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MIGRATION OF ThO_2 KERNELS UNDER THE INFLUENCE OF A TEMPERATURE GRADIENT

by

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ABSTRACT

BISO coated ThO_2 fertile fuel kernels will migrate up the thermal gradients imposed across coated particles during HTGR operation. Thorium dioxide kernel migration has been studied as a function of temperature (1300 to 1700°C) and ThO_2 kernel burnup (0.9 to 5.8% FIMA) in out-of-pile, postirradiation thermal gradient heating experiments. The studies were conducted to obtain descriptions of migration rates that will be used in core design studies to evaluate the impact of ThO_2 migration on fertile fuel performance in an operating HTGR and to define characteristics needed by any comprehensive model describing ThO_2 kernel migration. The kinetics data generated in these postirradiation studies are consistent with in-pile data collected by investigators at Oak Ridge National Laboratory, which supports use of the more precise postirradiation heating results in HTGR core design studies. Observations of intergranular carbon deposits on the cool side of migrating kernels support the assumption that the kinetics of kernel migration are controlled by solid state diffusion within irradiated ThO_2 kernels. The migration is characterized by a period of no migration (incubation period) followed by migration at the equilibrium rate for ThO_2. The incubation period decreases with increasing temperature and kernel burnup. The improved understanding of the kinetics of ThO_2 kernel migration provided by this work will contribute to an optimization of HTGR core design and an increased confidence in fuel performance predictions.

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INTRODUCTION

The basic heat generating component of large, high temperature, gas cooled reactor (LHTGR) fuel is the coated particle. Fissile fuel particles in the GA* design for a LHTGR will be TRISO coated UC_2 or WAR UC_xO_y** while the fertile fuel will be BISO coated ThO_2. There are, at present, extensive irradiation and out-of-pile fuel test programs in existence that are designed to evaluate fuel particle performance. The results of these test programs are used, in part, to develop and verify fuel particle performance models used to predict fuel behavior during irradiation testing or commercial reactor operation[1].

One performance limiting phenomenon identified early in the LHTGR fuel test program is kernel migration[2]. Kernel migration is the movement of a fuel kernel towards the hot side of a coated particle under the influence of a thermal gradient. Fuel coating failure can be caused by this phenomenon if a migrating kernel penetrates a structural coating layer to the point that the layer cannot support the stresses developed during irradiation. Although other failure mechanisms may operate, failures related to kernel migration could control fuel performance at elevated temperatures. Because of this, studies of the relationship between temperature, temperature gradient, irradiation exposure, and kernel migration are necessary for a complete description of HTGR fuel particle performance. Studies of UC_2 kernel migration have been reported elsewhere[3] and current results suggest that WAR UC_xO_y kernels do not migrate in a thermal gradient.

The purpose of this paper is to describe studies of ThO_2 kernel migration. The goal of the experimental program was to define the relationship between temperature, ThO_2 kernel burnup, and kernel migration rate. Until recently, ThO_2 kernel migration was only observed during irradiation. These in-pile data are invaluable but data analysis is complicated since (1) temperature varies continuously with time as reactor operating conditions change, and (2) ThO_2 kernel burnups change with time. The migration data reported below were obtained

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*GA = General Atomic Company, San Diego, California.

**UC_xO_y is used to define the stoichiometry of weak acid resin (WAR) derived UC_2-UO_2-C fissile fuel kernels.

from out-of-pile thermal gradient heating (OPH) tests conducted on irradiated BISO ThO_2. The out-of-pile tests have the advantage of (1) well known, constant test temperatures and temperature gradients, (2) constant kernel burnups during a test, which allows explicit studies of relationships between ThO_2 kernel migration and kernel burnup, and (3) the capability for periodic test interruption to determine the kernel migration behavior as a function of time.

MECHANISM FOR ThO_2 KERNEL MIGRATION

It was shown very early in experimental and theoretical studies of carbide kernel migration [UC_2, $(Th,U)C_2$, ThC_2] that solid state C diffusion through carbide kernels caused the kernel movement[2,3]. The C is taken into solution on the hot side of a kernel, diffuses through the kernel, and is rejected at the cool side kernel-buffer interface or precipitated within the kernel. As a result, the kernel advances toward the hot side of the particle.

The mechanism for oxide kernel migration is not as well defined as the carbide migration mechanism. Until recently, migration was only reported in irradiated oxide kernels [UO_2, $(Th,U)O_2$, ThO_2][4]. Since oxygen is released from irradiated oxide kernels, causing substantial CO overpressure in the coated particles, initial models assumed that oxide kernel migration was controlled by gas phase C transport resulting from the effects of a temperature gradient on the $CO\text{-}CO_2\text{-}C$ equilibrium[5]. Continued study showed, however, that, although a CO overpressure may be necessary for oxide kernel migration, the kinetics of migration are more consistent with the assumption that the migration rate is controlled by a solid state diffusion mechanism in the oxide kernels[4;6,7]. The kinetics of ThO_2 kernel migration are therefore best described by a kernel migration coefficient (KMC) analogous to the parameter used to define carbide kernel migration[2] where

$$KMC = \left(\frac{dx}{dt}\right)\left(T^2\right)\left(\frac{dT}{dx}\right)^{-1} = \beta e^{-\Delta H/RT} \qquad (1)$$

and $\frac{dx}{dt}$ = kernel migration rate (m/sec)

T = temperature (K)

$\frac{dT}{dx}$ = temperature gradient (K/m)

β = preexpontial constant ($m^2 \cdot K/sec$)

ΔH = heat of self-diffusion for the diffusing species (J/mol)

R = gas constant (8.313 J/mol•K)

EXPERIMENTAL

Materials

The BISO ThO_2 particles used in these experiments had been irradiated prior to the OPH tests. The kernels and coatings were prepared using standard fuel fabrication procedures and then irradiated in accelerated tests under conditions similar to those expected during LHTGR operation. A description of the coated particles used in the OPH tests is given in Table 1.

TABLE 1

General Description of Irradiated BISO ThO_2 Particles Used to Study ThO_2 Kernel Migration

Data Retrieval Number[a]	Irradiation Conditions				OPH Test Temperature (°C)
	Capsule	Fast Neutron Exposure[b] (10^{25} n/m^2)	Kernel Burnup[c] (% FIMA)	Time Average Temp[d] (°C)	
5466-149	FTE-14	1.2	0.9	1250	1323-1661
4252-02-015-5	HT-12	2.8	1.5	900	1293-1705
6542-17-013-3	HT-17	2.8	1.7	900	1518-1546
4413-75T	P13L	5.7	4.7	900	1548
4252-02-015-6	HT-13	6.1	5.8	900	1311-1666

[a] Nominal kernel and coating dimensions: kernel diameter = 500 μm, buffer thickness = 85 μm, outer PyC thickness = 75 μm.

[b] Equivalent fast fluence in a HTGR for neutrons having energy E > 29 fJ.

[c] % FIMA = percent fissions of initial metal atoms.

[d] Design.

Procedure

Kernel migration data were collected by heating particles under a thermal gradient and periodically examining the samples to determine the migration distance as a function of time. The thermal gradient was obtained by loading particles into a graphite crucible and then positioning the crucible between a graphite heating element and a cold furnace wall. The basic techniques used were essentially the same as those described for type 4 crucibles in Ref. 3. Slight modifications were made, however, in the crucible geometry and loading procedures to increase the confidence in the temperature and temperature gradients across the coated particles. Sample test temperatures were monitored using techniques described in Ref. 3. The range of test temperatures is shown in Table 1.

Contact X-radiographs of each crucible were prepared before heating and at intervals when tests were interrupted. Kernel migration distances were measured from the radiographs as a function of time. When possible, migration is defined by the change in distance between the hot side kernel-buffer interface and a reference point on the hot side of a coating. In these experiments, no reference point (i.e., the outer surface of the outer PyC layer) was consistently visible in the radiographs. Kernel migration was therefore defined as the change in distance from the original cold side buffer-kernel interface to the hot side kernel-buffer interface. The radiographs were also used to observe changes in the morphology of the advancing kernel surface.

Fission product inventories were monitored during testing. Metallic fission product release was detected by gamma counting loaded crucibles during test interruptions. Gaseous release was detected by periodically purging the furnace atmosphere and analyzing for Kr-85 by gamma counting. Particle failure was occasionally detected very early in these tests. The failure was attributed to interactions between particle coatings and the binder used to hold particles in the crucibles. Kernel migration data were not used from any crucibles after coating failure was detected.

Metallography and microprobe examinations were also done on selected samples to examine kernel microstructures and evaluate any detectable redistribution of kernel or coating constituents.

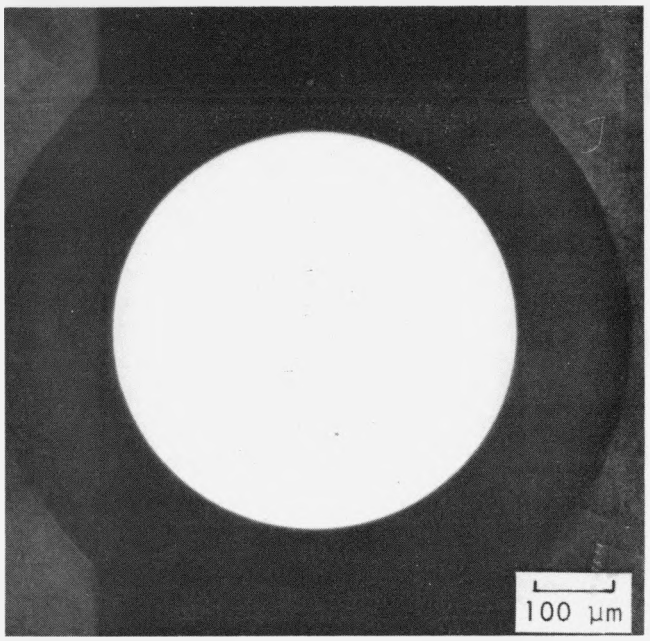
RESULTS AND DISCUSSION

Radiographic, Metallographic, and Electron Microprobe Observations

Periodic radiographic examinations of migrating kernels showed two forms for the advancing hot side kernel-buffer interface. Examples of each are shown in Figure 1. Type 1 migration is illustrated in Figures 1(a) and 1(c), which show pre- and post-OPH radiographs of a particle irradiated to 1.5% FIMA and then heated for 605 hours at 1559°C under a thermal gradient of 705°C/cm. In this example, the kernel elongated while migrating rather than maintaining its original spherical form. The interface between the migrating ThO_2 and the coatings on the hot side of the kernel remained sharp. Type 1 migration is typical of results obtained from tests conducted at 1300°C to 1705°C on samples having kernel burnups in the range 1.5 and 5.8% FIMA. Although burnup had no apparent effect on the type of migration, burnup did affect the shape of migrating kernels. As burnup increased, the fraction of kernel surface that elongated increased until at 5.8% FIMA, the distinct protrusion at the hot side kernel-coating interface was not visible and the migrating kernel appeared "egg" shaped.

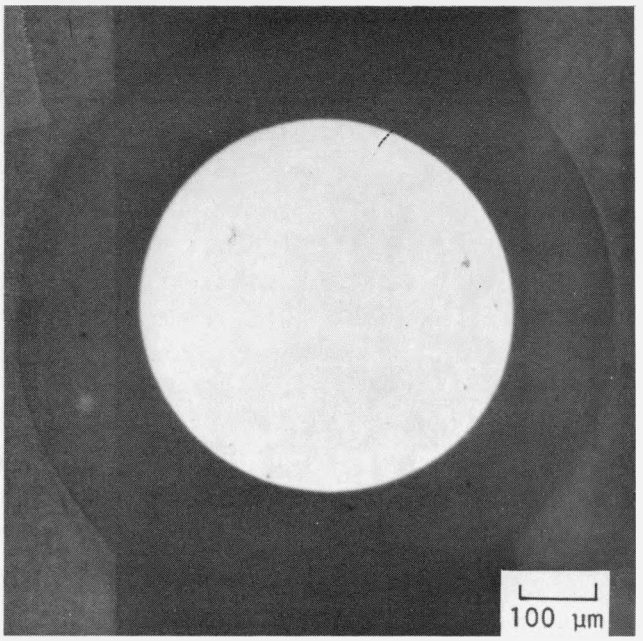
The second type of kernel migration is illustrated on Figures 1(b) and 1(d). Type 2 migration was characterized by a diffuse kernel-coating interface on the hot side of a particle [right side of Figure 1(d)]. The difference in the hot side kernel-coating interface for type 1 and type 2 migration is clearly visible in Figures 1(c) and 1(d). Type 2 migration began with the formation of a faint "halo" of heavy metal near the original hot side kernel-coating interface. The original hot side kernel-coating interface remained visible in radiographs during the initial stages of migration. As the migrating "halo" of heavy metal advanced into the coatings on the hot side and became more dense, the original hot side kernel-coating interface disappeared as shown on Figure 1(d).

A second characteristic of type 2 migration was the development of a low metal density region in the kernel near the original cool side kernel-coating interface [left side of Figure 1(d)] and some metal dispersion in the buffer coating at the cool side of the particle. The dispersed metal in the buffer layer on the cool side of the particle appeared late in the tests and will be identified during planned metallographic and electron microprobe studies. The



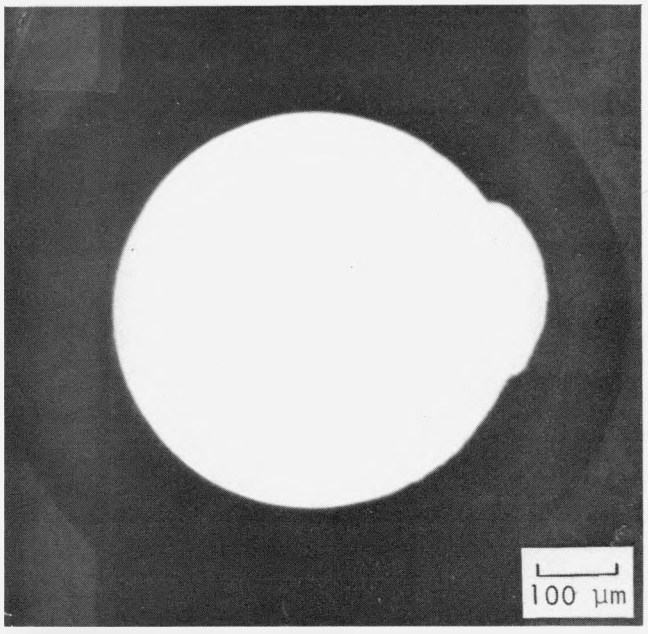
LA467

(a)



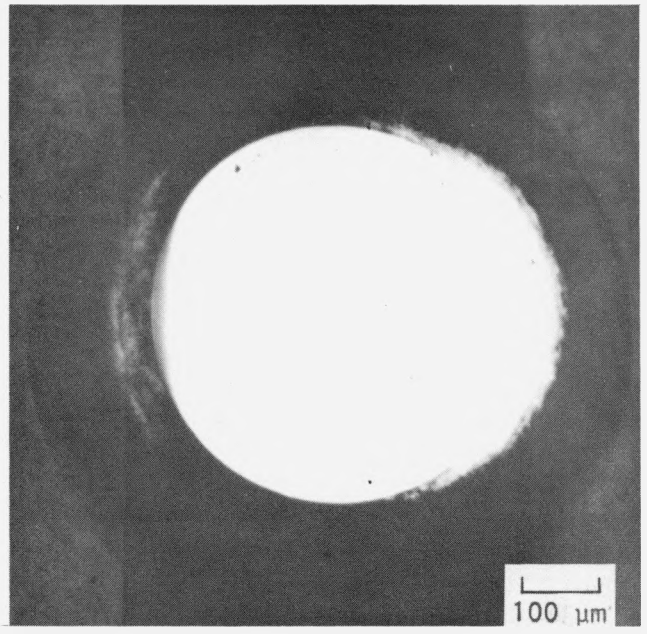
LB106

(b)



LA906 Type 1

(c)



TA85 Type 2

(d)

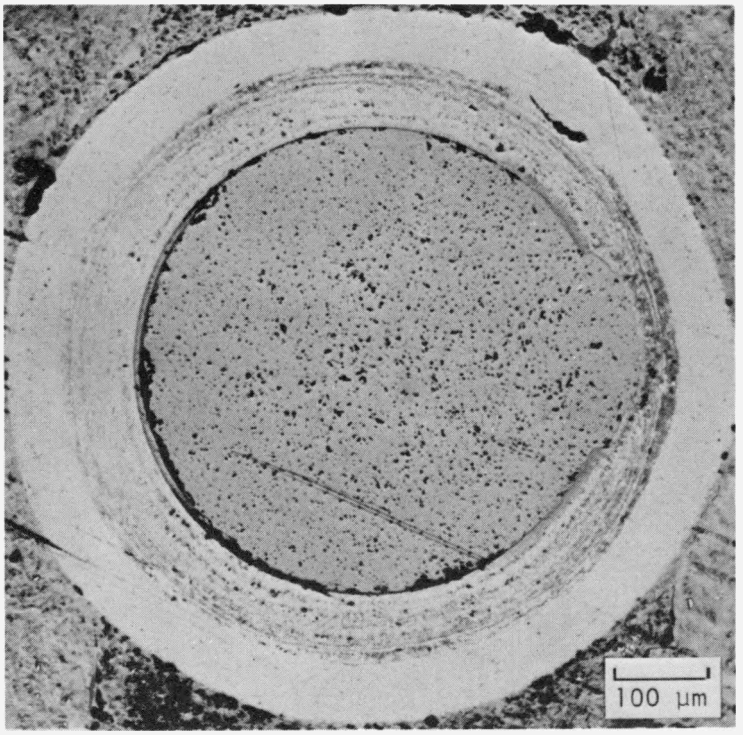
Figure 1. Pre- (a,b) and Post- (c,d) Thermal Gradient Heating Contact X-radiographs of Irradiated BISO ThO_2 Particles Showing Two Types of Kernel Migration

Batch Number	Burnup (% FIMA)	Heating Conditions	Figure
4252-02-015-5	1.5	605 hrs, 1559°C, 705°C/cm	(c)
5466-149	0.9	1933 hrs, 1450°C, 359°C/cm	(d)

low metal density region within the coolest portion of the ThO_2 kernel is typical of carbide kernel migration. In the case of carbide kernels, migration occurs as C diffuses from the hot side to the cool side of a kernel and is rejected at the cool side kernel-coating interface. This rejected C effectively pushes the kernel toward the hot side of the coatings and the cool side kernel-rejected C interface retains its original spherical form. The shape of the interface between the high and low density regions within the cool side of the kernel in Figure 1(d) suggests that type 2 ThO_2 migration is not the same as carbide migration. The fact that the interface between the high and low density regions within the cool side of the kernel in Figure 1(d) did not retain its original spherical shape suggests that Th is being removed from the cool side and presumably redeposited on the hot side of the kernel. From the appearance of the radiographic images, it is postulated that the Th may be moving to the hot side by surface diffusion over the kernel at the kernel-coating interface.

Type 2 migration was observed on samples irradiated to 0.9% FIMA before heating in the temperature range 1323 to 1661°C. It was also observed in a sample irradiated to 1.7% FIMA that was heated from 1200 to 2000°C over a period of 5 days in the absence of a thermal gradient before thermal gradient heating at 1546°C. However, a companion sample heated as irradiated at 1518°C under a thermal gradient showed type 1 migration. There is, at present, no explanation for the difference in observed migration behavior. It will be shown later that the particle to particle differences in appearance at the hot side kernel-coating interface has no impact on KMC values.

Metallographic examinations were done on ThO_2 samples heated at about 1550°C after irradiation to 1.5, 4.7, or 5.8% FIMA. Figure 2 is a metallographic cross section of the particle in Figure 1(c) showing type 1 migration. The apparent density of C in the buffer layer is higher on the cool side than on the hot side. This suggests that the kernel elongation could be related to extrusion of the kernel into a void on the hot side produced by gas phase transport of C from the buffer on the hot side to the cool side. There are, however, no indications of any flow patterns in the hot portion of the kernel which would be expected if kernel extrusion had occurred. This suggests that Th and O diffusion up the temperature gradient caused the elongation. It is not yet clear whether Th or O diffusion is the rate controlling step.



HOT
SIDE

R69171

Figure 2. Photomicrograph of a Particle Heated for 605 hr at 1559°C and 705°C/cm After Irradiation to 2.5% FIMA and 2.8 x 10^{21} n/cm^2 ($E \geq 29$ fJ)$_{HTGR}$

Close inspection of the cool side buffer-kernel interface on Figure 2 shows the presence of a dense phase. This phase is optically active under polarized light and has all of the characteristics of the rejected graphite layer observed behind migrating carbide kernels[3]. This is consistent with the hypothesis that C diffusion through the ThO_2 kernel is also related to the observed kernel migration.

There is no indication of grain structure in the kernel shown in Figure 2. Results of postirradiation thermal gradient heating metallography and electron microprobe studies from a sample with visible grain boundaries in the kernel after irradiation are shown in Figure 3. This sample was irradiated to 4.7% FIMA before thermal gradient heating. Although postirradiation metallography showed grain boundaries within the kernels, no extensive intergranular precipitation was noted. Post OPH metallography showed a precipitate in the kernel grain boundaries that was more pronounced on the cool side than the hot side of the kernel [Figure 3(a)]. Electron microprobe analysis showed conclusively that the precipitate in the grain boundaries was C. No Th, U, or metallic fission products were detectable in the precipitate [Figure 3(b), (c), (d)].

The kernel microstructures and surface morphologies shown in Figures 1, 2 and 3 are similar to microstructures reported for in-pile ThO_2 kernel migration[8]. This is taken as evidence that the mechanism for in-pile and out-of-pile ThO_2 kernel migration is the same.

Kernel Migration Data

Kernel migration data were collected at temperatures ranging from approximately 1300 to 1700°C on samples irradiated to kernel burnups of 0.9 to 5.8% FIMA. A typical example of kernel migration distance vs time is shown in Figure 4 for individual particles irradiated to 0.9, 1.5, or 5.8% FIMA before heating at approximately 1320°C under a gradient of about 370°C/cm. Particles irradiated to 5.8% FIMA began migrating immediately; particles irradiated to 1.5% FIMA began migrating after about 2500 hours at temperature; and particles irradiated to 0.9% FIMA had not yet begun to migrate when the test was terminated after 6568 hours. As shown on Figure 4, kernel migration distances increased linearly with time once migration began. The incubation period before the onset of kernel migration was estimated for each particle by extrapolating a least squares fit of migration

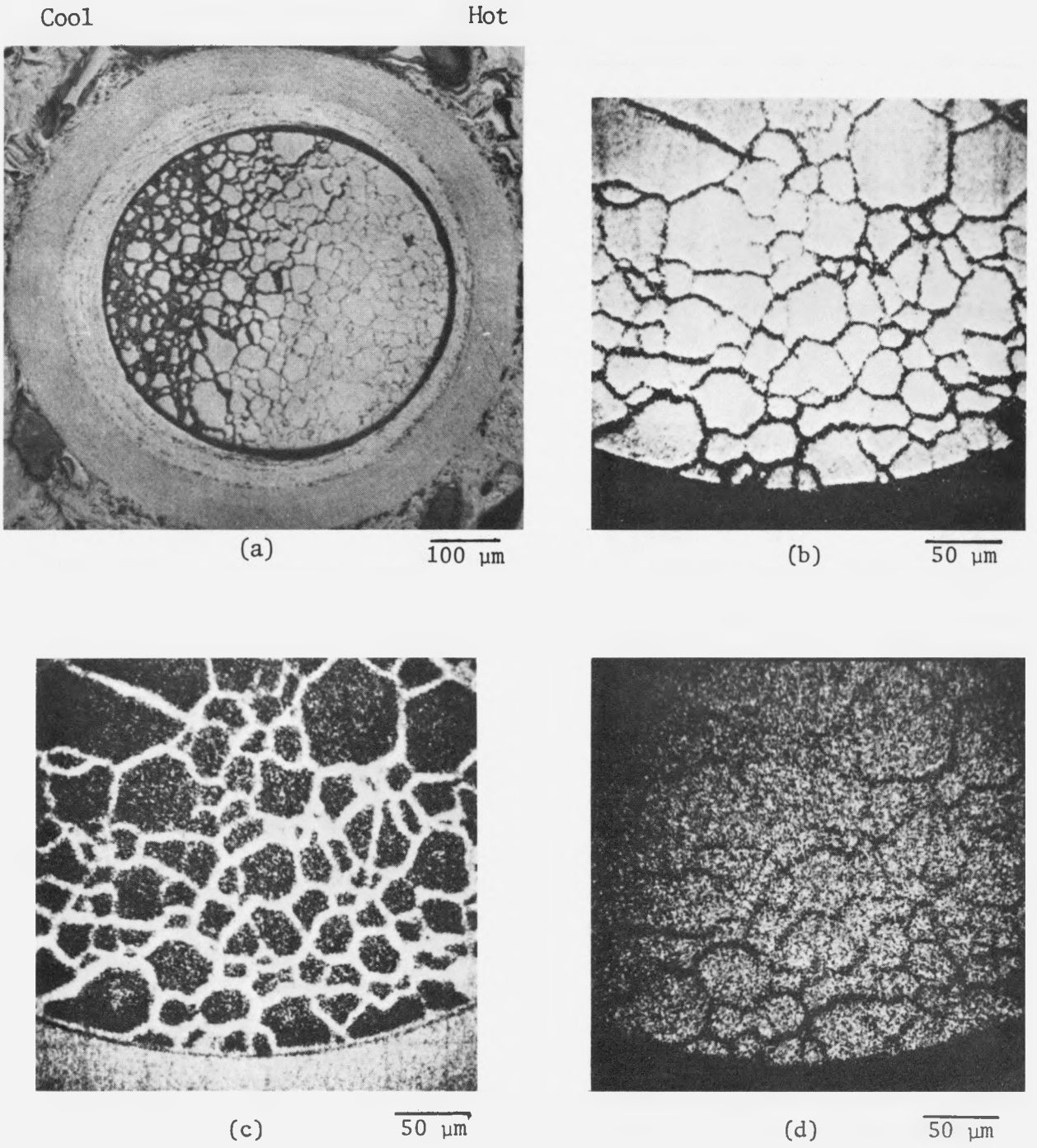


Figure 3. Post Thermal Gradient Heating Metallography (a) and Electron Microprobe Results (b-d) of a Sample from ThO_2 batch 4413-75T. The sample was irradiated to 4.7% FIMA before heating. The microprobe results show (b) back scattered electron and (c) characteristic C and (d) Th X-ray traces across the cool side of the kernel.

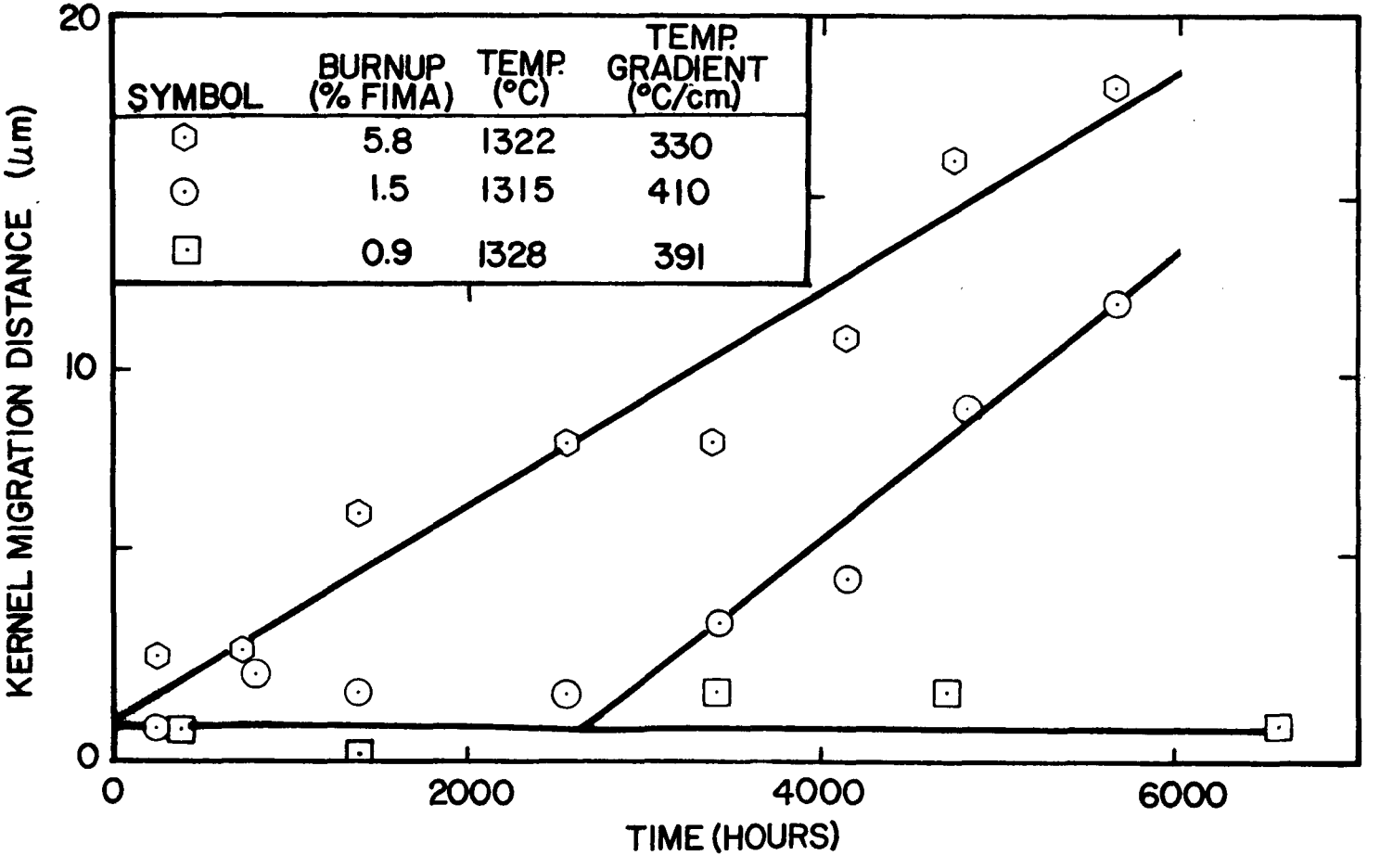


Figure 4. Variation in Kernel Migration Distance With Time During Postirradiation Thermal Gradient Heating at 1320°C of BISO ThO_2 Samples Irradiated to 0.9, 1.5, or 5.8% FIMA

distance versus time to zero migration. Migration distances were assumed to vary linearly with time and data were only used to estimate the length of the incubation period if kernel migration had clearly begun. The fits to the migration data on Figure 4 illustrate the method used to define the incubation time.

The concept of an incubation period is consistent with theoretical treatments of the kinetics of thermal gradient induced transport phenomena. The incubation period may be related to the time necessary to develop equilibrium concentration profiles in a temperature gradient which is given by[9]

$$\theta = \frac{na^2}{\pi^2 D} = \alpha e^{Q/RT} \qquad (2)$$

where θ = incubation time
n = constant
a = diffusion distance
D = self-diffusion coefficient for the diffusing species
α = preexponential constant (sec)
Q = heat of self-diffusion for the diffusing species (J/mol)
R = gas constant (8.313 J/mol•K)

The exponential dependence of θ on inverse temperature is also consistent with the form expected for most thermally activated processes.

The variation in incubation time with inverse temperature is shown on Figure 5 for kernels having burnups of 0.9 and 1.5% FIMA. A minimum incubation time of 2.36×10^7 sec at 1323°C was assumed in Figure 5 for 0.9% FIMA samples since no kernel migration had been observed when testing was terminated. The 0.9 and 1.5% FIMA data were intially fit separately to an expression having the form of Eq. (2). A students t-test was then used to show that values predicted for θ using the two sets of data independently were the same at the 0.05 significance level over the range of test conditions. The data were therefore combined for the single analysis shown in Figure 5.

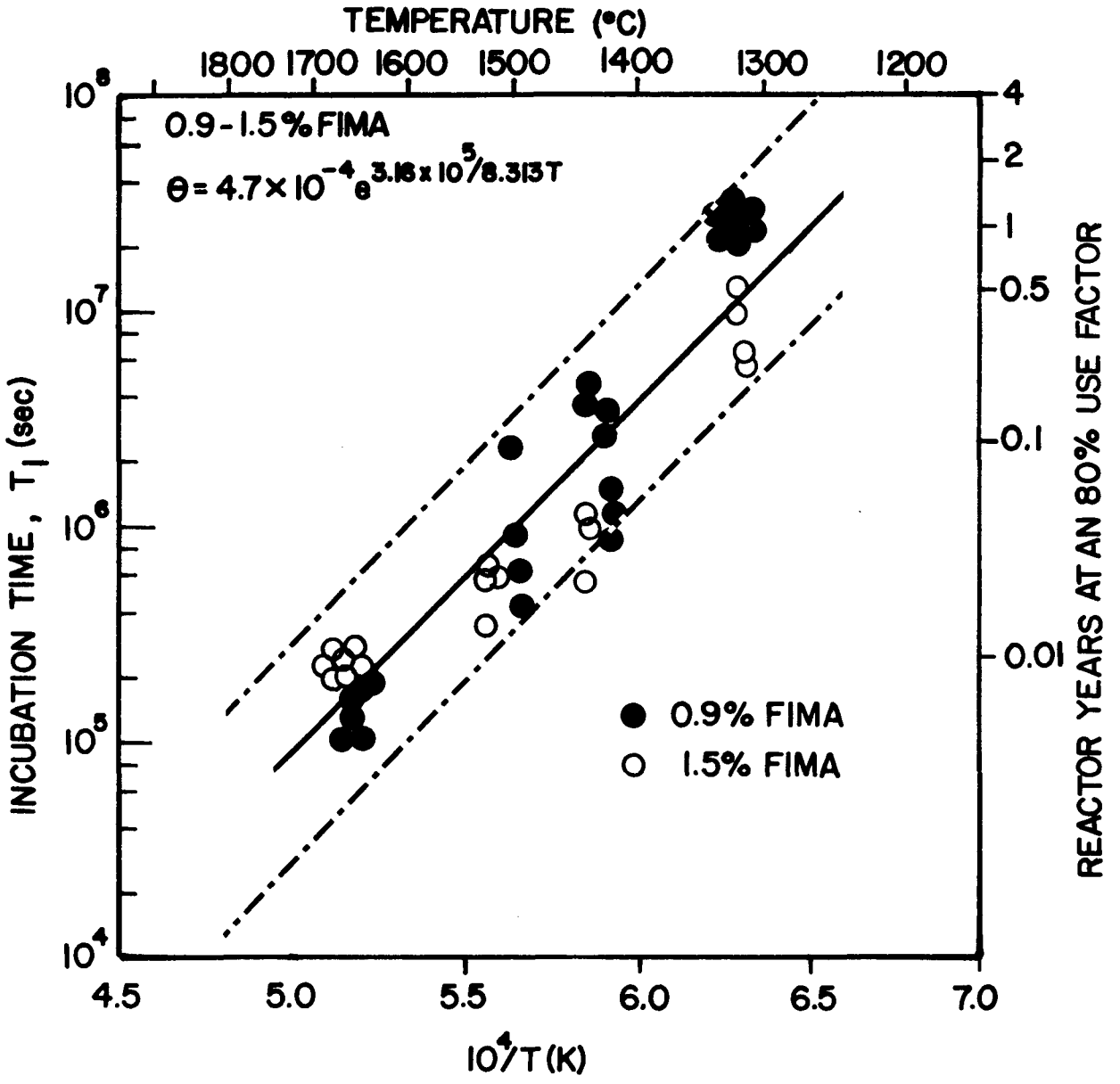


Figure 5. Variation in Incubation Time with Inverse Temperature for ThO_2 Kernels Having Burnups of 0.9 and 1.5% FIMA

Incubation periods were estimated for test samples having burnups as high as 5.8% FIMA. The data from the higher burnup samples showed qualitatively that the incubation time decreased with increasing kernel burnup; however, large uncertainties in the experimental values made it impossible to quantitatively define the relationship between θ, burnup, and temperature.

Investigators at ORNL[4] reported migration of unirradiated ThO_2 kernels during OPH tests conducted at 1810 to 1945°C. This is the only known evidence of unirradiated ThO_2 kernel migration. Kernel migration measurements made at about 150 hr intervals for times up to approximately 750 hrs gave no indication of an incubation period. Assuming an average temperature of 1878°C, Figure 5 suggests an incubation time of 6.2 hrs (2.2×10^4 sec). Because of the short predicted incubation time and the long time between kernel migration observations, the lack of a detectable incubation period in the ORNL tests is consistent with the results described here.

It will be shown later that ThO_2 KMC values are independent of burnup once migration begins. Given this, one would overestimate the migration of ThO_2 kernels if the temperature and burnup dependent incubation period were not accounted for (see Figure 4). Continued testing to define the relationship between ThO_2 kernel burnup, temperature, and incubation time is therefore necessary for accurate predictions of ThO_2 kernel migration during fuel service in an operating reactor.

Variation in ThO_2 KMC With Temperature

Data showing that ThO_2 migration is related to diffusion within the kernels and that migration distances increased linearly with time (Figure 4) are consistent with assumptions made in the derivation of Eq. (1). Eq. (1) was therefore used to calculate ThO_2 KMC values from observed kernel migration distances, test temperatures, and temperature gradients. The migration time in Eq. (1) was estimated by subtracting the incubation time from the total test time for each individual particle.

Separate estimates of KMC versus inverse temperature were made for kernels showing type 1 and type 2 migration (Figure 1) and for kernels having burnups in the range 0.9-1.7% FIMA or 4.7-5.8% FIMA. The students t-test was used to show that predicted values for ThO_2 KMC were independent of migration type and kernel burnup at the 0.05 significance level. The data were therefore combined for a general description of ThO_2 KMC that is independent of kernel burnup or migration type. The data and the least squares estimates for KMC are shown on Figure 6. The high and low burnup data are identified on Figure 6 to illustrate the lack of KMC dependence on kernel burnup.

The apparent activation energies for KMC [Eq. (1)] and the incubation time [Eq. (2)] are both defined as heats of self-diffusion. Comparison of the experimentally determined activation energies for θ and KMC should indicate if the incubation period and the migration phenomena have the same rate controlling steps. The apparent activation energies for the incubation period and KMC are, respectively, 3.16×10^5 J/mol and 2.96×10^5 J/mol. It was shown, using the students t-test, that these values are the same at a 0.05 significance level. This implies that the rate controlling steps for the incubation period and kernel migration are the same. The data do not point to a specific rate controlling step; however, they strongly suggest that the rate controlling step is thermal gradient induced diffusion of C, Th, or O within the ThO_2 kernel.

The data presented in this report will be used to predict ThO_2 kernel migration during irradiation. Final confirmation of the data will therefore include comparisons of out-of-pile and in-pile data. A comparison of available in-pile ThO_2 KMC data[4] and the range for ThO_2 KMC determined by OPH is shown on Figure 7. Agreement between the in-pile and out-of-pile data is good at temperatures less than 1500°C but at temperatures greater than 1500°C the in-pile data show lower KMC values than the out-of-pile data. The in-pile data on Figure 7 were not, however, corrected for the incubation period. The impact of an incubation period on calculated KMC values was examined for each in-pile test identified on Figure 7. The OPH data showed a near zero incubation time for ThO_2 with a burnup of 5.8% FIMA. The maximum incubation time for the ThO_2 in the HRB3-3A and HRB4-3A fuel rods would therefore equal the time to reach 5.8% FIMA. Since the burnups were, respectively, 13.5 and 14.0% FIMA, the

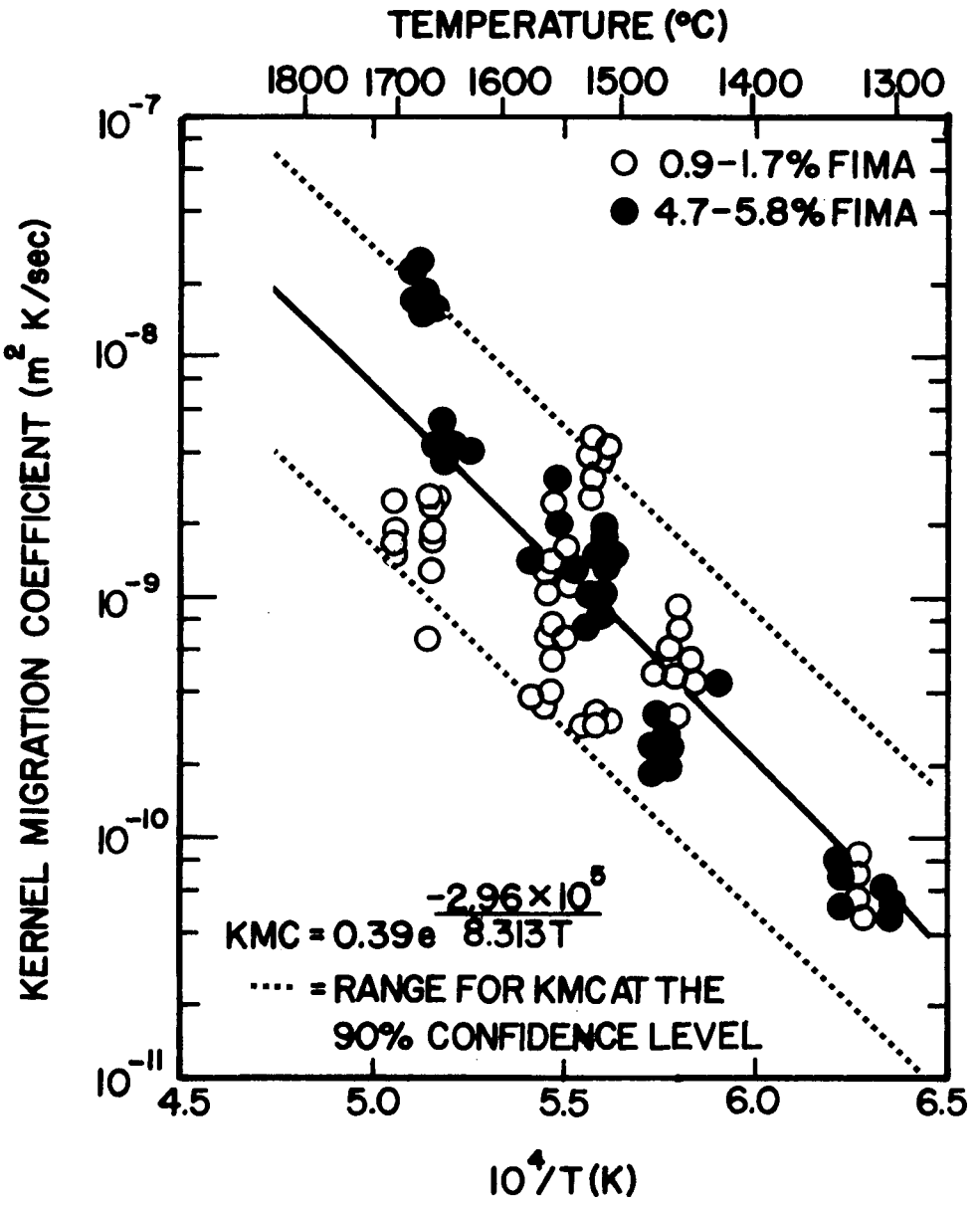


Figure 6. Variation in ℓn KMC with 1/T for ThO_2 Kernels Having Burnups in the Range 0.9 to 5.8% FIMA

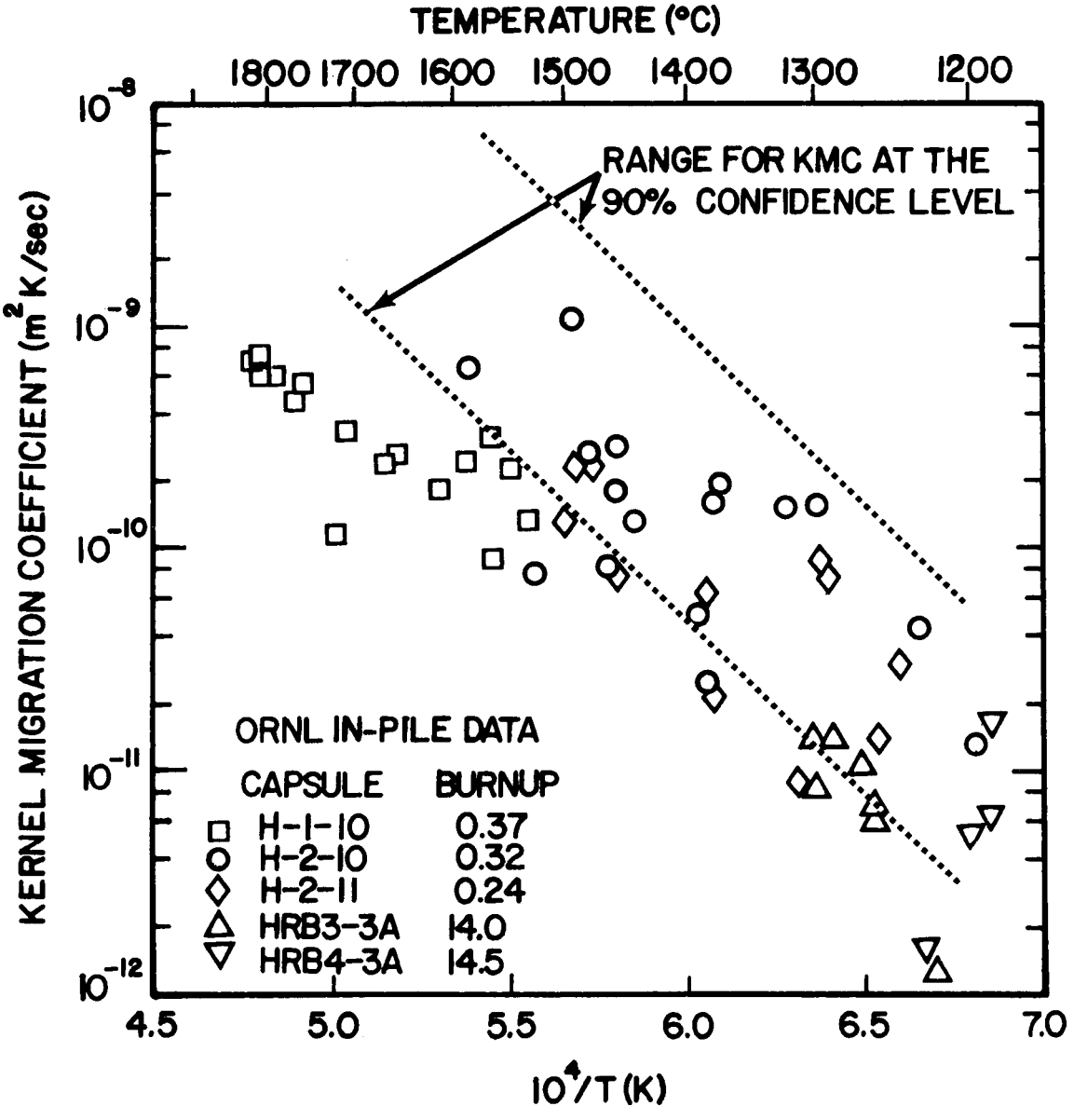


Figure 7. Comparison of In-Pile and Out-of-Pile ThO_2 KMC Data

minimum time during which migration occurred would be about 60% of the total irradiation time. The maximum KMC values would therefore be about 1.7 times the values shown on Figure 7. Agreement between the in-pile and out-of-pile data from these samples is therefore very good after considering the incubation period.

Treatment of the impact of the incubation period on KMC values from the H capsule tests is more difficult since no out-of-pile data are available for burnups as low as those of the H capsule ThO_2 samples. If one assumes the range of incubation times shown on Figure 5, the range of possible KMC values for observations made on H-2-10 are consistent with the out-of-pile data. The incubation period, however, is too short to affect KMC values from the H-1-10 sample and suggests no migration for most of the H-2-11 conditions. Uncertainties in capsule operating temperatures are the most probable source for the disagreement in the in-pile and out-of-pile results. Reported uncertainties in operating temperatures for the H capsules range from 200 to 300°C[8]. If one assumes that H-1-10 operating temperatures are 50 to 100°C less than reported and H-2-11 temperatures are 50 to 100°C higher than reported, the in-pile data are consistent with out-of-pile incubation times and KMC values. The in-pile and out-of-pile data can therefore be shown to agree when reported uncertainties are considered.

CONCLUSIONS

Migration of ThO_2 fuel kernels up a temperature gradient occurs during irradiation and during postirradiation, out-of-pile thermal gradient heating. Good agreement of kinetics data and microstructures of ThO_2 kernels after in-pile and out-of-pile kernel migration shows that the out-of-pile test data can be used to accurately describe ThO_2 kernel migration.

Radiographic and metallographic studies of samples tested out-of-pile showed two types of ThO_2 kernel migration that were characterized by kernel shape and the hot side kernel-coating interface. The kinetics of migration were not, however, related to the type of migration. It was also shown that solid state diffusion of C down the temperature gradient and Th and O up the temperature gradient takes place during ThO_2 kernel migration.

Migration of ThO_2 kernels occurs in two distinct stages. No migration is observed during the first stage. The time period during which no migration is observed (incubation time) decreases with increasing temperature and kernel burnup. An expression for the incubation time was supplied in the form

$$\theta = \alpha e^{Q/RT}$$

for ThO_2 kernels having burnups of 0.9 to 1.5% FIMA. Additional testing is required to characterize the relationship between burnup and incubation time in the range 0 to 1% FIMA and 2 to 7.5% FIMA. A complete knowledge of the variation in incubation time with temperature and kernel burnup is needed for accurate predictions of in-pile ThO_2 kernel migration.

Thorium dioxide kernels migrate at constant rates once migration begins. Values for ThO_2 KMC were found to be independent of kernel burnup. Migration distances also increase linearly with time for otherwise constant test conditions. The kinetics of ThO_2 kernel migration are described by

$$KMC[m^2K/sec] = 0.39e^{\frac{-2.96 \times 10^5}{RT}}$$

for R = 8.313 J/mol·K.

The apparent activation energies for the incubation time and for kernel migration were shown to be the same at the 0.05 significance level. This suggests that the rate controlling step for both phenomena is the same. The rate controlling step was not positively identified although the data indicate it to be thermal gradient induced diffusion of C down or Th or O up the temperature gradient across the ThO_2 kernels.

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