Science Policy

Thorium holds great promise for India's nuclear future

by Ramtanu Maitra

Thorium is several times more abundant in the Earth's crust than uranium. Most of the world's supply of thorium comes from a sandy mineral called monazite, and India has plenty of it. Monazite is essentially a mixture of phosphates of various rare earths, and is insoluble in water, so that grains of it tend to get concentrated along the banks of rivers and oceans when rock-bearing monazite mineral is eroded in water. In India, rich deposits of monazite are found in the beaches of Kerala State. India has little uranium, and the Indian nuclear program has all along been based upon making use of thorium. The element thorium (Th) was discovered by the Swedish scientist Jacob Berzelius in 1828. He named it after the nordic god Thor, who has been described in literature as a "powerful and helping god." The Indians hope that thorium may yet fulfill that promise.

Thorium, however, is not a fissile material, but it can be transformed into U^{233}, which is a man-made isotope, as is plutonium 239 (Pu^{239}). Both U^{233} and Pu^{239} are even better fission fuels than the naturally occurring U^{235}. Conversion of the fertile materials of Th^{232} and U^{238} to fissile materials through absorption of neutrons is a vital first step before energy can be derived from them. Th^{232}, which absorbs one neutron, goes through a series of transformations (radioactive decays). First, the unstable isotope Th^{233} is produced, which through beta decay (the emission of an electron) is transmuted into protactinium (Pa^{231}), and this in turn decays to U^{233}.

As a point of comparison, a similar sequence occurs when U^{238} is irradiated with neutrons. It absorbs one neutron to become U^{239}, and then decays to neptunium 239 (Np^{239}), and from there to Pu^{239}. (The emission of an electron transforms a neutron of virtually the same mass, although of course there is a shift in charge.) Nature is quite generous in providing us with both the fissile and fertile isotopes of uranium available in the natural state, but nature's generosity would have been more appreciated by nuclear engineers had U^{235} been available at 1.4% and not 0.7% in the natural state.

In order to use natural U^{235} as a fuel—which means generating a self-sustaining chain reaction—it is necessary either to use a moderator such as heavy water, or beryllium oxide or graphite in order to absorb neutrons; or, if light water is used as the moderator, one needs to enrich the U^{235} by about 2-3%. While U^{233} and Pu^{239} are better fuels than U^{235}, they do not appear naturally; however, U^{235} cannot be regenerated, and hence is only properly used for starting a cycle which would include reprocessing the fuel.

There are basically two fuel cycles, depending on whether thorium or U^{238} is the input fertile material. These are referred to as the Th^{232}-U^{233} cycle and the uranium-plutonium cycle. While the uranium-plutonium cycle is more suited for fast neutron systems, it has been established that the Th-U^{233} cycle is better adapted to thermal reactors because of the nuclear properties of the nuclei involved.

Less waste than with plutonium

One of the long-term advantages of the Th-U^{233} cycle not commonly known, is that the magnitude of long-lived actinide waste generated in this type of fuel is 1 million times less than that produced in the uranium-plutonium cycle. This is because, starting from U^{233}, five additional neutrons are required to be captured in succession in order to reach U^{238}. The probability of each additional capture decreases by a factor of 10, leading to an overall decrement of actinide wastes. This is of importance when nuclear power will be the main source of electricity, making it necessary to have adequate disposal of the actinide wastes, which have long half-lives.

A special feature of the Th-U^{233} cycle is the problem arising from the penetrating gamma radiation emitted by certain daughter products of U^{239}. This is an isotope of uranium which slowly accumulates in small quantities due to reactions occurring in the thorium fuel. U^{232} cannot be chemically separated from U^{233}, and itself does not emit any gamma radiation when it decays; however, a few weeks after the chemical separation of the uranium, levels of the energetic gamma-emitting daughter products, bismuth 212 and titanium 298, build up. Because of this, fabrication of U^{233} requires the use of heavily shielded facilities and remote fabrication methods. This complicates the process, and increases the overall cost of fuel fabrication in the Th-U^{233} cycle.

Use in heavy water reactors

India's interest in the thorium cycle is based upon establishing U^{233} as the fissile fuel for the CANDU-type heavy water reactors. At this point in time, pressurized heavy water reactors (PHWRs) appear to be particularly well-suited for operation on the Th-U^{233} fuel cycle. Extensive studies carried out in Canada, India, and other countries on the feasibility of fuelling PHWRs with thorium have shown very encouraging results.

Two types of calculations have been carried out. In the first type, it is assumed that adequate quantities of U^{233} are available to commence operation with an optimized Th-U^{233} fuel. In such a case, the study indicates that a self-sufficient equilibrium cycle mode of operation is feasible; but this would require lower fuel residence times, lower fuel discharge burn-ups, and more frequent fuel reprocessing, with consequent higher fuelling costs. It would seem that operation in the near-breeder regime with a conversion ratio of close to 0.95 may be economically justified, according to Indian researchers.

In the second type of calculation, it is assumed that we commence with a natural uranium cycle, but then into a few channels we introduce thorium bundles enriched with Pu^{239} to an extent of 2.5%, in order to produce some U^{233}. This U^{233} is then recycled back into the reactor with more thorium and plutonium. The proportion of thorium-fuelled channels is increased with time, until the entire reactor is switched over, after about 20 years, to a self-sufficient cycle mode.

The Indian work in irradiating thorium to generate fissile U^{233} took place in the CIRUS reactor in Trombay. The fabrication and reprocessing technology of thorium mixed oxide fuels (Th-Pu oxides) has been developed here and successfully tested for small-scale jobs. In general terms, a power reactor experiment to establish the thorium fuel cycle and demonstrate breeding, is well advanced.

In addition, the Indian reactor, Purnima II—the first of its kind to use U^{233}—only became critical at the Bhabha Atomic Research Center (BARC) in 1984, and has provided an enormous amount of research data. The reactor went critical with about 440 grams of U^{233}. This reactor is a unique one, in that it is the only operational reactor in the world using U^{233} as a fuel. A solution of uranyl nitrate in light water is used both as fuel and moderator. The reactor configuration is optimized for minimum critical mass using beryllium oxide as a reflector and a zircalloy core alloy. Due to the alpha activity of U^{233}, the entire system is enclosed in glove boxes, and the solution is transferred to the core vessel using a peristatic pump. The safety system is built around the Purnima (which means "full moon") facility, using the reflector drop and control blades as safety mechanisms.

The long-range program for nuclear power in the country is expected to be based on conversion of thorium to U^{233} and its uses in thermal or fast reactors. The research program at BARC has concentrated on problems associated with the fabrication, irradiation, and reprocessing of thorium, and on the experimental neutronics associated with the use of U^{233} in reactor systems. This reactor is therefore considered to be the first stage in the utilization of thorium.

A no-risk fuel

The importance of thorium as a future nuclear fuel is a subject of great importance in India; but it is also of strategic relevance, because by no stretch of the imagination could it produce a bomb-level fuel. A combination of high-temperature reactor design with the use of uranium-233 bred from thorium could short-circuit the present concerns about whether various nations are attempting to build nuclear generating facilities for dual-purpose, civilian-military use.

India is the place today where thorium breeding is most advanced. In this article, our New Delhi correspondent, Ramtanu Maitra, reports on the program there, in the context of the broader considerations which make the Indian program of world strategic import.

Thorium in fast breeder reactors

Another area of interest for the Indians is the use of the thorium cycle in fast breeder reactors (FBRs). Design strategy for FBRs centers on the selection of suitable coolant to remove the heat that is produced by the fast neutron chain reaction, as it will be when U^{233} is used.

An ideal coolant for FBRs is one which should enable the system to maintain a high breeding ratio and high specific power. Since it is essential to maintain the average energy of the neutrons carrying on the chain reaction as high as possible, common coolants such as water and heavy water rule themselves out of consideration. The coolants that deserve serious consideration are helium gas, liquid sodium, and sodium-potassium alloy. Liquid sodium has become almost the unanimous choice of FBR designers the world over, essentially because of its low neutron slowing-down power, low neutron capture, high boiling point, and excellent heat transfer properties.

Liquid sodium has yet another advantage. In case of a leak during reactor operation, the lower ambient temperature causes the sodium to freeze, automatically stopping the leak. Liquid sodium does have other problems, such as corrosion. Severe corrosion occurs if even a minute amount of oxygen finds its way into the dissolved sodium. Also, despite its slow neutron capture, sodium can become extremely radioactive,

emitting penetrating gamma rays. Finally, as is well known, sodium has a great affinity for water, and even a small amount of moisture in the atmosphere is enough to cause an instant fire.

Because the thorium-uranium cycle is superior in thermal reactors and satisfactory in fast ones, there are some advantages in introducing thorium, and possibly U^{233}, into fast reactors, particularly where a mixed system of fast reactors and high-conversion thermal reactors is anticipated.

Not well-suited for making bombs

In addition, the thorium-uranium cycle is preferred by some because it makes it difficult to convert recycled fuel for weapons. In order to dilute or "denature" it, measures have to be taken to dilute the fissile material either isotropically or radioactively. The first type of dilution is possible with only one of the fissile materials bred in a reactor. The most common material, plutonium, is considered as weapons material, and there is no suitable way of denaturing it isotropically—i.e., as U^{235} is naturally denatured by U^{238}. The alternative bred material, U^{233}, can be denatured by U^{238}. If the fissile content does not exceed 10% or so, conversion to weapons material requires an enrichment facility—even more of a commitment (at the present time) than reprocessing. Therefore, since the thorium-uranium cycle produces U^{233}, this cycle appears adaptable to a more proliferation-resistant form than the uranium-plutonium cycle. It should be kept in mind, however, that denaturing by U^{238} necessarily leads to production of some plutonium, which must be either used or disposed of.

It should also be kept in mind that by denaturing U^{233} with U^{238}, the fuel cycle is significantly altered. A pure thorium-uranium fuel cycle produces only U^{233} with no plutonium. The system could be started on highly enriched uranium (or even plutonium), and the principal recycle fuel is Th-U^{233}. Denaturing U^{233}, on the other hand, adds fertile U^{238}, which produces plutonium. How much? As a rough comparison with 3% enriched uranium, consider a U^{233}-U^{238}-Th^{232} mixture in a ratio of 12:88:300, so that U^{233} is 3% of the mixture. Ignoring cross-section differences, one would expect that about 23% as much plutonium will be produced as would be the case with the ordinary 3% U^{235}-97% U^{238} fuel. In fact, because of differences in cross-section, the amount of plutonium produced would be smaller, but it is still a significant amount.

Use in gas-cooled reactors

In the Indian context, there is recognition that the thorium-uranium cycle will be appropriate for high temperature gas-cooled reactors (HTGRs). However, there is no indication at this point that the Indian program is in the process of prioritizing the HTGR with the Th-U^{233} fuel cycle. The program remains to establish the Th-U^{233} cycle for thermal power reactors, and to develop breeder reactors using the same fuel cycle, while starting up the system with plutonium-uranium oxide fuel.

Nonetheless, there exists a host of literature on the HTGR using a thorium-uranium fuel cycle. In the 1970s, General Atomic, Bechtel, and Southern California Edison had a 4,000-megawatt design. Although the focus of the design was improvements in plant configuration—namely, in the design of the Prestressed Concrete Reactor Vessel support structure and fuel storage facility—the HTGR did take into consideration the thorium-uranium fuel cycle.

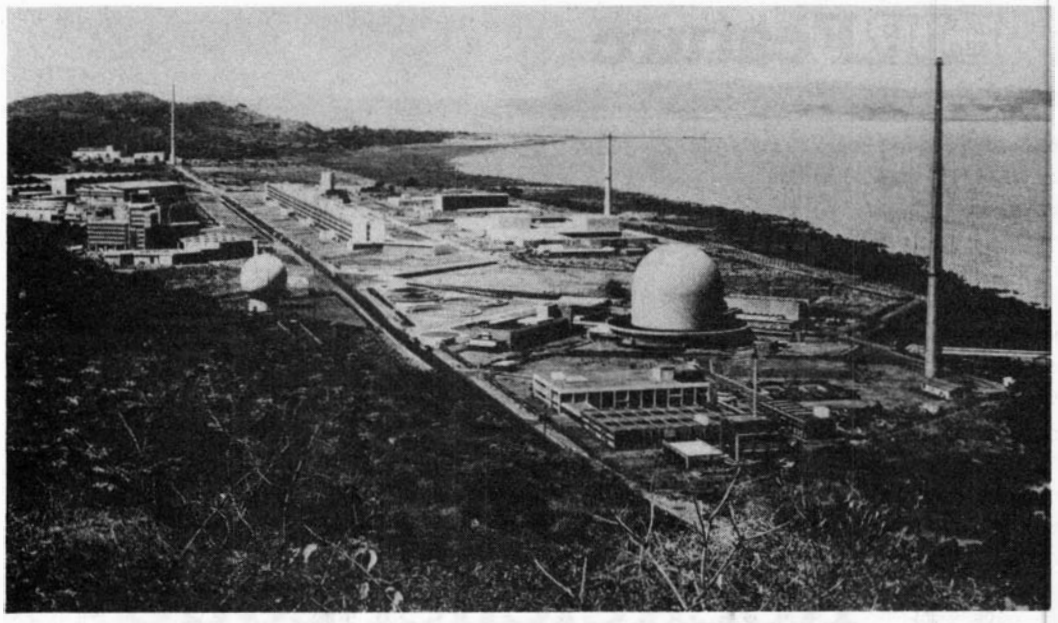
According to experts, the fuel cycle which results in the best uranium utilization and lowest fuel-cycle costs, is the high-enrichment uranium-thorium cycle. The use of fissile particles of high-enrichment uranium (HEU) and a thorium fertile particle, permits optimization to meet specific conditions. But the HEU used in the thorium cycle created potential front-end proliferation concerns. A variant of the thorium cycle, using medium-enriched uranium (MEU), e.g., 20%, and thorium appears to be a balance in meeting requirements for a low fuel-cycle cost, high proliferation resistance, and low U^{238} requirements. Intermediate enrichment of 30-40% is also being considered in order to improve resource utilization, while still being responsive to proliferation concerns.

Although the fuel development effort in the United States has focused on the HEU-Th cycle, most of the effort is also applicable to the MEU-Th cycle. Only the fissile particle is different: Fertile particles, fuel rods, and graphite remain the same. Thus, it appears that an incremental effort will be necessary to license and use MEU-Th fuels in the United States.

Several recycling options are available with the MEU-Th cycles: 1) Recycle only U^{233}, 2) recycle all uranium, and 3) recycle all uranium and plutonium. The U^{233} can be recycled either as bred in the thorium particle (highly enriched in U^{233}) or can be denatured by mixing it with U^{238} so that the enrichment is in the 10-15% range. These options exist because in the discharged fuel, the uranium particles, which contain unburned U^{238} and plutonium, can be mechanically separated from the thorium particles that contain all the bred U^{233}. The former could be stored indefinitely without chemical processing, while the latter could be processed to recover the fissile U^{233}.

Proliferation considerations

There are some inherent nonproliferation advantages in an HTGR using the MEU-Th cycle. In the discharged fuel, the plutonium is very diluted. About 200 elements would have to be processed in order to recover a critical mass of plutonium. Since the fuel elements are heavy and bulky, this would be a formidable diversion and chemical separation link. The particles themselves are a deterrent to diversion: It is a difficult process to crack open the coatings and recover the fissile fuel contained therein. As mentioned earlier, the plutonium-containing particles could be separated and stored



The Bhabha Atomic Research Center in Trombay.

without processing.

The presence of U^{233} is associated with strong gamma radiation—a consequence of the presence of a small amount of U^{232}. This activity level will deter diversion by making it easier to detect, and thus would facilitate safeguard procedures and greatly complicate bomb fabrication or assembly attempts. Moreover, the U^{233} could be denatured either *in situ* or in a reprocessing plant, making it an even less desirable nuclear material for clandestine processing.

Finally, total plutonium production is low. With 20% MEU, only about 60 kg of plutonium is discharged per GWe-year (gigawatt-electric-year; power multiplied by time is a measure of energy), and its enrichment is only 50-60%. If the enrichment of the initial fuel were increased to 40%, instead of 20%, the discharged plutonium would only be about 35-40 kg per year, and this would be distributed over more than 1,000 fuel elements. If refuelings were semiannual, rather than annual, then only about 15-20 kg would be contained in any single refueling batch—not much more than a a single critical mass—and over 500 fuel elements would need to be processed in order to recover it. This approaches an ideal situation from the proliferation risk standpoint.

For the thorium cycle in the HTGR, there are two schemes: the separable thorium cycle, and the non-separable thorium cycle. In the separable thorium cycle, the fissile and fertile materials are used as separate individual kernels; the optimum fuel system has coated fissile uranium carbide (93% enriched U^{235}) and coated thorium kernels. The silicon carbide layer in the uranium coating serves as a means of separating bred fissile U^{233} from the U^{235} and its activation product U^{236} during reprocessing. In the non-separable thorium cycle, kernels of mixed oxide or carbide with either coating are used.

Fabrication of coated particle fuels involves two steps: preparation of kernels and coating of the kernels. The preparation can be done through either a wet chemical process (sol-gel route) or a dry agglomeration method. The sol-gel and gel precipitation route is more suited for remote operations in the fabrication of recycled fuel. A stable thorium or uranyl nitrate solution containing a gelling agent is sprayed through a nozzle to form droplets, which are hardened by reaction first with gaseous ammonia. The gel spheres are then washed, dried, and sintered. If kernels of carbide or oxycarbide are to be made, adequate quantities of carbon black are added to the nitrate solution.

The most important aspect of assessing the irradiation performance of coated particle fuels is the integrity of the coating on the kernels. The mechanisms by which coatings may fail have been identified as: 1) mechanical failure of coating arising from internal build-up of fission gas pressure and from irradiation-induced stresses in the coating; 2) chemically induced failure of the coating due to fuel kernel migration toward the surface; and 3) chemically induced failure due to interaction of fission products with the coating.

The only operational Thorium High Temperature Reactor (THTR) was built, and since decommissioned, at Schmehausen in Germany. It was a 300 MWe prototype high-temperature gas cooled reactor, with a pebble-bed aftercore. The reactor reached criticality for the first time on Sept. 13, 1983, after the loading of some 200,000 spherical fuel and moderator elements. A further phase of fuel loading to the eventual full inventory of 675,000 elements, started later. The THTR project was begun in 1971, and was originally scheduled for completion in 1976. Delays, however, were caused mainly because of design changes called for by licensing authorities, and this project, along with the German SNR fast reactor, was subjected to repeated funding cuts.