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ABSTRACT

The thorium cycle HTGR typically employs fissile particles containing fully enriched uranium and fertile particles containing thorium. Small fuel kernels ($\sim 10^{-5}$ cm^3 in volume) are separated by coating layers and matrix material so that the mechanical and thermal properties of the fuel body are to a large extent independent of the fuel kernel properties. This feature gives rise to an advantageous flexibility in kernel selection. Both dioxide and dicarbide kernels of Th and U and solutions of the two have been developed. During irradiation carbide and oxide kernels undergo swelling and accumulate a significant fission product inventory. Interaction between the kernel and outer structural coatings is prevented by an intervening low-density buffer layer. Kernel migration into the coating up the thermal gradient is exhibited by both carbides and oxides. A limitation of coating penetration due to kernel migration in carbide fuels forms the basis for core thermal design. Thorium oxide is the most stable kernel material with little propensity for migration relative to other oxides and the carbides. Mixed oxide fuels have been favored in European HTR design. In the United States where a segregated thorium cycle is employed, fissile UC_2 and fertile ThO_2 kernels have been selected for HTGR fuel based on optimization of performance and predictability under operating conditions.

INTRODUCTION

The fuel for the High-Temperature Gas-Cooled Reactor (HTGR) consists of fuel microspheres about 10^{-5} cm^3 in volume, surrounded by a low-density sacrificial buffer carbon layer and a single pyrocarbon (BISO) or composite pyrocarbon, SiC (TRISO) structural coating layer (Ref. 1). The purpose of the buffer layer is to decouple the fuel phase from direct mechanical or chemical interaction with the structural coatings, which provide the primary barrier to fission product release. Because of the physical isolation of the fuel kernel, properties of the fuel body itself are to a large extent independent of the properties of the fuel phase. This allows for considerable flexibility in the choice of fuel type and fuel cycle in the HTGR system.

The two fuel cycles that are currently receiving the most attention are the low-enrichment cycle (Ref. 2) and the thorium cycle (Ref. 3). The thorium-cycle has significant economic advantages under United States conditions of enriched uranium supply and is being utilized commercially by GGA with support from the USAEC for recycle development. The development of the thorium cycle in the United States has progressed from complete mixing of fissile and fertile material in the Peach Bottom reactor through partial separation in Fort St. Vrain to complete separation in the commercial plants. The evolution of fuel kernel materials for the HTGR in the United States and the parallel developments in Europe are illustrated in Fig. 1.

In the course of HTGR fuel development, a wealth of knowledge about irradiation and thermal performance of kernel materials has been developed. The properties and performance of the principal kernel materials are discussed in the following sections.

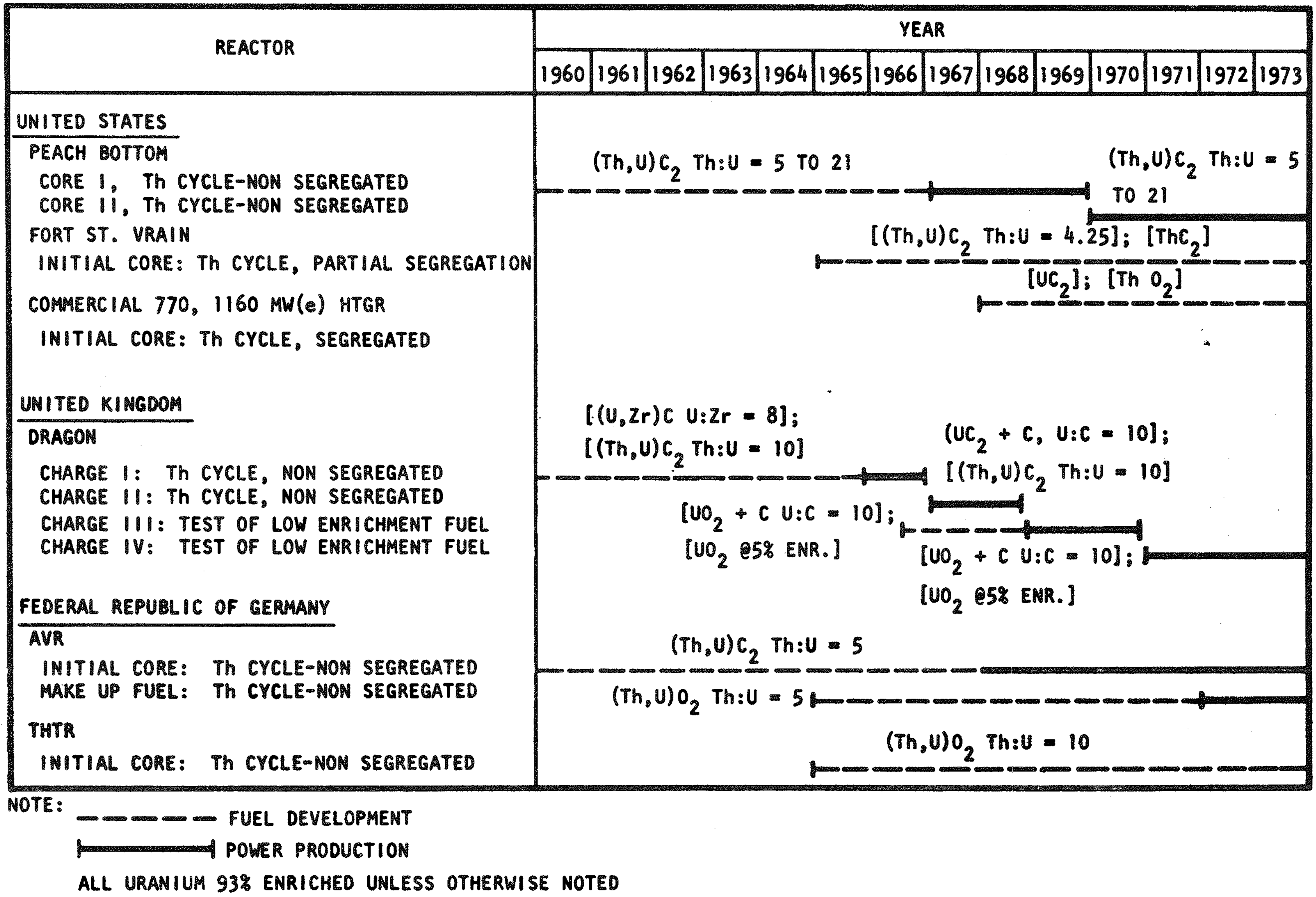

Fig. 1. Evolution of HTGR fuel kernel materials

CARBIDE FUEL KERNELS

The refractory nature and good thermal conductance of carbides, together with the availability of technology, favored the use of carbide materials in early HTGR development. For example, several types of coated carbide kernels were used in the first two core charges of the Dragon reactor in the United Kingdom and the initial charge of the AVR in the Federal Republic of Germany. The fissile kernels were diluted with Zr, C, or Th to restrict burnup and ensure coating survival. In the United States coated $(Th,U)C_2$ particles were used in producing the first electrical power from an HTGR in the Peach Bottom Nuclear Power Station on January 27, 1967. The fuel for the Fort St. Vrain HTGR contains fissile $(Th,U)C_2$ and fertile ThC_2 kernels. A good understanding of the performance of carbide coated particle fuels has developed from this background of experience.

FABRICATION AND GENERAL CHARACTERISTICS

The carbide fuels usually consist of ThC_2, UC_2, or mixtures of the two in solid solution. Kernels of this type can be produced by carbothermic reduction of oxide microspheres obtained by agglomeration or sol-gel techniques. However, the primary United States development effort has been conducted with kernels manufactured by a melting and spheroidization process (Ref. 4). Kernels produced by this latter method have diameters ranging from 100 to 500 μm and are near theoretical density. Kernels of this type have been used in the Peach Bottom, Fort St. Vrain, AVR, and Dragon reactors.

The carbide kernels are commonly fabricated with excess carbon which precipitates as proeutectic graphite on the kernel surface or in a eutectic structure within the kernel. The proeutectic graphite rejected during spheroidization on the kernel surface has the beneficial effect of providing limited protection of the carbide kernel from interaction with ambient moisture. However, the graphite shell, which is most prominent in UC_2, is not relied upon for long-term protection. Carbide kernels are handled and stored in inert gas until a coating of pyrocarbon is applied in the fuel production process.

IRRADIATION PERFORMANCE

Carbide fuel kernels that have been investigated in irradiation capsules have included fertile ThC_2 and fissile $(Th,U)C_2$ or UC_2 with 93% enrichment in U-235. During irradiation, up to 75% of the total uranium and 7% of the thorium after transmutation to U-233 are fissioned at a maximum neutron exposure of 8×10^{21} n/cm^2 ($E>0.18$ MeV). One of the most significant irradiation effects is kernel swelling resulting from gaseous and solid fission product generation. An example of the kernel diameter change with increasing irradiation temperature and burnup in ThC_2 and $(Th,U)C_2$ is shown in Fig. 2. Diametral swelling greater than 30% has not been observed even at a high burnup of 75% FIMA in UC_2. The multilayered coating concept of HTGR fuel provides a low-density sacrificial buffer coating adjacent to the kernel, which yields to the swelling kernel and minimizes mechanical or chemical interaction with the outer structural coatings under reactor operating conditions of time, temperature (<1250°C), and burnup.

THERMAL PERFORMANCE OF CARBIDES

The thermal characteristics of carbide kernels are well understood and the predictability of performance contributes to confidence in core thermal design. One predictable characteristic is kernel migration up an imposed thermal gradient. This phenomenon has been called the "amoeba effect" (Ref. 5). An example of the amoeba effect that took place in fertile particles in a fuel rod irradiated at high temperatures is shown in Fig. 3. Kernel displacement up the thermal gradient is effected by carbon taken into solution from the adjacent pyrocarbon coating on the hot side, transported across the fuel phase, and rejected as graphite on the cool side (Ref. 6). From theoretical consideration a kernel migration coefficient (KMC), which is a temperature and temperature gradient normalized migration rate, can be defined as follows:

$$KMC = \frac{dx}{dt} (T^2) \frac{1}{\Delta T/\Delta X} \quad .$$

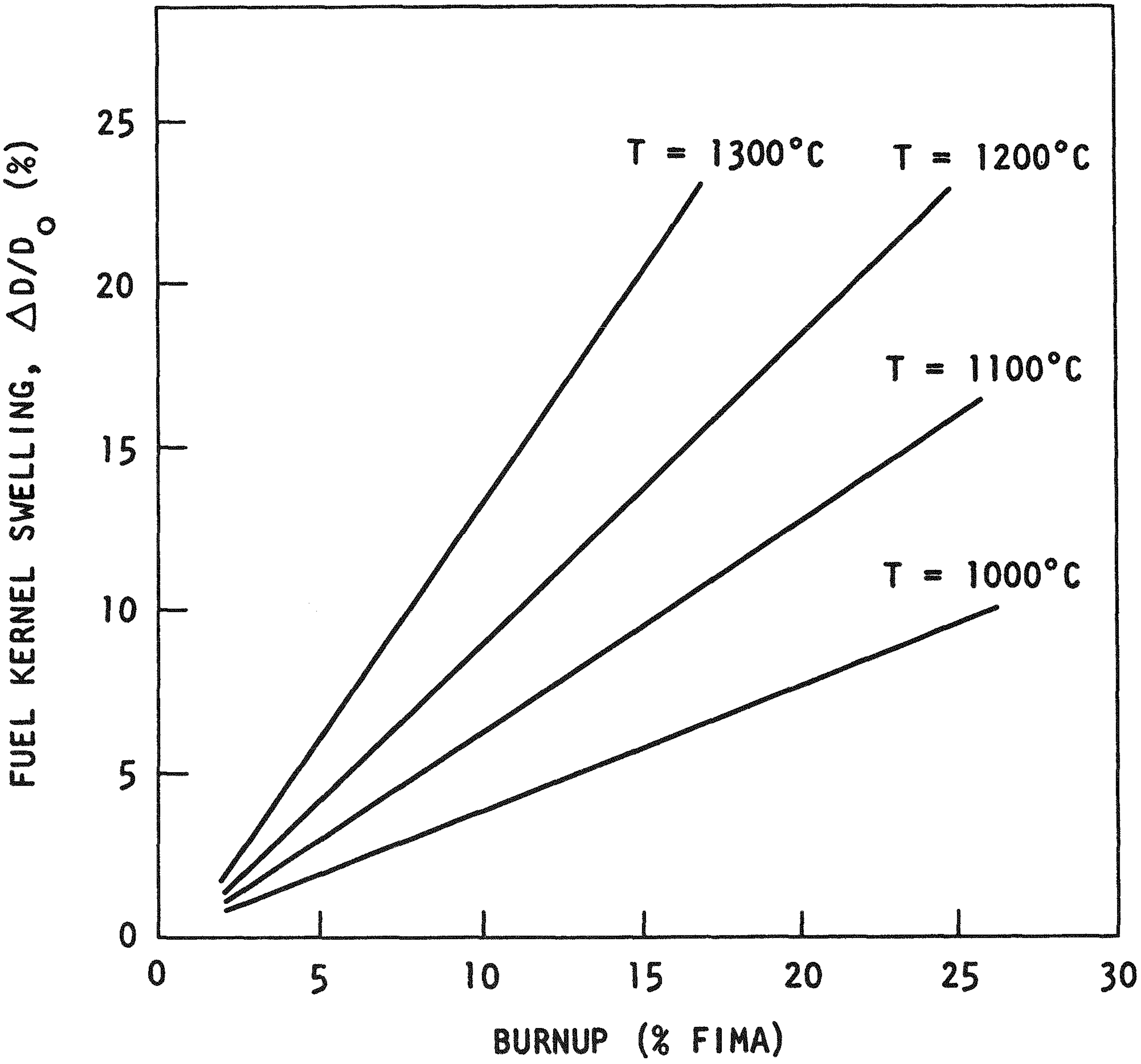

Fig. 2. Fuel kernel swelling in ThC_2 and $(Th,U)C_2$ as a function of burnup and irradiation temperature

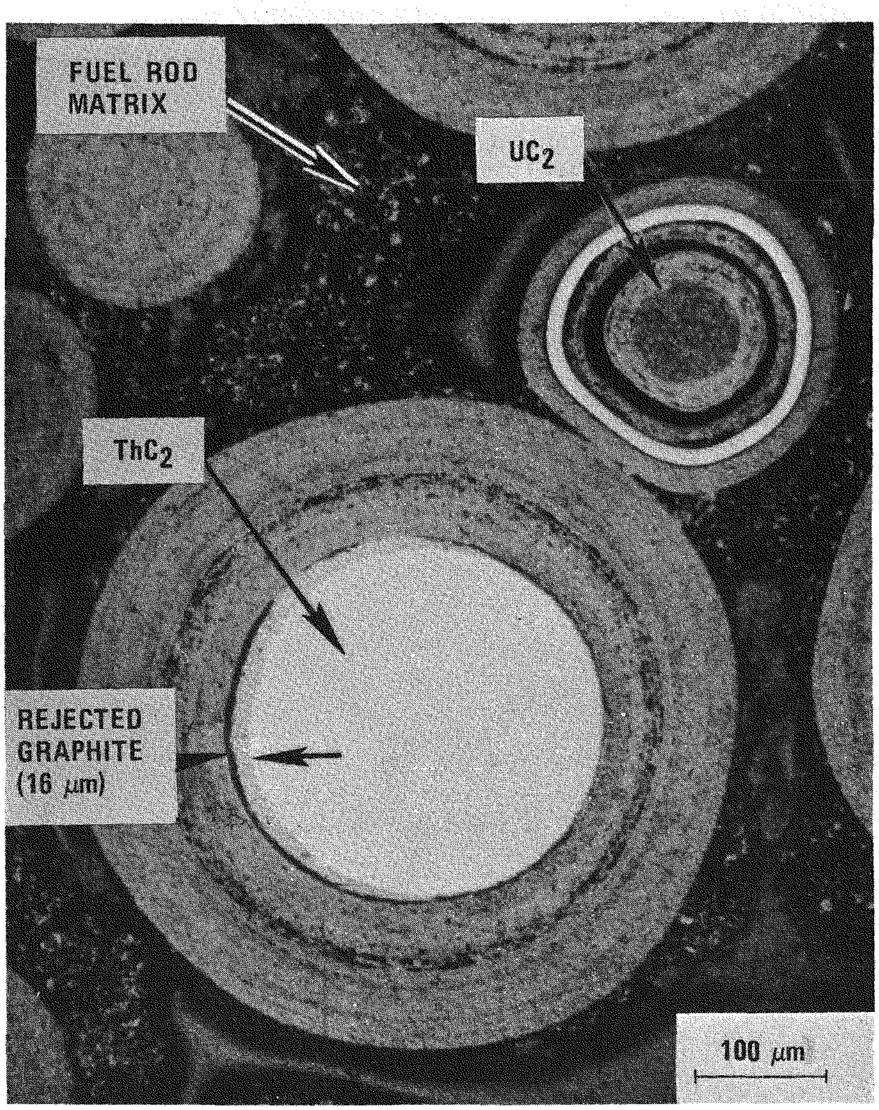

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Fig. 3. Coated fertile and fissile kernels irradiated to 4.4 x 10^{21} n/cm^2 ($E > 0.18$ MeV). The ThC$_2$ kernel (~3% FIMA) displayed kernel migration while an adjacent UC$_2$ kernel (50% FIMA) exhibited no detectable migration during irradiation at 1340°C, for 6215 hours under a thermal gradient of ~200°C/cm.

An Arrhenius relationship is predicted for the KMC, and empirical verification has been obtained with carbide kernel migration data gathered by heating irradiated and unirradiated coated particles in a known thermal gradient (Ref. 7). An example of the temperature dependence of KMC based on a least-squares fit of data for unirradiated ThC_2 is shown by the curves in Fig. 4. While some change in temperature dependence of KMC might be expected in the different equilibrium phases of ThC_2, the differences have not been detectable. Fig. 4 also includes in-pile and postirradiation heating data points for ThC_2 migration which show irradiation and burnup have no measurable effect on kernel migration rate (Ref. 7). Similar results have demonstrated that amoeba migration in UC_2 is not enhanced by irradiation even at very high burnups.

The thermal design of the HTGR assumes maximum migration rates (the upper 95% confidence boundary for KMC) and limits the total extent of kernel migration to a fraction of the coating thickness in the most extreme conditions of time-temperature and thermal gradient. This procedure ensures that no coating failure will take place as a result of kernel migration.

The rate of advance of the hot kernel surface under the same thermal conditions can be ranked in the following order: $ThC_2 > (Th,U)C_2 > UC_2$, with the rate for ThC_2 being 5 to 10 times greater than for UC_2. The tendency for migration in ThC_2 favored the development of another fertile material, ThO_2, as an alternative for ThC_2 in commercial HTGR fuel.

OXIDE FUEL KERNELS

Oxide fuel kernels have a number of potential advantages over carbides, including resistance to hydrolysis during fuel fabrication and irradiation, narrow size distribution, and relative ease of remotizing the fabrication process. For these reasons the performance of oxide coated particle fuels has been extensively studied both in the United States and in Europe since the mid-1960s.

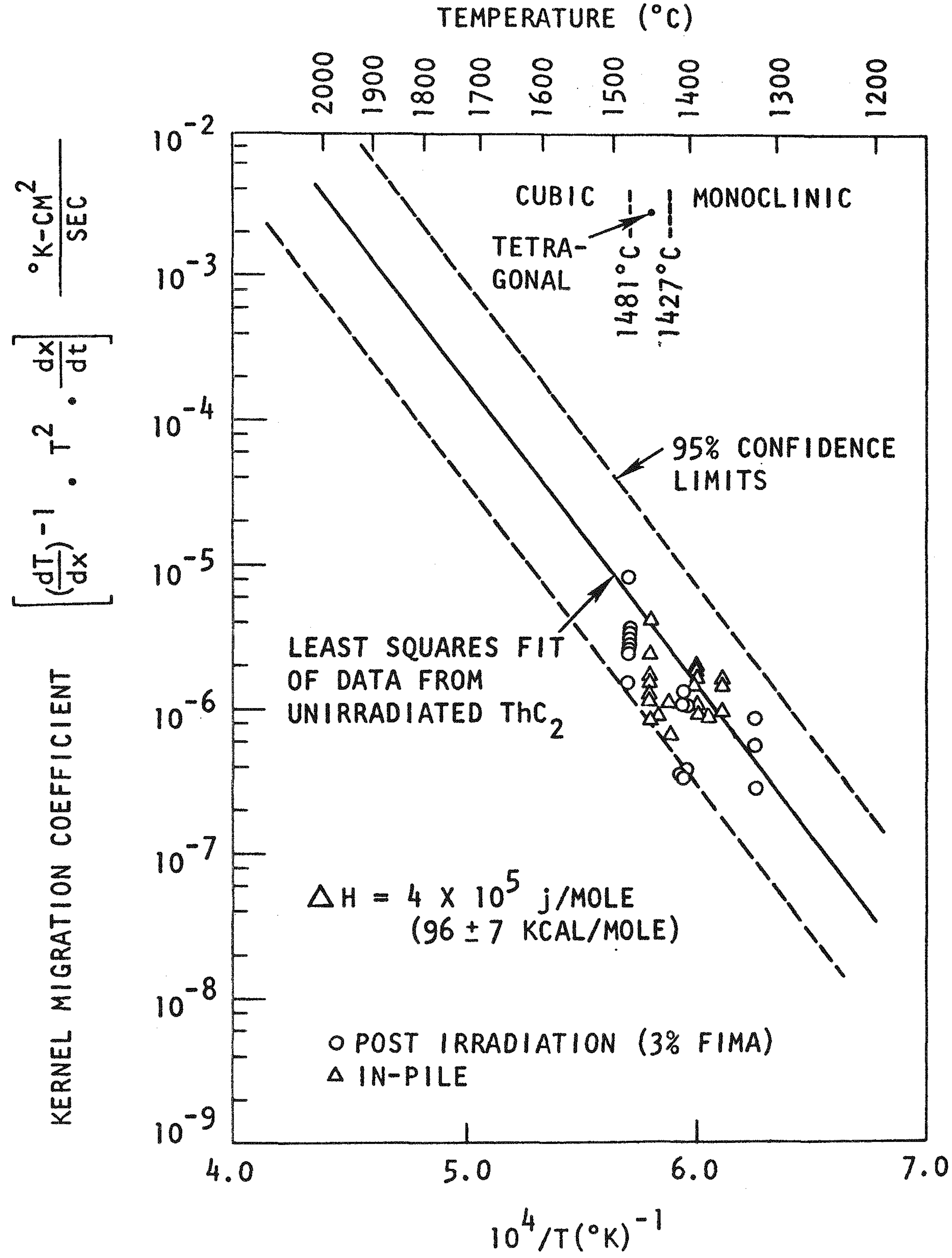

Fig. 4. Temperature and thermal gradient dependence of ThC_2 kernel migration showing independence from irradiation effects

FABRICATION AND CHARACTERISTICS OF OXIDE KERNELS

Oxide kernel development in the United States has centered around a modification of a gel-supported precipitation method of microsphere production (Ref. 8). In this process spherical droplets of a colloidal solution of ThO_2 and/or UO_2 are caused to harden while passing through a second phase of immisible gas or liquid. The hardened microspheres are dried and densified in subsequent heating cycles. Oxide microspheres have been produced with a final diameter of 100 to 500 μm and a density of >95% theoretical.

IRRADIATION PERFORMANCE

Irradiation of UO_2 produces voids in the kernel. The void formation is accompanied by kernel swelling that is accommodated by the buffer layer, thus preventing mechanical interaction between the kernel and the structural coatings. An example of UO_2 microstructure at high burnup (70% FIMA) is shown in Fig. 5. Highly reflective inclusions that are commonly observed within irradiated UO_2 kernels are rich in Ru, Mo, and Tc. Relative to UO_2 the microstructure of ThO_2 kernels is not severely altered during irradiation because of the low burnup (7% FIMA) and consequent low fission density. An example of the irradiated microstructure of ThO_2 is shown in Fig. 6.

THERMAL STABILITY

Kernel migration in a thermal gradient has been observed in irradiated UO_2 (Refs. 9,10) and an example of the effect is shown in Fig. 5. The relation of coating failure to migration rate has been empirically correlated with operating parameters of power, time, temperature, and gradient in low enriched UO_2 fuel by the Dragon Project (Ref. 10) for use in core thermal design. Kernel migration in ThO_2 is much slower than in UO_2 under comparable conditions and is not a limitation in thermal performance. In fact, kernel migration in ThO_2 or $(Th,U)O_2$ had not been observed even under severe conditions until recently (Ref. 11).

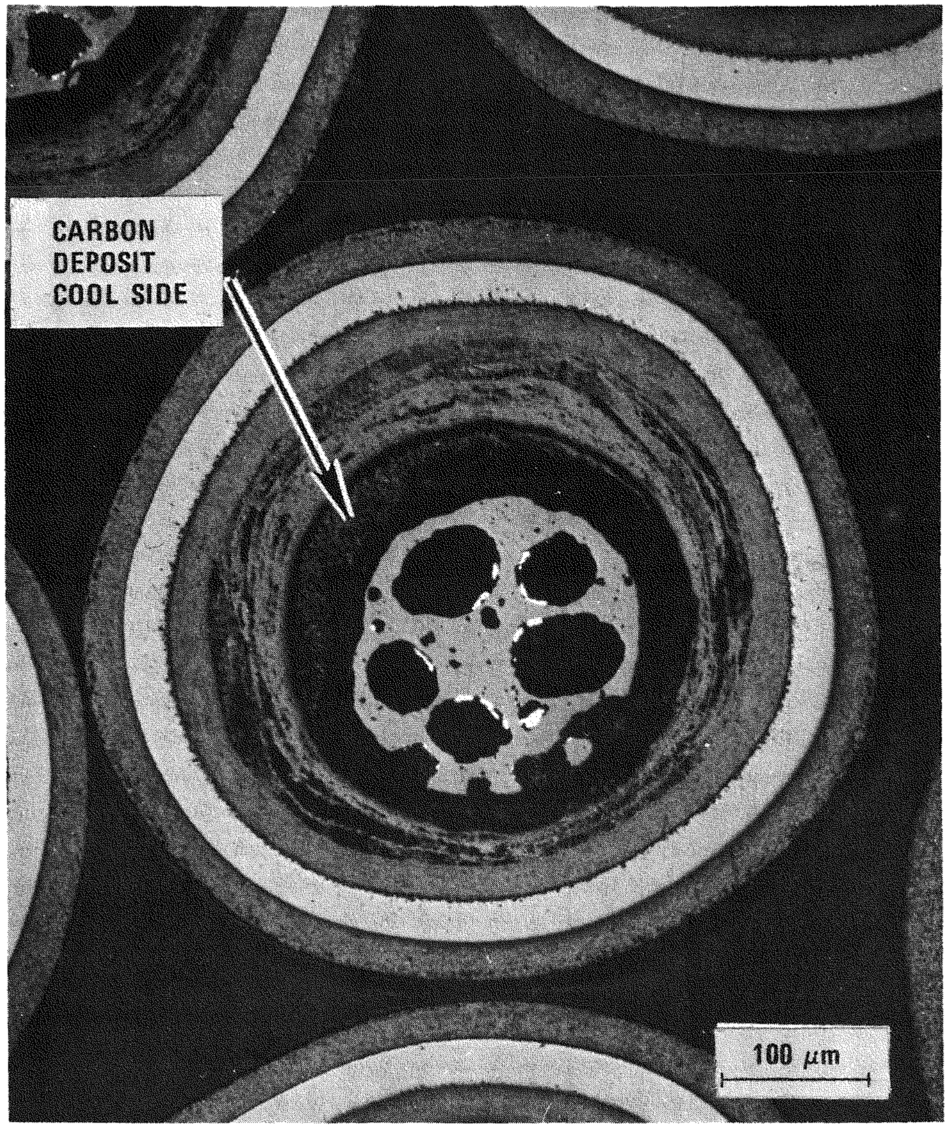

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Fig. 5. TRISO coated UO_2 kernel irradiated at 1150°C to 70% FIMA at 4.7 x 10^{21} n/cm^2 ($E > 0.18$ MeV). Carbon adjacent to the kernel on the cool side was transported from the hot side in conjunction with kernel displacement up the thermal gradient.

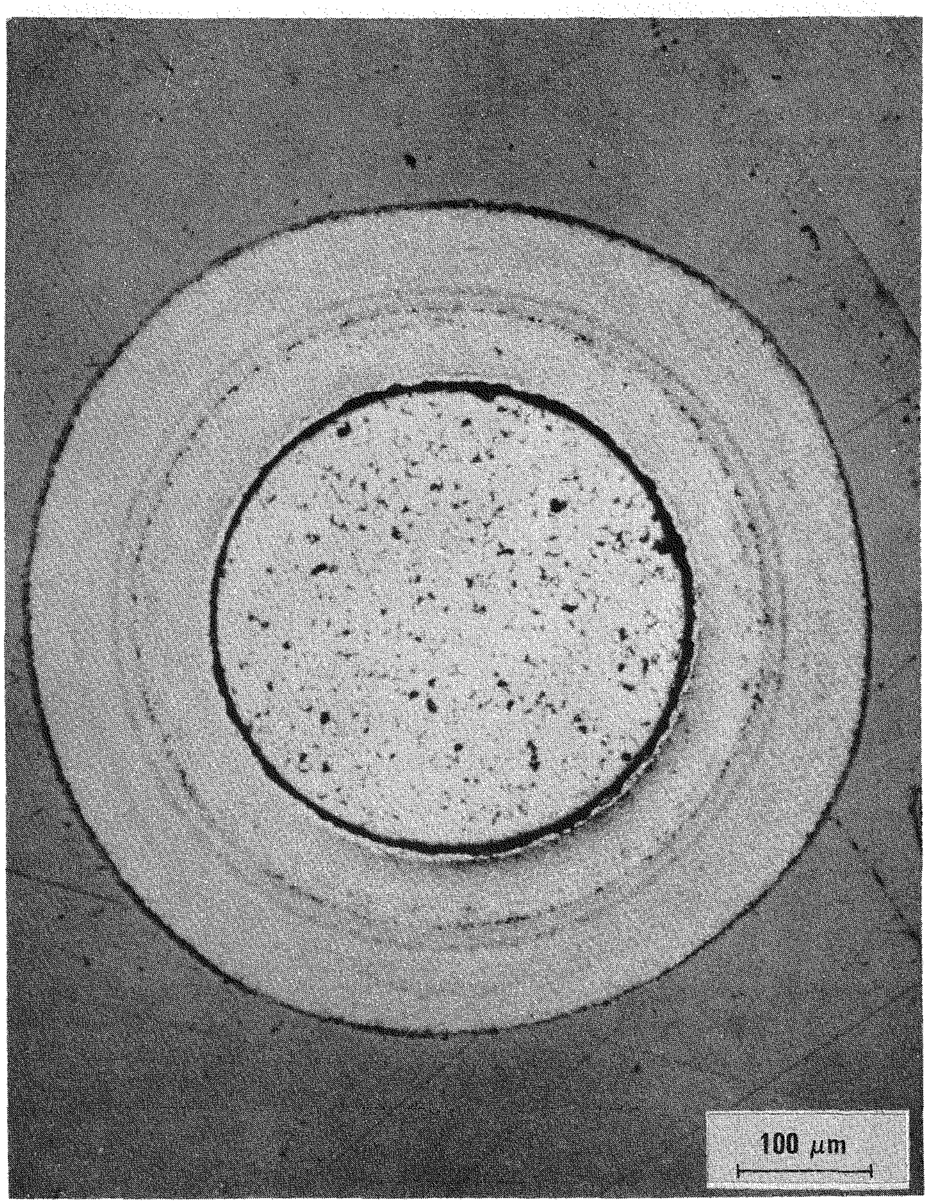

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Fig. 6. BISO coated ThO_2 kernel irradiated at 1150°C to 11% FIMA and 8 x 10^{21} n/cm^2 ($E > 0.18$ MeV). Irradiation effects on kernel microstructure and kernel-coating interaction are minimal.

A kernel migration mechanism has been proposed based on the reaction between CO-CO_2-carbon and thermal diffusion of gases in the buffer void volume (Ref. 12). Observations of UO_2 migration at temperatures less than ∿1500°C have not shown a strong temperature dependence, suggesting gas transport may be rate controlling. For example, in experiments between 900° and 1500°C an increase in thermal gradient from 300° to 900°C/cm compensated for a 600°C decrease in temperature with no resultant change in kernel migration rate (∿10^{-10} cm/sec).

While the CO/CO_2 gas pressure within the irradiated oxide particle is of concern with regard to the amoeba effect, the contribution of CO/CO_2 to total gas pressure is also an important parameter in design of the particle coatings. Internal gas pressures contained in the particle void volume (∿10^{-5} cm^3) are typically 200 to 600 atm at temperatures of 800° to 1400°C. Pressures are estimated from measurement of gas volume released upon cracking particles at high temperatures. They show that the release of oxygen during fission is no higher than the caluclated values of 0.13 oxygen atom/U-235 fission or 0.08 oxygen atom/U-233 fission (Ref. 12).

In coated UO_2 particles at high burnup, Si transport from the SiC coating to the kernel can occur if there is a breach in the intervening coatings. Stable compounds are formed in the kernel, presumably in the form of fission product silicates with low Si vapor pressure. The reaction is prevented by maintenance of an impermeable inner pyrocarbon coating which protects the SiC from oxygen liberated from the kernel. Since UO_2 is a potentially attractive kernel material, it is the subject of continuing characterization work under high burnup conditions in an attempt to achieve a level of predictability comparable to that of UC_2.

No interaction between kernels and coating has been observed in ThO_2 or $(Th,U)O_2$ particles irradiated at temperatures up to 1500°C and burnup of 3 to 30% FIMA. Irradiation of coated ThO_2 at 1150°C to 11% FIMA also produced no deleterious kernel coating interaction. Because of the extremely stable and refractory nature of ThO_2 during irradiation, it has been selected as the fertile kernel material for the large commercial HTGR in the United States.

CONCLUSIONS

Both oxide and carbide kernel materials have been developed for use in the first commercial HTGRs. Carbide kernels can be produced by several techniques but must be protected from reaction with moisture. Oxide kernels have been made by a gel-supported precipitation method, among others, and have potential advantages over carbides such as resistance to hydrolysis and relative ease of remotizing the fabrication process.

During irradiation, up to 75% of the total uranium and 7% of the thorium will be fissioned in the kernel at a maximum fast neutron exposure of 8×10^{21} n/cm^2 (E>0.18 MeV). A large fission product inventory and associated kernel swelling develop in both oxides and carbides, but a compliant buffer coating prevents interaction between the kernel and structural coatings. Carbide and oxide kernels migrate up an imposed thermal gradient, and the effect is accounted for in core thermal design. Thorium oxide is the most stable kernel material with little propensity for migration relative to other oxides and the carbides. Kernels of UC_2 show the least kernel migration in the carbide system.

The flexibility in kernel selection permits the HTGR to adapt to diverse technical and economic requirements. Mixed oxide fuels have been favored in European HTGR design. In the United States where a segregated thorium cycle is employed, fissile UC_2 and fertile ThO_2 kernels have been selected for HTGR fuel based on optimization of performance and predictability under operating conditions.

ACKNOWLEDGMENTS

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