give an overview of some MSR elements to show the broad on going research interest in MSR and fuel compound.

Reactor type	Neutron Spectrum	Application	Solute	Solvent
MSR-Breeder	Thermal	Fuel	7Li-BeF2	ThF4-UF4
	Fast	Fuel	7Li	ThF4-PuF4
	TNM	Secondary coolant	NaF-NaBF4	
MSR-Burner	Fast	Fuel	LiF-NaF	LiF-(NaF)-AnF4-AnF3
MSR-Burner	Fast	Fuel	LiF-(NaF)-BeF2	LiF-(NaF)-BeF2-AnF4-AnF3
MSR-Burner	Fast	Fuel	LiF-NaF-ThF4	
MS-Advanced High-Temp. Reactor	Thermal	Primary coolant	7LiF-BeF2	

Tab.1: Fuel and coolant salts composition for different thorium cycle reactor assembly

The renewed interest in the systems mentioned is related not only to new development in material science, surface treatment technologies, viability and advantages in energy economy but also to other aspects that justify the MSR technology as follows:

i.- Th-MSR has been proposed as one of the six innovative concepts by the prestigious "GEN-IV International Forum", it was selected on the basis of sustainability, economics, proliferation resistance and physical protection; ii.- the proven possibility to increase nuclear fuel resources by breeding ²³³U from thorium, *de facto* extending nuclear energy lifespan resources by two or more orders of magnitude; iii.- fuel utilization improvement; iv.- enrichment requirements are reduced significantly; v.- the build-up of Pu is not a primary objective; vi.- existing Pu stockpiles and long lived radio-toxic isotopes or other radioactive waste could be incinerated lowering considerably the environmental burden. These, among other observations, support the MSR technology to be considered one of the best ways to continue and spread nuclear energy generation, not only for a future Venezuelan nuclear program, but take the advantages of Th-MSR for emerging and developing countries in general.

In recent years the Venezuelan Government signed an agreement with the Russian Federation (RF) to develop nuclear activities related to the installation of a research reactor and electric

generation¹¹. It was announced officially and given diffusion by the media that Rosatom (RF) should install a NPP, probably of 1200MWe type Novovoronezh (VVER-1200). However, due to different causes including the Fukushima accident and its impact in the public opinion, it is highly probably that the program will be delayed at least few years.

Also, in the 1970s the Venezuelan Government received proposals from U.K. to install relatively small NPP to cope with the requirement of large amount of high-temperature heat by industrial plant of Petroleos de Venezuela SA, for example at Paraguana refinery complex producing 956,000 bbl/d (i.e. $152,000 \text{ m}^3/\text{d}$), or the Orinoco Basin oil extraction fields including areas for tertiary oil recovery.

On the other hand, the advantages of thorium fuel being more abundant in the Earth crust compared to natural U suggests that it is plentiful also because worldwide supply includes also byproduct of rare earth element production. Natural Th is almost 100% ²³²Th, so that costly fuel enrichment is avoided. However, fissile material must be added to the fuel to start a Th-MSR, e.g. weapons grade ²³⁵U or ²³⁹Pu from stockpiles and in this case a bonus of weapons dismantling could be provided at low cost.

Additionally the Th-Molten Salt Reactor will help to ensure the maintenance of nuclear weapons free Latin American-continent and so avoiding foreign powers interference with national nuclear programs. It will help also that most countries achieve energy independence and for several of them to make use of their own indigenous sources of thorium e.g. Venezuela occupying the sixth place for its Th resource at Cerro Impacto (World Nuclear org) and Brazil with proven large deposits, just to mention a few.

2 Preliminary studies on reactor dynamics

2.1 The sub-critical training assembly

Preliminary studies have been performed with aim to master computer codes related to the dynamical behavior of small assembly for a MSR structure. Yamamoto et al.,¹³ studied small MSR by steady-state analysis estimating the effects of fuel flow.

Based on their results, zero power structure for student training at the USB, was started as a project to assembly a small sub-critical system. This is designed to hold solid fuel having a salt composition with Th+Unat. The fuel, being natural element, is available on the market; our laboratory already received financial support for thorium rich mineral processing; it is planned to produce Th-salt fuel to feed the projected sub-critical assembly. It is devised to employ a radioisotope neutron source (either a 252 Cf, a purpose made Ra-Be, Am-Be source already being used for on-going applied projects). The possibility exists of employing a modified ion implanter, operating it with a deuteron beam on a titanium target to induce low energy D-D reaction and provide neutrons to experiment with a subcritical assembly. The produced neutrons are relatively of low energy ($E_n = 2.5 \text{ MeV}$) that by diffusion in a graphite assembly provides a facility for a broad reactor simulation and neutron related experiments. The sub-critical assembly main material is the moderator being a pirolytic graphite pile assembled with blocks of 20x20x60 cm³ (in the past employed as reflector to lower neutron leakage in research reactors). This material was donated by the Atomic Energy Research Institute AEKI of Budapest, Hungary, in the frame of collaboration with USB and the International Atomic Energy Agency (Vienna, Austria) from which originated the project VEN/8/014. Details of the sub critical assembly including the two strata biological shielding, can be seen in the schematic lay-out of figure 2.



Fig.2: Schematic lay out of zero power graphite assembly (a) has a radioisotope source (252Cf) that is positionable at different places indicated by the accessing holes (b). These are also suitable for fuel loading and neutron or gamma measuring devices. This assembly has an external biological paraffin shield (c) that covers the neutron absorber slabs Li-Polyethylene (d). For safety purpose against possible displacement due to an earthquake shake, a tightly holding non-ferrous structure (e) is included and it holds on place also the biological external shield. Spacing between loading holes is 20cm. Figure not to scale.

Neutron diffusion equation with two or more energy groups is often employed to determine reactor response time. The reactivity depends on the neutrons released from prompt fission and those from decay processes of precursors. The latter are delayed neutrons and without them the reactor could not reach criticality. Meaning that are important and have to be taken into account in the estimation of the neutron balance. Since fissions occur in a flowing media¹⁵, precursors may release delayed neutrons outside the moderating assembly and being lost without inducing new fissions. This means that the reactor volume (sub-critical assembly in our case. The number of neutron groups depends on the precursor life time and it is known that almost 240 precursors take part in the balance equation given below as follows:

$$\partial C(z,t) / \partial t + \partial (vC(z,t)) / \partial z = Q(z,t) - \lambda C(z,t), \partial t$$
(1)

where C is the precursors concentration, v is the fuel cycle speed, Q is the source function, and λ is the decay constant for the given group. It is common to consider six groups to determine neutron balance with fuel speed. Delayed neutron groups kinetic equation can be solved numerically with MATLAB as suggested by F. Reisch¹⁴. Following that procedure we determined the time behavior at zero power operation under the condition that fuel and moderator temperature have negligible increment. We obtained some indicative results that allow us to establish from the observable graphics (calculations are performed for zero power operation):

- the reactivity is changing as shown in figure 3



- the characteristics temperature change of the fuel and moderator for zero power operation, is given in Figure $4\,$



Fig.4: Temperature variation for 233U-fuel and moderator calculated employing MATLAB program. From these curves we learn fuel's thermal and neutron balance time constant for graphite moderator.

2.2 Thermal and Hydro dynamical effect on reactivity

As an excise we considered the case of a small size nuclear reactor where heath production is not neglected. Its transference to the moderator before reaching the steady state and its time variation with fluid speed is given by the following expression:

$$\partial (\rho h) / \partial t + \partial / \partial z (\rho v h) = Q$$
 (2)

where: ρ , h, v parameters indicate fuel density, enthalpy, and velocity respectively. The heath Q represent the sum of the following terms $Q_{fission} + Q_{delay} + Q_{graphite}$ i.e. $Q_{fission}$, due to fissile material in the fuel, the decay heat Q_{delay} , and the heat exchange between graphite and salt $Q_{graphite}$. The latter represents the heat release and its diffusion in the graphite pile including that it will be exchanged between graphite and fuel. The method of effective heat transfer coefficients determines the temperature gradient in the moderator. The ANSYS-CFX, computer code was employed for a larger assembly than that suggested for laboratory training. Results are given in figure 5. For the simulation of the reactor core, a duct of OD= 0,15m and 4,3m long was assumed and a fuel flow rate of 1m/s. Temperature of the salt was modeled by setting an energy balance considering heat production from delayed neutron in the salt, heat transfer through duct walls and the thermal capacity of the salt.



Fig.5: Delayed neutron number with varying flow speed. Segmented line indicates the moderator edge.

As the fuel speeds up the number of delayed neutrons are emitted increasingly out side of the reactor core. Independently of the speed values (limited by the precursor decay constant) always some delayed neutron will be emitted to induce further fission before leaving the core. However it is critical to maintain the delayed neutrons in a window of values to obtain fission rate at given values. For instance delayed neutron precursors changing the transit time from 8.13 s to 60 s induces a variation for neutron multiplication factor by about $0.03\%\Delta k/k$. That means that the neutron balance can be modified by changing the fuel flow velocity (see ref.: ORNL-LR-Dwg.75653 pag. 15).

Figure 6 shows heat transfer interactions at duct walls (into and from the graphite moderator). Figures 7 and 8 show temperature distribution over a plane at duct center and temperature evolution over duct axis respectively. As delayed neutron production varies with speed, so is the heat production term affecting the salt's energy balance and the temperature distribution inside the duct.



Fig.6: Heat transfer rate distribution along fuel housing tube axis.



Fig.7: Temperature variation over a plane located at tube middle longitudinal section.



Fig.8: Temperature variation along tube axis.

3 Conclusions

The future of energy production should rely on nuclear fission since other sources do not provide comparably high density energy within the present known technology. Based on reactor experiences of the early 1970s, a 1000 MW(e) MSR using the thorium cycle is at reach. In spite of low development for several decades, the Th-MSRs are now being positively reexamined suggesting that energy independence following this technology is not only possible but affordable with low risk due to the inherently advantages of thorium as fuel. Several countries have initiated Research and Development with molten salt assembly and a project is already at its final design stage as it is the miniFUJI-MSR. This could be built in a relatively short time scale to produce: electricity, space heating and desalinate water. We suggest that the nuclear programs of developing or even emerging countries should include the possibility to follow the MSR technology for electric energy generation. The first step is training young professionals and promoting further technological solution still required by MSR. Preliminary studies recently initiated at the USB, are encouraging and we are setting up a sub-critical assembly first a small one to be enlarged up to 120x120x180 cm³. To gain experience we have studied the heat transfer form fuel to moderator. The overall experience this induce us to continue promoting MSR with the aim to convince policy makers that the road to follow on short terms, is to accept the nuclear technology to satisfy future energy requirements.

Venezuela has an interesting potential in this way since we have important Thorium deposits and also experience with molten salt at industrial scale, specifically in the production of aluminum, and this experience can be transferred.

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