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High temperature reactors

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ABSTRACT

With the advent of high temperature reactors, nuclear energy, in addition to producing electricity, has shown enormous potential for the production of alternate transport energy carrier such as hydrogen. High efficiency hydrogen production processes need process heat at temperatures around 1173–1223 K. Bhabha Atomic Research Centre (BARC), is currently developing concepts of high temperature reactors capable of supplying process heat around 1273 K. These reactors would provide energy to facilitate combined production of hydrogen, electricity, and drinking water. Compact high temperature reactor is being developed as a technology demonstrator for associated technologies. Design has been also initiated for a 600 MWth innovative high temperature reactor. High temperature reactor development programme has opened new avenues for research in areas like advanced nuclear fuels, high temperature and corrosion resistant materials and protective coatings, heavy liquid metal coolant technologies, etc. The paper highlights design of these reactors and their material related requirements.

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1. Introduction

In the Indian context, development programme for high temperature reactors and its process heat utilisation are significant mainly for non-electric applications. Primarily the requirement for high temperature reactors has originated because of an urgent need to develop alternate energy carrier to substitute petroleum-based transport fuel, which has very small reserves in India and results in large import bills. Lately prices of petroleum-based products have shot up internationally increasing Indian import burden further. This trend is likely to continue due to dwindling resources and increasing demands of oil worldwide. It has become inevitable that India find an alternative to petroleum-based products for transport applications. Hydrogen is being considered as an attractive energy carrier for transport applications. It is an energy carrier like electricity – a way of transporting useful energy to consumers and is a versatile carrier because like oil and gas, it can be stored in large amounts and can be made from almost any energy source and used to provide almost any energy service and can be either used in internal combustion engines like petrol and diesel or easily converted to electricity using fuel cells. Thus it is envisaged to form an important component of future energy mix in India.

Hydrogen is available in abundance, but is not an energy source like oil, coal, wind, or sun. This is because it is not available in nature; the way other resources are found and is attached to other elements forming compounds. It must be freed from chemical compounds in which it is bound up. Therefore hydrogen production options basically deal with separating hydrogen from sources like fossil fuel, biomass, or water. All separation processes are highly energy intensive requiring energy in the form of either heat or electricity or both. Large quantities of hydrogen can be produced either by splitting water or from fossil fuels like coal and natural gas. Hydrogen is separated from fossil fuel or biomass by reforming processes using heat with or without electricity. Hydrogen can be obtained from water by splitting it either by electrolysis or by thermo-chemical processes. Table 1 shows process related parameters for some of the thermo-chemical processes, which have been researched in greater details. These processes are highly energy intensive and require temperatures generally exceeding 823 K, depending on the process used. These processes need either electricity or process heat at high temperatures, or both depending upon the process of hydrogen production selected. The efficiencies for these hydrogen-producing processes are higher at higher temperatures. Electrolysis, if carried out at high temperatures, saves electricity and is more efficient. Thermo-chemical processes for splitting water to produce hydrogen have a very high efficiency (40–57%), but needs process heat at 823–1173 K [1], depending on the thermo-chemical process selected. Iodine–sulfur based thermo-chemical process has the highest reported conversion efficiency but needs process heat at a temperature around 1173 K. The input energy needed for these processes can be provided by a suitable high temperature heat source. Due to long term sustainability of nuclear and solar energy, they have attracted a lot of attention worldwide, for providing required energy for hydrogen separation. More over these sources are environmentally benign. High temperature nuclear reactors capable of supplying process heat have a large potential for sustainably supplying energy for these hydrogen

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Table 1
Process related parameters for few thermo-chemical processes for hydrogen production

	Iodine–sulfur (IS) process	Calcium–bromine (Ca–Br) process	Copper–chlorine (Cu–Cl) process	Hybrid sulfur* (Hy-S) process
Efficiency (%)	57	40	41	40
Operating temperature (K)	1223	1003	773–823	1123–1223
Process streams	Liquid and gas	Liquid, gas and solids	Liquid, gas and solids	Liquid and gas
Development stage	Fully flow sheeted	Fully flow sheeted	Flow sheeted	Flow sheeted
Demonstration	Pre pilot plant	Pilot plant	Laboratory scale	Bench scale
Corrosion	High	High	Low	High

production processes at required high temperature conditions. Such reactors, even if used for electricity generation, would result in much higher efficiencies as compared to conventional nuclear reactors. Advanced and high efficiency generation technologies such as Magneto-hydro-dynamics or gas turbines can be integrated with such high temperature reactors.

2. Indian high temperature reactor programme

As stated earlier, the current Indian high temperature nuclear reactor programme is mainly based on requirements related to production of hydrogen from water by high efficiency thermo-chemical processes. BARC is currently developing concepts of high temperature nuclear reactors capable of supplying process heat at a temperature around 1273 K. These nuclear reactors are being developed with the objective of providing energy to facilitate combined production of hydrogen, electricity, and drinking water. The reject and waste heat in the overall energy scheme are utilised for electricity generation and desalination, respectively. Presently, technology development for a small power (100 kWth) Compact High Temperature Reactor (CHTR) capable of supplying high temperature process heat at 1273 K is being carried out. In addition conceptual details of a 600 MWth reactor supplying heat at 1273 K for commercial hydrogen production, are also being worked out.

3. Compact high temperature reactor

CHTR [1–3] is being developed as a platform to launch a focussed programme for the development and demonstration of technologies associated with high temperature reactors. The reactor has a modular design with shop fabrication of most of the modules. It is being designed to be compact in weight and size for ease in its delivery to remote locations for its use as a compact power pack. The reactor core consists of nineteen prismatic beryllium oxide (BeO) moderator blocks. These 19 blocks contain centrally located graphite fuel tubes. Each fuel tube carries fuel inside 12 equi-spaced longitudinal bores made in its wall. The fuel tube also serves as coolant channel. The fuel is based on TRISO coated particle fuel, which can withstand very high temperatures (1873 K) and provide very high burn-up. Eighteen blocks of beryllium oxide reflector surround the moderator blocks. These eighteen blocks have central holes to accommodate passive power regulation system. This system works on temperature feedback, and in case of rise of coolant outlet temperature beyond design value, inserts negative reactivity inside the core. Graphite reflector blocks surround these beryllium oxide reflector blocks. This part of the reactor is contained in a shell of a material resistant to corrosion against lead–bismuth eutectic alloy coolant, and suitable for high temperature applications. Top and bottom closure plates of similar material close this reactor shell. Lead–bismuth eutectic alloy, having low melting point (396 K) and high boiling point (1943 K), has been selected as the coolant. Above the top cover plate and below the bottom cover plate, plenums are provided for coolant leaving and entering the core respectively. These plenums have graphite flow guiding blocks, having passages for coolant flow, to increase the velocity of the coolant between the fuel tube and down comer tube provided for return of the cold coolant to lower plenum. The reactor shell is surrounded by two gas gaps that act as insulators during normal reactor operation and reduce heat loss in the radial direction. A passive system has been provided to fill the gas gaps with molten metal in case of abnormal rise in coolant outlet temperature. There is an outer finned steel shell, surrounded by heat sink. Nuclear heat from the reactor core is removed passively by natural circulation based coolant flow between the two plenums, upward through the fuel tubes and returning through the downcomer tubes. On top of the upper plenum, the reactor has multi-layer heat utilisation vessels to provide an interface to systems for high temperature heat applications. A set of sodium heat pipes is provided in the upper plenum of the reactor to passively transfer heat from the upper plenum to the heat utilisation vessels with a minimum drop of temperature. Another set of heat pipes transfers heat from the upper plenum to the atmospheric air in the case of any postulated accident. Cross-sectional layout of the reactor core is shown in Fig. 1.

To shut down the reactor, a set of seven tungsten shut-off rods has been provided, which fall by gravity in the central seven coolant channels. Appropriate instrumentation like neutron detectors, fission/ion chambers, various sensors, and auxiliary systems such as a cover gas system; purification systems, active interventions etc. are being incorporated in the design as necessary. Component layout of CHTR is shown in Fig. 2 and major design and operating parameters are shown in Table 2.

3.1. CHTR fuel

The CHTR fuel [4] is designed to operate at high temperatures, withstand high burn-up and has long core resident time. The important design parameters of the fuel are shown in Table 2. Thorium and burnable poison make the fuel temperature coefficient negative, thus making the reactor inherently safe. A typical CHTR fuel bed consists of prismatic BeO moderator block with centrally located graphite fuel tube carrying fuel compacts. Schematics of fuel particle, fuel compact, and single fuel bed are shown in Fig. 3.

A typical TRISO coated fuel particle has a kernel (500 μm diameter) comprising of fissile, fertile, and burnable poison materials followed by four coating layers. The functional requirements and proposed dimensions of these layers are given below:

(a) Low-density pyrolytic carbon (PyC) buffer layer: This porous layer (90 μm thick) acts as an absorber for fission recoils and provides volume to accommodate fission products and kernel swelling.
(b) Inner high-density PyC layer: This layer (30 μm thick) serves as a barrier to gross diffusion of fission products and fission gases. This is to protect integrity of subsequent SiC layer.
(c) Silicon carbide (SiC) interlayer: This layer (30 μm thick) contains gaseous fission products released by the kernel and thus acts like a pressure vessel. This also acts as an

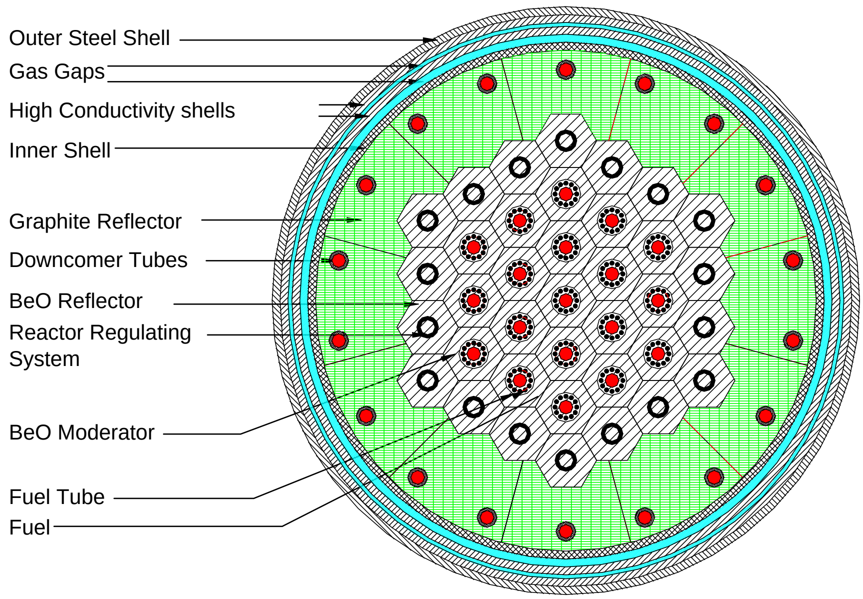

Fig. 1. CHTR core cross-sectional layout.

additional diffusion barrier to metallic fission products. The thickness needs to be adequate to withstand the developed pressure and corrosion attacks by fission products.

(d) Outer high-density PyC layer: This layer (50 μm thick) as well as inner PyC layer, on irradiation, puts SiC layer into compression to limit stresses. Additionally it provides chemical protection to SiC layer. It also provides bonding surface for making compacts.

3.1.1. R & D issues

Since very large number of TRISO particles (about 13.5 millions) comprise the core, manufacturing of fuel kernels, deposition of multilayered coatings on the particles, and characterisation of the coated as well as uncoated particles pose special challenges. As regards kernel fabrication [5], BARC has developed technologies related to oxide and carbide based micro sphere fabrication by Internal Gelation Process (IGP). The oxide micro spheres, are sintered to produce >99% TD micro spheres. The IGP flow sheet is modified for preparation of carbide micro spheres. Carbon powder is added in the feed solution prior to mixing of metal nitrate solutions. Vacuum furnace is used for conversion of heat-treated gel particles to carbide micro spheres. Sintering is carried out in high purity argon. As regards coatings development, coatings of PyC and SiC on fuel kernel are formed by chemical vapour deposition (CVD) technique. These are obtained by pyrolytic decomposition of hydrocarbon gas or methyl trichloro silane (CH_3SiCl_3) vapour in fluidised/spouted beds. Different coatings are prepared at different temperatures. Various process parameters like temperature, volume of the bed, composition and flow rates of the gases, etc. control the properties of the coatings. Experimental trials for these coatings on surrogate materials have been [6] carried out in BARC. Trials are continuing to optimise the parameters. Maintaining a stringent quality control would be important to reduce the probability of failed fuel particles. In addition, manufacturing of fuel compacts with isotropic properties and without damaging the constituent particles also pose special challenges. Characterisation techniques need to be developed and facilities to characterise the particles at different stages of its manufacture need to be setup. Besides manufacturing, analytical modelling and experimental studies for post irradiation and high temperature behaviour of particles open new avenues of R & D activities. These are important to understand the long-term behaviour of the fuel. It would be important to statistically demonstrate through analytical studies that the number of failed particles within the full core is less than the decided design limit.

3.2. CHTR core materials

CHTR core materials are exposed to extreme environmental conditions. They need to withstand neutron fluence, high temperatures, (around 1273 K) and corrosive environment of lead–bismuth eutectic alloy based coolant. CHTR core internal materials comprise BeO moderator and reflector blocks, graphite fuel and down comer tubes, and graphite reflector blocks. The BeO blocks required are high-density nuclear grade materials, hexagonal in shape with 0.135 m across the flats. Graphite reflector blocks are irregular shaped and are about 1.15 m long. Graphite fuel tube is a 1.4 m long cylindrical tube (0.035 m inner diameter with 0.020 m wall thickness) with 12 nos. of 0.010 m diameter – 1.05 m long bores made in thickness to accommodate fuel compacts. R&D issues related to these components are listed below:

3.2.1. Design related issues

For conventional nuclear reactors there are number of design rules and codes available for carrying out the design of core internal components and core support structures, e.g. Sections of ASME Boiler & Pressure Vessel Code. However, there is, as yet, no similar pre-established, authoritative, and sanctioned set of design rules to provide guidance for design of corresponding brittle graphite and BeO components. The issues involved in the design of such components are summarized below:

(a) In designing with brittle materials, two behavioural characteristics invalidate the usual deterministic design procedures – the statistical nature of its strength value and its large variation. In such a case statistical design techniques need to be resorted to. One of the most widely known methods is by the use of Weibull statistics, which can be used to calculate the survival probability in a given stress distribution.

(b) For multiaxial cases, the failure probabilities may be calculated on the basis of many different models. But none of the models exhibit all the behaviours shown by graphite or BeO.

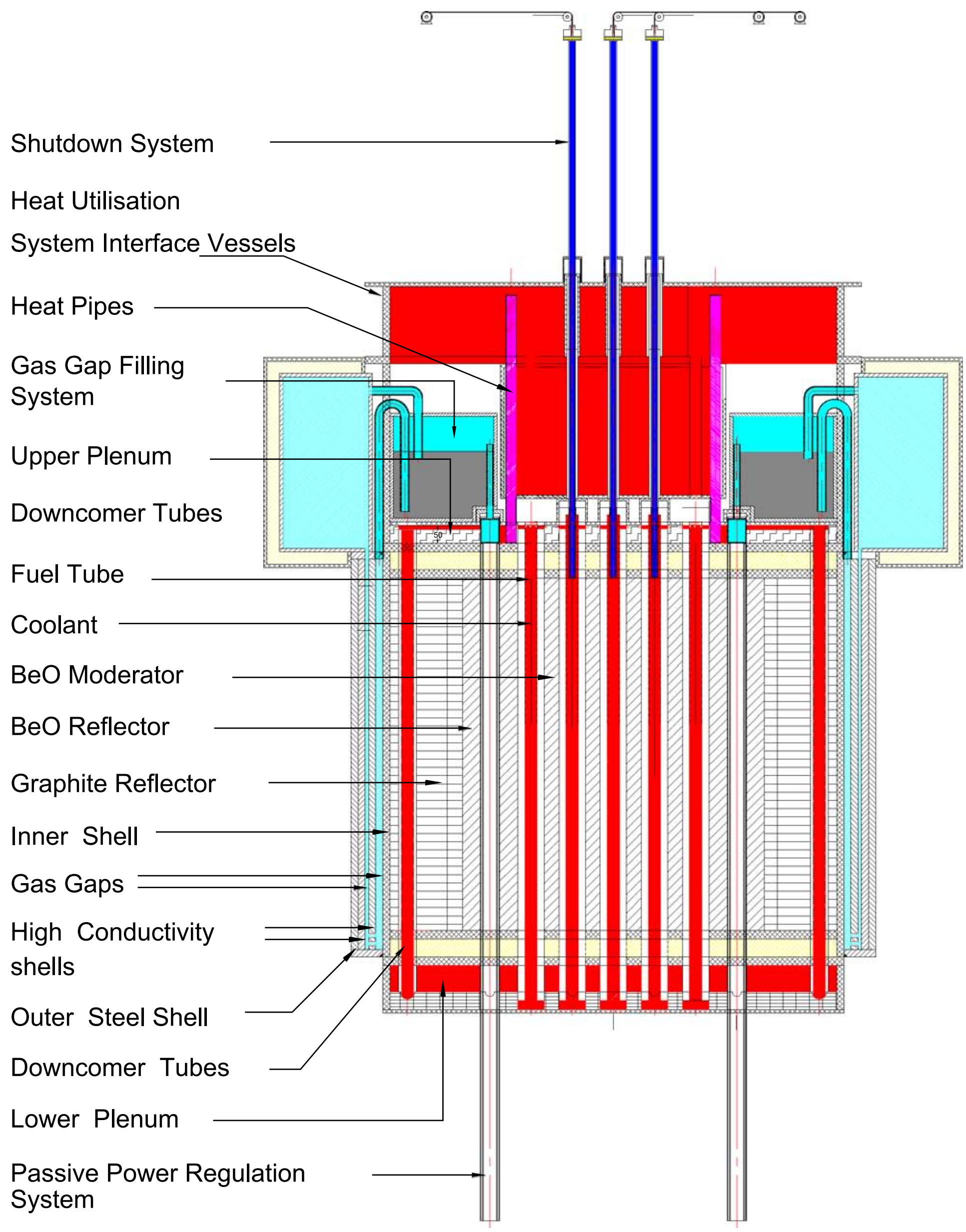

Fig. 2. Layout of CHTR components.

(c) To accurately determine the value of the various statistical parameters, a large number of samples need to be mechanically tested.

(d) Fatigue curves follow the same pattern as that of metals when drawn with homologous stress (peak stress divided by mean tensile strength) vs. number of cycles, but with a larger scatter in data.

Based on assessment of two draft codes, viz, ASME Section III Division 2 Subsection CE and German code KTA 3232, Ceramic internals for HTR pressure vessels, 1992, work [7] has been initiated to formulate design rules for such brittle components.

3.2.2. Compatibility issues

Inter-compatibility of BeO and graphite components with each other and also with lead–bismuth eutectic alloy based coolant at high temperatures is an important issue which might decide the useful life of the core components. Penetration of coolant into open porosities and its behaviour during thermal cycling especially during shut downs and cold start up might be another issue which needs to be thoroughly studied. Impervious and oxidation resistant coatings (PyC followed by SiC) on graphite and their behaviour when exposed to irradiation and thermal cycling is another issue requiring detailed study. A liquid metal loop [8] is being commissioned to carry out coolant-material compatibility studies.

3.2.3. Irradiation behaviour of BeO

Irradiation behaviour of BeO and degradation of properties after irradiation is another area requiring developmental activities. Changes in property under neutron irradiation, is needed to evaluate temperature and stresses prevalent in BeO at various stages during the lifetime of the reactor. Irradiation of BeO [9] under fast neutron flux causes two distinct effects. For lower fluence, the material remains coherent, but at higher fluence grain boundary separation takes place i.e. microcracking occurs. The fluence at which microcracking starts has been observed to be higher for higher temperatures. In addition, BeO of lower density and finer

Table 2
Major design and operating parameters of CHTR

Attributes	Design parameters
Reactor power	100 kWth
Core configuration	Vertical, prismatic block type
Fuel	$^{233}UC_2 + ThC_2$ based TRISO coated fuel particles shaped into fuel compacts with graphite matrix (Gd – only central fuel tube)
Fuel enrichment by ^{233}U	33.75 wt%
Refuelling interval	15 effective full power years
Fuel burn-up	≈68 000 MWd/t of heavy metal
Moderator	BeO
Reflector	Partly BeO and partly graphite
Coolant	Molten lead–bismuth eutectic alloy (44.5% Pb and 55.5% Bi)
Mode of core heat removal	Natural circulation of coolant
Coolant flow rate through core	6.7 kg/s
Coolant inlet temperature	1173 K
Coolant outlet temperature	1273 K
Loop height	1.4 m (actual length of the fuel tube)
Core diameter	1.27 m (including radial reflectors)
Core height	1.0 m (Height of the fuelled part and axial reflectors)
Passive reactor regulation system	18 B_4C elements of passive power regulation system
Passive reactor shutdown system	Seven mechanical shut-off rods

grain size withstand radiation better at both low and high temperatures. If microcracking does not occur, the strength, elastic constants, and thermal expansion either remain same or increase slightly. However, thermal conductivity is highly sensitive to neutron irradiation. Even in this case the change in thermal conductivity at elevated temperatures are considerably smaller than those obtained by irradiation at room temperatures. With the onset of microcracking, all these properties deteriorate. BeO also undergoes severe creep above 1373 K. Coarse grain structures are said to flow plastically above 1223–1273 K. The effects of high temperature creep are likely to increase due to irradiation creep and growth. High temperature creep behaviour needs to be established through experiments. Therefore dimensional changes, irradiation and thermal creep, and degradation of thermal properties are important factors to be considered in the design of the core components. Irradiation programme for samples of BeO developed in BARC is being worked out.

3.2.4. Irradiation behaviour of graphite

Irradiation data is needed in several areas as far as mechanical and thermal behaviour of graphite core components are concerned. Dimensional changes, irradiation creep, and degradation of thermal properties are important factors to be considered in the design of the core components. Irradiation behaviour of graphite has been very well studied internationally [10]. Efforts are being made to develop carbon–carbon composite materials with the help of other laboratories in India and carry out their irradiation trials in order to study their post irradiation behaviour.

3.3. Metallic materials

Reactor vessel of CHTR and other metallic components such as driver and control tubes of passive power regulation system and heat pipes are exposed to coolant at a temperature around 1273 K. Therefore the issues which are important for the structural material are chemical compatibility with coolant, oxidation resistance, irradiation behaviour, neutron absorption cross-section, activation of the alloying components and impurities, pre and post irradiation strength, Ductile to Brittle Transition Temperature of initial and irradiated material, and manufacturability. At such high temperatures and in contact with lead based coolant only refractory metals and their alloys have been reported to have good corrosion resistance. They have been reported to have high melting points, good thermal conductivities, high strength, low thermal expansion, and high resistance to swelling. Alloys like TZM (0.5%Ti, 0.1%Zr, trace carbon, balance Mo) and Nb-1%Zr are reported [11] to have good high temperature properties along with good corrosion resistance. But they show oxidation tendency in presence of air or oxidising atmosphere hence need to be protected by an oxidation resistant coating. Efforts are being made to develop these materials in-house as well as develop oxidation resistance coatings for such materials. In parallel efforts are being carried out to study fabrication, machinability, and irradiation behaviour aspects.

4. 600 MWth innovative high temperature reactor

Various options as regards fuel configuration and coolants were studied for a 600 MWth reactor for commercial hydrogen production. Proposed specification for this reactor is shown in Table 3. Proposed schematic of this reactor is shown in Fig. 4.

Initial studies carried out indicate selection of pebble bed reactor core with either lead or molten salt-based coolant. However this would be finalised after carrying out further studies. Many of the technologies developed for CHTR would be utilised for this reactor. There are many additional fuel and material related developmental activities, which would be initiated. Some of these are listed below:

(a) Design and development of facility for manufacture of TRISO coated particle fuel based on $^{233}UO_2$ and ThO_2.
(b) Design and development of facility for manufacture of pebble type fuel from TRISO coated particles.
(c) Design and development of experimental set-up for pebble fuel loading and unloading.
(d) Design and development of reprocessing facilities for pebble based fuel.

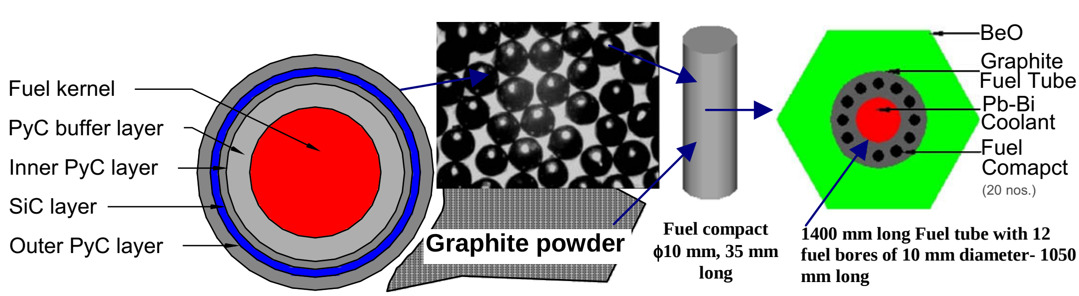

Fig. 3. Schematics of TRISO fuel particle, fuel compact, and single fuel bed for CHTR.

Table 3
Proposed general specifications of the reactor for commercial hydrogen production

Reactor power	600 MWth for following deliverables *Optimised for hydrogen production* Hydrogen: 80 000 Nm^3/h Electricity: 18 MWe Drinking water: 375 m^3/h
Coolant outlet/inlet temperature	1273 K/873 K
Moderator	Graphite
Coolant	Molten lead or molten salt
Reflector	Graphite
Mode of cooling	Natural circulation of coolant may be considered
Fuel	$^{233}UO_2$ and ThO_2 based high burn-up TRISO coated particle fuel
Control	Passive power regulation and reactor shutdown systems
Energy transfer systems	Intermediate heat exchangers for heat transfer to helium or other medium for hydrogen production + High efficiency turbo-machinery for electricity generation + Desalination system for potable water
H_2 production	High efficiency thermo-chemical processes

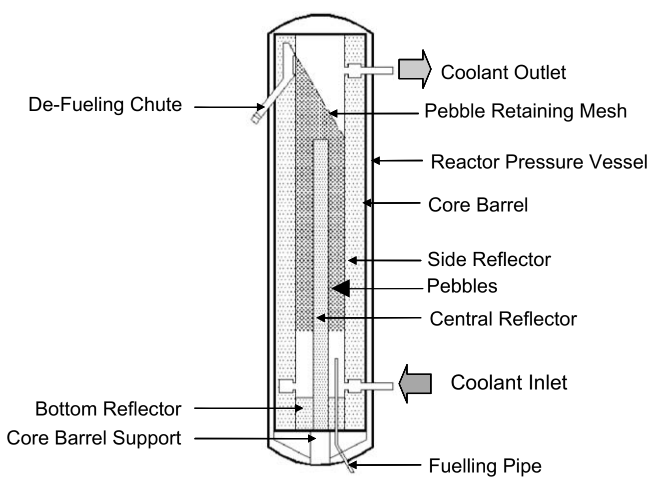

Fig. 4. Preliminary schematic of core configuration of 600 MWth pebble bed Indian HTR.

(e) Manufacture of large size components made of high-density, isotropic, and nuclear grade graphite.
(f) Design and development of experimental set-up for cooling of pebbles by coolants such as lead or molten salt.
(g) Design and development of heat exchangers for different coolant combinations.
(h) Design and development of structural materials for compatibility with lead/molten salt at high temperatures.
(i) Design and development of coatings for corrosion and oxidation resistance.
(j) Analytical and experimental safety related studies under various conditions such as air ingress, loss of coolant, loss of heat sink etc.
(k) Analytical and experimental seismic studies on components.
(l) Design and development work related to materials for systems integrating nuclear reactor and hydrogen production plant.
(m) Safety studies related to explosion of hydrogen and pressure wave produced and its effect on structures and materials.

5. Summary

Indian high temperature reactor and its associated programmes pose several fuel and material related challenges due to very high temperatures and aggressive environment. The R & D work encompasses selection/development of materials and their fabrication technologies for actual component manufacture, compatibility studies, oxidation and corrosion resistant coatings, joining technologies, development of high temperature high burn-up fuel, irradiation behaviour of fuel and materials, and development of characterisation techniques. These have opened many avenues of research in the field of advanced fuel and material related technologies. Developmental work has already been initiated in BARC to address many of these issues.

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