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Carbon based materials – applications in high temperature nuclear reactors

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Bhabha Atomic Research Centre (BARC) is currently developing concepts of high temperature nuclear reactors capable of supplying process heat at temperatures around 1000°C. These reactors would provide energy to facilitate production of hydrogen by a suitable high efficiency water splitting process. Currently a compact high temperature reactor is being developed as a technology demonstrator for associated technologies. Design and development has been also initiated for a 600 MWth Innovative High Temperature Reactor. Carbon based materials have important applications in these reactors. Nuclear fuel retaining elements in the form of fuel compacts and fuel pebbles, as well as neutron moderators and reflectors use carbon based materials. These materials are required to survive under extreme environmental conditions such as neutron irradiation, high temperatures around 1000°C and corrosive environment of liquid metal/salt based coolant. R & D issues involve design methodology for carbon-based components, compatibility with other materials and coolant, creep, and degradation under reactor conditions. Induced dimensional changes, irradiation creep, and degradation of thermal and mechanical properties are important factors to be considered in the design of these components. The paper highlights design of these high temperature reactors in general and their carbon based material related requirements in particular.

For India development programme for high temperature reactors and its utilisation for supplying process heat are important to develop alternate energy carrier to substitute petroleum based transport fuel, which has very small reserves in India and results in large import bills. Hydrogen is an attractive energy carrier for transport applications, and is expected to contribute to future Indian energy mix. Hydrogen production options deal with separating hydrogen from its source like water, fossil fuel, or biomass. All separation processes are highly energy intensive requiring energy in the form of either heat or electricity or both. Hydrogen can be obtained from water by splitting it either by electrolysis or by thermo-chemical processes. 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 in general are higher at higher temperatures. Thermo-chemical processes for splitting water to produce hydrogen has a very high reported efficiency (40-57%), but requires process heat at 550-900°C[1]. This energy for these processes can be provided by a suitable high temperature heat source. Nuclear and solar energy, have attracted a lot of attention worldwide, due to long-term sustainability. High temperature nuclear reactors have a large potential for sustainably supplying energy for these hydrogen production processes at required high temperature conditions.

Indian High Temperature Reactor Programme

BARC is carrying out development of concepts of high temperature nuclear reactors capable of supplying process heat at temperatures around 1000°C. These nuclear reactors are being developed so as to provide energy to facilitate combined production of hydrogen, electricity and drinking water. The reject and waste heat in the overall energy scheme are proposed to be utilised for electricity generation and desalination respectively. Currently, technology development for a small power (100 kWth) Compact High Temperature Reactor (CHTR) capable of supplying high temperature process heat at 1000°C is in progress. In addition, design and development of a 600 MWth Innovative HTR has also started.

Compact High Temperature Reactor

CHTR[1,2,5] is being developed as a technology demonstrator for the development and demonstration of technologies associated with high temperature reactors and associated systems. The core of the reactor consists of nineteen prismatic beryllium oxide (BeO) moderator blocks. These blocks contain centrally located fuel tube made of high-density

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isotropic nuclear grade carbon based material. Ideally the material shall be carbon-carbon composite, but based on current availability, high-density nuclear grade isotropic graphite has been chosen as fuel tube material. Each fuel tube carries fuel, in the form of fuel compacts, inside 12 longitudinal bores made in its wall. The inner bore of the fuel tube serves as coolant channel. The fuel is based on Tri-isotropic (TRISO) coated particle fuel, which can withstand high temperatures upto 1600°C without undergoing degradation and provide very high burnup. Six fixed and twelve movable blocks of beryllium oxide reflector surround the moderator blocks. High-density isotropic grade graphite reflector blocks surround these beryllium oxide reflector blocks. All these components are contained in a shell of a material resistant to corrosion against lead-bismuth eutectic alloy coolant, and suitable for utilisation at high temperatures.This reactor shell is covered by top and bottom closure plates of similar material. Major design and operating parameters of the reactor are shown in Table 1[5].

Lead-bismuth eutectic alloy, having low melting point (123°C) and high boiling point (1670°C), has been chosen as the coolant. Above and below the top and bottom cover plates, plenums have been provided for coolant leaving and entering the core respectively. These plenums have been provided with graphite blocks, having passages for coolant flow. Many passive systems have been provided for reactor safety, safe shutdown, and passive heat removal under all reactor-operating conditions. Cross-sectional layout of the reactor core is shown in Fig. 1. Schematic and 3D layout of the reactor is shown in Fig. 2.

TRISO coated particle fuel

The CHTR fuel[4,5] has been designed to operate at temperatures above 1000 ºC, withstand high burn up and has core resident time of about 15 years. The design parameters of the fuel are shown in Table 1. CHTR fuel bed is made up of BeO moderator block with centrally located graphite fuel tube carrying fuel compacts. Schematics of CHTR fuel is shown in Fig. 3.

TRISO coated fuel particle is made up of a centrally located kernel (500 µm diameter) comprising of fissile, fertile, and burnable poison materials followed by four coating layers. The

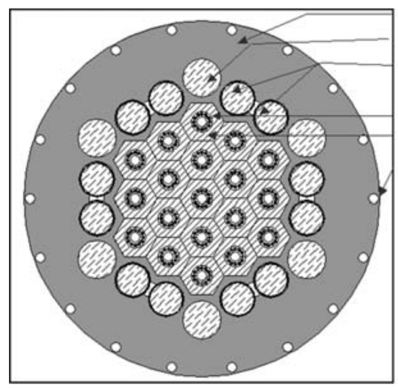

Fig. 1—Cross-sectional view of compact high temperature reactor

Table 1—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 Burnup	≈ 68000 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	900°C
Coolant outlet temperature	1000°C
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 primary reactor shutdown system	7 mechanical shut-off rods
Secondary shutdown system	Axial movement of movable BeO reflector blocks

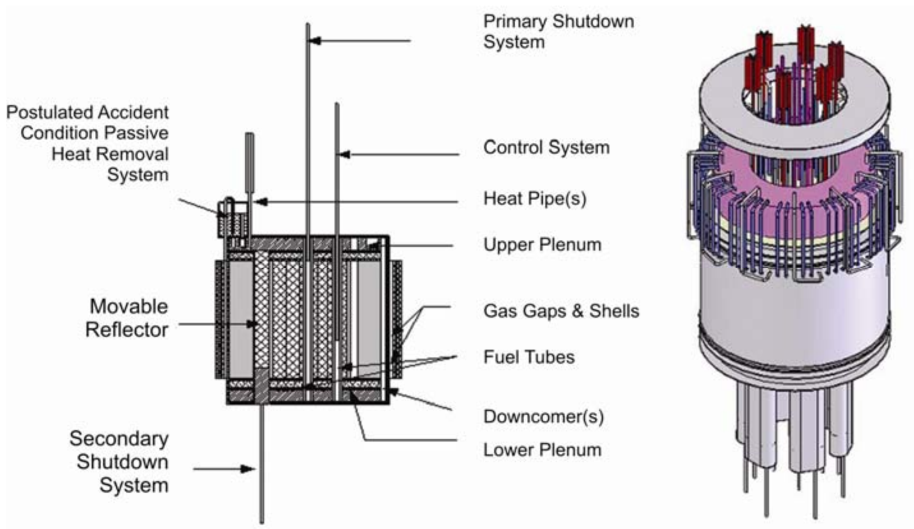

Fig. 2—Schematic and 3-dimensional layout of CHTR components

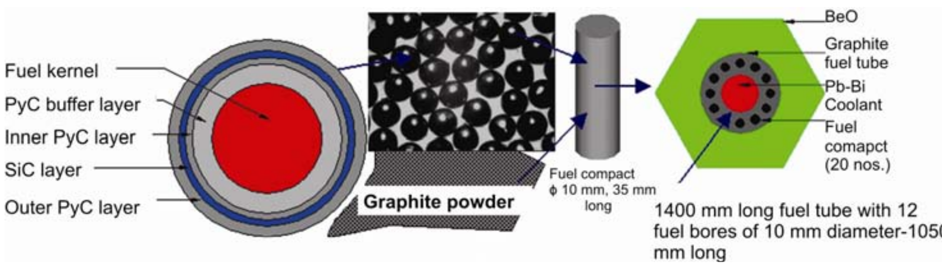

Fig. 3—Schematics of TRISO fuel particle, fuel compact and single fuel bed for CHTR

functional requirements and proposed dimensions of these layers are given below:

(i) Low-density pyrolytic carbon (PyC) buffer layer: This porous layer (90 µm thick) facilitates kernel swelling and accommodates fission products.

(ii) Inner high-density PyC layer: This layer (30 µm thick) acts as a barrier to diffusion of fission products and gases. This helps in protecting integrity of subsequent SiC layer.

(iii) Silicon carbide (SiC) interlayer: This layer (30 µm thick) retains gaseous fission products and thus acts like a pressure vessel. This also serves as an additional diffusion barrier to metallic fission products. The thickness is so chosen as to be adequate to withstand the developed pressure and corrosion by fission products.

(iv) Outer high-density PyC layer: This layer (50 µm thick) and the inner PyC layer, on irradiation, puts SiC layer into compression to limit tensile stresses developed in SiC. It provides chemical protection to SiC layer as well as a bonding surface for making compacts.

Technology challenges

The CHTR core comprises of very large number of TRISO particles (about 14 millions), manufacturing of fuel kernels, deposition of multilayered coatings on the kernels, and their characterisation pose special challenges. BARC has already developed and demonstrated technologies related to oxide and carbide based kernel fabrication[6] by internal gelation process (IGP). The oxide micro spheres, are sintered to produce > 99% theoretical density kernels. Carbide kernels are prepared by modifying the IGP flow sheet. Carbon powder is added in the feed solution prior to mixing of metal nitrate solutions. A high vacuum furnace is used for converting heat-treated gel particles into carbide kernels. Sintering of kernels is carried out in highly pure argon. For coatings development, coatings of PyC and SiC on fuel kernel are formed by chemical vapour deposition (CVD) technique. These coatings are obtained by pyrolytic decomposition of hydrocarbon gas for PyC and methyl trichloro silane (CH_3SiCl_3) vapour for SiC in fluidised/spouted beds. Different coatings need different temperatures. Process parameters like temperature, volume of the bed, composition and flow rates of the gases control the properties of the coatings. For coating technology development, presently these coatings have been made on surrogate materials in BARC[7]. Trials are being done to optimise the parameters. A stringent quality control would be important to reduce the quantity of failed fuel particles. In addition, manufacturing of fuel compacts also pose special challenges. Characterisation techniques are required to be developed and facilities to characterise the particles at different stages of its manufacture are required to be setup. Analytical modelling and experimental studies for post irradiation and high temperature behaviour of particles have opened new avenues of R & D activities, which are important to understand and predict long-term behaviour of the fuel.

Carbon based and other materials in CHTR core

Materials in CHTR core are exposed to extreme environmental conditions. They are required to withstand large neutron fluence, high temperatures (around 1000°C) and corrosive environment of the coolant. CHTR core materials comprise BeO moderator and reflector blocks, graphite fuel tube and reflector blocks. High-density nuclear grade BeO is required for the blocks, which are hexagonal in shape with 135 mm across the flats in dimensions. Graphite reflector blocks are irregular in shape and are about 1150 mm long. Graphite fuel tube is a 1400 mm long tube (with 35 mm inner and 75 mm outer diameter). This also has 12 nos. of 10 mm diameter - 1050 mm long bores made in thickness to accommodate fuel compacts. Major R & D issues related to these components are listed below:

Design methodolog[5]

Design rules and codes are available for carrying out the design of core internal components and core support structures for conventional nuclear reactors e.g., Sections of ASME Boiler & Pressure Vessel Code. However, there is, as yet, no similar pre-established, 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:

(i) For brittle materials design, two characteristics invalidate the usual deterministic design procedures - they are the statistical nature of its strength value and its large variation. Statistical design techniques are used in such cases. Use of Weibull statistics, is one of the most widely known methods, which can help in calculating the survival probability in a given stress distribution.

(ii) The failure probabilities for multiaxial cases, may be calculated on the basis of many available different models. Unfortunately none of the models include all the behaviours of graphite or BeO.

(iii) A large number of samples need to be mechanically tested so as to accurately determine the value of the various statistical parameters.

(iv) Fatigue curves exhibit the same pattern as by metals when drawn with homologous stress (peak stress divided by mean tensile strength) versus number of cycles, but with a larger scatter in data. Larger number of cycles is needed for failure when they become more compressive in nature.

Development work[8] has been initiated to formulate design rules for such brittle components. This is based on assessment of two draft codes, viz, German code KTA 3232, and ASME Section III Division 2 Subsection CE.

Material compatibility

Compatibility of BeO and graphite components with each other as well as their interaction with coolant at high temperatures is an important issue which might limit the useful life of the core components. Entry of coolant into open porosities and behaviour of penetrated cooling during thermal cycling might be another issue which needs to be studied in detail. Impervious and oxidation resistant PyC and SiC coatings on graphite and their behaviour when exposed to irradiation and thermal cycling other issues requiring detailed investigation. A lead-bismuth coolant based liquid metal loop[9] has been set-up in BARC to carry out these studies.

Behaviour under irradiation

Post irradiation data is important in several areas as far as mechanical and thermal behaviour of graphite core components are concerned. Important factors, which are usually considered in the design of components, include dimensional changes, irradiation creep, and change in thermal properties. Irradiation behaviour of graphite has been internationally[10] well studied. Efforts are also being made for development of carbon based composite materials with the help of other laboratories in the country and study their post irradiation behaviour.

600 MWth innovative high temperature reactor

BARC is carrying out design of a 600 MWth reactor for commercial hydrogen production. For this reactor various design options as regards fuel configurations, such as prismatic bed and pebble bed were considered for thermal hydraulics and temperature distribution analysis. Coolant options such as molten lead, molten salt and gaseous medium like helium were analysed. Besides these, other criteria such as ease in component handling, irradiation related material and fuel degradation, better fuel utilisation and passive options for coolant flow etc. were also considered. Studies carried out so far indicate selection of pebble bed reactor core with molten salt-based coolant. Table 2 shows proposed specification[11] for this reactor. Figure 4 shows schematic of 600 MWth Innovative HTR design. Many of the technologies developed for CHTR would be utilised for this reactor. There are plans to set-up engineering laboratories for carrying out research and development related to reactor components, coolant technologies, reactor safety, fuel and material development, and other aspects related to such high temperature reactors.

Many technologies developed for CHTR could be utilised for this bigger power reactor. There are many additional developmental activities, which would be initiated. Some of these are listed below[5]:

(i) Facility for manufacture of TRISO coated particle fuel based on $^{233}UO_2$ and ThO_2
(ii) Facility for manufacture of pebble type fuel from TRISO coated particles
(iii) Experimental set-up for pebble fuel loading and unloading
(iv) Reprocessing facilities for pebble based fuel
(v) Manufacturing facilities for large size components made of nuclear grade graphite
(vi) Experimental set-up for cooling system with pebbles and coolants such as molten salt
(vii) Heat exchangers for different coolant combinations

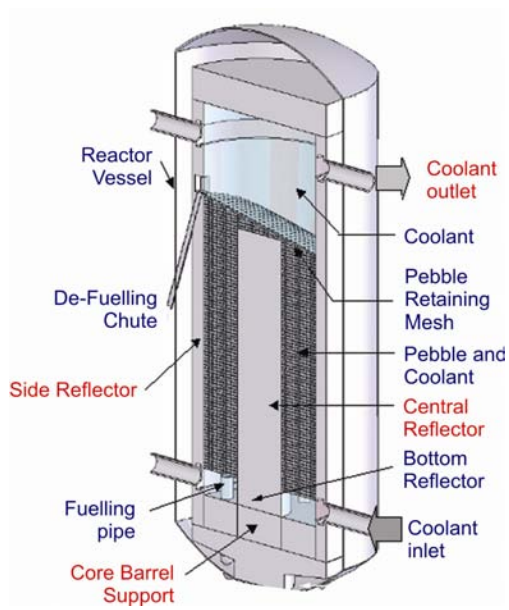

Fig. 4—Schematic of 600 MWth IHTR

Table 2—Proposed general specifications of the Innovative high temperature reactor

Reactor power	600 MWth for following deliverables – Hydrogen: 80,000 Nm^3/h – Electricity: 18 MWe – Drinking water: 375 m^3/h	Optimised for Hydrogen Production
Coolant outlet/ inlet temperature	1000°C/ 600°C	
Moderator	Graphite	
Coolant	Molten salt	
Reflector	Graphite	
Mode of cooling	Natural circulation of coolant	
Fuel	$^{233}UO_2$ & 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	

(viii) Structural materials for compatibility with molten salt at high temperatures

(ix) Coatings for corrosion and oxidation resistance

(x) Analytical and experimental safety related studies under various conditions such as air ingress, loss of coolant, loss of heat sink

(xi) Analytical and experimental seismic studies on components

(xii) Work related to materials for components and systems integrating nuclear reactor and hydrogen production plant

(xiii) Safety studies related to postulated conditions involving explosion of hydrogen and pressure wave produced and corresponding effect on structures and materials

Conclusions

Indian high temperature reactor and its associated programmes pose several fuel and material related challenges due to very high temperatures, neutron irradiation, and aggressive and corrosive coolant chemistry. The developmental work includes materials development along with their fabrication and joining technologies, compatibility studies with coolant and other materials, oxidation and corrosion resistant coatings and irradiation behaviour. As regards fuel, R&D includes development of high temperature high burn-up fuel, irradiation behaviour of fuel, and development of characterisation techniques. These have opened many avenues of research in the field of advanced fuel and materials. BARC has already initiated developmental work in most of these areas.

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