EXPERIENCE OF THORIUM FUEL DEVELOPMENT IN INDIA*

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Abstract. India has one of the largest resources of thorium in the beach sands of Southern India. Generation of nuclear power through utilization of thorium is the ultimate goal of India's three stage nuclear power strategy. Bhabha Atomic Research Centre (BARC) is actively pursuing research, development, fabrication, characterization and irradiating testing of ThO_2, ThO_2-PuO_2, ThO_2-UO_2 fuels in test and power reactors. Work related to developing the fuel fabrication technology including automation and remotization needed for ^{233}U based fuels is in progress. Use of ThO_2 fuel bundles for initial flux flattening in our PHWRs; recent criticality of KAMINI - a small neutron source reactor, with ^{233}U-Al alloy fuel; introduction of ThO_2 as axial and radial blankets in our Fast Breeder Test Reactor (FBTR) at Kalpakkam; proposed ThO_2-PuO_2 and ThO_2-$^{233}UO_2$ fuel for Advanced Heavy Water Reactor (AHWR) are some of the steps taken by us towards utilization of Thorium in India. The paper summarizes the present status of thorium fuel development in India.

1. INTRODUCTION

India has a relatively modest uranium resource (~50,000 tons) but is endowed with one of the largest deposits of thorium in the world (~ 360,000 tons) in the beach sands of Southern India. Any long term planning of the growth of nuclear power programme in India, therefore, has to be based on proper harnessing of energy potential of thorium. This was realized quite early by the founders of our nuclear energy programme who drew up a clear three stage power development profile with the generation of nuclear power through utilization of thorium as its ultimate goal. The first phase of the programme is based on Pressurized Heavy Water Reactors (PHWR) using natural uranium as fuel. The second phase is based on utilization of plutonium, generated as by-product from the first phase, in Fast Breeder Reactors (FBRs) for power generation and to enhance our fissile material inventory both in terms of ^{239}Pu and ^{233}U. The third phase is based on thorium fuelled thermal reactors. Several theoretical studies have been carried out [1] on thorium fuel cycles in Heavy Water Reactors (HWR).

As thorium plays such an important role in our nuclear power programme, it is natural that we have significant R&D Programmes devoted to thorium fuel cycle development. We are actively pursuing research & development programme in fabrication, characterization and irradiation testing of ThO_2, ThO_2-PuO_2 and ThO_2-UO_2 fuels in our test and power reactors. Fuel bundles containing high density ThO_2 fuel pellets are being used in all our new PHWRs for flux flattening in the initial Core. ThO_2 pins and sub-assemblies are also to be used as axial and radial blankets in our Fast Breeder Test Reactor (FBTR) operating at Kalpakkam. KAMINI, a neutron source reactor, is operating with ^{233}U -Al alloy fuel. ThO_2-PuO_2 and ThO_2-$^{233}UO_2$ are proposed as fuel for the Advanced Heavy Water Reactor (AHWR), the detailed design of which is being carried out in our Centre. Development of novel fuel fabrication processes and techniques related to automation and remotization needed for ^{233}U based fuel fabrication are under study.

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2. FUEL FABRICATION

2.1. ThO_2 Fuel

Fabrication of high density sintered ThO_2 pellets for the ThO_2 bundles used for flux flattening of the initial Core of PHWRs and as blanket in FBTR, is carried out by the conventional Powder Metallurgy technique of cold compaction and high temperature sintering either in reducing or in oxidizing atmosphere as shown in Figure 1.

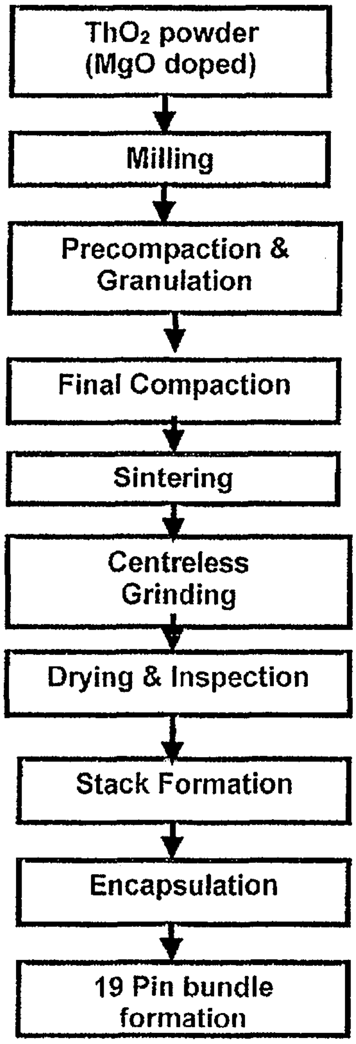

Figure 1. Flowsheet for ThO_2 fuel bundle fabrication.

The oxalate derived ThO_2 powder has a rectangular plate morphology and needs intensive milling to break the platelets and to increase the surface area and sinterability. To avoid caking of powder during milling, once-through dry nitrogen atmosphere is maintained in the enclosure around the pot mill/attritor. As the oxalate derived powder is not free- flowing, precompaction and granulation of powder is necessary to enhance flowability for ease of pneumatic powder/granule conveying and to obtain a uniform die fill during powder compaction.

ThO_2, being a perfectly stoichiometric compound with a high melting point (~3400°C), needs a sintering temperature of over 1800°C for obtaining high sintered density pellets (96% theoretical density). However, with the addition of 500-600 ppm of MgO dopant as sintering aid, high density pellets are presently being fabricated on an industrial scale using a sintering temperature of about 1650°C-1680°C under reducing atmosphere. The improved sintering characteristics of MgO doped ThO_2 pellets is due to enhanced volume diffusion of thorium brought about by formation of oxygen ion vacancies as Mg^{+2} is substituted for Th^{+4}. MgO doping is done in the form of $MgSO_4$ in the thorium nitrate solution before oxalate precipitation.

A method of low temperature sintering (~1250°C) of ThO_2 pellets in air, using 0.5w% Nb_2O_5 as additive, has also been developed [2] and test irradiation of low temperature sintered ThO_2 pellets are being planned shortly.

Tonnage quantity of ThO_2 pellets are now a days fabricated using glove box trains with local shielding around equipment and interim storage facilities for powder/granule/pellets to reduce dose to operating personnel, aerosol generation and air borne activity.

2.2. ThO_2-PuO_2 and ThO_2-$^{233}UO_2$ Fuels

Thorium does not have any naturally occurring fissile isotope. However it can be converted to highly fissile ^{233}U by using it in reactors in combination with either ^{235}U or ^{239}Pu. Once sufficient quantity of ^{233}U is accumulated, ThO_2-^{233}U O_2 fuel cycle can be adopted in the PHWRs with near breeder characteristics.

It is in this context that knowledge of fabrication technology of (ThO_2-PuO_2) and (ThO_2-^{233}U O_2) fuels for advanced fuel cycles of PHWRs assumes great significance.

ThO_2 has similar crystal structure as that of UO_2 and PuO_2; forms complete solid solution at all percentages with UO_2 and PuO_2 and has similar physical properties. Hence, fabrication procedure of mixed oxide fuels of ThO_2 with PuO_2 or UO_2 are similar to that of (UO_2-PuO_2) mixed oxide fuel for which experience exists in India. However, fabrication of ^{233}U bearing fuels in standard glove-boxes is normally not feasible due to the presence of high gamma radiation field associated with the daughter products of ^{232}U, which is always present along with ^{233}U as a minor constituent. Hence, a high degree of automation and remotization and thickly shielded hot cell facility is needed for fabrication of ^{233}U based fuels.

In general, the following techniques have been tried for ThO_2 based fuel fabrication for PHWR:
(a) Cold pressing of powder mixture of ThO_2-PuO_2 or ThO_2-UO_2 followed by high temperature sintering.
(b) Vacuum impregnation of partially sintered low density (~70-80%T.D.) ThO_2 pellets with uranyl nitrate or plutonium nitrate solution followed by drying and final sintering.
(c) Sol-gel derived microsphere pelletization (SGMP) followed by sintering.

2.2.1. Cold Pressing and Sintering Route

This process is essentially the same as the process followed for the fabrication of ThO_2 fuel. The process consists of co-milling the ThO_2, PuO_2 or ThO_2 and UO_2 powders, cold pressing of the powder mixture into green pellets and sintering of the green pellets to get high density. This method needs handling of very fine and non-free- flowing powder and it generates highly radioactive dust which settles on the equipment and glove box/ hot cell surfaces necessitating their frequent decontamination in order to keep personnel exposures to a minimum. Moreover, this method of fuel fabrication is least amenable to automation and remotization. However, we have some experience of fuel fabrication by this method.

2.2.2. Impregnation Method

In this process (Figure 2), partially sintered, porous (density in the range of 70-80% T.D.) ThO_2 pellets, fabricated in a conventional ceramic fuel fabrication plant, are transferred to a shielded facility where the pellets are vacuum impregnated with Uranyl nitrate solution, dried and then sintered. The most attractive feature of this process is its amenability to automation and remotization and possibility of separating most of the equipment for pellet production from shielded facility where ^{233}U solution is handled.

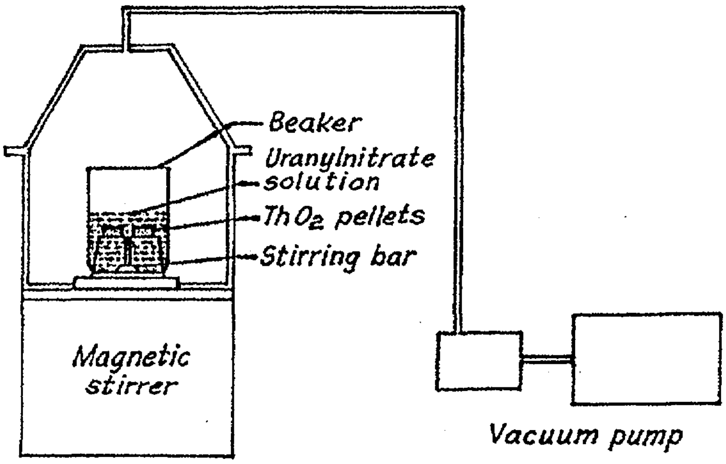

Figure 2. Schematic diagram of ThO_2 pellet impregnation set-up.

The limitations of this processes are:

(a) by this technique only 2-3w% ^{233}U can be introduced in ThO_2;
(b) multiple cycle of impregnation and drying are needed even to introduce 2-3w% uranium in ThO_2 as needed for PHWR pellets;
(c) it is very difficult to get a uniform distribution of UO_2 in ThO_2 over the whole cross section of the PHWR pellet.

In BARC, ThO_2-2w% nat.UO_2 pellets have been fabricated by this technique with homogeneous and uniform uranium distribution over the whole cross section of the pellet [3].

.2.3. Sol-Gel Microsphere Pelletization (SGMP) Process

Sol-gel microsphere pelletization process, popularly known as SGMP technique, utilizes sol-gel derived dust-free and free-flowing soft microspheres of $(Th-U)O_2$ [either by internal gelation process or by external gelation process], in the size range of 100-600 microns in diameter, which are cold compacted and sintered to high density pellets the same way powder pellets are fabricated. The general flow-sheet of fuel fabrication by SGMP is shown in Figure 3. Because of the free flowing and dust-free nature of the microspheres, the fuel fabrication process is amenable for automation and remotization needed for hands-off plant operation philosophy. SGMP process is being vigorously pursued in BARC for the fabrication of UO_2, $UO2-PuO_2$ and ThO_2-UO_2 fuel pellets.

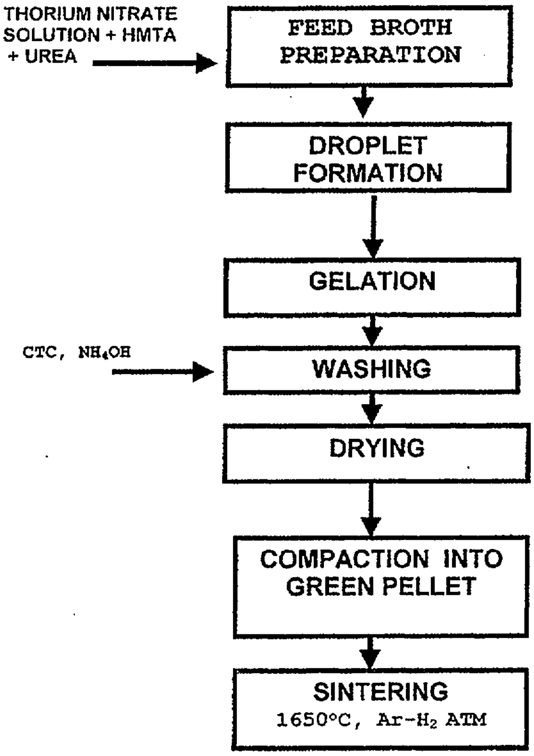

Figure 3. Flowsheet for fabrication of ThO_2 pellets by SGMP technique.

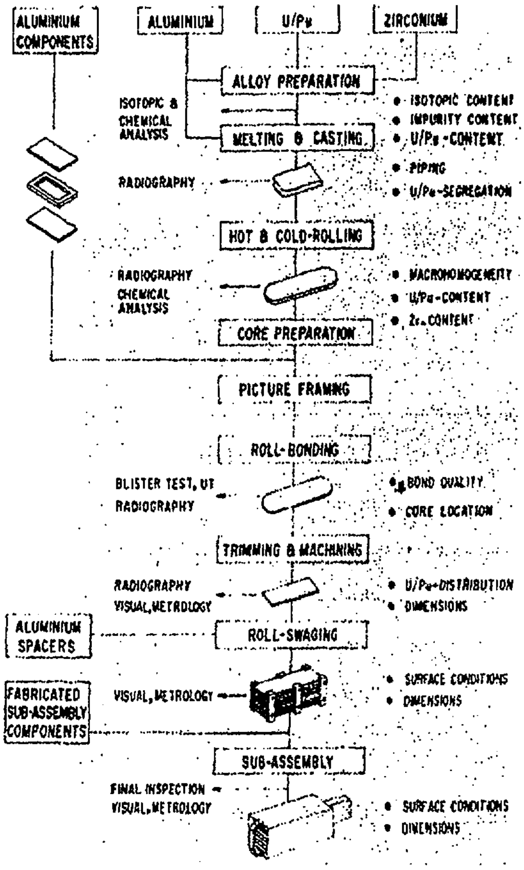

Figure 4. Flowsheet for KAMINI fuel fabrication.

3. FABRICATION OF Al-^{233}U PLATE FUEL

KAMINI, a 30 kW (thermal) light-water cooled and moderated, compact research reactor, is operating at our Indira Gandhi Centre for Atomic Research (IGCAR). BARC has gained substantial experience in fabrication of ^{233}U based fuel by supplying the Al-clad Al-20wt% ^{233}U plate type fuel assemblies for the reactor. The fabrication flow-sheet (Figure 4) consisted of preparing the master alloy using aluminium & uranium as feed materials, remelting and casting of the fuel alloy ingots, rolling, picture framing and sandwiching the fuel between thin aluminium sheets, roll-bonding, core location by radiography, trimming & machining to final dimensions. The detailed procedure of fuel fabrication has been described elsewhere [4].

4. THERMOPHYSICAL PROPERTY EVALUATION

A data base of thermal conductivity and hot hardness of ThO_2 fuel with temperature and PuO_2 and/or UO_2 content as variable is being generated to theoretically predict and model prediction of in-pile central temperature of these fuels and their performance. The general methods followed and description of the instruments used have been reported elsewhere [5, 6]. The results of thermal conductivity and hot hardness with temperature for ThO_2-2wt% UO_2 and ThO_2-4% PuO_2 pellets are reproduced here in Figures 5 & 6.

5. IRRADIATION TESTING OF (ThO_2 . PuO_2) ASSEMBLIES IN REACTORS

A six pin cluster consisting of (ThO_2-4%PuO_2) fuel pellets produced by powder pellet route has undergone irradiation testing in the pressurized water loop (PWL) of our research reactor CIRUS upto a burn up of 18.4 MWd/kg and is awaiting post irradiation examination. The loop test conditions, pellet details and irradiation data are shown in Tables I, II & III respectively. Two additional six pin clusters, containing high density ThO_2 and ThO_2-6.75% PuO_2 pellets clad in collapsible Zircaloy-2 tubes, (similar to our PHWR) are presently undergoing irradiation in the PWL - CIRUS and have accumulated a burn up of about 13 MWd/kg to-date. ThO_2 based fuel pellets fabricated by SGMP technique and low temperature sintering technique are being planned for future irradiation in our test loops.

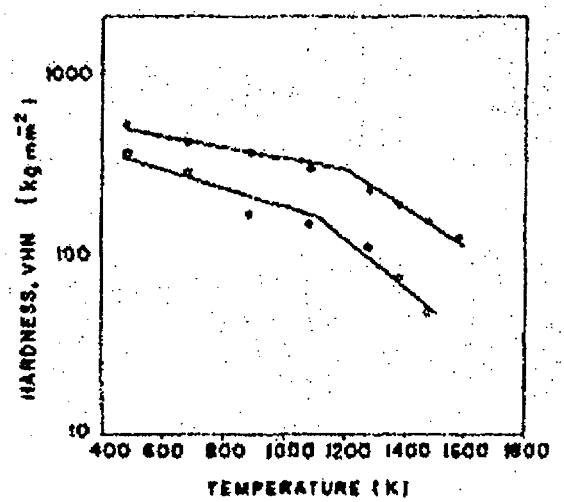

Figure 5. Hardness vs temperature plots for (•) ThO_2 - 2% UO_2 and (o) ThO_2 - 4% PuO_2 sintered pellets.

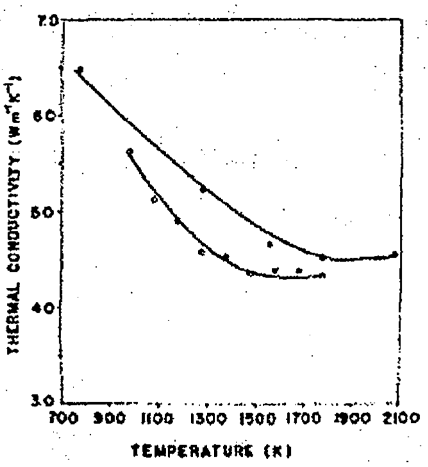

Figure 6. Thermal conductivity vs temperature for (•) ThO_2 - 2% UO_2 and (o) ThO_2 - 4% PuO_2 sintered pellet (corrected to 100% T.D.).

Table I. Loop Test Conditions

Test Section ID	57.4mm
Neutron Flux (Thermal)	5×10^{13} n/cm^2/sec
Coolant/pH	Demineralized Water/9.5-10.5
Coolant Flow Rate	16899 kg/h
Pressure	105 kg/cm^2
Temperature	204°C

Table II: Details of pellets for tests 1 & 2.

	Test 1	Test 2
Enrichment (PuO_2%)	4%	6.75%
Diameter	12.22 mm	14.4 mm
Density	92-94% T.D.	> 96% T.D.
Stack Length	435 mm	471.5 mm
Cladding Outer Diameter	14.3 mm	15.23 mm
Cladding Wall Thickness	0.8 mm	0.38 mm
Cold Plenum Length	20 mm	NIL

Table III: Irradiation data

	Test 1	Test 2
Peak Linear Power	385 w/cm	435 W/cm
Peak Burn-Up	18.4 MWd/kg	13.00 MWd/kg
Number of Power Cycles (> 30% Full Power)	100	177
Fuel Surface & Centre Temp.	462°C/1980°C	--

6. CONCLUSION

We have made a modest beginning in utilizing thorium for power generation and are planning for large scale utilization of thorium based fuels in future. Development of fabrication technologies, generation of data base for thermophysical properties and irradiation testing of thorium based fuels are being actively pursued and will be further intensified in future so that thorium can play an important role in the growth of nuclear power in India.

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