

PREFACE

STATUS OF METALLIC MATERIALS DEVELOPMENT FOR APPLICATION IN ADVANCED HIGH-TEMPERATURE GAS-COOLED REACTORS

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HISTORICAL DEVELOPMENT OF GAS-COOLED REACTORS

The first gas-cooled reactor power station was built at Calder Hall in the United Kingdom and began operation in 1956. In this Magnox reactor type, CO₂ was used as the coolant and natural metallic uranium as the fuel. Magnox power plants with a combined electrical rating in excess of 8000 MW are now running and have achieved over 500 reactor-yr of operation.¹

The advanced gas-cooled reactor (AGR) was developed in the United Kingdom as an extension of the Magnox line, retaining CO₂ as the primary coolant, using enriched UO₂ fuel to allow an increase in coolant temperatures from 360 to 650°C. At the present time, AGR systems with a combined electrical rating of almost 9000 MW are in operation or under construction.²

The high-temperature gas-cooled reactor (HTGR) represents a further development to increase coolant temperatures. The higher operating temperatures (<750°C) require changes in the fuel/core design and the substitution of helium for CO₂ as the primary coolant. This leads to a more economical plant with a higher power density core and a low radioactive environment within the primary loop. The technological development of the system has proceeded steadily with regard to:

1. development of fuel particles, which are coated with layers of ceramics to retain fission products
2. use of an all-graphite core construction
3. use of helium gas as a coolant.

Table I shows the existing variants of HTGR design with some of their characteristics. The development of the HTGR has proceeded in two directions:

1. the pebble bed concept in the Federal Republic of Germany (FRG) (Ref. 3)
2. the prismatic core in the United States and the United Kingdom.⁴

The fuel elements for the pebble bed system consist of 60-mm-diam spheres made up of a carbon outer zone and an inner 50-mm-diam region containing coated fuel particles uniformly dispersed in a graphitic matrix. The prismatic fuel element consists of a machined hexagonal graphite block ~750 mm long and 350 mm across the flats. Fuel and coolant channels are drilled in a hexagonal array. Fuel rods, which consist of coated particles bonded in a random close-packed array by a carbonaceous matrix, are contained in the fuel channels. Although fuel elements in the two

HTGR designs differ substantially, the basic fuel-containing unit, the coated particle, is essentially the same. Comprehensive surveys of the international development of coated fuel particles and the different HTGR fuel elements were published in the 1977 *Nuclear Technology* special issue.⁵

It is generally accepted that materials for the core—the fuel elements as well as structural graphite—are suitable for use at the highest temperatures envisaged in the advanced HTGR concepts. The spherical fuel elements have shown themselves particularly suitable for very high temperature service.

THE HTGR

Experimental Reactors

Part A of Table I shows some features of the three HTGR experimental reactors that have been constructed:

TABLE I
High-Temperature Gas-Cooled Reactors

A. Experimental Reactors				
	Peach Bottom	Dragon	AVR	
Operational	1967-1974	1968-1975	1967-	
Thermal/electric power (MW)	115/40	20/-	46/15	
Fuel element type	Prismatic	Prismatic	Spherical	
Helium inlet/outlet temperature (°C)	377/750	350/750	270/950	
Mean helium pressure (bar)	25	20	10	
B. Prototype Reactors				
	Fort St. Vrain		THTR	
Operational	1976		1985	
Thermal/electric power (MW)	842/330		750/300	
Fuel element type	Block		Spherical	
Helium inlet/outlet temperature (°C)	404/777		270/750	
Mean helium pressure (bar)	45		39	
C. HTGR Projects, Advanced HTGR Systems, and Projected Commercial Plants				
	PNP	HHT	HTR-500	Modular HTGR
Thermal/electric power (MW)	750/-	1240/600	1250/500	200
Fuel element type	Spherical	Spherical	Spherical	Spherical
Helium inlet/outlet temperature (°C)	300/950	450/850	280/700	250/750
Mean helium pressure (bar)	39	50	47	50
	SC/C	VHTR	VGR-50	VG-400
Thermal/electric power (MW)	2240/872	50/-	136/50	1060/300
Fuel element type	Block	Prismatic	Spherical	Spherical
Helium inlet/outlet temperature (°C)	319/686	395/1000	296/810	350/950
Mean helium pressure (bar)	72	40	40	50

1. In June 1967 the first HTGR to produce electricity, Peach Bottom 1, began operation in the United States. This 40-MW(electric) developmental power plant ran for a total of 1349 full-power days and produced more than 1.4 billion kWh of electricity before being shut down in October 1974 as scheduled.⁶

2. The international Organization for Economic Cooperation and Development Dragon Project at Winfrith in the United Kingdom developed the 20-MW(thermal) Dragon reactor, which was in operation from 1968 until 1975 (Ref. 7).

3. The 15-MW(electric) Arbeitsgemeinschaft Versuchs-Reaktor (AVR) at Jülich in the FRG with a pebble bed core reached criticality in 1967. This reactor, which is still in operation, had produced 1.2 billion kWh during 3360 full-power days up to the end of February 1983 (Ref. 8).

The Peach Bottom and Dragon reactors operated with primary coolant temperatures of $<750^{\circ}\text{C}$; the AVR has proved since 1974 that with a pebble bed core, gas temperatures of $\sim 950^{\circ}\text{C}$ can be achieved and maintained over long periods (Ref. 8). It should be noted, however, that in this steam cycle reactor the maximum operating temperature of the steam generator components is only $\sim 530^{\circ}\text{C}$.

Prototype Reactors

The highly successful operation of the three experimental reactors was a major factor in continuing the development of the HTGR, which led to the construction of two steam cycle prototype reactors, the 330-MW(electric) Fort St. Vrain Reactor (United States) and the 300-MW(electric) thorium high-temperature reactor (THTR) at Schmehausen, FRG (Table I, part B). The first of these has a prismatic core, while the second has a pebble bed core design. Both core designs share the high thermal efficiency of the HTGR and its inherent safety advantages, which result from the relatively low power density in the core compared with light water reactors (LWRs) and fast breeder reactors (FBRs), the large thermal capacity of the core, the absence of coolant phase changes, and the rapid negative temperature coefficient.

The Fort St. Vrain Reactor was reaching criticality at just about the time that Peach Bottom operations were winding down. The plant contains a number of design features that were new to power reactor systems in the United States, including silicon-carbide-coated fuel particles, once-through modular steam generators, and prestressed concrete reactor vessels. Fort St. Vrain reached 68% power in January 1978 and, after a series of delays, started ascent to full power in early 1981 (Refs. 6 and 9).

The THTR is still under construction and operation is scheduled to begin in 1984 (Ref. 10).

Advanced HTGR Systems and Expected Commercial Plants

The current generation of HTGRs operates on a conventional steam cycle. However, advanced HTGR designs are under intensive development in the FRG (Refs. 10, 11 and 12), in the United States,^{6,13} and in Japan¹⁴ because of their potential for electricity generation based on a direct cycle gas turbine and for the supply of heat for chemical processes, such as the gasification of coal and methanol production. It is here that the full potential of the HTGR can be realized with the supply of primary coolant at temperatures in the 850 to 1000°C range. Countries that have been involved in the past or that have a more modest ongoing effort include Austria, France, and Switzerland. Since 1970 the Soviet Union has also been active in the development of HTGRs for advanced applications.¹⁵⁻¹⁷ The advanced HTGR concepts and commercial plants under development are described in Table I, part C.

In the FRG, the advanced HTGR development program comprises three projects, two of which have commercial prospects:

1. project PNP [prototype HTGR plant for nuclear process heat (Table I, part C)], a process heat HTGR providing heat via a secondary helium circuit for the steam gasification of coal using a gasifier heat exchanger in a fluidized bed
2. project NFE (long distance energy transport system), a process heat HTGR for methane reforming in a tube furnace heated directly by the primary coolant, the product gas being used for hydrogasification of lignite (project PNP) or for long distance heat transport by means of chemical latent heat
3. project HHT (Table I, part C), an HTGR with direct cycle helium turbine for electricity production; although this project has been terminated the materials development work is continuing but with relatively low priority
4. for electric power and process steam generation, a commercial plant based on an improved THTR design—the HTR-500—is considered to be competitive with water reactors (Table I, part C)
5. with the modular HTGR (Table I, part C) small units for electric power generation, process steam and process heat applications are proposed, each unit having a metallic pressure

vessel instead of prestressed concrete; it is proposed that several units could be combined to form a nuclear power station.

In Japan a demonstration plant for nuclear process heat supply is under development: very high temperature reactor (VHTR) project (Table I, part C). It was originally intended for steelmaking, using the direct reduction of iron ore, but has been recently changed to more general high-temperature applications in chemical processes.

Efforts in the United States are now concentrated on a steam cycle/cogeneration (SC/C) HTGR for the production of electricity and process steam (Table I, part C). It is hoped that this 2240-MW(thermal) plant can begin operation in the middle 1990s. In addition to this primary effort, work is under way on assessment of advanced HTGR systems and applications and on the technology necessary for commercialization. A restricted effort is being applied to nuclear process heat applications or modular HTGRs.

In the Soviet Union R&D work on pilot gas-cooled reactors for electrical power generation and for nuclear heat application is being carried out. For an experimental chemical energy plant using the VGR-50 reactor, the design parameters are given in Table I, part C. This plant will provide experience in the design, construction, and application of HTGRs with spherical fuel elements and demonstrate the design feasibility of principal primary loop components.

The industrial prototype energy plant using VG-400 (Table I, part C) is being designed for the combined production of high-temperature heat and electrical energy. At the present stage, a method is being developed for using the VG-400 HTGR in an industrial chemical plant mainly for the production of ammonia. The VG-400 reactor serves as a prototype for future industrial process heat plants.

QUALIFICATION OF METALLIC MATERIALS

As mentioned above, core materials and design have been developed to the stage where the feasibility of core outlet temperatures up to 1000°C is proven.

For the steam cycle HTGRs (Fort St. Vrain and THTR), the highest metal temperatures are around 750°C; therefore, the high-temperature iron-based Incoloy alloy 800 is used. This alloy is qualified for nuclear applications at temperatures up to 760°C, and design codes are available.

In order to take full advantage of the process heat capability of the HTGR, alloys of higher strength than Alloy 800 are required for the components operating in the 800 to 1000°C temperature range, which are

1. intermediate heat exchanger (IHX) tubes
2. hot gas header

3. baffle plates
4. reformer tubes.

An important aspect of the qualification of alloys for HTGR applications is the effect of the working environment on mechanical behavior. The first investigations of the effects of impure helium simulating the primary coolant gas of an HTGR on the properties of alloys were started in the early 1970s by the OECD Dragon Project.¹⁸ The stress rupture and corrosion behavior of various alloys in simulated HTGR helium were investigated. The results of these tests showed a significant influence of the gas composition and temperature on corrosion effects (e.g., at 850 to 1000°C, internal oxidation and severe carburization were found). However, an effect of corrosion on the stress rupture properties was not clearly observed. The Dragon Project had also carried out a well-defined R&D program in close collaboration with Commissariat à l'Énergie Atomique (France) and GA Technologies (United States) for component testing. The work was completed and the test facilities are still available for use in advanced HTGR projects.

Following the close of the Dragon Project, interest was focused on process heat applications, and it became clear that extensive materials evaluation and development programs were required before metallic materials could be reliably designed for and applied at operating temperatures of 800 to 1000°C. Such programs were initiated in the FRG, the United States and Japan.^{19,20} At first in the FRG and the United States, large numbers of commercial alloys were subjected to screening tests to assess suitability for HTGR applications.²² In Japan the major effort from the beginning was on the development of new alloys especially designed for HTGR service conditions.²³

The selection of candidate alloys for the advanced HTGR projects was based on long-term creep rupture properties, structural stability, and fabricability. For the highest operating temperatures, solid solution-hardened nickel-base alloys were therefore chosen. To qualify these alloys for nuclear applications, extensive test programs have been initiated, the principal tasks of which are:

1. creep-rupture testing of the basic alloys and weldments
2. gas/metal reaction kinetics and associated transport phenomena
3. determination of the changes in mechanical and physical properties after long-term exposure under simulated service conditions, particularly ductility (end-of-life properties)
4. high-and low-cycle fatigue testing

5. influence of high-temperature corrosion in simulated service environments on mechanical and physical properties
6. fracture mechanics studies with special emphasis on the effects of service environments on crack initiation and propagation
7. creep fatigue interactions
8. fracture toughness characteristics
9. fretting, friction, and wear
10. structural design code, rules for temperatures above 800°C
11. constitutive equations
12. damage accumulation and estimation of service life
13. transferability of mechanical properties data to multiaxial loadings and complex geometries
14. hydrogen and tritium permeation
15. development of coatings
16. development of decontamination methods for maintenance and possible repair operations
17. influence of irradiation on materials behavior (metallic absorber rod cladding)
18. development of procedures for nondestructive testing (NDT) of components
19. weldability, and postweld and postfabrication heat treatments.

In addition to the extensive research and test programs for the qualification of commercial alloys for HTGR process heat applications, efforts have been expended to develop new alloys designed specifically for HTGR components. Generally, the primary goals of these developments are improved high-temperature creep and fatigue strength, better corrosion resistance in the HTGR primary coolant, and improved performance under high-temperature irradiation.

For the development of new alloys for heat exchanging components with very high operational temperatures, the metallurgical tools are limited. The trends in developmental materials for turbine blades, which lead to working temperatures of more than 1000°C, cannot transfer to large heat exchanging components. In addition to the high creep strength and corrosion resistance in HTGR helium, workability and weldability are very important factors. These boundary conditions restricted the field of development on solid solution strengthened Ni-Cr-Fe-base alloys. The expected improvement of creep strength is marginal;

the HTGR corrosion resistance, however, should be markedly improved. A new hardening method can be achieved in Ni-Cr-W-base material due to alpha-tungsten precipitation. These materials give a certain potential for the improvement of high-temperature creep strength.

For improvement of the irradiation behavior of control rod materials, thermal mechanical treatment of austenitic steels should have the best potential beside those developments, trying to improve the high-temperature capability of ferritic materials.

ORGANIZATION OF THE SPECIAL ISSUE

Although results of the different investigations in a more or less detailed form are available in the different reports of the HTGR partners (especially in the FRG, Japan, and the United States) and for specific items in proceedings of national or international HTGR conferences (see, e.g., Refs. 19, 20, and 21), the complete status of the high-temperature alloy development itself has not been comprehensively treated in a single publication. This special issue of *Nuclear Technology* is intended to fill that gap by providing a compilation of papers that are representative of the current state of the art in high-temperature alloys for application in advanced HTGRs. The continuing, strongly international nature of the development and testing of high-temperature materials is reflected in the papers in this issue, written by scientists from seven countries and many organizations. The papers are divided into the following categories:

1. selection, development, and production of alloys for HTGR components
2. characterization of microstructure
3. mechanical properties (creep rupture, fatigue and short-term properties, and fracture mechanics)
4. gas/metal reactions
5. friction and wear
6. hydrogen and tritium permeation
7. irradiation behavior
8. structural design codes and life prediction
9. NDT.

These categories illustrate the scope and depth of the ongoing work devoted to materials for both steam cycle and advanced HTGRs. All aspects of materials technology deemed important to the commercialization of the HTGR are covered.

SUMMARIZING REMARKS ABOUT THE STATUS OF THE QUALIFICATION AND DEVELOPMENT OF HIGH-TEMPERATURE ALLOYS

In this section, some of the highlights of the contributed papers are discussed in an attempt to put the current state of the technology in perspective and to indicate directions for future work. For convenience, this special issue has been structured in different categories of R&D; however, there is substantial interaction among these various areas.

Selection, Production, and Development of Alloys for HTGR Components

Because of the considerable time required to develop and qualify new alloys specifically designed for HTGR applications, the first advanced HTGR plants will have to be constructed from available commercial alloys. A thorough investigation of the fabricability of candidate alloys to the various product forms required in an HTGR, e.g., IHX tubing and reformer tubes, is therefore required as part of the materials assessment and qualification program. Of particular interest in this context is the weldability of the alloys and the mechanical properties of weldments.

Although available commercial alloys will have to be used at first, it has been realized that improvements in the performance of the commercial alloys are desirable and possible in view of the unique operating conditions in an HTGR, especially long service times, high temperatures, and low oxygen potential service environments. In addition, extensive efforts are being devoted to the development of new alloys specifically designed for HTGR service. These efforts are being concentrated on improvements in corrosion resistance and creep rupture strength over and above the candidate commercial alloys with adequate formability and weldability behavior. The direction of alloy development can be grouped as follows:

1. (Fe)-Ni-Cr alloys with chromium content above 20%, adjusting manganese and silicon and removing aluminum and titanium for better corrosion resistance in a specified HTGR test helium, using molybdenum and/or tungsten for solid solution hardening
2. (Fe)-Ni-Cr alloy with chromium content less than ~12%, using optimized contents of aluminum and titanium for improving very high temperature carburization resistance in specified HTGR test heliums with molybdenum and/or tungsten additions to confer high creep resistance
3. a new alloy system, first proposed within the alloy development program of the Engineering

Research Association of Nuclear Steelmaking, is Ni-Cr-W with alpha-tungsten precipitates for high-temperature strengthening.

The results of the various alloy development programs are very promising, several experimental alloys showing significant improvements over the available commercial alloys.

Characterization of Microstructure

To understand the mechanical properties and long-term behavior under service conditions, a detailed examination of microstructure and microstructural stability is necessary for defining materials specifications for nuclear applications. For some commercial alloys (e.g., Incoloy-800H, Hastelloy-X, Inconel-617), provisional time/temperature/precipitation diagrams are available, but it seems necessary to increase the effort devoted to microstructural examinations to obtain reliable long-term information on structural stability.

Mechanical Properties

The main effort in the evaluation of design-related basic data is concerned with

1. creep and creep/rupture properties
2. fatigue properties
3. short-term properties, mainly after long-term exposure
4. fracture mechanics.

The interaction of creep/fatigue and the problems concerning constitutive equations and damage accumulation rules are summarized together with an analysis of structural design problems.

Some papers present sets of the basic data for $\frac{1}{4}$ Cr-1Mo ferritic steel, Hastelloy-X, and Incoloy-800 for parent metal and weldments in steam cycle application. At the Japanese Atomic Energy Research Institute, a considerable data base for the Cr-Mo steel to be used for small reactor vessels, including irradiation effects, is available. The data of the materials for advanced HTGRs are given in detail, and the basic data needed for turbine blade material Inconel alloy 713 LC are summarized for HHT helium turbine application, demonstrating that the alloy can satisfy the requirements of HHT service conditions.

Creep Properties

For the steam cycle application, the creep/rupture behavior in simulated HTGR helium in comparison to air is discussed on the basis of experimental results up to 550°C for ferritic steels and up to 870°C for Incoloy-800H and Hastelloy-X, with test times of more than 20 000 h.

For the evaluation of design values for commercial alloys Inconel-617, Hastelloy-X, Nimonic-86 in the temperature range up to 1000°C, extensive experimental results up to 20 000 h with individual points extending beyond 30 000 h are now available. The evaluation of weldments needs further effort. For the modified Hastelloy-XR, the materials data for temperatures between 800 and 1000°C up to 10 000 h for bars, tube wall, and plates have been obtained.

The creep behavior in simulated HTGR helium seemed to be dependent on gas impurity levels and on the surface condition of the specimen, but the differences in the behavior in air and under carburizing or decarburizing atmospheres are marginal. Using a scatter-based evaluation, all rupture points fall in the same scatterband. It is well demonstrated, however, that the behavior of pre-carburized specimens differs from that of specimens subject to continuous carburization during creep testing.

Fatigue Properties

Both high and low-cycle fatigue properties are described. The high-cycle fatigue properties of Incoloy-800 in "wet" HTGR helium seem to be slightly better than in air. Up to 650°C, there seemed to be a fatigue endurance limit, but at higher temperatures, there is no evidence for an endurance limit. For higher temperature and HTGR helium impurity levels expected in advanced applications, no fatigue limit in high-cycle fatigue tests can be defined. There is some evidence that in HTGR helium the number of cycles to failure is higher than in air for Inconel-617, Nimonic-86, and Hastelloy-X.

Low-Cycle Fatigue

In simulated HTGR helium, the commercial alloys in the as-received condition show improved resistance to low-cycle fatigue failure; with decreasing strain rate, the number of cycles to failure also decreases. The impure helium atmosphere leads to a behavior of as-received material more similar to that in vacuum than that in air. Hold time in tension reduces the number of cycles to failure. For very high temperatures, it is critically discussed whether the low-cycle fatigue test with hold time is the correct simulation of creep/fatigue interaction.

Following the proposal of the American Society of Mechanical Engineers (ASME) Code Case N 47, the experimental basis for the evaluation of design curves has been obtained. The experimental basis for weldments is, as yet, insufficient to define safety margins.

Short-Term Properties

All highly creep-resistant materials for high-temperature applications tend to exhibit a loss of room temperature ductility, indicated by a low elongation in

the tensile test and a low notched impact strength after long-term thermal exposure. With increasing test temperature, the deformability values for most materials increase, but for some specific heats, low values at operating temperatures above 400°C have been observed. For the design and treatment of upset conditions, impact strengths are not needed. Using the expected loading history, a critical assessment of the ductility levels that are required in service must be made.

Fracture Mechanics

In ASME Code Case N 47, there is no rule for using fracture mechanics arguments for the design and treatment of loading conditions for the components. Nevertheless, it would be very helpful if fracture mechanics methods can be introduced into structural design procedures. It is postulated that the pure fatigue crack growth behavior of as-received material can be described by a linear elastic fracture approach up to high temperatures; fatigue crack growth can be used for the consideration of components behavior with flaws.

The results on creep crack growth are not sufficient for establishing a design criterion. There are, however, indications that creep crack growth in impure HTGR helium may be marginally higher than in air. For creep crack growth and for creep/fatigue crack growth, a physically useful concept must be developed.

Gas/Metal Reactions

The heat-transferring components of an HTGR plant are subjected to corrosive gases at high temperatures in the primary circuit as well as in subsequent loops for long periods of time. Detailed knowledge of the reactions between these gases and metallic surfaces are required in order to derive a quantitative description of damage processes due to corrosion for strength calculations in HTGR component design.

Extensive experimental results on the corrosion behavior of alloys under steam cycle conditions, in process gas, and especially in the simulated primary circuit helium of advanced HTGRs are now available. The corrosion behavior in the oxidizing gases of dual-cycle systems is largely understood and can be basically quantified. However, a quantitative description of corrosion processes in primary circuit helium is much more complicated. The HTGR helium is characterized by a very low oxidation potential, and test programs carried out so far have shown that minor variations in gas composition, especially of the water content, may lead to highly different corrosion phenomena, especially at temperatures above 900°C. Carburization as well as decarburization is possible in

addition to oxidation, depending on the material, gas composition, and gas flow rate.

In the recent past, model conceptions have been developed on the basis of thermodynamic and kinetic data, by means of which the ranges of gas composition in which clearly different corrosion phenomena occur can be specified. Past experiments have shown that only very low carbon transfer occurs between the material and the atmosphere in those ranges of gas composition featuring a protective coating even after long periods of exposure.

The following subjects are dealt with in current and future programs of investigation for determining viable rules that allow for corrosion in component design:

1. quantification of model concepts of corrosion in order to determine ranges of gas composition for various materials in which different corrosion phenomena occur
2. determination of time laws for corrosion in the different ranges of gas composition, also under dynamic operating loads
3. influence of carburization, decarburization, and internal oxidation on mechanical properties and on crack initiation and crack growth.

Friction and Wear

In HTGR helium, protection against excessive friction and wear is needed over a wide temperature range for many regions of different HTGR components, such as parts of control rod sheaths and tube/spacer contact points in heat exchangers. For temperatures up to 750°C, the experiences of pilot plants and prototype projects seemed to satisfy the needs of the designer. For advanced HTGRs, the development and screening programs are described. Coating layers of oxides, combined oxides and nitrides, or carbides produced by different coating methods are being investigated in long-term tests under static and dynamic conditions, in model-type tube-to-space vibration tests, and in sliding tests at high temperatures up to 1000°C in simulated HTGR helium. The materials examined are graphite, ferritic steel, austenitic steel, Incoloy-800H, Hastelloy-XR and Inconel-625. Promising results have been obtained for duplex ZrO₂ coatings on Hastelloy-XR and on austenitic steels. However, the existing know-how required for coatings is not described in detail in the contributions of this special issue. The data provide an understanding of the ongoing work. A clear and better understanding of coating microstructural features and their influence on wear-resisting properties is still needed.

Also reported in this part are some effects of pre-oxidation on the corrosion behavior of austenitic steels

in advanced CO₂-cooled reactors and some ideas on thermal barriers for HTGRs.

Hydrogen and Tritium Permeation

Hydrogen and tritium permeation play a role that cannot be neglected, especially in HTGR plants for process heat application. It is well known that the primary gas of an HTGR contains small quantities of tritium originating from uranium fission and nuclear transformation of lithium in graphite, of boron in shutdown/control rods, and of ³He in the helium coolant gas. The sources of tritium release mentioned and the gas purification system produce a characteristic high-temperature partial pressure as a significant absorption sink in primary helium, causing the tritium to permeate through the heat exchanger tube walls into the secondary circuits. It passes, for instance, into the product gas of a coal gasification plant whose permissible limits for tritium activity are extremely low for reasons of radiation protection. On the other hand, hydrogen may get into the primary circuit from the secondary or tertiary atmosphere, e.g., from the product gas of methane reforming in a tube reformer. It would enhance graphite corrosion in the core as well as radiolytic methane formation.

Extensive measurements, especially in the German but also in the Japanese HTGR program, have revealed that permeation is only decisively inhibited by oxide coatings. Metallic barrier walls with bare surfaces feature unacceptably high permeation rates irrespective of alloy composition. Previous development has meanwhile led to the availability of coatings inhibiting hydrogen and tritium up to factors of 1000, as compared to bare metal. The fabrication of such coatings is based either on the formation of an oxide layer prior to using the tube systems, e.g., by pre-oxidation of the tube surfaces involving the growth of stable and adhesive oxides, or by an "in situ layer formation." The latter process is particularly attractive due to the self-healing of cracks in coatings during plant operation. Particular attention will be paid in future work to the following:

1. definition of fabrication and characterization of coatings and correlation of coating properties with permeation measurements
2. study of permeation and corrosion mechanisms
3. quantitative description of hydrogen and tritium permeation using mathematical and physical models
4. stability of coatings under mechanical and temperature cycling operational loads
5. behavior of permeation-inhibiting oxide coatings against corrosion, such as carburization and sulfidizing.

Irradiation Behavior

The influence of neutron irradiation on mechanical properties of steels and alloys at elevated temperatures has been examined for FBR components. This topic has received relatively little attention for HTGRs as metallic materials are subjected to only very low levels of neutron irradiation, and therefore irradiation effects can be neglected. There are, however, some important components of an HTGR that are exposed to neutron irradiation during operation, such as control rods, near-core thermal barriers, liner structures, and, to a very limited extent for small reactors, the metallic pressure vessels. In comparison to the FBR, the neutron spectrum in an HTGR is basically of the lower energy range, but does depend on the core design. At temperatures above 500 to 600°C, although the atomic displacement effects of fast neutrons can be neglected, thermal neutrons can cause significant levels of nuclear transmutation reactions, e.g., neutron and alpha reactions to produce helium atoms.

For low alloy steels, considerable irradiation data exist from the LWR pressure vessel experiments. In addition to a survey of literature data, the room and elevated temperature embrittlement due to thermal neutrons is described for a $2\frac{1}{4}$ Cr-1Mo steel irradiated at 400°C up to 1×10^{19} n·cm⁻². The results demonstrate that the stress has a more important effect on embrittlement than neutron irradiation. The tendency for embrittlement is also dependent on phosphorus content. For a ferritic liner steel (A 537, manganese 1.32%, copper 0.26%, silicon 0.26%, nickel 0.21%, chromium 0.14%), the influence of irradiation at 60°C with high thermal neutron spectrum ($\phi_{thermal}/\phi_{fast} \sim 1000/1$) has been examined and a mathematical description for the correlation between fluence and ductile/brittle transition temperature has been established. Low alloy steels can be used for some HTGR structures with low operation temperature, and the information needed for design is available.

The work on austenitic stainless steels, such as X8CrNiMoNb1616, and on Fe-Ni-Cr alloys, such as Incoloy-800H and Hastelloy-X, demonstrates that in dependence on irradiation history, the tensile and creep ductility at test temperature can be markedly decreased. This effect must be considered in the design and estimate of allowable operation time. The main transmutation reactions to be considered are $^{10}\text{B}(n, \alpha)^7\text{Li}$ or $^{58}\text{Ni}(n, \gamma)^{59}\text{Ni}(n, \alpha)^{56}\text{Fe}$.

The loss in total elongation increases with increasing test temperature, and failure is attributed to enhanced intergranular cracking and therefore a decrease in load-bearing capability.

The post irradiation loss of deformability is more pronounced in nickel-base alloys than in austenitic steels. A thermomechanical treatment of austenitic steels helps to reduce the loss of ductility in the test temperature range between 600 to 850°C.

The irradiation behavior of control rod materials is sufficiently known for the different core designs. The metallurgical controlling mechanisms for high-temperature helium embrittlement, however, are not yet fully understood. Irradiation experiments on control rod materials are still in progress.

Structural Design Codes and Life Prediction

The status of structural design codes for metallic HTGR components is described for the operating temperature range of time-independent and time-dependent properties. The temperature limit for time-independent strength analysis is $\sim 370^\circ\text{C}$ for ferritic steels and $\sim 425^\circ\text{C}$ for austenitic steels. The data, design values, rules, and procedures of ASME Code Case N 47 satisfy the needed high-temperature structural engineering data with only a few limitations up to temperatures of $\sim 800^\circ\text{C}$. The status for a very high temperature design code ($>850^\circ\text{C}$), required for the application of HTGRs for process heat, illustrated that there remains much work in establishing safety margins, lifetime prediction rules, and constitutive equations. From the structural mechanical point of view, the experimental and theoretical efforts should be oriented to the confirmation of the behavior of basic materials and weldments in long-term testing, to the determination of load history for better evaluation of HTGR special lifetime prediction rules, and to simplified inelastic procedures for describing the component behavior under primary and secondary HTGR loading conditions. The transferability of mechanical properties data to multiaxial loading and complex geometries is also required for a better definition of safety margins.

A worldwide effort and cooperation for the establishment of the basis for high-temperature nuclear design codes could help to improve the commercial application of HTGRs to electric power generation and process heat. This special issue is a welcome opportunity to survey the existing data, which may form the basis of an HTGR structural design code.

NDT

Nondestructive testing methods are continually being improved, and considerable efforts have been made for LWR components. From the metallurgical point of view, the materials of the very high temperature application pose great difficulties in the interpretation of ultrasonic and eddy current test results, due to the coarse-grained microstructures, the presence of weldments, and corrosion effects such as carburization and internal oxidation. Only restricted and preliminary experiences with HTGR material are reported. The effort on NDT methods should be increased for typical HTGR components; however, the

procedures used for light water pressure vessels (full-size inspection methods) should not be duplicated without careful consideration of the HTGR safety concept and the high degree of inherent safety of the HTGR system.

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