

Thorium utilization in India's nuclear energy programme

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Ever since the Civil Nuclear Cooperation between India and the United States was first mooted in July 2005, there were detailed discussions in the print and electronic media on the requirement of raw materials, such as natural uranium for India's nuclear power programme, as well as on the Separation Plan, IAEA Safeguards, etc. It emerged that the present natural uranium (NU) resources in our country are limited and would be able to sustain about 10,000 MWe of nuclear power through Pressurized Heavy Water Reactors (PHWRs). The official target was set at 20,000 MWe by AD 2020, comprising a mix of PHWRs, Fast Breeder Reactors (FBRs) and imported Light Water Reactors (LWRs). On the other hand, it was often noted that India has abundant reserves of thorium (Th). An impression prevails that early induction of Th in our nuclear energy programme would be the most promising solution to get over the problem of fuel availability and achieve growth in nuclear power. A basic difference between NU and Th is usually overlooked. It arises due to the isotopic composition of NU, in that it has a small fraction of U-235 that undergoes fission with thermal neutrons. On the other hand, the mono-isotopic Th is purely a fertile material and fissile U-233 needs to be generated from it. The object of the present note is to examine the utilization of Th in a proper perspective.

At the dawn of India's nuclear energy programme, Homi Bhabha understood the importance of making use of Th to produce U-233. He envisaged a three-stage plan in which nuclear power reactors fuelled with NU would form the first stage. This was to be followed by chemical reprocessing of spent nuclear fuel to recover plutonium (Pu). In the second stage of the programme, the Pu so obtained would then be used as the nuclear fuel in FBRs. In such FBRs, a Th blanket could be deployed to breed U-233. Following chemical reprocessing, the recovered U-233 would serve as the fuel material for the third stage. Th would continue to be used as the breeding material in that stage also.

It needs to be emphasized that when Bhabha was articulating his strategy in

the early fifties, the nuclear behaviour of Th in a fast neutron environment was not well understood. Over the next decade (later half of 1950s and early 1960s), studies revealed the nuclear constraints that are inherent to Th. They are as follows.

1. The number of U-233 atoms generated in the Th blanket of a FBR is not significantly greater than the number of Pu-239 atoms consumed. This is called the breeding ratio. IAEA also published a technical document¹. Discussing the breeding characteristics in Russian large-power BN-type fast reactors, the document (table 3.4.4 (p. 54)) gives a breeding ratio of 1.04–1.10 for a mixed oxide fuel of U-233–Th, depending on fuel loading (kg/MWe). Corresponding BR range for Pu–U combination that regenerates Pu-239 was given as 1.25–1.35. Paranjpe² points out that 'Lower threshold for fast fission in U-238 compared to Th also leads to better breeding ratios in U-238-based systems as compared to Th-based systems in comparable chemical form'. Chemical reprocessing losses in a closed Pu–Th cycle might bring the final yield of U-233 to less than unity, thus losing the meaning of a breeder.

2. At the fast neutron energies prevalent in FBRs, a parallel nuclear reaction ($n, 2n$) with Th leads to the formation of undesirable U-232. By itself it is a worse radiation hazard than U-233 and the strong gamma radioactivity of its daughter products further complicates the situation and requires remote handling in all stages of latter operations.

3. During the formation of U-233, a long-lived (27.4 days) intermediate isotope, protoactinium-233 (Pa-233) is formed. Its chemistry complicates the subsequent chemical reprocessing. To avoid this and to maximize the yield of U-233, a cooling period of about 140 days might be required.

4. Pa-233 also has an affinity for absorbing a neutron during the time of its residence in the reactor, leading to the formation of Pa-234. According to Rastogi³, it is a double disadvantage. An atom of Pa-233 (hence an atom of U-233) is lost along with a valuable neutron. This could be mitigated by having a low neutron flux, but at the cost of reducing the formation of U-233.

5. Deploying Pu-239–Th combination in the thermal neutron region entails a penalty. Pu-239 has a large neutron capture cross-section (nearly thrice that for U-235), leading to the formation of Pu-240. This pathway has implications for fissile material inventory and neutron flux.

6. It has been pointed out by Rastogi³ among others, that the thermal neutron absorption cross-section for Th-232 is about three times more than that for U-238. It implies that when Th is used in a thermal reactor system, the requirement of fissile material such as Pu-239 would go up threefold. Kamala Balakrishnan and Kakodkar⁴ countered this argument by drawing attention to the much higher reactivity worth of U-233 in thermal reactors and its substantially slower fall in reactors containing Th.

7. Over the last two decades, a positive point for Th deployment emerged. Part of the U-233 generated during a fuel cycle in a thermal reactor could be burned *in situ*. This concept has relevance to the design of new nuclear reactors like the Advanced Heavy Water Reactor⁵.

Sixty years ago, a readily exploitable source for natural uranium in India was monazite from the beach sands of Kerala. While monazite is one of the high-grade sources of Th, only low amounts of associated U are present in it. Since uranium was the primary (first) requirement, there was no alternative available in the early 1950s, except to process large quantities of monazite sand to recover uranium tetrafluoride as a by-product. This automatically resulted in the accumulation of large quantities of high-purity Th as thorium nitrate from the very early stages, needed only for the second and third stages of Bhabha's vision.

From 1965–66 onwards, apart from Pu-239, depleted U (DU), depleted in terms of its U-235 content, became available from chemical reprocessing of spent NU nuclear fuel from the CIRUS reactor. In subsequent decades the availability of DU increased in a big way due to the reprocessing of spent fuel from PHWRs. The superiority of DU as a breeding material in FBRs gives it an advantage over Th.

In November 1985, Raja Ramanna underscored this plus-point for DU by re-

phrasing the second and third stages of India's nuclear energy development. Delivering the 20th Shri Ram Memorial Lecture, Ramanna⁶ mentioned:

'Phase II. Construction of FBRs which utilize plutonium and depleted uranium, the by-products of the phase I reactors'.

'Phase III. Use of thorium by converting it to uranium-233'.

Against the above-mentioned background, the following is a brief summary of the progress on Th utilization in India as collated from published information.

1. Thorium nitrate was converted to the oxide form and high-density pellets were made for loading in J rod positions of the CIRUS research reactor to generate U-233.

2. A pilot plant was set up to chemically reprocess the irradiated Th oxide pellets and U-233 was recovered.

3. U-233 in the form of its aqueous solution of nitrate was used as the fuel in the PURNIMA-2 critical assembly to study neutron multiplication in homogeneous systems.

4. U-233-Al alloy fuel was fabricated for loading in KAMINI (Kalpakkam Mini reactor). The design was validated through PURNIMA-3 at Trombay. This is being used as a high-flux neutron source for component testing by neutron radiography.

5. After the Fast Breeder Test Reactor at Kalpakkam reached mature operation, several kilograms of Th-oxide pellets were loaded in an outer axial blanket ring. Nickel filters were placed between the core and Th to modify the neutron spectrum, so that the neutron energy falls below the threshold for the Th-232 ($n, 2n$) and Th-231 reaction⁷. This would substantially reduce the contamination of U-233 by U-232. If this concept proves successful on a large scale, it might ease the shielding requirements.

6. A new thermal reactor of 300 MWe to make use of Th has been designed. It is the Advanced Heavy Water Reactor (AHWR). The fuels for this are mixed oxides of (a) plutonium and thorium, and (b) U-233 and thorium. Test facilities are being set up and the required computer codes are getting firmed up. In addition

to the fissile materials loaded initially, the design envisages *in situ* burning of a part of U-233 generated during a fuel cycle. This aspect was emphasized by Saha and Sinha⁵ and also by the Department of Atomic Energy⁸.

7. A test cluster having mixed oxides of Pu and Th was fabricated and irradiated in the pressurized water loop of CIRUS reactor to a burn-up of 18,000 MWd/T under conditions of temperature and pressure, similar to those in power reactors⁹. The cluster performed well.

8. Fuel containing U-233 is needed for AHWR testing programme. For this purpose, some PHWRs are being utilized. To achieve initial flux flattening in PHWRs, DU is generally employed. In a number of PHWRs, thorium oxide has been substituted so as to generate U-233. After chemical processing, the recovered U-233 would be used to fabricate the required test fuel clusters for AHWR⁷.

9. Pal and Jagannadhan¹⁰ have given the physics design of an Advanced Thorium 'Breeder' Reactor. This 600 MWe reactor proposes to burn *in situ*, a part of the U-233 generated from Th. Thoria rods seeded with 70% fissile Pu and seedless thoria rods constitute the core. To quote the authors, 'the thoria rods which were initially receiving neutrons from the inner seed fuel would become the supplier of neutrons and also take major share of power within the fuel cluster'. The study indicates a requirement of about 2.2 tonnes of 70% fissile Pu for one-third of core loading. It means that the total Pu core inventory would be over 6.5 tonnes and illustrates the need for substantial amounts of driver fuel for starting large-scale utilization of Th.

From the above account, it is clear that the use of thorium in FBRs will have to overcome the constraints due the inherent nuclear properties of Th, as well as the competition from DU and subsequent issues in chemical reprocessing to recover U-233. Balakrishnan and Kakodkar⁴ have aptly commented in general on the use of Th. 'One should not confuse the question of growth with energy potential. For growth we need to breed fissile material fast... When talking

about Th we are concerned with the latter'. Thus while growth would be achieved via DU, long-term sustenance has to be through Th utilization.

To achieve the second objective in a commercial sense, a span of 30–40 years would be required since, as noted earlier, Th utilization depends on the availability of large amounts of Pu, which in turn requires much larger quantities of natural uranium. Apart from India, several other countries have active programmes for Th utilization¹. Studies in Russia indicate that a core of metallic Th and Pu oxide and a blanket of metallic Th in BN-800-type FBRs could give a breeding ratio as high as 1.27. USA is looking at LWRs for exploiting the energy potential of Th. Development of new materials and technologies over the next few decades would certainly lead to the much sought after objective.

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