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CHEMICAL ENGINEERING IN THE HTGR

by

R. H. Simon, M. Gitterman, and G. B. Webb

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GENERAL ATOMIC
DIVISION OF
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JOHN JAY HOPKINS LABORATORY FOR PURE AND APPLIED SCIENCE

P.O. BOX 608, SAN DIEGO, CALIFORNIA 92112

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CHEMICAL ENGINEERING IN THE HTGR

The High-Temperature Gas-cooled Reactor (HTGR) system is an advanced type of nuclear power system employing helium cooling and graphite fuel elements without metallic cladding. It has some features that are of particular interest to chemical engineers. As will be seen, these features have resulted from the application of the same basic chemical engineering principles as are used in chemical process plants. The HTGR system has been under development since early 1957 at General Atomic Division of General Dynamics Corporation in San Diego, California. The U.S. Atomic Energy Commission and 58 electric power companies have cooperated in these development programs.

PEACH BOTTOM PLANT

The first HTGR power plant, located at Peach Bottom, Pennsylvania, on the Philadelphia Electric Company System, is now undergoing shakedown and start-up tests. This plant has a design core outlet temperature of 1380°F , a steam temperature of 1000°F , and a power output of 40,000 kw(e). Because of the high reactor coolant temperature full advantage can be taken of the high-temperature steam cycles and turbines developed for conventional generating plants, that is, with steam temperatures in the range 1000° to 1100°F . The large temperature difference between the helium and the steam is an important factor in reducing the size of the steam generators.

The net cycle efficiency of the Peach Bottom plant is 34.6%; larger HTGR plants will have efficiencies of well over 40%. This high cycle efficiency, coupled with good neutron economy, results in excellent fuel utilization. Figure 1 is a schematic flow diagram of the Peach Bottom plant.

Fig. 1--Schematic flow diagram of Peach Bottom plant

Helium was selected as the coolant for the HTGR primarily because of its chemical inertness, which is desirable when graphite-clad fuel elements are used, as in the case of the HTGR. Its inertness is also valuable in eliminating any possible reaction with fuel, vessel, ducting, circulators, or other components. Secondly, in common with other gases, it has a very low macroscopic neutron cross section. Thus, valuable neutrons are not absorbed in the coolant. Thirdly, it does not undergo a phase change.

Graphite was chosen as the moderator and reflector for the HTGR because of its low neutron absorption cross section and its high-temperature properties. The moderator and reflector are maintained at a temperature substantially above the temperature required to eliminate such effects as the energy storage associated with irradiated graphite at lower temperatures. The high heat capacity of graphite provides an excellent heat sink, which is effective in impeding rapid temperature changes and results in very stable and safe operation.

The fuel element is of the solid semihomogeneous type in which graphite serves as the moderator, cladding, fuel matrix, and structure. As shown in Figure 2, the fuel element for the Peach Bottom HTGR consists of an upper reflector section, a fuel-bearing middle section, a bottom reflector section, and an internal fission-product trap. The sleeve, which extends the length of the element, is made of low-permeability graphite and contains the fuel bodies, a portion of the bottom reflector, and the fission-product trap. Within the fuel element, annular fuel compacts are stacked on a cylindrical graphite spine. These compacts contain a mixture of graphite and fissile and fertile materials.

The uranium and thorium within the fuel compacts are in the form of a carbide, uniformly dispersed as particles in a graphite matrix. The $(U, Th)C_2$ particles are 150 to 400 μ in diameter. Each particle is pyrolytically coated with a 50 to 60 μ thickness of dense carbon. This coating protects the fuel mixture from oxidation during fabrication and increases the retention time of fission products during reactor operation. Although

Fig. 2--Fuel element for Peach Bottom reactor

beyond the scope of this paper, the preparation of the low-permeability graphite and the various steps involved in fabricating the fuel particles are the end products of laboratory and pilot-plant development using classical chemical engineering techniques.

By incorporating the graphite moderator with the fuel elements, the extensive moderator surface is made available for heat transfer. Moreover, since the graphite imposes no temperature limitation, large temperature differences between the helium coolant and the graphite cladding become practical. In addition to removing heat from the fuel element outer surfaces, conduction of heat within the fuel to these surfaces must also be accomplished. Fortunately, the high temperature levels permitted by the carbide-graphite fuel compacts allow heat transfer by radiation as well as conduction, so that close tolerances within the fuel element are not necessary for high thermal ratings of fuel.

The dispersal of the $(U, Th)C_2$ fuel particles in a graphite matrix minimizes radiation damage to the fuel body. The U^{235} and thorium atoms constitute only about 1% of the total atoms in the fuel compact. In an in-pile loop, which served as a pilot plant for the Peach Bottom reactor, burnup of over 70,000 Mw-days per tonne of uranium plus thorium was achieved.

Good fuel-cycle economy dictates that a power reactor must operate with a significant amount of neutron absorption in fertile material, i. e., material from which new fissile material is created. Since thorium yields the fissile isotope U^{233} , which has very good nuclear characteristics in thermal reactors, U^{235} -Th fuel has been used for the HTGR. Newly formed U^{233} builds up within the fuel element and a large fraction is fissioned in situ. In this manner, a large part of the total energy produced is obtained from the inexpensive fertile material.

OVER-ALL PLANT HEAT AND MATERIAL BALANCE

In the design of the Peach Bottom plant, the component design criteria were established before the detailed heat and material balance was prepared.

This is different from the design of conventional chemical plants, in that preparation of an over-all heat and material balance is usually the first consideration. In the case of Peach Bottom, however, this was practical because various plant design parameters determined as a result of independent studies, together with the required plant output, established the design requirements of the components making up the nuclear steam supply system. Preparation of the over-all plant heat and material balance for the Peach Bottom reactor was therefore undertaken to predict the actual operating performance of the plant over the full-load range (12 to 40 Mw(e)), taking into account the performance characteristics of the components specified for the plant. Because of the large number of cases to be investigated, it was decided to program the heat and material balance for calculation by an IBM-7090 computer. The primary coolant system is shown schematically in Figure 3.

The basic problem of interest was to determine the interaction between the steam generators and the main helium circulators as a function of the available heat-transfer surface in the steam generators. The steam generators are designed for a 30-year life, and because of anticipated high radiation levels, maintenance will consist only of occasional acid cleaning to remove water-side scale and plugging of any tubes that may develop leaks. The steam generator design therefore called for an arbitrary allowance of 5% excess surface to allow for tube plugging over the life of the plant, in addition to a fouling factor allowance. Furthermore, the required steam generation capacity was established at a time when it was planned that the helium circulators would be driven by steam turbines (the drives were later changed to electric motors, with variable speed, fluid couplings). The net result is a substantial oversize in the heat-transfer surface, at least during the early years of plant operation. The presence of this excess area, particularly in the superheater section of the steam generator, requires that the inlet helium temperature be lower and the helium flow rate be higher than design values, in order to produce the required amount of steam at 1500 psia and 1002^oF (1465 psia and 1000^oF at the turbine throttle).

Fig. 3--Primary coolant system flow diagram

Details of the steam generators are shown in Figure 4. The superheater is essentially in co-current flow, while the economizer and evaporator sections are countercurrent. Hot helium entering the steam generator is distributed along the length of the tube bundle by means of a perforated inlet baffle plate.

Calculation of the heat-transfer characteristics of the steam generator is complicated by the radial helium flow pattern through the outlet pass (up-pass) of the superheater. Rather than attempt to determine a single crossflow area characteristic of the outlet pass, it was decided to calculate the performance of each row of tubes. For this purpose, the superheater inlet pass was taken as four rows deep, the outlet pass as eight rows deep, and the evaporator and economizer inlet and outlet passes each as four rows deep. Lengthwise, the tubes were divided into six sections. Thus, the steam generator was divided into 168 increments for calculation purposes.

Heat-transfer coefficients were calculated, using conventional equations (the Dittus-Boelter equation for the water-side coefficient and the Colburn equation for the helium-side coefficient). A correction factor was applied to the helium coefficient in the inlet (down) pass of the superheater to allow for the fact that this part of the tube bundle is only four rows deep.⁽¹⁾ Flow through the remainder of the tube bundle was assumed to be turbulent enough for the heat-transfer equation to apply. The mean temperature for crossflow was calculated by the method of Bowman, et al.,⁽²⁾ assuming that each steam generator increment could be considered as single pass with one fluid (helium) mixed and the other unmixed.

Results of these calculations are presented in Table 1, where the calculated steam generator operating conditions are compared with the design conditions. Note that although the required steam production for a net plant output of 40 Mw(e) is ~7% less than the design steam flow, the required helium flow is about the same as the design flow. The inlet helium temperature, however, is substantially less than the design value. This is a direct consequence of the excess heat-transfer surface provided

Fig. 4--Steam generator cross section

Table 1
STEAM GENERATOR DESIGN AND OPERATING CONDITIONS

	Calculated Operating Conditions ^o		Design Conditions	Design Steam Flow with Clean Steam Generator, No Tubes Plugged
	Clean Steam Generator with No Tubes Plugged	Steam Generator with Design Fouling and 5% of Tubes Plugged		
Steam flow, lb/hr	184,200	184,200	197,500	197,500
Helium flow, lb/hr	232,800	231,700	234,200	254,800
Inlet helium temp., °F	1,278	1,315	1,367	1,274
Outlet helium temp., °F	587	621	628	597

in the steam generator (primarily in the superheater section) as an allowance for fouling and for tube plugging. The last column of Table 1 shows, for comparative purposes, the helium flow and temperatures required to produce the design steam flow, if the steam generator were clean and had no tubes plugged.

In the chemical industry, it is standard practice to include safety factors in equipment specifications to allow for various types of design uncertainties. The resulting oversize of equipment items may add to the required plant investment, but usually has no deleterious effect on plant operability. In fact, plant operation is frequently enhanced by the presence of excess capacity. In the present case, however, the situation is complicated by the very close interrelationship between the steam generators and the helium circulators. Provision of excess heat-transfer surface in the steam generators requires increased capacity in the circulators, if the desired final steam temperature is not to be exceeded. Considerations of this type, which lead to selection of suitable design safety factors, are normal chemical engineering functions, and make chemical engineers particularly valuable in nuclear power plant design.

HELIUM PURIFICATION SYSTEM

Purification of the helium coolant is required for two reasons. First, it is necessary to keep chemical impurities, particularly oxidizing ones, at a low level to prevent excessive reaction with the graphite of the fuel elements and reflector. Second, it is desirable to eliminate radioactive impurities, particularly the rare gas precursors of solid radioactive products, in order to make the maintenance of equipment easier.

Chemical impurities originate in several ways. Initially, and whenever the coolant system is opened for maintenance or refueling, air is bound to be introduced. Graphite in the reactor and metal walls of the pressure vessel, piping, and other parts of the coolant system will undergo

degassing when the plant is first started up. In case of a steam generator tube rupture there will be a temporary large leak of steam (or water).

The oxidizing impurities, such as O_2 , CO_2 , and H_2O , react with the graphite to cause chemical erosion and to form CO as a product. The CO , if not removed, reverts, partially at least, to CO_2 in cooler parts of the coolant system according to the reaction

The CO_2 thus formed reacts with more graphite in the reactor by the reverse reaction. A mechanism is thus established for mass transport of carbon from the reactor graphite to cooler parts of the coolant system.

The gaseous fission products, some radioactive, diffuse through the coating of the fuel particles and the graphite sleeves of the fuel elements into the helium coolant. In the Peach Bottom design, in order to reduce the diffusion through the sleeves, the sleeves are made of low-permeability graphite and a small purge stream of helium flows from the primary coolant stream through each element and through the internal fission-product traps to the helium-purification system. The internal traps, consisting of charcoal, remove the high-temperature volatile products by adsorption, and the radioactive gases going to the helium-purification system consist mainly of isotopes of the rare gases krypton and xenon.

Helium purification is based on adsorption at several temperature levels and with several adsorbents. Some of the adsorption beds are run saturated with the impurities and are not regenerated. These beds are called "delay" beds and their purpose is to delay certain radioactive isotopes long enough to provide substantially complete decay of these isotopes. The other adsorption beds are periodically regenerated, either immediately after or before breakthrough of a particular impurity.

The theory of delay by adsorption has been treated in a number of published articles (see, for example, Refs. 3 and 4). In simple terms,

the average delay time of a gaseous isotope in an adsorption bed of volume V and voidage fraction ℓ is given by

$$t = \frac{V(\phi + \ell)}{F}$$

where F is the volume flow rate of the carrier gas and ϕ is the adsorption coefficient of the gaseous isotope at the temperature and pressure of the bed. ϕ is defined as

$$\phi = \frac{\text{moles of isotope per unit volume of adsorbent}}{\text{moles of isotope per unit volume of carrier gas}}$$

Quite often the voidage fraction is negligible compared with the adsorption coefficient.

Figure 5 is a simplified flow diagram of the helium-purification system of the Peach Bottom plant. The primary feed to the system consists of the collected purge streams through the reactor fuel elements; this stream enters the fission-product trapping section of the system. A second feed stream, combining the purge streams from the steam generators, enters the chemical cleanup section.

The purge helium flow from the reactor enters the fission-product trapping section through a water-cooled heat exchanger, which removes the sensible heat from the gas. The cool gas then enters a water separator, which is used to remove water that may have condensed out in the heat exchanger following a major steam leak. The helium stream then enters a condensibles trap, directly cooled by water, for the removal of iodine and other non-noble-gas fission products that might be present. The helium then enters a precooler, before entering a charcoal delay bed. No direct cooling is provided for this bed; i. e., the cool process gas is used to remove fission-product decay heat generated within this delay bed. After leaving this delay bed, the process helium enters another identical combination precooler and delay bed.

Leaving the second delay bed, the process gas then enters a regenerative feed-effluent heat exchanger, which pre-cools the gas before it enters

Fig. 5--Flow diagram of helium purification system

a series of five low-temperature charcoal delay beds, which operate with inlet temperatures between -96° and -112° F. Exchangers remove the decay heat generated in the delay beds and picked up by the helium stream. These exchangers are cooled by a circulating Freon "brine" which in turn is cooled by a Freon refrigeration system.

A computer program has been used to predict the steady-state and transient performance of the krypton-xenon delay beds. Output of the computer program includes: (1) activities in each bed; (2) helium temperature leaving each bed; (3) krypton and xenon delay in each bed; (4) krypton and xenon activities leaving the system; and (5) resulting primary coolant system activities for each time interval desired.

The process gas, which is now essentially free from any radioactive isotopes except Kr^{85} , is rewarmed by the feed-effluent exchanger, compressed, cooled, and passed through a mist eliminator and oil filter. Most of the gas is then returned directly to the reactor primary coolant system; lesser amounts are returned indirectly through services requiring clean helium.

A small side stream is taken through a liquid-nitrogen-cooled adsorption system which is used to remove Kr^{85} , the only noble-gas fission product whose half life is too long to permit its removal by a system of delay beds. This liquid-nitrogen-cooled system also removes chemical impurities, in particular, nitrogen, argon, and carbon monoxide.

The adsorption beds in this side stream system are not operated as delay beds; i. e., they are not run at saturation. The flow rate has been selected sufficiently high to maintain the level of Kr^{85} activity in the primary coolant system below the design value. This, coupled with the transient characteristics of the preceding delay beds, makes it possible to run with the liquid-nitrogen-cooled system off line for long periods of time without exceeding allowable levels of Kr^{85} . The traps are provided with internal electric heaters for regeneration.

A nitrogen recondenser provides the refrigeration for the liquid-nitrogen-cooled traps. A feed-effluent exchanger cools feed helium against effluent helium and thus conserves on refrigeration.

Filters are provided in the helium-purification system to remove charcoal dust and aerosols.

The steam generator purge gas enters the chemical cleanup section through a plateout trap, which removes condensible activity. This section has an oxidizer in which CO and H₂ are converted to CO₂ and H₂O; a condenser and a knock-out drum, which remove the bulk of the water; and two dehydrator (molecular sieve) beds in series, which remove any remaining H₂O and essentially all of the CO₂. The effluent from this section is taken through a portion of the fission-product trapping section in order to remove any residual fission-product activity. Additional chemical purification is accomplished by the liquid-nitrogen-cooled traps.

The oxidation of H₂ and CO in the oxidizer is accomplished in a copper oxide bed. Any oxygen present is removed by the elemental copper in the oxidizer. The oxidation bed is regenerated by taking it off line and passing air through it that has been appropriately diluted with nitrogen from an inert gas generator. At the end of the normal regeneration cycle, a specified quantity of CO is passed through the bed to ensure oxygen removal when the oxidizer is put back on stream.

The concentration of CO₂ and water in the effluent from the leading dehydrator bed is monitored. When the breakthrough point is reached for either CO₂ or water, the first adsorption bed is taken off line for regeneration. This bed is regenerated by heated purified helium that warms the bed and drives off any adsorbed CO₂ and water. If the adsorbate is mainly water, the effluent helium from regeneration is passed through a condenser and water separator and then returned to the process stream. However, if the

bed contains appreciable CO_2 , the effluent helium from regeneration is further processed through a caustic scrubber, which removes the carbon dioxide. This helium from the caustic scrubber is then returned through a water scrubber to the steam generator purge gas stream at a point just upstream of the oxidizer condenser. After being regenerated, the dehydrator bed is put back on line downstream of the other bed.

The helium purification system just described applies a number of well known chemical engineering principles. Heat transfer, adsorption, oxidation, and drying operations are encountered in many process plants. The application of "delay" adsorption is an extension of adsorption to removal of impurities which have a relatively short life. The main difference between a nuclear power plant system of this type and similar systems or units in process plants is that the design for a nuclear plant must allow for the problems of radioactivity containment and of operation and maintenance of radioactively contaminated equipment.

SUMMARY

We have reviewed briefly the High-Temperature Gas-cooled Reactor with emphasis on some of the aspects which are of particular interest to chemical engineers. The concept is undergoing further development to enhance its efficiency as a nuclear power reactor and to improve the economics. Advancements are taking place in all phases of the design, including the fuel elements, the steam generators, and the helium purification system, which have been discussed. Chemical engineers have played an important part in the development of the HTGR and will continue to play a major role in the advancement of the concept.

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