PREFACE

COATED PARTICLE FUELS

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Coated particle fuel was developed to provide fission-product containment at relatively high fuel surface temperatures of the high-temperature gas-cooled reactor (HTR). Coated particle is a generic term applied to miniature spherical fuel elements up to ~1 mm in diameter. The fuel "kernel" is commonly an oxide or carbide of uranium (with various enrichments), thorium, or plutonium. Coatings in most general use are pyrolytic carbon (PyC) and silicon carbide (SiC), although a variety of other materials were or are being examined. With the variety of kernel and coating materials available, coated particles can be optimized for a particular application. This was accomplished and, in general, current work is focused on a few well-defined fuel systems that are discussed in detail later in this Preface.

Under the impetus of providing a clean primary circuit for HTRs, coated particle fuel technology developed rapidly as a broad international effort from its beginnings in about 1960. The international nature of the effort was reflected in the makeup of the first symposium on the subject, sponsored by the U.S. Atomic Energy Commission at Battelle Memorial Institute in 1962 (Ref. 1), where papers from six countries were presented. Papers from a subsequent American Nuclear Society Symposium on Coated Particle Fuels[2] demonstrated the rapidly increasing level of sophistication of the technology. Papers in this special issue describe many of the advances that were made in the intervening period.

In the following paragraphs, the current state of coated particle development is briefly reviewed to provide an introduction for the specialized papers that follow.

THE HTR

The HTR utilizes an all-ceramic core, a graphite core structure, and ceramic-coated particle fuels. The use of refractory core materials combined with a single-phase inert helium coolant allows high-coolant temperatures and results in a number of significant advantages including high efficiency and high tolerance for transient high temperatures. In addition, low-power density and large thermal mass of the core mitigate safety concerns during postulated loss-of-forced-circulation events. These features ease reactor siting constraints by reducing both cooling water requirements and the consequences of postulated accidents.

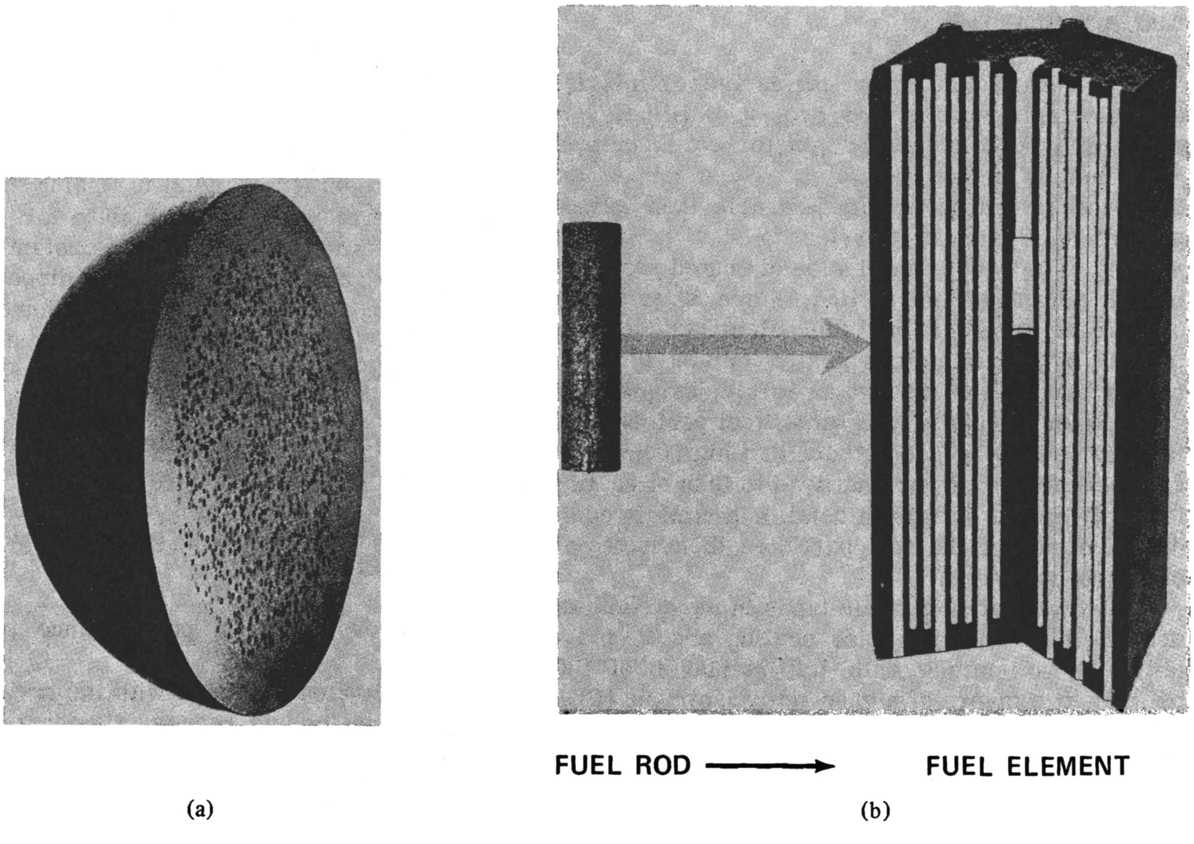
By 1967, a mere 11 yr after the idea of the HTR was conceived by a group at the United Kingdom Atomic Energy Authority (UKAEA), and ~7 yr after the initial development of coated particle fuel, three demonstration or experimental HTRs were operating with early designs of coated fuel particles. The highly successful operation of these reactors, the Dragon Reactor Experiment in the U.K., the Arbeitsgemeinschaft Versuchsreaktor (AVR) in the Federal Republic of Germany (FRG), and the Peach Bottom 1 Reactor in the U.S., was a major factor in continuing interest in the HTR.

The development of the HTR has proceeded in two directions: (a) the pebble bed concept in the FRG (Ref. 3), and (b) the prismatic core in the U.S. and the U.K. (Ref. 4). Schematic diagrams of the fuel elements for the two systems are shown in Fig. 1. The fuel elements for the pebble bed system consist of 60-mm-diam spheres made up a fuel-free carbon outer zone and an inner-fueled region with coated 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. Alternate fuel and coolant holes are drilled in a hexagonal array. Fuel rods, consisting of coated particles bonded in a random close-packed array by a carbonaceous matrix, are stacked in the fuel holes.

Although fuel elements in the two HTR designs differ substantially, the basic fuel-containing unit, the coated particle, is essentially the same, and coated particle fuel development proceeded as an international effort quite independent of differences in reactor design. Development of both HTR designs is now approaching final demonstration with the impending commercial operation of the 330-MW(e) Fort St. Vrain (FSV) reactor built by General Atomic Company for the Public Service Company of Colorado, and the scheduled operation in 1979 of the 300-MW(e) thorium high-temperature reactor (THTR) at Schmehausen in the FRG. The first of these utilizes a prismatic core, while the second utilizes a pebble bed core design.

Pebble bed and prismatic core designs share the high thermal efficiency of the HTR and its inherent safety advantages resulting from the low-power density and large thermal capacity of the core, the absence of coolant phase changes, and the prompt negative temperature coefficient. Some of the characteristics of the various HTRs are summarized in Table I.

The current generation of HTRs operates on a conventional steam cycle. However, advanced designs with direct-cycle helium turbines and reformers for industrial process heat are under active development both in the FRG and in the U.S., and development of an HTR for process heat is in progress in Japan. Development of nuclear process heat is of particular interest in the FRG and in certain other countries with limited indigenous resources of high-grade fossil fuels. Both direct cycle and process heat HTRs become more attractive with higher core outlet temperatures than those typical of steam cycle designs. Higher primary coolant temperature can be achieved with current fuel technology through engineering design trade-offs. In fact, sustained operation of the AVR at outlet helium temperatures of 950°C (1223 K) (Ref. 5) demonstrated that it is within the



(a) (b)

Fig. 1. The HTGR fuel elements: (a) cross section of a spherical fuel element for the pebble bed HTGR, and (b) fuel element for the prismatic core HTGR.

TABLE I

Characteristics of Experimental and Demonstration HTGRs

	Peach Bottom	Dragon	AVR	FSV	THTR
Date of first operation	1966	1967	1967	---	---
Thermal/electric power, MW(e)	115/40	20/-	46/15	851/330	750/300
Helium inlet/outlet temperature, °C[a]	377/750	350/750	290/950	405/784	270/750
Mean helium pressure, bar	25	10	10	45	39
Fuel element type	Tube	Tube	Pebble bed	Prismatic	Pebble bed
Fuel kernel type, s	$(Th,U)C_2$	Various (UO_2 driver fuel)	$(Th,U)C_2$, $(Th,U)O_2$	$(Th,U)C_2/ThC_2$	$(Th,U)O_2$
Coating type	Biso[b]	Triso[c]	Biso[b]	Triso[c]	Biso[b]

[a] $t_K = t_C + 273$.

[b] Two-layer coating of a relatively dense, impermeable PyC layer over a low-density "buffer" coating.

[c] Four-layer coating consisting of a composite structural coating, made up of two relatively dense PyC layers encasing a SiC layer, over a buffer coating.

realm of current technology. However, development of coated particles with improved high-temperature capability is an important factor in making the HTR even more attractive for future applications.

COATED PARTICLE FUEL

Coated particles are in themselves miniature fuel elements on the order of a millimeter in diameter. A commercial reactor core contains $\sim 10^{11}$ individual fuel particles. The coatings, and to a lesser extent the fuel kernels themselves, provide the primary barrier to fission product release. The very small size of coated particles is an advantage in testing, since statistically significant numbers of "fuel elements" can be tested. Individual tests typically contain 10^3 to 10^5 coated particles. Through properly designed fuel development and test programs, fuel performance in-service can be predicted with a high degree of confidence. Processes that lead to loss of coating integrity can be defined in detail and minimized by design either of the fuel particles themselves or of the reactor core.

The two coated particle types in most common use are the two-layer Biso coating and the four-layer Triso coating with its interlayer of SiC between two layers of high-density isotropic (HDI) PyC (Fig. 2). Seal coats of dense, relatively anisotropic PyC are sometimes used between the buffer and HDI layers in Biso particles. This is primarily to minimize reactions between oxide kernels and carbon coatings during processing at high temperatures. Both Biso and Triso particles are capable of essentially complete retention of gaseous fission products with properly designed and specified coatings. Intact Triso particles also provide nearly complete retention of metallic fission products at current peak HTR design temperatures. However, diffusional release of certain metallic fission products, particularly cesium, strontium, and silver does occur at elevated temperatures from Biso coatings. Biso coatings are generally selected where fuel temperatures are relatively low, or with relatively low burnup oxide fuel where the kernel provides sufficient retention of metallic fission products.

Fuel development is complete for HTRs now under construction. However, substantial development programs are in progress directed toward optimization of the fuel. Most current coated particle fuel development has one or more of the following objectives:

1. development of completely separable thorium-cycle fuel

2. simplifications in processes and optimization of specifications

3. improvements in performance predictions

4. development of fuels with improved temperature capability and reduced fission product release.

One of the attractive features of the HTR is its flexibility in use of different fuel cycles. The thorium cycle with both separable and mixed fuel,

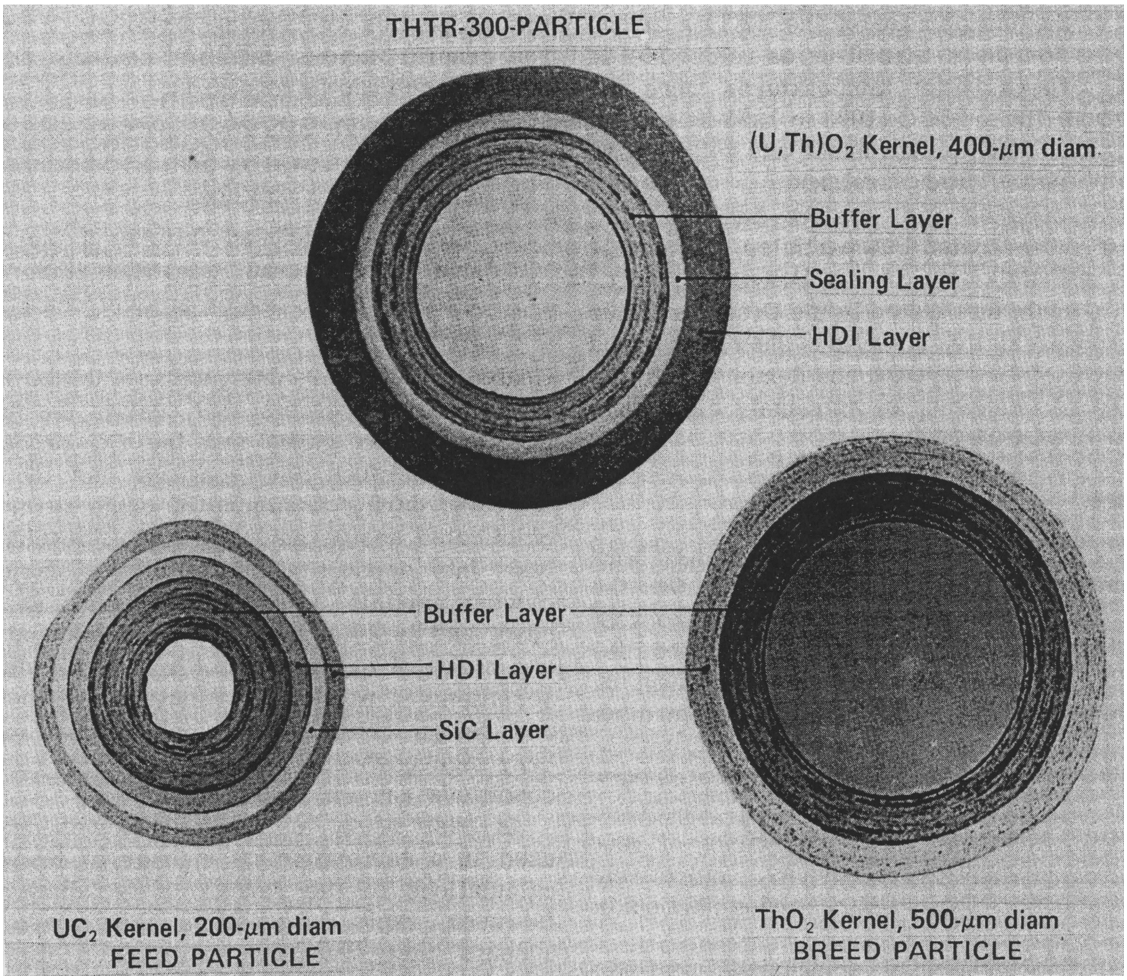


Fig. 2. Coated particles for the U/Th fuel cycle.

the low-enriched uranium cycle, and even cycles based on plutonium fissile particles are feasible. Fuel particles characteristic of those required for each of these cycles have been successfully tested in one or more of the prototype HTRs as well as in Materials Test Reactors.

Much of the fuel development effort over the last several years, particularly in the U.S., has been directed toward the development of completely separable fuel for the thorium fuel cycle.[6] This type of fuel optimizes fuel economics and resource conservation by allowing essentially complete separation, during reprocessing, of the ^{233}U bred in the thorium-containing particles from the neutron poison, ^{236}U, generated in ^{235}U fissile particles. The optimum fuel system that was selected is a Triso fissile particle with a UC_2 kernel 93% enriched in ^{235}U, and a Biso fertile particle with a ThO_2 kernel. The use of Triso coatings on the fissile particles and Biso coatings on the fertile particles allows separation of fissile and fertile species during reprocessing by a burn-leach process. Performance of this type of fuel was demonstrated in a large number of irradiation experiments over the entire range of conditions anticipated for the current generation of HTRs. Completely separable fuel represents an evolutionary advance over the fuel used in the initial core loading of the FSV reactor, which contained fissile particles of mixed uranium and thorium carbide (4-to-1 ratio) and fertile particles containing only thorium carbide. In both the completely and partially separable systems, fissile and fertile particles are uniformly blended in the fuel rods to obtain spatially uniform heat generation.

A second type of thorium-cycle coated particle under active development is the "mixed" $(Th,U)O_2$ particle with either Biso or Triso coatings.[7] Although this fuel is not optimized from the standpoint of resource conservation, it has substantial advantages of simplicity in fabrication and in reprocessing. It can also be used to advantage in advanced pebble bed cores that do not require variations in the thorium-to-uranium ratio. This type of fuel was also extensively tested, and will be used in the initial core of the THTR. It has been in use for more than 6 yr as reload fuel in the AVR, where outlet temperatures were increased from 750 to 950°C (1023 to 1223 K) without deterioration of fuel performance and with limited increase in fission product release.

The low-enriched fuel system for the HTR, employing Triso-coated UO_2 particles, was developed by the UKAEA and the Dragon Project.[8] Production-scale fuel tested in the Dragon reactor at fully representative conditions showed the good performance of this system and demonstrated the possibility of achieving an extremely clean primary circuit with an HTR.

Coating Materials

The PyC is a unique material that has been central to coated particle development from the earliest days. The pioneering work of Bokros[9] showed that the structure and irradiation behavior of PyC coatings are highly dependent on deposition conditions, which in turn determine coating properties such as density and crystalline anisotropy. Current work in this area is emphasizing the optimization of deposition processes and specifications and the development of improved characterization techniques. Anisotropy and permeability of PyC's are receiving particular attention.

Early PyC development emphasized coatings deposited from methane at relatively high temperatures [1800 to 2100°C (2073 to 2373 K)], often referred to as high-temperature isotropic (HTI) coatings. More recent work emphasized coatings deposited at lower temperatures [1200 to 1400°C (1473 to 1673 K)] from a variety of hydrocarbons such as propane, propene, propene mixed with acetylene, and butane. These are conventionally referred to as low-temperature isotropic (LTI) coatings. The LTI coatings are preferable from the manufacturing standpoint and have much better irradiation performance than HTI coatings. However, intact LTI coatings are significantly less retentive for the volatile metallic fission products such as cesium than comparable HTI coatings. In spite of this disadvantage, the superior irradiation stability of LTIs has led to widespread use.

The SiC is a much simpler coating material than PyC because its properties are independent of the production process when deposited under optimum conditions. It provides mechanical strength to Triso-coated particles and near-complete retention of fission products.

Coated Particle Performance

Once the coating materials are specified to ensure that particle performance is not limited by the properties of coating materials themselves, two general factors that limit coated particle fuel performance can be identified:

1. mechanical failure of coatings due to internal gas pressure and irradiation-induced effects in the coatings
2. chemically induced failure of the coatings due to fuel kernel migration (the "amoeba effect") or to fission product/coating interactions.

Coated particles are designed as miniature spherical pressure vessels that contain the pressure due to fission gases and the CO produced by reaction between the coatings and the oxygen liberated during fissioning of oxide kernels. Sophisticated design models[10,11] were developed that take into account a multiplicity of factors including particle dimensions, internal gas pressure, and irradiation-induced dimensional change and creep of PyC coatings. In general, statistical variations of particle parameters are considered and design criteria are established to ensure a very high probability of survival under all normal conditions and anticipated transients.

In past work, fuel performance models were used in a semiempirical manner by normalizing calculations to the results of irradiation tests. However, rapid progress is being made, and it appears likely that performance calculations from first principles will be feasible in the future.

Thermochemical aspects of fuel performance are of particular interest in the development of fuel for advanced HTR systems since they are generally controlled by highly temperature-dependent thermally activated processes. Considerations of the thermal performance of coated particle fuels were dominated by the amoeba effect, a phenomenon in which fuel kernels migrate into the carbon coatings under the influence of a temperature gradient. The amoeba effect was shown to be a significant factor in the high-temperature performance of both carbide and oxide fuels. The mechanism of the amoeba effect in carbide fuels is controlled by thermal diffusion of carbon in the fuel phase. The mechanism is less well defined in the case of oxide fuels. However, current research is yielding excellent data both on amoeba migration kinetics and on the mechanism of migration in oxides.

More recently, attention was focused on reactions between fission products and the SiC layer as a performance-limiting factor at very high temperatures. An important example is the reaction involving the lanthanide-group fission products in carbide fuels. This particular reaction is generally not observed in oxide fuel, apparently because the lanthanide fission products are retained in the fuel kernel as stable oxides. Similar reactions involving the palladium-group metals were observed in low-enriched oxide fuels. Work is in progress at a number of sites to accurately define the kinetics of these processes.

Both carbide and oxide fuels are used in HTR applications. The selection for a particular application depends on a combination of factors including resistance to amoeba migration, resistance to fission product/coating reactions, retention of metallic fission products such as strontium, reactions with moisture, and ease of manufacture. There was recently considerable interest in oxycarbide fuel kernels, which appear to combine many of the advantages of both kernel types. The weak acid resin kernel fabrication route, developed by Oak Ridge National Laboratory (ORNL) (Ref. 12), provides a particularly convenient means of obtaining oxycarbide kernels and is also amenable to use in remote refabrication of recycle fuel kernels.

The HTR core design is optimized for each type of fuel system by using kinetic data on amoeba migration, fission product/coating reactions, and other performance-limiting phenomena to establish limits on peak temperatures, temperature gradients, and time-at-temperature. Simple criteria, such as those used to control clad melting in light water reactors, are not applicable since the graphite and PyC that serve as "cladding" in the HTR do not melt and, in fact, retain their integrity to temperatures well beyond those anticipated for HTR transients. Also, there is no phase change in the coolant that could lead to a rapid degradation of heat transfer characteristics. All coated particle fuel failure mechanisms lead to only a gradual increase in failure as the fuel temperature is increased beyond the normal range. Several experiments, including the large-scale "high-temperature experiment" in the Dragon Reactor,[13] demonstrated that only limited fuel failure occurs at temperatures as high as 1800 to 2000°C (2073 to 2273 K) [~500 to 700°C (~773 to 973 K) above normal operating temperatures].

Fission Product Release from Coated Particle Fuels

The ultimate goal of all coated particle research is to define and minimize fission product release. The fission product isotopes of most interest from the standpoint of plant maintenance and potential safety considerations are those of the fission gases (krypton and xenon), iodine, and the metals cesium and strontium. Silver can also be important to maintenance considerations, particularly in the low-enrichment system where the fission yield is relatively large.

Properly specified and manufactured coatings of both Biso and Triso types are impermeable to fission gases and iodine. Consequently, fission gas release occurs only from failed fuel or tramp fuel not protected by coatings. Fission product gases of major radiological significance generally have relatively short half-lives. Only those short-lived fission product atoms lying very near the surface of the kernel can be released by diffusion before they decay; the exact distance is a function of the half-life. Consequently, fission recoil becomes an important release process; depending on geometrical factors, several percent of the fission products will be released from the fuel kernel by fission recoil. Recoiled atoms tend to stop in surrounding solid material, such as carbon coatings, from which they are released by diffusional processes. Consequently, diffusion is the dominant release process even for recoiled fission products. The competition between diffusion and decay results in a dependence of release on the square root of the half-life at temperatures where diffusional processes dominate. Only for very long half-lives does the release approach unity. The decay process is so significant in the release of relatively short-lived isotopes that the fractional release of important isotopes such as ^{133}I, ^{88}Kr, and ^{133}Xe is only on the order of 1% for failed fuel under reactor conditions. This is demonstrated in Fig. 3, which shows the release of short-lived isotopes from an experiment (GAIL III B) containing 100% failed fuel.[14]

The release of metallic fission products is more complex than release of the gases since diffusional release through intact Biso coatings must also be considered at high temperatures. There is also evidence that ^{110m}Ag can be released from apparently intact Triso coatings at high temperatures. Many of the metallic isotopes of interest (e.g., ^{90}Sr and ^{137}Cs) are very long lived.

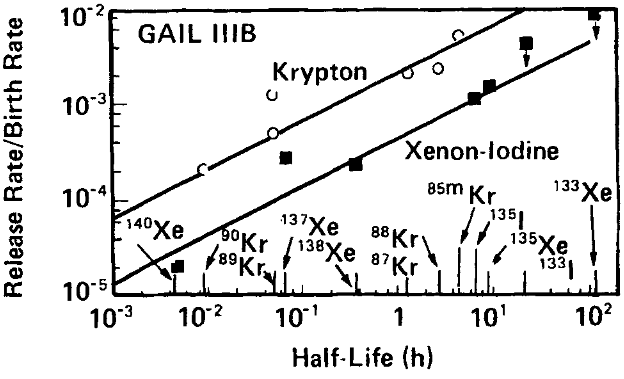


Fig. 3. Release of short-lived fission products from the GAIL III B experiment containing 100% failed fuel [normalized to 1100°C (1373 K)].

Consequently, radioactive decay is of less importance than for the fission gases, and diffusional release from the fuel kernel is often more important than release by fission recoil.

The fuel kernel plays an important role in the release of metallic, as well as gaseous, fission products. Strontium, which forms a stable oxide, is strongly bound in oxide fuel kernels, while cesium and silver, which do not form stable simple oxides at reactor temperatures, are much more mobile. Carbide kernels are much less retentive of metallic fission products than oxide kernels. The diffusivity of the fission product metals in PyC also varies widely, with the following relationship being applicable: $D_{Ag} > D_{Sr} > D_{Cs}$. The situation is further complicated by an apparent structure dependence of diffusivity in PyC with HTI PyC's significantly more retentive for cesium than LTI PyC's.

Coated Particles for the Future

The development of HTRs for direct cycle or process heat applications is aimed not only at obtaining higher coolant outlet temperatures, but also at minimizing radioactive contamination of the primary circuit to facilitate inspection and maintenance of primary circuit components. Consequently the development of improved fuels has two major goals: improvement in fuel temperature capability and reduction in fission product release (particularly metallic fission products).

With the detailed understanding of fuel performance and fission product release that was developed over the last 15 yr, the ways in which improvements can be made are becoming apparent. The first line of defense against fission product release is the fuel kernel. It is known that the high oxygen chemical potential of oxide and oxycarbide kernels results in good retention of many of the metallic fission products through the formation of stable oxides. The retention of certain metallic fission products can be further improved by the addition of materials that form even stronger compounds.[a] This area of research, originated by workers at the Kernforschungsanlage[15] offers promise for reducing plated-out fission product activities in the HTR primary circuit.

Improved coatings are being developed to increase the strength and integrity of the particle, to allow use of thinner coatings for high conversion ratio HTR designs, to reduce the diffusional release of metallic fission products, and to improve the overall chemical stability and temperature capability of coated particles. Two of the most promising areas of research on improved coatings are the use of co-deposited PyC-SiC (or ZrC) alloys as potential replacements for Triso coatings, and ZrC as a replacement for the SiC in Triso coatings. These developments hold considerable promise for application to advanced HTR systems.

Organization of the Special Issue

Although there were periodic meetings on gas-cooled reactor technology,[16-18] coated particle fuel technology itself has not been comprehensively treated in a single publication since the two symposia in the early 1960's. This special issue of *Nuclear Technology* is intended to fill that gap by providing a compilation of papers, from a variety of sources, that are representative of the current state-of-the-art in coated particle fuel development. Although no effort such as this can be completely comprehensive, an attempt was made to include a broad range of papers to put the current state of coated particle fuel technology in perspective and to provide a point of departure for future work.

A concluding paper is provided in which some of the most important developments presented in the contributed papers are briefly discussed.

The continued strongly international nature of coated particle fuel development is reflected in the papers in this issue. Workers from six countries and many organizations are represented as authors. Most of the papers fall into five general categories:

1. fabrication
2. properties and characterization of PyC and SiC coatings
3. performance and performance modeling
4. fission product release
5. advanced or improved fuels and applications.

These categories represent the most active areas of coated particle development at the current time, and they form the structure around which this special issue of *Nuclear Technology* is organized.

ACKNOWLEDGMENTS

The organizers of this special issue gratefully acknowledge the many members of the HTR community who have contributed to this project by contributing papers and participating in reviews. The efforts of J. Coobs in coordinating the contributions from ORNL are particularly acknowledged. Finally, we

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[a] The addition of SiO_2 and Al_2O_3 stabilizes cesium and strontium in complex aluminosilicate compounds.

acknowledge our debt to the many early workers in the coated particle field, who through innovation, enthusiasm, and hard work established the basis for what is now a mature technology.

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