

MINERALOGY AND GEOCHEMISTRY OF NUCLEAR FUELS: RELATIONSHIP TO SOURCE, HOST-ROCKS, AGE, MINERALIZATION-PROCESSES AND ORE PROCESSING, WITH INDIAN EXAMPLES

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ABSTRACT

Nuclear fuels, from which nuclear power is generated, are Uranium (U) and Thorium (Th), and products derived from them, viz., Plutonium (^{239}Pu) and ^{233}U , formed, respectively, due to neutron-absorption by ^{238}U and ^{232}Th . India's 3-stage nuclear power programme envisages (i) natural U-fuelled Pressurized Heavy Water Reactors (PHWR) in the 1st stage, (ii) Pu-fuelled fast breeder reactors in the 2nd stage and (iii) ^{233}U -fuelled (derived from natural Th) breeders in the 3rd stage. The radio-elements of U and Th occur in ~200 discrete 'primary' (e.g., uraninite and thorite) and 'secondary' (e.g., uranophane and thorogummite) radioactive minerals. U and Th in small contents occur in a large number of rock-constituting accessory minerals such as zircon and monazite. Furthermore, U, as a substituting ion, occurs in ~20 U-niobates, -tantalates and -titanates. Besides, U, in adsorbed state, is associated with clays, carbonaceous matter, hydrated Fe-oxides, etc. Radioactive ores contain a combination of some of the above minerals, along with gangue and other ore minerals. From these ores, U- and Th-minerals are first to be concentrated by physical beneficiation, using appropriate techniques that are selected based on the mineralogy, nature of association of minerals and geochemistry of the ores, followed by cost-effective extraction of U and Th for use as nuclear fuels. Selection of extractive techniques depends again upon the bulk chemistry of the ore, mineralogy and textures of U-/Th-minerals and their chemistry, which, in turn, are controlled by diverse parameters of which source, host-rock, age and mineralization-processes are important. Relationship of these parameters and U-Th-ore processing to mineralogy and geochemistry of nuclear fuels is presented in this paper, with some Indian examples.

INTRODUCTION

India, in its pursuit for energy-security to meet its ever increasing demand for energy, is actively pursuing diverse modes of energy-generation. These include the renewable ones such as solar, wind and wave energy, biomass (wood, sugar), geothermal and hydro, and non-renewable ones like fossil fuels (coal, oil and natural gas) and nuclear, with fossil-fuels presently providing predominant component (~80%) of energy. As (i) fossil fuels are fast depleting with cost-escalation and uncertainty in supply from some of their major producing-countries of middle east (Libya, Algeria, Egypt, Saudi Arabia and Oman) due to recent political problems, besides draining major chunk of foreign exchange for their import; (ii) generation of hydro-power being localized and depends much on good monsoon; and (iii) other renewable energy resources like solar, wind, tidal, bio-mass and geothermal are being diffuse and intermittent with limited potentiality, generation of nuclear power in India, like in many developed and developing countries of the world, is an absolute necessity to meet the

ever increasing energy-demand. This led to the Indo-US Nuclear Agreement of 2008-2009 that facilitates the establishment of many nuclear power plants in India in collaboration with countries like U.S.A., Russia and France, besides allowing import of Uranium fuel to meet the requirement of nuclear power plants, presently under operation by the Nuclear Power Corporation of India Limited (NPCIL) of the Dept. of Atomic Energy. Some of the highly appealing aspects of nuclear power are its (i) *very high heat value*, as compared to other fuels (e.g., Natural Uranium in light water reactor: 500,000 Mega Joules (MJ)/kg as against 16 MJ/kg of firewood, 9-30 MJ/kg of different qualities of coal, 45-46 MJ/kg of crude oil), thereby requiring much less volume of fuel, (ii) *clean nature* of power with no emission of greenhouse gases like CO₂, methane and CFCs, all of which are contributing to global-warming that is associated with catastrophic consequences and (iii) *dependability*. India is one of the few countries in the world, which operates, with indigenous technology, the complex 'nuclear fuel cycle'. This encompasses from the 'front end' of exploration – mining – milling – conversion – fabrication of fuel to the 'back end' of treatment of spent fuel and radioactive waste by storage – recovery – recycling – disposal, through the most important stage of production of electricity in a nuclear power reactor.

Nuclear fuels, required for generation of nuclear power, are Uranium (U) and Thorium (Th), and products derived from them, viz., Plutonium (²³⁹Pu) and ²³³U, formed, due to neutron-absorption, respectively, by ²³⁸U and ²³²Th. As India is having moderate resources of U [0.1248 Million tonnes (Mte) of U₃O₈, as on Jan. 1, 2009; AMDER, 2009] and more resources of Th (0.9-1.0 Mte of ThO₂, contained in 10.2 Mte of Th-bearing monazite; Chandrasekharan, 2007), the Indian Nuclear Power Program, as envisaged by late Dr. Homi J. Bhabha, the father of this program, is of 3-stage, viz., (i) *first stage* involving natural U-fuelled Pressurized Heavy Water Reactors (PHWR), (ii) *second stage* with Pu-fuelled fast breeder reactors and (iii) *third stage* with ²³³U-fuelled (derived from natural Th) breeder reactors (Fig. 1). To operate these stages, the nuclear fuels of U and Th are to be extracted cost-effectively from their respective ores. In an ore, such extraction depends upon the mineralogy of U-/Th-minerals and their chemistry (see next section), which, in turn, are controlled by diverse parameters of which source, host-rock, age and mineralization-process are important. Relationship of these parameters and processing of U- and Th-ores to mineralogy and geochemistry of nuclear fuels is presented in this paper, with a few Indian examples.

Fig. 1. near here.

BRIEF ACCOUNT OF MINERALOGY AND GEOCHEMISTRY OF U AND Th

Minerals of U and Th are radioactive. They comprise both discrete minerals and others in which either U or Th or both occur in notable quantity like the minerals of Rare Metals (Nb-Ta, Li, Be, Sn and W) and Rare Earth Elements (REE: La to Lu and Y). Geochemical processes play an important role in the formation of these minerals. Geochemistry of U is more complex than Th. This is due to: (i) different oxidation states, mainly U⁴⁺ and U⁶⁺; (ii) variable large size cell units permitting U⁴⁺ substituting for many elements of similar ionic radii, largely Th and to a limited extent Ca, Zr, Ti, W, Mo, Nb, Ta and REE; (iii) U-phases hydrolyze in the presence of solutions; (iv) high chemical reactivity of U⁶⁺ that forms complexes with a variety of anion in aqueous solutions; (v) relative solubility of U⁶⁺-compounds in aqueous solutions, compared to the relative insolubility of U⁴⁺-compounds; and (vi) adsorption tendency of U on clay, organic and other particles or on certain hydroxides of Fe, Ti and Zr. Due to these geochemical processes, U forms a major constituent in many minerals of its own, with bulk of them being of U⁶⁺-compounds, formed in oxidizing environment, and only a few are of U⁴⁺-compounds, formed in reducing

environment; the latter group transforms into the former with increasing oxidation potential, while the former changes to the latter in the presence of reductants like sulphides and carbonaceous matter. In contrast to U, Th-mineralogy is relatively simple as only one principal oxidation state of thorium (Th^{4+}) is of importance. Th commonly replaces (i) Zr in zirconium minerals; (ii) Y, Ce and other REE in most REE-minerals like monazite; and (iii) U in certain U-minerals under conditions of relatively high temperature. The minerals of U and Th are broadly divided into primary and secondary discrete minerals, and nearly 200 such minerals are known. The primary minerals are those formed directly from magmas, hydrothermal solutions and groundwater. Secondary minerals are formed due to remobilization of elements from primary minerals, their transportation in solution and later precipitation due to over-saturation in oxidizing or supergene environment. The most common primary minerals of U are uraninite (pitchblende, if microcrystalline) (oxide), coffinite (silicate) and brannerite (complex oxide with Ti). In these, U occurs mostly in U^{4+} -state, besides some U^{6+} in pitchblende. Secondary minerals of U are many (~180) in which U occurs entirely in the U^{6+} -state. These include various oxides (gummite), silicates (uranophane), vanadates (carnotite, tyuyamunite), phosphates (autunite, torbernite), carbonates, sulphates, molybdates and arsenates, and their complex derivatives. The most common primary minerals of Th are thorianite/thorouraninite (oxides) and thorite/uranothorite (silicates), whereas the most common secondary mineral of Th being thorogummite. Furthermore, U and Th in notable contents (≥ 1 wt. %) occur in a large number of rock-constituting accessory minerals like zircon, monazite, xenotime, apatite, allanite, sphene and rutile. U, as a substituting ion, occurs in ~20 U-niobates, -tantalates and -titanates, the notable ones of which are betafite, davidite, euxenite, samarskite, columbite-tantalite and pyrochlore-microcline. Besides, U, in adsorbed state, is associated with clays, carbonaceous matter, hydrated Fe-oxides, etc. (Dhana Raju, 2009, pp. 271-292). Radioactive ores contain a combination of some of the above minerals, along with other gangue and ore minerals. From these ores, U and Th are to be extracted cost-effectively for use as nuclear fuels. In an ore, such extraction depends upon the mineralogy of U-/Th-minerals and their chemistry, which, in turn, are controlled by diverse parameters of which source, host-rock, age and mineralization-process are important. Relationship of these parameters and ore processing to mineralogy and geochemistry of nuclear fuels is presented in the following.

RELATION OF SOURCE, HOST-ROCKS, AGE, MINERALIZATION-PROCESSES AND ORE PROCESSING TO MINERALOGY-GEOCHEMISTRY OF U AND Th

Source: Geochemically, both U and Th belong to the group of ‘Large Ion Lithophile Elements’ (LILE) or ‘Incompatible Elements’. Hence, during the evolution of the Earth, they, like other LILE and incompatible elements – K, Rb, Cs, Sr, Ba, REE, Zr, Hf, Nb-Ta, etc., -- were expelled from the mantle and core, and concentrated in the shallower crust. The average abundance of U and Th in the Earth’s crust, respectively, is, ~2 ppm and ~8 ppm, with the value for Th/U being between 3 and 4. In common rock types, both U and Th prefer (i) acidic, especially alkali-rich magmatic rocks; (ii) carbonaceous and phosphatic sedimentary rocks; and (iii) pyritiferous quartz-pebble conglomerate and low-medium grade metamorphic rocks like phyllite and schist (for U) and high-grade metamorphic rocks like granulite (for Th), with Th/U value generally ranging from <1 to 5. These rocks generally constitute the source for these elements. Thus, the major *source* for U and Th in the (i) magmatic system are acidic – ultra-acidic rock types (rhyolites, granitoids and pegmatites), in which the common U-Th minerals are high-temperature Th-bearing uraninite, uranothorite, thorianite and thorite, which are associated with refractory ‘Rare Metal and Rare Earth (RMRE) minerals’ like betafite, samarskite, brannerite, davidite and columbite-tantalite, as

noted in the granitoids of Kullampatti in Tamil Nadu (Roy *et al.*, 2004), Kanigiri in Andhra Pradesh (AP) (Thirupathi *et al.*, 1996), quartz-syenites in the Dhurakanatagiri in Meghalaya and Binda-Nagnaha in Bihar (Dhana Raju, 1989); (ii) sedimentary system are black shale and phosphatic rocks, in which U is generally in the adsorbed state, associated with organic matter, clays and phosphatic minerals, as recorded in the phosphorites of Mussorie in Uttarakhand, Sonrai in Uttar Pradesh and phosphatic dolostone (with collophane) in the Tummalapalle – Giddankipalle in AP (Dhana Raju *et al.*, 1993; Roy and Dhana Raju (1997,1999); and (ii) metamorphic system, low-medium grade metamorphic rocks (schists) for U, as in the Singhbhum shear zone in Jharkhand (Rao and Rao,1983), and high-grade metamorphic rocks for Th, like granulites that constitute the source for Th-U-bearing minerals of monazite and zircon, which occur in exploitable grade in placer mineral-sands of the East and West coasts of India (see papers in EARFAM, 2001; Dhana Raju, 2007).

Host-Rocks: The *host-rocks* for major types of U-mineralization in India are diverse. These include (i) low-medium grade schists with major minerals of chlorite-quartz-sericite-biotite, apatite-magnetite-tourmaline, quartzite and conglomerate in the Singhbhum shear zone in Jharkhand (Rao and Rao, 1980 and 1983; Sarkar, 1995) and the Shahabad limestone and its underlying biotite granitoid in Gogi in Karnataka (Achar *et al.*, 1997; Pandit , 2002; Pandit *et al.*, 2002; Dhana Raju *et al.*, 2002) for *hydrothermal type*; (ii) biotite granitoid and its overlying quartzite in the Lambapur – Peddagattu – Chitrial - Koppunuru in AP for *unconformity-proximal type* (Shrivastava *et al.*, 1992; Sinha *et al.*, 1995; Jeyagopal *et al.*, 1996); (iii) quartz/feldspathic arenite in the Gomaghat - Domiasiat – Wakhyn area in Meghalaya for *sandstone type* (Saraswat *et al.*, 1977; Dhana Raju *et al.*, 1989; Kaul and Varma, 1990; Kumar *et al.*, 1990; Sen *et al.*, 2002); (iv) impure (phosphatic, siliceous) dolostone in the Tummalapalle – Giddankipalle and adjoining area in AP for *stratabound carbonate type* (Vasudeva Rao *et al.*, 1989; Dhana Raju *et al.*, 1993; Roy and Dhana Raju, 1997); (v) albitite, quartz-albitite, albititized schist and albititized pyroxenite in the Rohil-Ghateswar area in Rajasthan for *albitite type* (Yadav *et al.*, 2002); (vi) quartz-pebble conglomerate (QPC) and meta-arenite (mica-quartz schist), respectively, in the Walkunji (Aurora, 1985; Viswanath *et al.*, 1988 a) and Arbail-Dabguli (Viswanath *et al.*, 1988 b; Varma *et al.*, 1988) areas in Karnataka for *paleo-placer type*; and (vii) A-type granitoids in the Kullampatti in Tamil Nadu (Roy and Dhana Raju, 1999; Roy *et al.*, 2004) and Kanigiri in AP (Banerjee *et al.*, 1983; Thirupathi *et al.*, 1996; Dharma Rao and Reddy, 2007) for *syn-magmatic type*. The major U-minerals in these are medium-low temperature Th-poor uraninite, pitchblende and coffinite in the first four types, whereas in the last three types they are high-temperature Th (up to 10 wt. %)-bearing uraninite, uranothorite, brannerite, davidite and fergusonite (Dhana Raju, 2005, 2007 and 2009). Generally, the grain-size of primary U-minerals in the paleo-placer, syn-magmatic and albitite types is relatively of bigger size (0.05 to 0.2 mm) as compared to that (a few microns to ~0.02 mm) in the other types like hydrothermal, stratabound dolostone-hosted and sandstone types (Dhana Raju, 2009, p. 383). Chemically, pitchblende and coffinite from the hydrothermal type in limestone from Gogi contain the highest contents of U – 80 and 78 wt. %, respectively, whereas uraninite from the paleo-placer type analyzed the highest contents of Th (15.2 wt. %) and Pb (18.8 wt. %) (Dhana Raju, 2010). The high-medium temperature uraninite is characterized by high-medium value for its unit-cell dimension (a_0 , 5.44 - 5.47 Å) and low oxidation grade (OG,

UO_{2.0 - 2.3}) in contrast to low-temperature pitchblende, marked by low a_0 value (5.40 - 5.43 Å) and high OG (UO_{2.3 - 2.65}) (Chaki, 2010).

Age: Both U and Th, due to their radioactive property, continuously decay with emission of α - and β -particles, and γ -rays, in a series of stages, to yield ultimately stable, radiogenic Pb. Thus, ²³⁸U, ²³⁵U and ²³²Th, respectively, decay to yield finally ²⁰⁶Pb, ²⁰⁷Pb and ²⁰⁸Pb. Due to this, with increase in the age of U- and Th-minerals, there should be a decrease in their contents of U and Th, and corresponding increase in their radiogenic Pb content. Accordingly, *age-wise*, the oldest (late Archean/early Proterozoic) paleo-placer QPC type U-mineralization in Walkunji, Karnataka, in the form of Th-bearing uraninite, contains the highest content of radiogenic Pb (6.5 to 18.8 wt. %), in contrast to the youngest (Upper Cretaceous and Neogene) sandstone type in Meghalaya and Siwaliks of the Himalaya like at Loharkar, Himachal Pradesh, in the form of coffinite, which contains very low content of Pb (1.2 to 0.0 wt. %), while other types of U-mineralization like syn-magmatic, hydrothermal and stratabound, dolostone-hosted syn-/dia-genetic types of mostly Proterozoic age, in the form of uraninite, pitchblende, coffinite and U-Ti complex, contain an overall intermediate content of radiogenic Pb (0.0 to 11.3 wt. %) (Dhana Raju, 2010).

Mineralization-Processes: Amongst many diverse *mineralization-processes*, the important ones for U-mineralization are: syn-magmatic, syn-/dia-genetic sedimentary, epigenetic, metamorphic-metasomatic, placer and remobilization - transportation - precipitation. The syn-magmatic, Na-metasomatic and placer types are characterized by high-temperature, Th-/RE-bearing uraninite, brannerite, uranothorite, fergusonite, samarskite, betafite, columbite - tantatite and xenotime. The epigenetic type mineralization in unconformity-proximal, hydrothermal and stratabound, syn-/dia-genetic dolostone-hosted types is manifested as medium-low temperature minerals of Th-poor uraninite, pitchblende, coffinite, U-Ti complex and a host of low-temperature, secondary U-minerals like uranophane and autunite. The remobilization – transportation - precipitation process of U-mineralization in the sandstone-type is manifested in the form of low-temperature pitchblende and coffinite, besides a host of secondary U-minerals (Dhana Raju, 2007). The economically-exploitable Th-mineralization in India is mainly in the form of placer type monazite, recorded in the coastal mineral-sands (see papers in EARFAM, 2001), derived mainly from granulites, and also in the riverine (eluvial, colluvial and alluvial) placers, derived from granitoids-pegmatites in the Siri river basin area, Jashpur district of Chattisgarh ((Ramesh Babu, 1999) and the Deo river area in the Gumla district Jharkhand (Sinha, 1999).

Ore Processing: Mineral processing of radioactive ores involves 3 main stages, viz., *ore preparation* (crushing and grinding of Run-Of-Mine (ROM) ore, and its sizing and classification) to liberate minerals of U and Th; *physical beneficiation* to concentrate these minerals by methods of gravity separation, flotation and magnetic-/electromagnetic-separation; and *hydrometallurgy* for extraction of the metal (U) by different leaching-techniques (heap, percolation, pressure, puchuka, hot-agitation, acid-/alkali-leach), solvent extraction, filtration, ion (cationic or anionic) exchange and precipitation. Selection of specific techniques for a particular ore depends to a large extent on bulk chemistry, mineralogy, morphological characters of ore minerals and textures, and association of ore and gangue minerals in an ore (Dhana Raju, 2009, p. 382-386). To extract nuclear fuels of U and Th from their respective ores, the type and nature of occurrence of their U-/Th-minerals, nature of host-rock and the associated gangue and other ore minerals in the ores all make every ore unique and it would be prudent to study the mineralogy of the ore before even starting ore-processing studies (Padmanabhan and Sreenivas, 2010). Thus, the type or route of cost-effective extraction of U and Th from their respective ores depends to a large extent

on both the nature of gangue of the ores and their constituent U-Th minerals. For example, extraction of U from the U-ores with silicate-gangue, like the hydrothermal type in schists in the SSZ, Jharkhand, unconformity-proximal type in granitoid in the Lambapur – Peddagattu – Chitrial - Koppunuru area in AP and the sandstone type in the Wahkyn - Lambapur area in Meghalaya requires the acid-route, using sulphuric acid, whereas the alkali route, using sodium-carbonate and –bicarbonate, is required for the carbonate ores like that in dolostone-hosted U-mineralization in the Tummalapalle – Giddankipalle area in AP (Dhana Raju, 2009, p. 358-359). U-minerals from which U can be extracted easily by either of the above routes include uraninite, pitchblende, coffinite and secondary U-minerals (some of which are even water-extractable, as in the sandstone type ore in Domiasiat-Wahkyn in Meghalaya), whereas it is difficult to extract U from the refractory minerals like brannerite, davidite, betafite, samarskite, columbite - tantalite, pyrochlore - microcline, etc., all of which require very high acid-concentration and/or high-temperature/pressure. Similarly, U that is adsorbed with clays, phosphates and organic matter, and/or associated with lighter minerals like micas and carbonate minerals (bromoform-/methylene iodide-lights) requires special conditions for extraction, e.g., alkaline pressure-leaching technique, filtration on a continuous belt filter and counter current washing of the filter cake for the carbonate U-ore from the Tummalapalle - Giddankipalle area in AP (Padmanabhan and Sreenivas, 2010). Importance of process mineralogy (involving bulk chemistry of ore, identification, nature of association and quantification ore- and gangue-minerals, and study of morphological features of ore minerals and textures of ore- and gangue-minerals) on pre-concentration of U-minerals and extraction of U is documented in case studies on U-ores of Jaduguda and Narwapahar in the Singhbhum shear zone, Domiasiat in Meghalaya and Tummalapalle in AP by Padmanabhan and Sreenivas (2010), and Rohil in Rajasthan by Madakkaruppan *et al.*, (2010). Beneficiation of Th-bearing monazite from heavy mineral-sands and gravels, derived from granite-pegmatites, involves mainly gravity and electro-magnetic separations under varying conditions.

CONCLUSIONS

1. Nuclear fuels, from which nuclear power is generated, are Uranium (U) and Thorium (Th), and products derived from them due to neutron-absorption, viz., Plutonium (^{239}Pu) and ^{233}U . India's 3-stage nuclear power programme envisages (i) natural U-fuelled Pressurized Heavy Water Reactors (PHWR) in the 1st stage, (ii) Pu-fuelled fast breeder reactors in the 2nd stage and (iii) ^{233}U -fuelled (derived from natural Th) breeders in the 3rd stage.
2. U and Th occur in ~200 discrete 'primary' and 'secondary' minerals, and a large number of rock-constituting accessory minerals. Besides, U, as a substituting ion, occurs in ~20 U-niobates, -tantalates and -titanates, and in adsorbed state, is associated with clays, carbonaceous matter, hydrated Fe-oxides, etc.
3. The major *source* for U and Th in the (i) magmatic system are acidic – ultra-acidic rock types; (ii) sedimentary system are black shales and phosphatic rocks and (iii) metamorphic system, low-medium grade phyllites and schists for U, and high-grade granulites for Th-U-bearing minerals of monazite and zircon in placer coastal mineral-sands.
4. *Host-rocks* for the major types of U-mineralization in India are diverse and include (i) low-medium grade schists, limestone and biotite granitoid for *hydrothermal type*, (ii) biotite granitoid and quartzite for *unconformity-proximal type*, (iii) quartz/feldspathic arenite for *sandstone type*, (iv) impure (phosphatic, siliceous) dolostone for *syn-/dia-genetic, stratabound carbonate type*, (v) albitite, quartz-albitite, albititized schist and albititized

pyroxenite for *albitite type*, (vi) quartz-pebble conglomerate (QPC) and meta-arenite, for *paleo-placer type* and (vii) A-type granitoids for *syn-magmatic type*. The major U-minerals in these are medium-low temperature Th-poor uraninite, pitchblende and coffinite in the first four types, whereas in the last three types they are high-temperature Th (up to 10 wt. %)-bearing uraninite, uranothorite, brannerite, davidite and fergusonite.

5. *Age-wise*, the oldest (late Archean/early Proterozoic) paleo-placer QPC type U-mineralization contains the highest radiogenic Pb (6.5 to 18.8 wt. %) and the youngest (Upper Cretaceous and Neogene) sandstone type contains very low Pb (1.2 to 0.0 wt. %), while that of the Proterozoic age contains intermediate contents of Pb.

6. Amongst many *mineralization-processes*, the important ones for U-mineralization are syn-magmatic, epigenetic, syn-/dia-genetic, metamorphic - metasomatic, placer and remobilization - transportation - precipitation, and for Th-mineralization is placer.

7. *Ore processing* of the U- and Th-ores involves ore preparation for liberation, physical beneficiation for pre-concentration and hydrometallurgy for extraction; selection of techniques for each stage is dependent on bulk chemistry, nature and association of ore- and gangue-minerals, and their morphological characteristics and textures.

8. Relationship of the above *parameters* to the mineralogy and geochemistry of the nuclear fuels of U and Th is presented, with some Indian examples.

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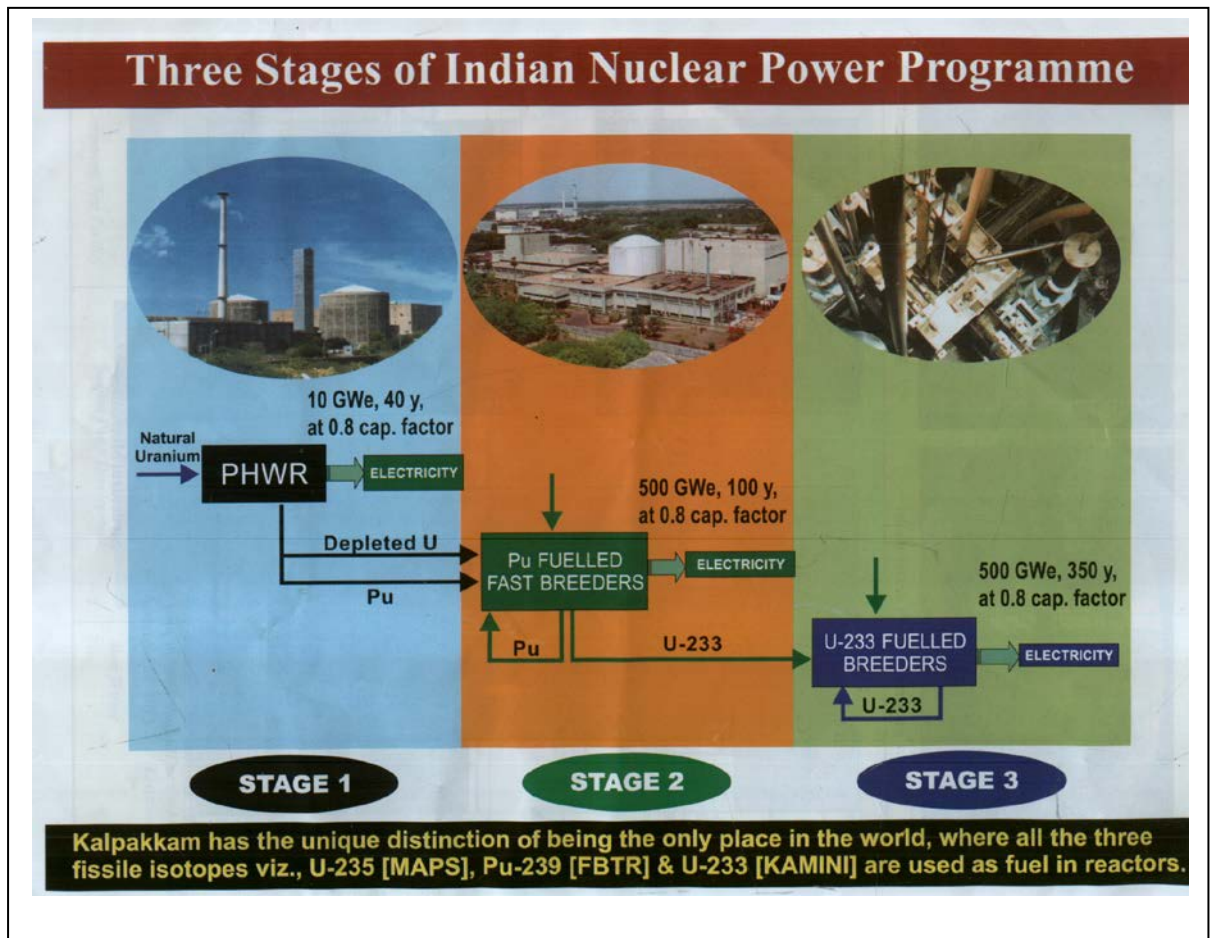


Fig. 1. Three stages of the Indian Nuclear Programme (Source: 'Nuclear India', p. 6, v. 38, nos. 3-4, Sept.- Oct. 2004, Dept. of Atomic Energy, Govt, of India, Mumbai-1).

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