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PERSPECTIVES OF THE THORIUM FUEL CYCLE

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1. WHY THORIUM ? A BRIEF HISTORICAL INSIGHT (N° 1-3)

We can consider 3 development periods in the past nuclear energy history, at first in the USA:

1.1.1945-1958:
the follow-ups of the Manahattan Project, at the leading US laboratories, Brookhaven, Oak Ridge and Los Alamos : U-233 bred from thorium is considered as a potential weapon of gun-type model, easier to manufacture than plutonium weapons. About 55 kg U-233 were available in 1958.

1.2.1955-1975:
the Atoms for Peace Program unfurls a flurry of projects initiated at first under cooperation of USAEC and US industry. Postwar optimism reigns, nuclear energy is seen as going to have a formidable development, in view of the massive needs of energy in the world for the coming decades.

Thorium, which is deemed to be available in more abundance than N° 4 uranium, will play an important role, as can be seen from INFCE's predictions in 1980. About 1500 kg U-233 are separated in (N° 5) the USA during that period, from 900 tonnes thorium. Many reactor prototypes are built and operated as we shall see. Thorium extraction plants are built in many countries. US, Germany, France have each separated about 2000 tonnes of thorium, part of it is still available.

1.3.1985-today.
Following the anti-reprocessing policies of Presidents Ford and Carter (1974, 1975) the success of the water reactors, especially light water reactors, less quick development of nuclear energy is anticipated and, for other causes analyzed further, disinterest for thorium crops up and this line is progressively abandoned. India (N° 6), however, steadily continues on the plans set at the time by late Dr. Bhabha: energy self independence for 1 billion inhabitants has to come by nuclear energy principally, but the uranium resources in India (50 000 t) are not sufficient, even with aid of breeder reactors, and the large thorium deposits (360 000 t) in India will permit a durable development.

1.4 All predictions (N° 7) today coincide to confirm for the next 30 years the world energy demand foreseen at the time of INFCE in 1980. We are also confronted with the problem of greenhouse effect.

These challenges (N° 8) will foster nuclear energy again and in the longer term thorium will have its place.

2. THORIUM AS A NUCLEAR FUEL(N° 9)

2.1. PARALLEL BETWEEN URANIUM AND THORIUM (N° 10)

Generally speaking, we observe a striking parallel between natural uranium containing 99.3 % U-238, and thorium almost exclusively composed of Th-232. We can see that the fertile isotopes are U-238 and Th-232, and that the fissile isotopes are U-235, 0.7 % of natural uranium present in nature and the artificial fissile isotopes comparable to U-235 which are Pu-239 for uranium and U-233 for thorium. We note in passing that U-235 is our sole natural nuclear match available, and that we would be well inspired to use it with parcimony.

2.2. ADVANTAGES OF THE THORIUM CYCLE

- (N° 11) U-233 bred from thorium is, seen from a neutronic standpoint at least, the best of the 3 nuclear fuels, U-235, Pu-239, U-233, and this for all neutron energies practically envisaged for power reactors, either thermal ones, or epithermal, or fast ones. As a matter of fact, the eta ratio (*) of neutron yield per fission, to neutrons absorbed, is higher to that of U-235 or of Pu-239. This means that U-233 will be a good fuel in any reactor type.

- (N° 12) Moreover, from the respective position of uranium and thorium in the Mendeleiev's table, the long-lived minor actinides resulting from fission are in much lower quantity with the thorium cycle, especially compared with the plutonium cycle. This ecological advantage is an important argument brought forward these days.

- Finally, quadrivalent thorium and its compounds are very stable and among the highest known refractories. Thorium oxide ThO2 melts around 3300°C (UO2 : 2700-2800°C). This stability authorizes high burn-ups and high temperatures. It does complicate somewhat, however, the chemical treatments for the preparation of thorium compounds or their dissolution for reprocessing.

Moreover, U-233 also keeps its good neutronic properties with high temperatures, better than either U-235 or Pu-239. These properties have led naturally to recommend the thorium cycle for high temperature reactors.

2.3. DRAWBACKS OF THE THORIUM CYCLE (N° 13)

- The first remark is that a reactor fuelled with thorium only will not diverge. Thorium needs a "match" which, today, could only be U-235 or plutonium. Here is indeed an excellent way to use up the excess plutonium stocks on the market these days.

- It appears from what precedes that sooner or later U-233 formed should be separated to be incorporated in efficient nuclear fuel elements. This means that reprocessing is an integral part of a sustainable thorium fuel cycle.

- In a reactor, Th-232 by neutron absorption produces first Th-233 and Pa-233 which has a 27-day half-life, to produce U-233 :

Th-232 + n ----> Th-233 -(22m)--> Pa-233 -(27d)--> U-233 (1.5x10^6y)

This rather long half-life of Pa-233 results in a reactivity surge after reactor shutdown due to U-233 production, and this must be taken into account.

- We have mentioned that ThO_2 dissolution is not as easy as that of UO_2, but this problem can be overcome with a buffered fluoride addition in the dissolver solution.

- (N° 14) Finally, one of the principal drawbacks of the thorium cycle today is the presence of hard gamma emitters (2 to 2.6 MeV) among the descendents of the thorium-232 isotope, and especially of U-232, an alpha emitter of 72 years half-life which is always present along U-233 at concentrations ranging from some tenths to some hundreds ppm. This obliges to manufacture U-233 based fuels completely remotely in gamma-shielded environment, a very expensive technique (N° 15) which only starts to be mastered with MOX UO_2-PuO_2 fuel elements fabrication.

3. PREINDUSTRIAL DEVELOPMENTS (N° 16)

3.1. MANY INTERESTING PROJECTS INVOLVING INDUSTRY

It is remarkable that, starting around 1955-1960, a great many industrial prototypes based on thorium were built in parallel with those based on uranium, involving main industrial companies with the scientific knowledge of the research laboratories, among which we should single out Oak Ridge in the US. General Atomics, Babcock and Wilcox, Allis Chalmers, Westinghouse and others in the USA have taken an active role in this industrial development. Germany, the UK, France, Russia have followed suit. Today Japan and China continue, the case of India being quite special.

3.2. HIGH TEMPERATURE REACTORS

The most remarkable applications have been in view of developing high temperature reactors aiming at 700-800°C in the cooling fluid, helium, selected for its chemical inertness and good heat conductivity properties. Helium pressure is about 30-50 kg/cm². Experience with the former gas-cooled graphite reactors was readily available to start with.

High temperatures were sought to : a) use conventional turbogenerating plants known with flame boilers, b) obtain good thermodynamic yields, c) aim at chemical syntheses like ammonia, hydrogen, etc...)

The fuel elements (N° 17) of these reactors are small pellets of about 1 mm diameter, of enriched uranium/thorium carbide coated with tight pyrolitic carbon layers (BISO type), sometimes a silicon carbide overcoat (TRISO type), which will keep the fission gases in and the recoil fission products inside too. These particles are embedded into amorphous graphite into what is called "compacts".

Fabrication of these "compacts" is an expensive and lenghty process. (N° 18) Could it possibly be streamlined and remotized more easily than the "traditional" fuel element fabrication ? This is an important question.

Note that similar HTGR fuel elements can also be made out of UO_2/ThO_2 microspheres, and in view of the difficulties encountered with carbides ulterior reprocessing, utilization of oxides is also something to examine in detail.

The fuel elements which have been used are of two types (N° 19): hexagonal vertical graphite fuel elements, or graphite balls of 60 mm diameter which flow slowly down the reactor core.

3.3. EXPERIMENTAL H.T. REACTORS

The first prototypes of a few megawatts have all run very regularly, which has led their designers to count on fruitful developments :

- PEACH BOTTOM, (40 MW) of General Atomics, from 1966 until 1972.

- The DRAGON experimental reactor (20 MW) (1966-1973) which was built and operated successfully as a cooperation OECD- EURATOM venture in England, involving also Sweden, Norway, Switzerland, an excellent example of international cooperation.(N°20) Dragon tested different types of fuel elements, some with thorium, some without. All of them, however, somewhat complex. It produced temperatures higher than 1000°C butthere the graphite had a tendency to shrink. Burn-ups of 100 000 MWd/t were demonstrated.

- The German experimental pebble-bed AVR in Jülich built with Siemens and operated between 1967 and 1969, has been a very remarkable (N° 21) and quite exceptional reactor due to its revolutionary design. Its core contained 80 000 pebbles; the maximum power being 2.4 kW for a ball at 1350°C maximum temperature. The residence time was 2 to 6 years in the reactor and the burn-ups reached 100 000 MWd/t.

Operation was very stable with passive features, and an experiment was even performed to introduce water in the core, simulating a heat exchanger tube rupture, without damage or accident.

3.4. INDUSTRIAL PROTOTYPE H.T. REACTORS

Happy with these results, General Atomics and Siemens have (N° 22) extrapolated their designs of Peach Bottom and AVR to build power reactors of 300 MWe, respectively Fort St Vrain (1976-1989) and THTR (1985-1989) at Schmehausen. This last one should have been followed by a more advanced MODUL concept which was not built.

Unfortunately, these industrial prototypes have been too quickly extrapolated and have suffered costly technological incidents, coming at a time when nuclear industry was prey to ecologists, supported by coal and petroleum industry.

It is important to mention that these technical failures do not affect (N° 23) the nuclear principles of these reactors which can reach very high burn-ups and which could be used as "converters" to generate supplementary fissile material.

It is worthwhile to note that General Atomics in the eighties had clocked nearly 10 orders for large 1000 MWe High Temperature Reactors (that the new mother company Gulf asked to cancel). It is true, however, that the fabrication of fuels for high temperature reactors presents complications and high costs, especially if thorium is involved, and that reprocessing of these otherwise excellent fuels is not solved satisfactorily for the time being.

(N° 24) JAERI in Japan finishes the construction of its HTTR prototype, something like an advanced 30 MWe Dragon, which is due to diverge at the end of this decade. The Chinese and Russia have teams working on this type of reactor, the latter to burn their weapons' plutonium. Netherlands also works on a small HTR project.

3.5. OTHER REACTOR TYPES (N° 25)

India goes on with its Heavy Reactor programme, also using some thorium fuel elements to flatten the flux in the core. This permits to extract U-233, and a small experimental MTR-type reactor named KAMINI of 30 kW using light water and Al-clad U-233 oxide fuel is due to diverge at Kalpakkam in one or two years from now.

Prototypes of light-water reactors using thorium have been built and operated in the USA :

- ELK RIVER, a 24 MWe BWR (1963-1968), built by Allis- Chalmers(N° 26), using "classical" U-Th fuel elements clad in zircalloy. This small reactor gave entire satisfaction but its limited power caused its closure. A cooperation project involving Elk River fuel elements was started between Italian CNEN and Allis Chalmers at Trisaïa in the 1970s.

- INDIAN POINT, a 285 MWe PWR (1962-1980) has also given good results.

- An interesting case is that of the ancestor of all our PWRs, the SHIPPINGPORT reactor (100 MWe), started in 1957, which today has been entirely dismantled. At the end of the 60s, USAEC (N°27) has used it as test bench for a thermal breeder with thermal or epithermal neutrons using special U-233 hexagonal fuel elements in a so-called "feed-blanket" array, whereby the more enriched "seed" element penetrates into the less enriched "blanket element", as a sort of control rod. This particular arrangement has permitted to demonstrate that U-233 production was actually slightly higher than that burnt, at the cost, however, of some complication of the fuel elements.

- India has a program of building some 500 MWe fast breeder sodium-cooled reactors which will use thorium in part of the blanket. Engineering loops are under demonstration.

- EC JRC at ISPRA has studied a thorium version of the ORGEL reactor. It is clear that any type of reactor can be run using thorium, as was demonstrated by Siemens in collaboration with Brazil in the 1980s.

- A small "homogeneous" slurry reactor of 1 MWt using UO2/ThO2 microparticles, named SUSPOP or KSTR has been operated by KEMA in Holland between 1974 and 1977.

ORNL (N° 28) in USA has operated during 6 years (1964-1969) the Molten Salt Reactor Experiment, (MSRE), of 7.5 MWth, a truly homogeneous reactor using molten fluorides, an advanced concept which, at least theoretically, presents many advantages : excellent neutronics, inherent safety, breeding capabililities, in line venting of fission gases, in-line purification by fluoride volatilization processes,high thermal yields and a relatively simple design. Corrosion and erosion problems were encountered at the time, which could possibly be mitigated with more modern materials.

4 - DISINTEREST, OTHER PRIORITIES (N° 29)

The interest for thorium started to decrease around 1980 due to a number of reasons that we shall not analyze in detail. Among them we can cite :

- A growing resistance to nuclear acceptance by pacifist and ecologist movements which prompted Presidents Ford and Carter to ban reprocessing, a direct blow to the thorium cycle.
- The foreseen technical difficulties and costs associated with U-233 fuel fabrication and carbide fuels reprocessing. Such difficulties could today be largely overcome, but the question of the costs remains.

- General Atomics, the pioneer in HGTRs, was bought by Gulf.

- USAEC's decline was a blow to ORNLs activities.

- The great success of the Light Water reactors with good availability of uranium and relatively straightforward fabrication of UO2 fuels lead to abandon thorium and high temperature reactors.

India now stands alone, for reasons of national independence, and continues on its thorium projects. We believe that it would be desirable to cooperate somehow with India on this subject, if possible.

5. PRELIMINARY CONCLUSIONS (N° 30)

Without being overly optimistic for the near future, it seems that in Europe a steady, if limited, interest should be devoted to thorium to prepare for the longer term. We wish to cite a few reasons by decreasing order of priorities and increasing order of importance :

- Contributing to burn excess plutonium,

- Generating less long-lived waste,

- Going to higher burn-ups,

- Going to high temperatures,

- Sustainability, with our limited uranium reserves.

- Very important seems to us the interest in high temperature reactors which was initiated with thorium reactors, but which can be developed with uranium fuel as well, with enrichments below the fatidic 20 % U-235, or even with plutonium.

At a time when the economics of the PWRs are put in question due to their higher costs caused by elaborate safety systems, compared to the modern thermal dual-cycle power plants, modern high temperature reactors may help tap the immense potential of nuclear energy which so far has been quite poorly implemented, from a total yield point of view :

A PWR, for example, will use 0.6 - 0.8 % of the potential energy of the uranium mined, with 32 % thermal efficiency.

At least theoretically, but former industrial examples show us the way, we can cite some of the interesting features of what could be a modern high temperature reactor using modern refractory materials :

- Enhanced passive safety and "forgiveness",

- Reliability of operation during long periods of time between reloads (5-6 years),

- Good thermal yields (50-55 %),

- High fuel burn-ups, hence better fuel economy,

- Possibility to generate high temperatures for chemical or petrochemical cogeneration, including hydrogen production.

We feel that time has come to start giving another hard look at what has been perhaps too quickly set aside twenty years ago and start anew with fresh ideas. The accelerator-driven reactor is one of them.