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Kernel Migration for HTGR Fuels from the Th-U-C-O-N System

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T. B. Lindemer and R. L. Pearson

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Table of Contents

	<u>Page</u>
Abstract	1
I. Introduction	1
II. General Experimental Methods	2
III. Determination of Rate-Controlling Mechanisms	5
(1) Dicarbide Fuels	6
(2) Dioxide Fuels	9
IV. Data and Analysis	17
(1) UO_2 + Fission-Product Oxides	18
(2) $\text{UO}_2\text{-U}_2\text{N}_3$ Solid Solution	18
(3) ThO_2	20
(4) $\text{Th}_{1-z}\text{U}_z\text{O}_2$ + Fission-Product Oxides	24
(5) UC_2	24
(6) $\text{UC}_{1-z}\text{N}_z$	28
(7) ThC_2	28
(8) $\text{Th}_{0.84}\text{U}_{0.16}\text{C}_2$	31
V. Discussion	31
(1) Carbide Results	31
(2) Oxide Results	33
(3) Application of Data to HTGR Fuel Design	35
VI. Conclusions	36
Acknowledgments	37
References	38
Appendix A. Coated Particle Characteristics	43
Appendix B. Original In-Reactor Data Used for KMC Calculations	47

KERNEL MIGRATION FOR HTGR FUELS FROM THE Th-U-C-O-N SYSTEM*

T. B. Lindemer and R. L. Pearson

Abstract

Nuclear fuels for the high-temperature gas-cooled reactor (HTGR) consist of spherical kernels of actinide compounds contained within gastight pyrolytic carbon and SiC. This fuel is subjected to a significant in-reactor temperature gradient that leads to migration of the fuel kernel up the temperature gradient and into the coating layers. This phenomenon has been studied both in the laboratory and in-reactor for fuel kernels from the Th-U-C-O-N system. Application of theory to the analysis of in-reactor migration data indicates that a solid-state diffusion process operating across the kernel controls the migration rate in fissioned $\text{Th}_{0.84}\text{U}_{0.16}\text{C}_2$, UO_2 , ThO_2 , and $\text{Th}_{0.8}\text{U}_{0.2}\text{O}_2$ particles. The theoretically-based kernel migration coefficient (KMC), $(\text{cm/sec})^\circ\text{K}^2 (\text{K/cm})^{-1}$, is thus used to correlate the data. The in-reactor KMC values were apparently not dependent on the extent of fission, the fission of either ^{233}U or ^{235}U , or the presence of a SiC coating layer. Laboratory KMC values were obtained from unirradiated dicarbide particles; these values were in excellent agreement with previously published laboratory KMC data and did not appear to be inconsistent with in-reactor dicarbide KMC values. Laboratory KMC values for ThO_2 agreed with in-reactor values. The irradiation performance of nitrogen-containing oxide or carbide kernels was no better than that of analogous nitrogen-free compounds.

I. Introduction

Fuel elements for high-temperature gas-cooled reactors (HTGRs) are made from rigid assemblies (fuel rods or compacts) of coated fuel particles distributed appropriately in graphite holders that contain coolant passages and constitute both moderator and core structure.¹⁻³ The coated fuel particles consist of small spherical oxide or carbide kernels that

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are each coated with successive layers of pyrolytic carbon ("BISO" particles) and sometimes an intermediate layer of SiC ("TRISO" particles). The pyrolytic carbon layers absorb damaging fission fragments and retain gaseous fission products and their precursors, while the SiC layer improves the retention of metallic fission products and fuels.^{4,5} These fuels are currently designed to operate from 700°C to a nominal maximum of 1550°C, the latter temperature being sustained for times that are much smaller than the core lifetime.

When a sufficient temperature gradient is imposed across the particle for an adequate time at an appropriately high temperature, one observes a migration of the fuel kernel up the temperature gradient and into the coatings, the so-called "amoeba effect." This phenomenon is one of several that can limit fuel performance and is observed with carbide and oxide kernels of thorium and uranium, as well as with solid solutions of either the carbides or the oxides. The migration rates are sufficiently large to exclude the use of some kernel compounds as U.S. HTGR fuels; therefore, measurement of the migration rates is quite important. Typical microstructures demonstrating the results of this phenomenon have appeared in the literature.⁶⁻⁹ This paper will review and apply the previously published theories and analyses that were developed to explain and to properly correlate the experimental data and will then present the extensive data obtained in the research and irradiation programs at Oak Ridge National Laboratory.

II. General Experimental Methods

In-reactor kernel migration data were obtained from several sources. Kernel migration, y , was obtained from measurements on 40X to 150X photographs of metallographic cross-sections of irradiated particles contained in fuel rods. The location, x , of each particle along the radius of the fuel rod was also measured so that the particle temperature and temperature gradient could be calculated from the temperature equation for the rod. The temperature equations are based on the assumption of uniform internal heat generation in the fuel rods. In the case of cylindrical fuel rods, the equation is:

$$T = T_{\max} - bx^2; \quad (1)$$

for annular cylindrical fuel rods with no heat flux across the inner surface, the equation is:

$$T = T_{\max} - b(x^2 - r_{\text{in}}^2 [1 + 2 \ln (x/r_{\text{in}})]), \quad (2)$$

where $T_{\max}$ is the maximum temperature in °K of the fuel rod, b is a proportionality constant having the units °K cm⁻², and r_{in} is the inner radius in cm of the annulus.¹⁰ The term b for cylindrical rods is more fundamentally equal to the linear heat generation rate of the fuel rod in watts per cm of length divided by the product $(4\pi)(\text{thermal conductivity in watts cm}^{-1} \text{ °C}^{-1}) (\text{square of the rod radius in cm}^2)$. Temperature gradients at x values are obtained from the absolute value of the derivative with respect to x of either Eq. (1) or (2).

Laboratory kernel migration studies were performed by employing another technique on unirradiated particles. Coated particles were bonded into a 1.27-cm-diam. by 0.13-cm-thick graphite disc. The graphite disc was made by molding particles into a mixture of 67 wt % graphite flour and 33 wt % of a fluid composed of 10 wt % maleic anhydride in furfural alcohol. The particles were distributed over the midplane of the disc perpendicular to the cylindrical axis. Each disc contained a central row of ten BISO-coated, 475-μm-diam., fully-dense UC₂ kernels. These kernels, which were oriented parallel to the temperature gradient, served as an internal temperature and temperature gradient standard because the KMC values for UC₂ were already known from previous work at the General Atomic Company (GAC).⁷ Approximately 40 particles containing other kernels were randomly distributed throughout the rest of the disc. In order to detect the particle-graphite matrix interface in a radiograph, all the particles were dipped into a concentrated solution of zirconyl chloride and then dried before being placed in the disc. Each disc was subsequently cured in air at 75°C for 16 h, buried in graphite flour, and heated in vacuum at 1600°C for 4 h. One to three discs were mounted across the 0.95-cm-wide gap in the graphite holder (Fig. 1) with a graphite flour--furfural alcohol--maleic anhydride paste. The assembly was cured at 75°C in air, and the

ORNL DWG. 75-14996 RI

Fig. 1. Apparatus for producing a temperature gradient in sample discs containing coated particles.

paste was repeatedly added until the gap between the disc and holder was filled with a solid bond of graphite. The holders, mounted as shown in Fig. 1, were heated in a graphite-resistance furnace in which a known temperature and temperature gradient were maintained. Temperatures were read with a disappearing-filament optical pyrometer calibrated against a NBS strip lamp system. Calibrated temperature corrections were also made for the furnace sight window. Other calibrations led to the conclusion that no temperature corrections were necessary for sighting down the 0.185-cm-diam. sight holes (Fig. 1) or for the graphite emissivity being other than unity at temperature. Several sight holes were drilled to different depths into a disc containing no particles, and measurements showed that the temperature from one side of the disc to the other decreased linearly with distance. It was assumed that the temperature gradient across the kernels was the same as in the disc.⁷ The range of the gradients could be altered from 200 to 575 °C/cm, depending on the maximum temperature of the run and whether insulation had been placed next to the cold wall inside the furnace. Kernel movement was determined by radiographing the mounted discs before and after heating and then measuring the migration of the hot side of the kernel from the radiographs. Migration data were used only when $y \geq 5 \mu\text{m}$ in a minimum of $\sim 33\%$ of the particles that lay above a given temperature; this criterion prevented the generation of spurious y data.

III. Determination of Rate-Controlling Mechanisms

One of three classes of mechanisms - solid-state diffusion, gas-phase diffusion, or a reaction - should control the mass transport rate, and hence the kernel migration rate, in HTGR particles at a given temperature. It is important to determine which of these mechanisms is controlling because each one has a specific theoretically-based parameter to be used for data correlation. In this section, experimental data will be used to identify the controlling mechanism in dicarbide and dioxide fuels.

(1) Dicarbide Fuels

The migration theory for dicarbides in the Th-U system was historically the first to be developed and was published by Gulden,¹¹ whose analysis was based on the principles of irreversible thermodynamics.¹² Gulden¹¹ concluded that the dicarbide migration rate was controlled by the solid-state diffusion of carbon down the temperature gradient and through the dicarbide kernel. The experimental data could be correlated with the aid of the theoretically-based KMC,

$$\text{KMC} = T^2 \frac{dy}{dt} \left(\frac{dT}{dx} \right)^{-1} = \beta \exp (-\Delta H/RT) \quad (3)$$

in which T is temperature in $^{\circ}\text{K}$; dy/dt is the observed kernel migration rate in cm/sec obtained by dividing the observed migration, y , by the time, t , at temperature; dT/dx is the temperature gradient in $^{\circ}\text{K}/\text{cm}$ across the particle; and R is $1.98 \text{ cal mol}^{-1} \text{ }^{\circ}\text{K}^{-1}$. It should be noted that the T^2 term is primarily a scale factor on the KMC; the Arrhenius-type temperature effect is equivalent to that for $\Delta H \approx 5,000 \text{ cal/mol}$. A plot of the KMC vs T^{-1} permits an empirical determination of β , which is a product containing thermodynamic, geometric, and diffusion terms, and ΔH , the apparent activation energy in cal/mol of the rate-controlling mechanism. In this case, ΔH is the enthalpy of carbon self-diffusion in UC_2 .^{7,11}

In many irradiations the maximum temperature varied by hundreds of degrees during the course of a run; therefore, it was necessary to calculate an average, $\bar{T}_{\text{max}}$, that could be used to calculate the correct KMC value. Integrating Eq. (3) with respect to time and solving for y leads to the identity

$$\frac{\beta}{r} \sum_i \left[t_i T_i^{-2} (\Delta T)_i \exp (-\Delta H/RT_i) \right] = \frac{\beta}{r} t \bar{T}_{\text{max}}^{-2} \bar{\Delta T} \exp (-\Delta H/R\bar{T}_{\text{max}}). \quad (4)$$

Here T_i is the average experimental T_{max} during time t_i , $\sum_i t_i = t$, $(\Delta T)_i$ is the temperature difference across the rod of radius r during time t_i and is a result of the approximation $(dT/dx)_i \approx (\Delta T)_i/r$, $\bar{\Delta T} = [\sum_i t_i (\Delta T)_i]/t$,

and the ΔH value is that determined from constant-temperature experiments for the particular kernel compound. In most cases, $(\Delta T)_i$ is constant and the ΔT terms cancel on both sides of Eq. (4). The left-hand side of Eq. (4) was evaluated from the irradiation time-temperature records, and $\bar{T}_{\max}$ was determined by a Newton-Raphson iteration method.¹³ In-reactor, time-dependent fuel temperatures were calculated by computer codes;¹⁴ these results were used to evaluate $\bar{T}_{\max}$ and b for calculation of KMC values for each irradiation reported below. It is important to note that the computer-calculated temperatures are uncertain by a minimum of $\pm 100^\circ\text{C}$; this often has the net effect of preventing a precise temperature comparison of KMC data from different irradiations.

The theoretically-based techniques developed in Refs. 6 and 15 for establishing the rate-determining kernel migration mechanism can be validated in part with recent dicarbide data. This validation increases the confidence in the use of these theoretical techniques for oxide data correlation. In carbide particles, solid-state diffusion has been established as the mechanism controlling the kernel migration rate by other arguments.¹¹ Thus, an analysis of the migration data for $\text{Th}_{0.84}\text{U}_{0.16}\text{C}_2$ by the rate-determining methods given earlier^{6,15} should again indicate control by a solid-state diffusion mechanism. The analytical method^{6,15} requires a plot of the migration for each kernel vs the radial location of the particle in the fuel compact, as shown in Fig. 2. These data were obtained from fuel compacts 16 and 18 in the GAC Ell-07 irradiation in the 40 MW(e) Peach Bottom gas-cooled reactor operated by Philadelphia Electric Company. The average kernel migration behavior as a function of radial location is shown as a solid line in this figure. The theoretically-derived kernel migration behavior is based on Eq. (3) and is calculated from Eq. (4) of Ref. 15 to give

$$\frac{y_x}{y_{\bar{x}}} = \frac{T_x^2 \left(\frac{dT}{dx} \right)_x \exp(-\tau \Delta H/R)}{T_{\bar{x}}^2 \left(\frac{dT}{dx} \right)_{\bar{x}}} \quad (5)$$

Fig. 2. $\text{Th}_{0.84}\text{U}_{0.16}\text{C}_2$ kernel migration in fuel compacts from the Ell-07 irradiation in the Peach Bottom reactor. The theoretical behavior was calculated from Eq. (5) for $\Delta H = 76$ kcal/mol and $\bar{x} = 2.85$ cm.

where $\bar{x}$ is the radial midpoint in the fuel compact, x ranges from the minimum to the maximum value of the fuel compact radius, and $\tau = T_x^{-1} - T_{\bar{x}}^{-1}$. It is obvious that the ratio in Eq. (5) is 1 when $x = \bar{x}$. For the $\text{Th}_{1-z}\text{U}_z\text{C}_2$ system, $\Delta H = 76$ kcal/mol,⁷ and the temperatures and temperature gradients were calculated from Eq. (2) and the applicable T_{max} and b values for the "PBell-07" compacts listed in Table I. The resulting theoretically-calculated behavior is shown as the dashed line in Fig. 2; here Eq. (5) has been multiplied by a scale factor of 20 to bring the actual and theoretically calculated curves into compliance for comparison. The agreement of the shapes of the two curves indicates that the migration rate of the dicarbide kernel is indeed controlled by a solid-state diffusion mechanism. More importantly, since Eq. (3) and (5) are valid for solid-state diffusion in both carbide and oxide systems,⁶ this exercise demonstrates the usefulness of the analytical methods developed previously for determining whether or not a solid-state diffusion mechanism controls the rate of kernel migration in dioxide fuels.^{6,15}

(2) Dioxide Fuels

The determination of the rate-controlling process in oxide fuels resulted from the theoretical analyses provided by several workers. Flowers and Horsley¹⁶ presented an initial analysis that considered the effects of gas-phase carbon transport resulting from temperature-dependent gradients of CO and CO₂ pressures in a coated particle. Here the pressures of these gases are a function of the temperature and pressure dependence of the CO-CO₂-C equilibrium, the amount of actinide burnup, the fission-yield spectrum of the fissioning isotope, and the imprecisely known temperature and burnup dependence of the oxidation state of the fuel plus fission-product mixture.¹⁶ Lindemer and de Nordwall⁶ extended this analysis to include consideration of rate control by either gas-phase CO-CO₂ diffusion mechanisms, gas-solid and solid-solid reactions, or solid-state diffusion processes in the oxide kernel.

Application of the theoretical developments to the analysis of in-reactor migration of UO₂ kernels containing ~30% fission-product oxides resulted in the conclusion that a solid-state diffusion process operating

Table I. HTGR Particle Irradiation Characteristics

Kernel type and irradiation No.	$\bar{T}_{\max}$ (°K)	b (°K/cm)	Time (days)	FIMA (%)	Range of observed kernel migration (μm)*	Footnotes**
<u>UO₂</u>						
HRB-7-4	1575	710	184.0	84.68	13-72	a, h, i
HRB-7-5	1600	735	184.0	84.58	20-65	b, h, i, l
HRB-7-6	1620	760	184.0	84.54	37-63	h, i, l
HRB-7-7	1640	780	184.0	84.57	47-90	h, i
HRB-8-4	1520	710	228.0	84.81	8-15	h, i
HRB-8-5	1505	690	228.0	84.63	30-82	b, h, i, l
HRB-8-6	1495	670	228.0	84.56	28-42	h, i, l
HRB-8-7	1515	690	228.0	84.57	50-105	h, i
<u>ThO₂</u>						
H-1-10	2273	1488	39.3	0.37	12-62	g
H-2-11	2033	1600	39.3	0.24	4-36	g
HRB-3-3A	1695	793	274.0	14.00	4-9	g
HRB-4	1569	561	244.0	13.50	3-9	g
<u>Th_{1-z}U_zO₂</u>						
HRB-7-12	1700	810	184.0	46.32	7-64	c, h, i
HRB-8-12	1605	730	228.0	48.05	7-42	c, h, i
HRB-7-8	1675	775	184.0	33.53	6-47	d, h, i
HRB-8-8	1560	695	228.0	35.83	5-30	d, h, i
H-2-11	2033	1600	39.3	12.71	60-200	d, g
H-1-10	2273	1488	39.3	8.70	50-262	e, g
H-2-10	1938	1512	39.3	8.78	16-240	e, g
H-1-4	1693	967	97.1	6.82	2-43	e, g
HRB-6-3A	1383	413	161.0	24.70	3-24	e, g
HRB-6-3B	1368	413	161.0	22.80	7-20	b, e, g
HRB-7-2	1558	673	184.0	20.53	8-25	e, h, i
HRB-7-9	1683	805	184.0	23.40	5-19	e, h, i, l
HRB-7-11	1695	820	184.0	23.40	14-68	e, h, i
HRB-7-19	1605	740	184.0	19.95	7-22	e, h, i
HRB-8-2	1458	672	228.0	22.40	5-20	e, h, i
HRB-8-11	1603	740	228.0	26.21	10-37	e, h, i
HRB-8-19	1510	740	228.0	21.64	5-18	e, h, i
OF1-1-1	1423	310	381.0	19.98	2-8	e, g, k
OF1-2-2	1423	310	381.0	18.44	2-17	e, g, k
OF1-3-2	1423	310	381.0	22.39	3-10	e, g, k
OF1-4-1	1423	310	381.0	19.98	3-40	e, g, k
OF1-5-3	1423	310	381.0	22.68	2-8	e, g, k
HRB-7-10	1695	800	184.0	17.84	5-38	f, h, i, l
HRB-8-10	1600	725	228.0	20.98	10-65	f, h, i, l
<u>ThC₂</u>						
H-1-9	1873	1203	39.3	0.42	50-350	g
<u>Th_{0.84}U_{0.16}C₂</u>						
PBE11-07 C16	1588	30	701.0	8.95	3-42	h, j
PBE11-07 C18	1574	29	701.0	8.53	7-41	h, j

* All migration less than 2 μm was ignored.

** The footnotes may be explained as follows:

- a. 1% UC₂ added; b. ²³³U; c. z = 0.50; d. z = 0.33; e. z = 0.20; f. z = 0.11;
g. Eq. (1); h. Eq. (2); i. R_{in} = 0.16 cm; j. R_{in} = 2.2 cm, x_{max} = 3.49 cm;
k. nominal $\bar{T}_{\max}$ and b; l. TRISO coated.

across the oxide kernel was controlling the kernel migration rate.^{6,15} In addition, the theory demonstrated that Eq. (3) should be used to correlate the UO_2 kernel migration data.⁶

An analysis of the migration behavior of ThO_2 can also be performed. Use of the KMC to correlate the ThO_2 data has not been previously substantiated by the method just described because the data from a given fuel rod were always insufficient to determine the rate-controlling mechanism. Therefore, the use of the KMC was assumed for the ThO_2 data. However, auxiliary data can be used to demonstrate that other rate-controlling mechanisms such as a proposed carbon-transporting, CO-CO_2 diffusion mechanism^{6,16} does not control the ThO_2 migration rate, thus further justifying by default the use of the KMC. The equation for carbon transport by the CO-CO_2 diffusion mechanism has been rigorously derived and is given by Eq. (B27) of Ref. 6 as

$$y = \frac{\epsilon'}{q'} k_l t \frac{dT}{dx} \frac{1}{f} \int_0^f \frac{P_{\text{CO}} S_1}{S_2 S_3} df \quad (6)$$

in which $\epsilon'/q' =$ ratio of the observed diffusion coefficient in porous media to that in free space (dimensionless); $f =$ fraction FIMA (Fissions per Initial Metal Atom); $S_1 = P_{\text{CO}} + P_{\text{CO}_2}$ in atm; $S_2 = P_{\text{CO}} + P_{\text{CO}_2} + P_g + P_{fg}$ in atm, where P_g is the pressure of residual coating gases such as He in the coated particle and P_{fg} is the pressure of the Kr and Xe fission-product gases; $S_3 = (1 + 2 P_{\text{CO}_2} P_{\text{CO}}^{-1})^2$; and $k_l =$ rate constant in cm of carbon $\text{sec}^{-1} (\text{°C/cm})^{-1} \text{ atm}^{-1}$ for gas-phase diffusion in the CO-CO_2 system at a given temperature and pressure. The temperature-dependent value of k_l can be calculated⁶ from $\ln(k_l) = -18.04 + 43,200/RT$. Recent measurements of burnup-dependent $P_{\text{CO}} + P_{\text{CO}_2}$ and P_{fg} have been performed¹⁷ at $1100^\circ\text{C} \leq T \leq 2000^\circ\text{C}$ for BISO-coated ThO_2 particles irradiated to $0.03 \leq f \leq 0.18$. The $P_{\text{CO}} + P_{\text{CO}_2}$ measurements were converted to the more fundamental oxygen-release-per-fission (o/f) values, which were fitted to give the expression $\text{o/f} = (0.594 + 21.8f) \exp(-10,900/RT)$. The Kr + Xe release per fission at STP could be expressed as $(\text{Kr} + \text{Xe})/f = 3.4f$ up to $f = 0.09$ and as $(\text{Kr} + \text{Xe})/f = 0.306$ for $f > 0.09$; these results are in close agreement with measurements made by Janvier et al.¹⁸ It was

further assumed that $P_g = P_{CO} + P_{CO_2} = 1 \text{ atm}$ for as-coated particles, and that $\epsilon'/q' = 1$. Consequently, all terms in Eq. (6) were given, with in-particle pressures being calculated from the equations for o/f, the thermodynamic equilibrium for the C-CO-CO₂-O₂ system, (Kr + Xe)/f, the ideal-gas law, and an in-particle free volume for the gases that was typically one-half of the original ThO₂ volume. Finally, Eq. (6) was couched in units equivalent to those for the solid-state KMC by multiplying both sides of Eq. (6) by $T^2 t^{-1} (dT/dx)^{-1}$. The results of the calculations are shown in Fig. 3. Two distinguishing features are immediately apparent; the calculated KMC increases with burnup at a given temperature and decreases with increasing temperature at a given burnup. The basic reason for this behavior is that the proportion of CO₂ increases as $P_{CO} + P_{CO_2}$ increases with temperature at a given burnup. (Similar calculated results based on less-well-known $P_{CO} + P_{CO_2}$ behavior were indicated in Figs. 11-13 of Ref. 6.) Comparison of the actual KMC values for ThO₂ (to be described later, see Fig. 7) with the data in Fig. 3 clearly shows that the CO-CO₂ gas-phase diffusion mechanism cannot be controlling the ThO₂ kernel migration rate.

Recent results for fissioned Th_{0.8}U_{0.2}O₂ can also be analyzed to establish the rate-controlling mechanism for kernel migration. Figure 4 shows kernel migration data obtained from seven different fuel rods in the HRB-7 and -8 irradiations.¹⁹ This figure requires explanation because it is different in some aspects from that for Th_{0.84}U_{0.16}C₂ (Fig. 2) and that for UO₂ plus fission-product oxides in Ref. 15. The migration data are plotted in terms of $y/\bar{y}$, where $\bar{y}$ is the average migration distance observed in a particular fuel rod. This permits data from different fuel rods to be displayed on an equivalent basis even though $\bar{y}$ varied from 10 to 31 μm . Such a plot is valid as long as the radial particle distribution from rod to rod is equivalent, as was the case with these data. The actual radial dependence of $y/\bar{y}$, as shown in Fig. 4, was obtained by averaging all the $y/\bar{y}$ data within each 0.1-cm increment of the radial position. Theoretical calculations from the equations for the different rate-controlling models could also be performed even though T_{max} and b were different for each rod. It was shown that the shapes of

Fig. 3. Calculated KMC values for in-reactor ThO_2 migration controlled by the CO-CO_2 gas-phase diffusion rate.

Fig. 4. Normalized $\text{Th}_{0.80}\text{U}_{0.20}\text{O}_2$ kernel migration in fuel rods from the HRB-7 and -8 irradiations. Equation (5) was used to calculate migration behavior controlled by solid-state diffusion through the kernel at $\Delta H = 36$ kcal/mol. Equation (7) was used to calculate the migration behavior controlled by CO-CO_2 gas-phase diffusion at $\Delta H = -43.2$ kcal/mol (A) and $\Delta H = -20$ kcal/mol (B).

the theoretical curves calculated from Eq. (5) and from each of Eqs. (1-4) of Ref. 15 were insensitive to $\bar{T}_{\max}$ and only moderately sensitive to the b value. Consequently, the $\bar{T}_{\max}$ and b values for these seven fuel rods, Table I, were each averaged to give $\bar{T}_{\max} = 1587^\circ\text{K}$ and $b = 741^\circ\text{K}/\text{cm}^2$. Also, $\bar{x} = 0.435$, the average radial location of all the particles, was used. Equations (2) and (5) were used to calculate the behavior shown in Fig. 4 for solid-state diffusion control in fissioned $\text{Th}_{0.8}\text{U}_{0.2}\text{O}_2$ at $\Delta H = 36,000$ cal/mol (Table II). When CO-CO_2 diffusion controls the migration rate, the following expression for $y/y_{\bar{x}}$ is derived from Eq. (6):¹⁵

$$\frac{y}{y_{\bar{x}}} = \frac{\left(\frac{dt}{dx}\right)_x I_x}{\left(\frac{dt}{dx}\right)_{\bar{x}} I_{\bar{x}}} \exp (+ 43,200 \tau/R) \quad (7)$$

where I_x and $I_{\bar{x}}$ denote the appropriate integral in Eq. (6) and $-43,200$ cal/mol is the ΔH value for k_ℓ . Equation (7) was used for two different situations. In the first, $I_x/I_{\bar{x}}$ was assumed to be temperature-independent, and the curve in Fig. 4 for $\Delta H = -43,200$ cal/mol was obtained. In the second, the Arrhenius-type temperature dependence of the $k_\ell I$ product was assumed to be identical to that for ThO_2 at 15 to 20% FIMA, or $\Delta H = -20,000$ cal/mol. This assumption for I_x is reasonable because the o/f and Kr + Xe release data for $\text{Th}_{0.81}\text{U}_{0.19}\text{O}_2$ at $\sim 22\%$ FIMA and $1100 \leq T \leq 2000^\circ\text{C}$ were consistent with that obtained for fissioned ThO_2 .¹⁷ The origin of the value $\Delta H = -20,000$ cal/mol can be derived. In a coated particle, $P_{\text{CO}_2} \ll P_{\text{CO}}$ under most of the conditions of interest here; thus, in Eq. (6), $S_3 = 1$ and $S_1 = P_{\text{CO}}$. Also, $P_{\text{fg}} > P_{\text{CO}}$ and S_2 does not reflect an appreciable Arrhenius-type temperature dependence, effectively leaving P_{CO}^2 in the numerator of the integral as the only such temperature-dependent term. The P_{CO} value is calculated from the o/f value; thus the $k_\ell I$ product has an effective $\Delta H = -43,200 + 2(10,900) \approx -20,000$ cal/mol. Equation (7) was used to calculate the curve for $\Delta H = -20,000$ cal/mol in Fig. 4. The experimental average behavior agrees much more closely with the calculated behavior of the solid-state mechanism than with the calculated behavior of the CO-CO_2 diffusion mechanism. Finally, consider the calculated x - y behavior resulting from rate control by a reaction which

Table II. Coefficients for Equation (8)

Kernel	α (log [cm ² T ² (dT/dx) ⁻¹])	α_1 [ΔH (2.303R) ⁻¹]	ΔH (kcal/mol)
<u>Laboratory Data</u>			
UO _{1.65} N _{0.25}	-2.49 ± 0.47**	5,280 ± 890**	24.3
ThO ₂	-0.31 ± 0.61	10,200 ± 1280	47.1
ThC ₂	3.51 ± 0.27	15,600 ± 530	71.8
UC ₂	1.96 ± 0.51	13,100 ± 1030	60.4
<u>In-Reactor Data</u>			
UO ₂ *	-2.77 ± 0.43	4,800 ± 580	22.1
Th _{0.8} U _{0.2} O ₂ *	-0.82 ± 0.28	7,830 ± 420	36.0
ThO ₂ *	-0.85 ± 0.47	8,900 ± 790	41.0

* Final fission-product oxide concentrations approximately equal to percent FIMA, Table I.

** One standard deviation.

has been given in Fig. 1 of Ref. 15. Such control always indicates that y decreases as x increases simply because the reaction rate decreases with temperature; the experimental average behavior in Fig. 4 does not indicate such behavior. Thus, a solid-state diffusion process operating across the kernel appears to control the rate of kernel migration in the fissioned $\text{Th}_{0.8}\text{U}_{0.2}\text{O}_2$ particle.

IV. Data and Analysis

Many sources of data are used to calculate KMC values. The first U.S. in-reactor KMC data for oxide kernels were obtained at ORNL in mid-1973 from the H-1 and H-2 irradiations.^{20,21} Subsequent irradiations at ORNL and GAC have added to the in-reactor KMC for oxide and carbide kernels. The migration results, the temperature conditions, and the particle and kernel dimensional data are available for each particle measured at ORNL to date.^{22,23} In addition, comprehensive reports for most of the irradiations are also available (see the list of references). All oxide and carbide kernels were $\sim 97\%$ of theoretical density and were BISO-coated unless specified as TRISO-coated. The uranium contents were 93% enriched ^{235}U unless stipulated otherwise. Kernel migration measurements were always obtained from the penetration of the higher-temperature kernel interface into the coating layers; the observed range of migration values for each irradiation is given in Table I.

Unirradiated, as-coated particles were also used in laboratory experiments to determine KMC values; such values are called "laboratory data" in this report. The least-squares fit of the KMC data to the equation

$$\log_{10} (\text{KMC}) = \alpha - \alpha_1 T^{-1} \quad (8)$$

was performed for specific kernel compositions. The values of $\alpha = \log_{10} \beta$, $\alpha_1 = \Delta H(2.303 R)^{-1}$, and their standard deviations²⁴ are given in Table II for the cases where KMC data existed over a temperature range sufficient to calculate a meaningful ΔH value. Confidence limits were computed for the KMC data such that one is 90% confident that a single future determination of a KMC value will lie between the limits.²⁴

(1) UO₂ + Fission-Product Oxides

Migration measurements are available from four irradiations. In-reactor data for 69 particles from the H-1-2 irradiation²⁰ to 30% FIMA have been reported previously;¹⁵ a least-squares fit of the H-1-2 data is shown in Fig. 5. Also shown are recent data at 85% FIMA from the HRB-7 and -8 irradiations.¹⁹ The fission products Zr, Ba, Sr, Y, La, and the rare earths form oxides and collectively yield ~ 1 mol of oxides per mol of UO₂ that fissioned. Thus, burnups of 30 to 85% FIMA result in similar percentages of fission-product oxides, the balance of the kernel oxide being UO₂. The KMC values are obviously for kernels of this composition. TRISO-coated particles were used in HRB-7-5 and -6, and in HRB-8-5 and -6; ²³³UO₂ kernels were used in HRB-7-5 and HRB-8-5 particles. GAC has also reported TRISO-particle KMC data that agree with the present data.²⁵ Collectively, these results apparently do not indicate a significant difference in KMC values originating from BISO-vs-TRISO particles, ²³³UO₂ vs ²³⁵UO₂ kernels, or burnup. The H-1-2, HRB-7, and HRB-8 data were thus fitted to Eq. (8) to give the parameters shown in Table II.

Several laboratory experiments on as-coated UO₂ particles from 1200 to 1900°C failed to produce kernel migration at times and temperature conditions that should have been sufficient to promote observable migration.

(2) UO₂-U₂N₃ Solid Solution

The solid solution of UO₂ and U₂N₃ was investigated as a possible HTGR fuel. At HTGR temperatures, extensive amounts of U₂N₃ can dissolve in the face-centered-cubic UO₂ lattice.^{26,27} In a pyrocarbon-coated HTGR particle this establishes the equilibrium $[U_2N_3] + 4 CO \rightleftharpoons 2[UO_2] + 1.5 N_2 + 4 C$, where the brackets indicate the single-phase solid solution. It was postulated from the form of the equilibrium constant that operation of this equilibrium during burnup would result in CO pressures (P_{CO}) substantially lower than those generated in fissioning UO₂ particles.²⁸ Laboratory KMC values obtained for as-coated UO_{1.65}N_{0.25} are shown in Fig. 5; at the indicated temperatures, in-particle P_{CO} and P_{N_2} were

Fig. 5. Laboratory KMC values for as-coated UO_{1.65}N_{0.25} (particle batch A6590-55P) and in-reactor KMC values for UO₂ plus fission-product oxides irradiated either to ~ 85% FIMA or to ~ 30% FIMA (H-1-2). The 90% confidence limits for irradiated UO₂ are for all the KMC data from H-1-2, HRR-7, and HRB-8. Coated-particle batch numbers (such as "OR-2097H") given in this and subsequent figure legends can be used to identify the specific characteristics of coated particles described in the references.

each ~ 0.5 atm.²⁸ Applicability of the KMC is assumed here so that a comparison could be made with the in-reactor KMC values for UO_2 . The parameters for the least-squares fit of Eq. (8) are given in Table II for the laboratory data. It appears from Fig. 5 that the migration behavior in the two systems is comparable. Typical in-reactor behavior¹⁹ at 80% FIMA of a kernel with the initial composition $\text{UO}_{1.50}\text{N}_{0.38}$ is shown in Fig. 6; at this burnup, P_{CO} and P_{N_2} were calculated to be ~ 0.5 and ~ 350 atm, respectively. Penetration of the kernel material into the coating layers was randomly oriented toward the direction of the temperature gradient. However, the extent of migration was approximately that expected for a UO_2 kernel for the particular T , dT/dx , and t conditions. This is again consistent with the KMC values in Fig. 5 and indicates that the migration properties of the fissioned $\text{UO}_2\text{-U}_2\text{N}_3$ solid-solution kernel are not an improvement over those for fissioned UO_2 kernels.

(3) ThO_2

In-reactor KMC data obtained from several irradiations are shown in Fig. 7. Burnup values ranged from 0.24 to 0.35% FIMA in the H-1 and H-2 irradiations,^{20,22} to 14.0% FIMA in the HRB-3 irradiation,²⁹ and to 13.5% FIMA in the HRB-4 irradiation.³⁰ No apparent burnup effect was evident in the KMC values. It should be noted that the high-burnup ThO_2 kernels contained ~ 2.5 to 3.0% of bred-in U;^{6,17} ThO_2 particles in the HTGR would contain similar amounts.

The laboratory KMC data obtained on as-coated ThO_2 kernels agree with the in-reactor data (see Fig. 7 and Table II). A photomicrograph of an as-coated ThO_2 kernel after migration (see Fig. 8) exhibits the cold-side graphitic deposit typically seen behind the kernel in in-reactor specimens.^{20,29,31} Laboratory KMC data were also obtained from 1310 to 1480°C by GAC on previously irradiated ThO_2 particles;³² these data agree with those in Fig. 7.

The laboratory migration data for ThO_2 were also analyzed to demonstrate a linear y - t dependence. Seven particles heated at 1325 to 1345°C in one specimen at $dT/dx = 280^\circ\text{C}/\text{cm}$ migrated an average of 4.4, 6.7, 10.1,

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Fig. 6. Kernel migration in fissioned $\text{UO}_{1.50}\text{N}_{0.38}$ at $\sim 1150^\circ\text{C}$, $\sim 650^\circ\text{C}/\text{cm}$, and $\sim 80\%$ FIMA in the HRB-7 irradiation.

Fig. 7. Laboratory KMC values for as-coated ThO_2 and in-reactor KMC values for ThO_2 irradiated from 0.24 to 14.0% FIMA. The 90% confidence limits are for the in-reactor data.

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15 30 MICRONS 90
0.001 750X INCHES 0.003 0.004

Fig. 8. BISO-coated ThO_2 heated in the laboratory for 551 h at $\sim 1800^\circ\text{C}$ and $280^\circ\text{C}/\text{cm}$. The crescent-shaped layer is carbon deposited on the cold side of the ThO_2 kernel during kernel migration; the ThO_2 kernel is shown at the right.

and 20 μm in 159, 309, 435, and 731 h, respectively. Another group of seven particles heated at 1810 to 1935°C in a second specimen migrated an average of 4.4, 6.9, and 10.4 μm in 143, 365, and 551 h, respectively. A plot of the average y values for each specimen vs time appeared both to be linear and to pass through the origin. On the other hand, GAC workers noted that in laboratory studies made at $\sim 1500^\circ\text{C}$ the onset of migration of previously irradiated ThO_2 kernels was delayed for a period of time that decreased with increasing burnup and temperature.³² The ORNL and GAC results are probably consistent, but further work is needed in this area.

(4) $\text{Th}_{1-z}\text{U}_z\text{O}_2$ + Fission-Product Oxides

Extensive in-reactor KMC data were obtained for kernels having initial compositions in the UO_2 - ThO_2 solid solution. As was the case for irradiated UO_2 , the fission-product oxide percentage in the kernel is approximately equivalent to the percent FIMA. The KMC data for $\text{Th}_{0.8}\text{U}_{0.2}\text{O}_2$ irradiated from 6.8 to 26% FIMA are given in Fig. 9 and Table II. A TRISO coating was used in HRB-7-9,¹⁹ while ^{233}U was used (instead of ^{235}U) in HRB-6-3A³³ and OF-1-5-3. No detectable effect of ^{233}U content, burnup, or TRISO coatings was noted on the KMC values. In-reactor data for $\text{Th}_{0.5}\text{U}_{0.5}\text{O}_2$, $\text{Th}_{0.67}\text{U}_{0.33}\text{O}_2$, and $\text{Th}_{0.98}\text{U}_{0.11}\text{O}_2$ are given in Fig. 10. The KMC data for these compositions, which lie within the range of KMC values for irradiated $\text{Th}_{0.8}\text{U}_{0.2}\text{O}_2$, indicate no discernible effect of the Th/U value on the KMC behavior. The HRB-7-10 and HRB-8-10 particles had TRISO coatings; again, no KMC differences are apparent for BISO- and TRISO-coated particles.

Laboratory experiments from 1500 to 1900°C on as-coated $\text{Th}_{0.8}\text{U}_{0.2}\text{O}_2$ particles were not successful in producing kernel migration.

(5) UC_2

The KMC data from laboratory studies performed on as-coated particles containing fully-dense UC_2 kernels are given in Fig. 11 and Table II. The KMC behavior is considered to be identical to that determined by Stansfield

Fig. 9. In-reactor KMC values and their 90% confidence limits for $\text{Th}_{0.8}\text{U}_{0.2}\text{O}_2$ irradiated from 6.8 to 26% FIMA.

Fig. 10. In-reactor KMC values for $\text{Th}_{1-z}\text{U}_z\text{O}_2$ having z values of either 0.50 (squares), 0.33 (circles), or 0.11 (triangles).

Fig. 11. Laboratory KMC values for as-coated UC₂; the GAC 90% confidence limits⁷ for as-coated UC₂ are used to illustrate the agreement of the GAC and ORNL data. Also shown is the upper-limit KMC for UC_{0.28}N_{0.72}; these kernels did not migrate. The shaded area is the range of the maximum KMC values that are permissible for the present GAC HTGR fuel design.³⁷

et al.⁷ since the present data lie essentially within the 90% confidence limits of the GAC results. This agreement and other data given below also indicate that differences in the configuration⁷ of the ORNL and GAC laboratory experiments had no effect on the KMC.

(6) UC_{1-z}N_z

Laboratory experiments on as-coated particles containing UC_{0.43}N_{0.57} kernels were first described by Naoumidis;⁸ subsequently, Naoumidis and Thiele³⁴ reported data for four particles with a KMC value of $\sim 6 \times 10^{-6}$ at 1675 to 1730°C. The present authors attempted to produce migration in particles containing UC_{0.28}N_{0.72} kernels. (The kernels were prepared by reacting UC₂ with N₂ for 48 h at 1700°C in flowing N₂ at P_{H₂} = 0.5 atm.) No migration was observed in the experiments; however, an upper limit for the KMC was calculated by assuming that the minimum detectable migration was $y = 2 \mu\text{m}$. This plus the t , T , and dT/dx values for the experiments led to the upper KMC limit shown in Fig. 11.

In-reactor tests of TRISO-coated UC_{0.4}N_{0.6} were performed. Irradiation in HRB-10 to 80% FIMA at a maximum temperature of 1500°C resulted in the microstructure shown in Fig. 12. The extensive and deleterious cold-side interaction with the pyrocarbon and SiC was typical. This interaction, which is also characteristically observed in TRISO-coated UC₂ particles irradiated at these conditions, is associated with the presence of fission-product elements Nd, Ce, Pr, and La in the pyrocarbon and SiC.^{19,32,35} Thus, the UC-UN solid solution is apparently not a viable HTGR fuel kernel.

(7) ThC₂

The ORNL in-reactor KMC data for ThC₂ obtained in irradiation H-1-9²⁰ are shown in Fig. 13. It is apparent that the in-reactor results do not entirely agree with the laboratory results on as-coated particles. This discrepancy is analyzed in the Discussion section.

The laboratory KMC data for as-coated particles furnished by GAC from four different coating runs are given in Fig. 13 and Table II. These KMC

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Fig. 12. Bright-field photomicrograph of TRISO-coated $\text{UC}_{0.4}\text{N}_{0.6}$ irradiated to $\sim 80\%$ FIMA at $\sim 1400^\circ\text{C}$ and $\sim 650^\circ\text{C}/\text{cm}$ in the HRB-10 capsule. Cold-side interactions with the coatings are typically associated with the La, Pr, Ce, and Nd fission products.

Fig. 13. Laboratory KMC values for as-coated ThC_2 and in-reactor ThC_2 KMC values. The H-1-9 values are probably atypical since the kernels completely penetrated the particle coatings during irradiation. The GAC 90% confidence limits⁷ for as-coated ThC_2 are shown to illustrate the agreement of the laboratory data obtained at ORNL and at GAC.

values are consistent with the confidence limits reported by Stansfield et al.⁷

(8) Th_{0.84}U_{0.16}C₂

In-reactor KMC data were obtained for Th_{0.84}U_{0.16}C₂ from fuel element Ell-07, which had been irradiated to ~ 9% FIMA in the Peach Bottom reactor. Two annular fuel compacts were examined metallographically and the kernel migration measured. The time-dependent thermal analysis was provided by GAC,³⁶ and Eq. (4) was used to calculate the $\bar{T}_{\max}$ shown in Table I. The resulting KMC values, shown in Fig. 14, appear to be higher than the most recent GAC confidence limits for as-coated Th_{1-z}U_zC₂ particles.³⁷ This discrepancy is analyzed in the Discussion section.

Laboratory KMC data were obtained on as-coated Th_{0.84}U_{0.16}C₂. An archive fuel compact identical to those irradiated in the Peach Bottom reactor was available. A core drill was used to remove a 1.27-cm-diam. cylinder of matrix and fuel particles from the compact, and a low-speed diamond saw was used to slice a 0.09-cm-thick disc from the cylinder. This disc was mounted as shown in Fig. 1; heating resulted in the KMC data shown in Fig. 14. Again, these data are consistent with the most recent GAC confidence limits for Th_{1-z}U_zC₂.³⁷

V. Discussion

(1) Carbide Results

The in-reactor dicarbide data require further general comment. The ThC₂ KMC data from fuel rod H-1-9 (Fig. 13) clearly disagree with the laboratory data for as-coated ThC₂. One might first suspect the accuracy of the thermal analysis. However, the accuracy appears reasonable since the thermal analyses for all the H-1 and H-2 fuel rods were performed simultaneously and the KMC values obtained from H-1 and H-2 fuel rods containing UO₂, ThO₂, Th_{0.8}U_{0.2}O₂, and Th_{0.67}U_{0.33}O₂ particles appear to be consistent with KMC data from other irradiations. The primary difference between the H-1-9 specimens and others was that the ThC₂ kernels from

ORNL DWG 75-8953R1

Fig. 14. $\text{Th}_{0.84}\text{U}_{0.16}\text{C}_2$ KMC values for particles in a fuel compact irradiated to $\sim 9\%$ FIMA and for particles from the same coating batches in a comparable archive compact. The most recent GAC 90% confidence limits³⁸ are used here.

H-1-9 had generally completely penetrated the particle coatings and often had progressed into the fuel rod matrix. For this reason alone, the H-1-9 data are to be considered atypical.

The KMC data for $\text{Th}_{0.84}\text{U}_{0.16}\text{C}_2$ particles from the Ell-07 fuel element (Fig. 14) straddle the upper confidence limit of the laboratory KMC data. Given the probable temperature uncertainty of $\pm 100^\circ\text{C}$ and a probable uncertainty of $\sim 30\%$ in dT/dx , one could not definitely conclude that the in-reactor KMC data were inconsistent with the laboratory data. Thus, further irradiation results would be necessary before one could substantiate the conclusion of Stansfield et al.⁷ that the in-reactor and laboratory KMC values for the dicarbides are indistinguishable.

(2) Oxide Results

Control of the oxide kernel migration rate by a solid-state diffusion mechanism is firmly indicated. Analyses of the in-reactor $\text{Th}_{1-z}\text{U}_z\text{O}_2$ and UO_2 migration data (Fig. 4 and Ref. 15) clearly favor the solid-state diffusion mechanism. Also, workers both at GAC³¹ and at ORNL²⁰ have observed intergranular graphitic deposits within the cooler portion of ThO_2 particles migrating in-reactor; a similar deposit has been noted in UO_2 .³⁴ This phenomenon is not inconsistent with a solid-state transport process operating across the kernel. The few ThO_2 kernels that displayed observable migration in the HRB-3 irradiation were located at approximately two-thirds of the fuel rod radius,²² which is again in accordance with migration control by a solid-state diffusion mechanism.

Several observations can be used to argue against control of the migration rate by gas-phase, CO-CO_2 diffusion. The calculated in-reactor migration behavior for ThO_2 resulting from control by the CO-CO_2 gas phase diffusion mechanism (see Fig. 3) differs significantly from the actual KMC behavior (see Fig. 7). Sayers et al.³⁸ have used the same reasoning to conclude that UO_2 kernel migration behavior is inconsistent with control by the CO-CO_2 mechanism. The CO plus CO_2 overpressures and, thus, the predicted gas-phase transport rates differ by an order of magnitude or more both in as-coated ThO_2 vs irradiated ThO_2 and in the as-coated $\text{UO}_2\text{-U}_2\text{N}_3$

solid solution vs irradiated UO_2 . Nevertheless, one obtains apparently interconsistent KMC values within the ThO_2 system as well as within the UO_2 -based system.

The presence of in-particle gas atmospheres may nevertheless play a significant role in establishing chemical potential gradients and lattice defect structures for solid-state diffusion in oxide particles. Workers at GAC noted that oxide kernel migration ceased when the coating failed and postulated that this was a consequence of the loss of the kernel atmosphere.³¹ The oxygen chemical potential gradient may be involved in the driving force for the solid-state diffusion of the undetermined rate-controlling species and can be calculated by methods suggested by Aitken's work.³⁹ In this case the driving force, X_{O_2} , is $-T \text{ grad}(\Delta F_T^{\text{O}}/T + 2R \ln P_{\text{CO}})$, where the Gibbs free energy change for the equilibrium is $\Delta F_T^{\text{O}} \approx -56,500 - 40T$. Thus, X_{O_2} is relatively insensitive to P_{CO} , which may partially explain why the KMC for oxides is insensitive to burnup even though P_{CO} apparently increases significantly with burnup. On the other hand, a significant decrease of P_{CO} by the addition of second-phase carbides to the UO_2 and $\text{Th}_{1-z}\text{U}_z\text{O}_2$ kernels has also led to the apparent elimination of kernel migration. Sayers et al.³⁸ reported that SiC was an effective additive, while Naoumidis and Thiele³⁴ and Nickel and Balthesen⁴⁰ reported that UC_2 and $\text{Th}_{1-z}\text{U}_z\text{C}_2$ were effective. Wagner-Löffler et al.⁴¹ have also proposed TiC and ZrC additions. These additions establish a specific oxide-carbide equilibrium, such as $\text{UO}_2 + 4\text{C} \rightarrow \text{UC}_2 + 2\text{CO}$ with $X_{\text{O}_2} = T \text{ grad} (-238,000/T + 43.9)$. These equilibria have P_{CO} values that are orders of magnitude smaller than those in 100% oxide particles at normal HTGR temperatures. However, it can be shown that X_{O_2} values calculated from the two quantitative expressions given immediately above have the same sign and magnitude. Thus, these driving forces are apparently not significantly affected by large changes in P_{CO} , or equivalently, μ_{O_2} . Thus, one must look elsewhere to explain the effect of μ_{O_2} on the solid-state diffusion mechanism. It appears that a likely explanation lies in the μ_{O_2} dependence of the interaction of the lanthanide fission-product oxides in extensive solid solution with the uranium oxide to form UO_{2+x} . (Collectively, Y and the lanthanide fission products from

^{233}U and ^{235}U in a thermal neutron flux have a yield of $\sim 55\%$.) In 100% oxide particles, the CO generated during fission results in μ_{O_2} values in the range of $-120 \leq \mu_{\text{O}_2} \leq -90$ kcal/mol for $1000 \leq T \leq 1500^\circ\text{C}$.^{6,17} Thermodynamic data at these μ_{O_2} values for the U-La-O,^{42,43} U-Y-O,⁴³ U-Ce-O,⁴⁴ and U-Nd-O⁴⁵ systems indicate O/U ratios in the range 2.001 to 2.2 would occur.¹⁷ It is known that increasing hyperstoichiometry significantly increases the self-diffusion coefficient of oxygen;⁴⁶ presumably this would similarly affect the diffusion of other "anions" such as solid-solution carbon. The migration observed in the as-coated $\text{UO}_2\text{-U}_2\text{N}_3$ solid solution (see Fig. 5) may also be a direct result of a lattice defect structure that is comparable to that for the UO_2 plus lanthanide oxide solid solution. The lack of the lanthanide oxides and hyperstoichiometry in as-coated UO_2 may also explain why as-coated UO_2 does not migrate. Finally, addition of UC_2 to the UO_2 kernel would lower the μ_{O_2} sufficiently to preclude any hyperstoichiometry in the UO_2 -lanthanide oxide system, thus significantly reducing both the solid-state diffusion rate in the anion sub-lattice and the in-reactor kernel migration rate.

It is clear that a more precise understanding of the thermodynamic and kinetic factors affecting oxide kernel migration is desirable. To a great extent this future understanding will be dependent on information on as-yet uninvestigated thermodynamic and kinetic phenomena at HTGR fuel temperatures and μ_{O_2} values in actinide oxides containing typical amounts of fission products. Future quantitative kernel migration theories based on this information could then be applied to the kernel migration data given in Refs. 22 and 23.

(3) Application of Data to HTGR Fuel Design

Gulden et al.³⁵ have discussed the GAC HTGR fuel design criteria that affect the selection of kernel compositions. These criteria were expressed in terms of maximum acceptable KMC values by setting y at a constant value which represented an acceptable total amount of kernel migration, and by properly incorporating t and the time-dependent T and dT/dx characteristics of the fuel design. This range of maximum KMC

values is shown as the shaded area in Fig. 11. Actual KMC values lying below the design-based KMC were acceptable, while those within or above were not. By this means, fully-dense UO_2 and $\text{Th}_{1-z}\text{U}_z\text{O}_2$ kernels were found to have unacceptably high KMC values and hence were not considered further for the GAC HTGR design. The KMC values for thorium and all the dicarbide fuel kernels were acceptable. Other HTGR fuel designs may have maximum acceptable KMC values different from those in Fig. 11; for example, if dT/dx in another design were 0.1 of the GAC dT/dx values, then the shaded area would lie at KMC values that were higher by a factor of 10. Consequently, the KMC values for $\text{Th}_{1-z}\text{U}_z\text{O}_2$ and UO_2 would also be acceptable.

VI. Conclusions

Several conclusions may be drawn from the data and analyses presented here. These are as follows:

- (1) The previously published technique^{6,15} for determining the in-reactor, rate-controlling mechanism for kernel migration has been validated with $\text{Th}_{0.84}\text{U}_{0.16}\text{C}_2$ data. This reinforces the conclusion^{6,15} that the kernel migration rate for UO_2 plus fission-product oxides is controlled by a solid-state diffusion mechanism operating across the kernel.
- (2) In-reactor migration of fissioned $\text{Th}_{0.8}\text{U}_{0.2}\text{O}_2$ appears to be controlled by a solid-state diffusion mechanism.
- (3) The ThO_2 kernel migration rate does not appear to be controlled by the rate of carbon transport resulting from in-particle CO-CO_2 diffusion. Circumstantial evidence suggests that solid-state diffusion is the controlling mechanism.
- (4) The oxide KMC derived from solid-state diffusion theory appears to correlate the migration data for UO_2 plus fission-product oxides, ThO_2 and ThO_2 plus fission-product oxides, and $\text{Th}_{1-z}\text{U}_z\text{O}_2$ plus fission-product

oxides. No dependence of burnup, BISO-vs-TRISO coatings, or ^{233}U vs ^{235}U content on the KMC values was discernible. The KMC values for $\text{Th}_{1-z}\text{U}_z\text{O}_2$ appeared to be independent of composition in the range $0.11 \leq z \leq 0.50$.

- (5) The kernel migration rate for as-coated ThO_2 as determined in the laboratory appears to be linear with time at 1810 to 1945°C.
- (6) The laboratory KMC data obtained by ORNL and by GAC for dicarbide kernels appear to be in excellent agreement. In view of the uncertainties for in-reactor temperature calculations, the in-reactor and laboratory KMC values do not appear to be inconsistent.
- (7) The irradiation performance of nitrogen-containing oxide or carbide kernels was no better than that of analogous nitrogen-free kernel compounds. Consequently, nitrogen additions to HTGR fuels do not appear to be warranted.

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Appendix A. Coated Particle Characteristics

Table A-I. Description of ORNL Fissile and Fertile Oxide Particles

Initial kernel composition	Batch No.	Kernel diameter (μm)	Buffer thickness (μm)	Inner LTI thickness (μm)	SiC thickness (μm)	Outer LTI thickness (μm)	In-Reactor experiment No.
UO_2	OR-2089 H	196	75	37	27	77	HRB-7 Rod 6
UO_2 -1% UC_2	OR-2097 H	194	68	None	None	78	HRB-8 Rod 6
							HRB-7 Rod 4
UO_2	OR-2114 H	196	85	None	None	41	HRB-8 Rod 4
							HRB-7 Rod 7
UO_2^*	PU-303 H	227	85	35	27	41	HRB-8 Rod 7
							HRB-7 Rod 5
$\text{UO}_{1.50}\text{N}_{0.38}$	OR-2098H	193	71	None	None	76	HRB-8 Rod 5
							HRB-7 Rod 3
$\text{UO}_{1.65}\text{N}_{0.25}$	A6590-55 P**	497	29	None	None	53	None ^{††}
ThO_2	J-489	495	80	None	None	77	None [§]
ThO_2	OR-1365	450	47	None	None	73	H-1 Rod 10
							H-2 Rod 10
							H-2 Rod 11
ThO_2	OR-1562	398	55	None	None	76	HRB-3 Rod 3A
ThO_2	OR-1849	501	69	None	None	77	None ^{§§}
ThO_2	OR-1856	488	88	None	None	79	HRB-4 Rod 3A
$\text{Th}_{0.5}\text{U}_{0.5}\text{O}_2$	OR-2090 H	246	79	None	None	84	HRB-7 Rod 12
							HRB-8 Rod 12
$\text{Th}_{0.67}\text{U}_{0.33}\text{O}_2$	OR-2094 H	282	85	None	None	89	HRB-7 Rod 8
							HRB-8 Rod 8
$\text{Th}_{0.69}\text{U}_{0.31}\text{O}_2$	PR-67	353	78	None	None	142	H-2-11
$\text{Th}_{0.80}\text{U}_{0.20}\text{O}_2$	J-263	354	101	None	None	92	HRB-7 Rods 2 and 19
							HRB-8 Rods 2 and 19
							OF-1 Rods 1-1, 2-2, 3-2, and 4-1
$\text{Th}_{0.80}\text{U}_{0.20}\text{O}_2$	OR-1909	366	97	None	None	93	HRB-6 Rod 3B
$\text{Th}_{0.80}\text{U}_{0.20}\text{O}_2$	OR-2111 H	360	95	None	None	95	HRB-7 Rod 11
							HRB-8 Rod 11
$\text{Th}_{0.80}\text{U}_{0.20}\text{O}_2$	OR-2116 H	360	98	39	34	43	HRB-7 Rod 9
$\text{Th}_{0.80}\text{U}_{0.20}\text{O}_2$	PR-57	355	80	None	None	135	H-1 Rod 4
							H-1 Rod 10
							H-2 Rod 10
$\text{Th}_{0.80}\text{U}_{0.20}\text{O}_2^*$	PU-291	351	105	None	None	85	HRB-6 Rod 3A
$\text{Th}_{0.89}\text{U}_{0.11}\text{O}_2$	GGA-6155-00-020 [†]	502	106	32	34	42	OF-1 Rod 5-3
							HRB-7 Rod 10
							HRB-8 Rod 10

* ^{233}U .

** Number of T. B. Lindemer's laboratory notebook.

† Supplied by GAC.

†† Used in laboratory runs 14, 18, 20, 24, and 30.

§ Used in laboratory run 32.

§§ Used in laboratory run 23.

Table A-II. Description of GAC-Supplied Fissile and Fertile Carbide Particles

Initial kernel composition	Batch No.	Kernel diameter (μm)	Buffer thickness (μm)	Inner LTI thickness (μm)	SiC thickness (μm)	Outer LTI thickness (μm)	Laboratory run No.
UC ₂	6151-00-035	201	87	33	27	37	19
UC ₂	A6590-58 P ^{*,**}	454	85	None	None	43	11, 12, 13, 14 19, 22, 29, 32
UC _{0.28} N _{0.72}	A6590-46P ^{*,**}	420	18	None	None	48	22 ^{††}
ThC ₂	4000-225	379	59	None	None	73	19A, 29
ThC ₂	4261-00-011	358	54	35	25	38	33
ThC ₂	4413-127	578	68	None	None	61	33
ThC ₂	9T-980-BL	371	52	None	None	65	18, 33 [§]
Th _{0.84} U _{0.16} C ₂	Peach Bottom [†] Core-2	300, 400	40, 50	None	None	60, 85	20 ^{§§}

* Supplied by ORNL.

** Number of T. B. Lindemer's laboratory notebook.

† A mixture of two different particle sizes.

†† Also used in in-reactor experiment HRB-10 Rod-18.

§ Also used in in-reactor experiment H-1 Rod-9.

§§ Also used in in-reactor experiment Ell-07 Compacts 16 and 18.

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Appendix B. Original In-Reactor Data Used for KMC Calculations*

*Also see Ref. 22 for previous data.

IN-REACTOR SPECIMEN NUMBER HRB-7-4, UO2-1XUC2, (OR-2097H)
 $T(K) = 1575. - 710. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.96
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=84.68 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0013	0.500	1397.	711.	2.16E-07
0.0033	0.500	1397.	711.	5.70E-07
0.0031	0.356	1485.	505.	8.52E-07
0.0072	0.513	1388.	729.	1.21E-06

IN-REACTOR SPECIMEN NUMBER HRB-7-5, 233UG2, (PU-303H)
 $T(K) = 1600. - 735. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 2.17
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=84.58 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0048	0.523	1399.	769.	7.68E-07
0.0020	0.351	1510.	515.	5.57E-07
0.0065	0.312	1528.	459.	2.08E-06
0.0020	0.409	1477.	601.	4.57E-07
0.0050	0.409	1477.	601.	1.14E-06

IN-REACTOR SPECIMEN NUMBER HRB-7-6, UO2, (OR-2089H)
 $T(K) = 1620. - 760. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 2.25
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=84.54 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0050	0.356	1524.	541.	1.35E-06
0.0037	0.488	1439.	741.	6.59E-07
0.0047	0.566	1376.	861.	6.50E-07
0.0063	0.381	1510.	579.	1.56E-06

IN-REACTOR SPECIMEN NUMBER HRB-7-7, UO2, (OR-2114H)
 $T(K) = 1640. - 780. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 2.76
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=84.57 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0062	0.533	1418.	832.	9.43E-07
0.0054	0.447	1484.	697.	1.07E-06
0.0065	0.414	1506.	646.	1.44E-06
0.0060	0.526	1424.	820.	9.34E-07
0.0047	0.508	1439.	792.	7.80E-07
0.0080	0.439	1489.	685.	1.63E-06
0.0090	0.495	1449.	773.	1.54E-06

IN-REACTOR SPECIMEN NUMBER HRB-8-4, UO2-1XUC2, (OR-2097H)
 $T(K) = 1520. - 710. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.96
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=84.81 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0008	0.569	1290.	808.	7.84E-08
0.0010	0.566	1292.	804.	1.05E-07
0.0015	0.213	1488.	303.	5.56E-07

IN-REACTOR SPECIMEN NUMBER HRB-8-5, 233UC2, (PU-303H)
 $T(K) = 1505. - 690. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 2.17
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=84.63 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0030	0.190	1480.	263.	1.27E-06
0.0082	0.432	1376.	596.	1.33E-06

IN-REACTOR SPECIMEN NUMBER HRB-3-6, UO2, (OR-2089H)
 $T(K) = 1495. - 670. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 2.25
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=84.56 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0028	0.592	1260.	793.	2.80E-07
0.0042	0.521	1313.	698.	5.33E-07

IN-REACTOR SPECIMEN NUMBER HRB-8-7, UC2, (OR-2114H)
 $T(K) = 1515. - 690. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 2.76
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=84.57 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0105	0.551	1305.	761.	1.19E-06
0.0050	0.155	1498.	214.	2.67E-06
0.0090	0.216	1483.	298.	3.37E-06
0.0087	0.508	1337.	701.	1.13E-06
0.0075	0.330	1440.	456.	1.73E-06
0.0060	0.607	1261.	838.	5.78E-07
0.0082	0.594	1271.	820.	8.25E-07
0.0100	0.602	1265.	831.	9.78E-07

HRB-4 FUEL ROD NUMBER 3A, THO2
 $T(K) = 1569. - 561. * X^{**2}$, DAYS= 244.0
 RATIO, FREE VOLUME /OXIDE VOLUME= 0.76
 INITIAL BURNUP, PERCENT FIMA= 0.0
 FINAL BURNUP, PERCENT FIMA=13.50

Y, CM	X, CM	T, K	K/CM	KMC
0.0003	0.549	1400.	616.	5.29E-08
0.0000	0.544	1403.	610.	7.66E-09
0.0009	0.450	1456.	504.	1.69E-07
0.0003	0.442	1459.	496.	6.11E-08
0.0003	0.422	1469.	473.	5.41E-08
-0.0010	0.185	1550.	208.	-5.75E-07
0.0000	0.325	1510.	365.	1.48E-08
-0.0009	0.399	1480.	447.	-1.97E-07

IN-REACTOR SPECIMEN NUMBER HRB-7-12, (TH,U)O₂, (OR-2090H)
 $T(K) = 1700. - 810. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.71
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=46.32 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0014	0.264	1644.	428.	5.56E-07
0.0001	0.259	1646.	420.	4.06E-08
0.0064	0.566	1440.	918.	9.10E-07
0.0015	0.564	1442.	913.	2.15E-07
0.0007	0.259	1646.	420.	2.64E-07
0.0021	0.518	1483.	839.	3.46E-07
0.0035	0.630	1379.	1020.	4.10E-07
0.0025	0.432	1549.	700.	5.39E-07
0.0042	0.554	1452.	897.	6.28E-07
0.0020	0.323	1616.	523.	6.28E-07
0.0039	0.577	1431.	934.	5.38E-07
0.0018	0.338	1608.	547.	5.50E-07

IN-REACTOR SPECIMEN NUMBER HRB-8-12, (TH,U)O₂, (OR-2090H)
 $T(K) = 1605. - 730. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.71
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=48.05 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0030	0.396	1490.	579.	5.85E-07
0.0035	0.424	1474.	619.	6.23E-07
0.0020	0.551	1383.	805.	2.41E-07
0.0013	0.351	1515.	512.	2.85E-07
0.0002	0.196	1577.	286.	1.11E-07
0.0022	0.231	1566.	337.	8.30E-07
0.0030	0.483	1435.	705.	4.45E-07
0.0030	0.483	1435.	705.	4.45E-07
0.0007	0.378	1500.	553.	1.55E-07
0.0010	0.272	1551.	397.	3.08E-07
0.0042	0.559	1377.	816.	5.01E-07
0.0025	0.577	1362.	842.	2.80E-07
0.0010	0.391	1493.	571.	1.98E-07
0.0037	0.429	1471.	627.	6.57E-07

IN-REACTOR SPECIMEN NUMBER HRB-7-8, (2TH,U)O₂, (OR-2094H)
 $T(K) = 1675. - 775. (X^{**2} - RIN^{**2} (1 + 2LN (X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.56
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=33.53 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0007	0.368	1570.	571.	2.04E-07
0.0006	0.249	1627.	386.	2.59E-07
0.0047	0.571	1422.	886.	6.75E-07
0.0029	0.444	1522.	689.	6.03E-07
-0.0035	0.297	1607.	461.	-1.23E-06
0.0041	0.556	1435.	862.	6.24E-07
0.0044	0.493	1487.	764.	8.01E-07
0.0040	0.505	1477.	783.	7.01E-07
0.0038	0.480	1496.	744.	7.29E-07

IN-REACTOR SPECIMEN NUMBER HRB-8-8. (2TH,U)02. (OR-2094H)
 $T(K)=1560.-695.(X^{**2}-RIN^{**2}(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.56
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=35.83 .RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0030	0.569	1335.	791.	3.43E-07
0.0020	0.561	1341.	780.	2.34E-07
0.0018	0.551	1349.	766.	2.11E-07
0.0015	0.541	1357.	752.	1.86E-07
0.0010	0.282	1505.	392.	2.93E-07
0.0015	0.564	1339.	784.	1.74E-07
0.0012	0.330	1484.	459.	3.05E-07
0.0019	0.569	1335.	791.	2.17E-07
0.0015	0.564	1339.	784.	1.74E-07
0.0005	0.193	1534.	268.	2.23E-07
0.0025	0.500	1386.	696.	3.51E-07

IN-REACTOR SPECIMEN NUMBER HRB-7-2. (4TH,U)02. (J-263)
 $T(K)=1553.-673.(X^{**2}-RIN^{**2}(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.44
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=20.53 .RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0014	0.511	1383.	687.	2.45E-07
0.0012	0.320	1489.	431.	3.89E-07
0.0009	0.366	1468.	492.	2.48E-07
0.0003	0.290	1502.	390.	1.09E-07
0.0008	0.444	1425.	598.	1.71E-07
-0.0002	0.198	1532.	267.	-1.11E-07
0.0010	0.251	1515.	338.	4.27E-07
0.0025	0.541	1361.	728.	4.00E-07
-0.0008	0.208	1529.	280.	-4.20E-07

IN-REACTOR SPECIMEN NUMBER HRB-7-9. (4TH,U)02. (OR-2116H)
 $T(K)=1633.-805.(X^{**2}-RIN^{**2}(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.34
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=23.40 .RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0015	0.335	1593.	540.	4.43E-07
0.0015	0.516	1469.	830.	2.45E-07
0.0006	0.444	1524.	716.	1.22E-07
0.0005	0.493	1483.	793.	7.90E-08
0.0002	0.371	1572.	597.	1.95E-07
0.0012	0.457	1515.	736.	2.35E-07
0.0019	0.547	1441.	893.	2.81E-07

IN-REACTOR SPECIMEN NUMBER HRB-7-11. (4TH,U)02. (OR-2111H)
 $T(K)=1695.-820.(X^{**2}-RIN^{**2}(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.28
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=23.40 .RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0068	0.455	1520.	746.	1.34E-06
0.0026	0.505	1486.	829.	4.35E-07
0.0030	0.437	1539.	716.	6.23E-07
0.0017	0.363	1584.	604.	4.31E-07
0.0014	0.193	1604.	317.	7.71E-07
-0.0002	0.221	1655.	362.	-1.19E-07
0.0034	0.485	1502.	756.	6.06E-07
0.0046	0.424	1547.	696.	9.96E-07
0.0015	0.490	1493.	804.	2.63E-07

IN-REACTOR SPECIMEN NUMBER HRB-7-19, (4TH,U)02, (J-263)
 $T(K)=1605.-740.(X^{**2}-RIN^{**2}(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.44
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=19.95 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0012	0.559	1374.	827.	1.72E-07
0.0007	0.351	1514.	519.	1.95E-07
0.0007	0.559	1374.	827.	1.01E-07
0.0007	0.290	1543.	429.	2.27E-07
0.0016	0.452	1454.	669.	3.28E-07
0.0017	0.546	1384.	808.	2.54E-07
0.0005	0.290	1543.	429.	1.75E-07
0.0015	0.559	1374.	827.	2.15E-07
0.0022	0.478	1436.	707.	4.04E-07

IN-REACTOR SPECIMEN NUMBER HRB-8-2, (4TH,U)02, (J-263)
 $T(K)=1458.-672.(X^{**2}-RIN^{**2}(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.44
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=22.40 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0010	0.587	1227.	789.	9.69E-08
0.0005	0.554	1252.	744.	5.35E-08
0.0013	0.561	1246.	754.	1.31E-07
0.0007	0.264	1411.	355.	1.85E-07
0.0015	0.508	1285.	683.	1.84E-07
0.0005	0.391	1355.	526.	8.87E-08
0.0010	0.554	1252.	744.	1.07E-07
0.0015	0.579	1233.	778.	1.49E-07
0.0010	0.523	1274.	703.	1.17E-07
0.0008	0.348	1377.	468.	1.54E-07
0.0012	0.417	1341.	560.	2.04E-07
0.0008	0.254	1415.	341.	2.23E-07
-0.0002	0.201	1431.	270.	-9.64E-08
0.0010	0.399	1351.	536.	1.73E-07
0.0000	0.196	1432.	263.	4.72E-13
0.0007	0.320	1389.	430.	1.71E-07
0.0020	0.444	1325.	597.	2.98E-07
0.0015	0.391	1355.	526.	2.66E-07
0.0005	0.295	1400.	396.	1.26E-07
0.0005	0.384	1359.	515.	9.10E-08

IN-REACTOR SPECIMEN NUMBER HRB-8-11, (4TH,U)02, (OR-2111H)
 $T(K)=1603.-740.(X^{**2}-RIN^{**2}(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.28
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=26.21 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0025	0.554	1376.	819.	2.93E-07
0.0027	0.378	1497.	560.	5.59E-07
0.0010	0.432	1465.	639.	1.70E-07
0.0037	0.490	1425.	726.	5.33E-07
0.0025	0.516	1406.	763.	3.29E-07
0.0015	0.536	1390.	793.	1.86E-07
0.0025	0.422	1471.	624.	4.40E-07
0.0020	0.516	1406.	763.	2.63E-07
0.0013	0.368	1503.	545.	2.63E-07
0.0018	0.307	1533.	455.	4.59E-07
0.0020	0.353	1511.	523.	4.43E-07
0.0002	0.173	1581.	256.	1.24E-07
0.0025	0.452	1452.	669.	4.00E-07
0.0015	0.378	1497.	560.	3.05E-07
0.0027	0.450	1453.	665.	4.43E-07
0.0022	0.452	1452.	669.	3.60E-07

IN-REACTOR SPECIMEN NUMBER HRR-3-19, (4TH,U)02, (J-263)
 $T(K)=1510.-740.(X**2-RIN**2(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME / OXIDE VOLUME= 1.44
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=21.64 ,RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0008	0.300	1444.	444.	1.79E-07
0.0010	0.340	1424.	504.	2.04E-07
0.0015	0.348	1420.	515.	2.93E-07
0.0010	0.566	1273.	838.	9.81E-08
0.0005	0.566	1273.	838.	4.90E-08
0.0010	0.554	1283.	819.	1.02E-07
0.0010	0.389	1398.	575.	1.73E-07
0.0010	0.536	1297.	743.	1.08E-07
0.0008	0.465	1350.	688.	1.01E-07
0.0012	0.566	1273.	838.	1.23E-07
0.0000	0.259	1460.	383.	1.35E-12
-0.0037	0.414	1383.	613.	-5.94E-07
0.0018	0.531	1301.	786.	1.92E-07
0.0005	0.554	1283.	819.	5.10E-08

IN-REACTOR SPECIMEN NUMBER OFI-1-1, (4TH,U)02, (J-263)
 $T(K)=1423.-310.*X**2$, DAYS= 381.0
 RATIO, FREE VOLUME / OXIDE VOLUME= 1.44
 INITIAL BURNUP, PERCENT FIMA= 0.0
 FINAL BURNUP, PERCENT FIMA=19.93

Y, CM	X, CM	T, K	K/CM	KMC
0.0002	0.063	1422.	39.	2.61E-07
-0.0005	0.485	1350.	301.	-9.25E-08
0.0008	0.249	1404.	154.	3.25E-07
-0.0003	0.597	1313.	370.	-4.74E-08
-0.0003	0.541	1332.	335.	-5.38E-08
0.0	0.343	1387.	213.	0.0
0.0002	0.564	1324.	350.	2.55E-08
0.0003	0.632	1299.	392.	4.38E-08
-0.0002	0.617	1305.	383.	-2.26E-08
-0.0002	0.452	1360.	280.	-3.36E-08
-0.0002	0.244	1405.	151.	-6.64E-08
0.0	0.262	1402.	162.	0.0
0.0	0.345	1386.	214.	0.0
0.0	0.574	1321.	356.	0.0
0.0007	0.632	1299.	392.	9.76E-08
-0.0005	0.559	1326.	346.	-7.75E-08
0.0005	0.594	1313.	369.	7.15E-08
0.0005	0.076	1421.	47.	6.53E-07

IN-REACTOR SPECIMEN NUMBER OFI-2-2, (4TH,U)02, (J-263)
 $T(K)=1423.-310.*X**2$, DAYS= 381.0
 RATIO, FREE VOLUME / OXIDE VOLUME= 1.44
 INITIAL BURNUP, PERCENT FIMA= 0.0
 FINAL BURNUP, PERCENT FIMA=18.44

Y, CM	X, CM	T, K	K/CM	KMC
0.0007	0.378	1379.	235.	1.65E-07
0.0013	0.505	1344.	313.	2.35E-07
0.0010	0.310	1393.	192.	3.08E-07
0.0002	0.056	1422.	35.	2.97E-07
0.0012	0.592	1314.	367.	1.68E-07
0.0013	0.531	1336.	329.	2.21E-07
0.0017	0.411	1371.	255.	3.75E-07
0.0012	0.473	1352.	296.	2.20E-07
0.0010	0.470	1355.	291.	1.92E-07
0.0008	0.521	1339.	321.	1.41E-07
0.0007	0.620	1304.	384.	9.01E-08
0.0003	0.175	1413.	109.	1.87E-07

Continuation of OF-1-2-2

0.0010	0.556	1327.	345.	1.56E-07
0.0013	0.180	1413.	112.	7.27E-07
0.0002	0.058	1422.	36.	2.84E-07
0.0007	0.505	1344.	313.	1.17E-07
0.0003	0.627	1301.	389.	4.43E-08
0.0008	0.500	1345.	310.	1.48E-07
0.0013	0.564	1324.	350.	2.04E-07
-0.0002	0.594	1313.	369.	-2.38E-08
0.0008	0.518	1340.	321.	1.42E-07
0.0005	0.140	1417.	87.	3.54E-07
0.0012	0.351	1385.	217.	3.14E-07
0.0012	0.394	1375.	244.	2.76E-07
-0.0002	0.538	1333.	334.	-2.71E-08
0.0012	0.556	1327.	345.	1.82E-07
0.0012	0.544	1331.	337.	1.87E-07
0.0007	0.589	1315.	365.	9.64E-08
0.0010	0.424	1367.	263.	2.17E-07
0.0007	0.274	1400.	170.	2.34E-07
-0.0008	0.328	1390.	203.	-2.42E-07
0.0012	0.544	1331.	337.	1.87E-07
0.0008	0.432	1365.	268.	1.77E-07
0.0012	0.424	1367.	263.	2.53E-07
0.0005	0.348	1335.	216.	1.36E-07
0.0	0.190	1412.	118.	0.0

IN-REACTOR SPECIMEN NUMBER OF 1-3-2, (4TH,U)02, (J-263)
 T(K)=1423.- 310.*X**2, DAYS= 381.0
 RATIO, FREE VOLUME /OXIDE VOLUME= 1.44
 INITIAL BURNUP, PERCENT FIMA= 0.0
 FINAL BURNUP, PERCENT FIMA=22.39

Y, CM	X, CM	T, K	K/CM	KMC
0.0005	0.574	1321.	356.	7.48E-08
0.0	0.597	1313.	370.	0.0
0.0000	0.300	1395.	186.	1.02E-12
0.0000	0.236	1406.	146.	1.31E-12
0.0005	0.317	1392.	197.	1.50E-07
0.0007	0.505	1344.	313.	1.17E-07
0.0005	0.320	1391.	198.	1.49E-07
0.0000	0.394	1375.	244.	7.52E-13
0.0003	0.561	1325.	348.	5.14E-08
0.0005	0.531	1336.	329.	8.27E-08
0.0010	0.599	1312.	372.	1.41E-07

IN-REACTOR SPECIMEN NUMBER OF 1-4-1, (4TH,U)02, (J-263)
 T(K)=1423.- 310.*X**2, DAYS= 381.0
 RATIO, FREE VOLUME /OXIDE VOLUME= 1.44
 INITIAL BURNUP, PERCENT FIMA= 0.0
 FINAL BURNUP, PERCENT FIMA=19.98

Y, CM	X, CM	T, K	K/CM	KMC
0.0003	0.597	1313.	370.	4.74E-08
0.0005	0.508	1343.	315.	8.74E-08
0.0017	0.531	1336.	329.	2.76E-07
0.0010	0.229	1407.	142.	4.26E-07
0.0022	0.414	1370.	257.	4.84E-07
0.0008	0.594	1313.	369.	1.19E-07
0.0040	0.493	1348.	306.	7.26E-07
0.0010	0.422	1368.	261.	2.19E-07
0.0010	0.429	1366.	266.	2.14E-07
0.0023	0.536	1334.	332.	3.81E-07
0.0005	0.503	1345.	312.	8.85E-08

IN-REACTOR SPECIMEN NUMBER DF1-5-3. (4TH,233U)02, (PU-291)
 T(K)=1423.- 310.*X**2, DAYS= 381.0
 RATIO, FREE VOLUME / OXIDE VOLUME= 1.44
 INITIAL BURNUP, PERCENT FIMA= 0.0
 FINAL BURNUP, PERCENT FIMA=22.68

Y, CM	X, CM	T, K	K/CM	KMC
0.0002	0.498	1346.	309.	2.99E-08
0.0003	0.564	1324.	350.	5.11E-08
0.0	0.439	1363.	272.	0.0
0.0007	0.582	1318.	361.	9.81E-08
0.0003	0.424	1367.	263.	7.23E-08
0.0002	0.546	1331.	339.	2.66E-08
0.0000	0.437	1364.	271.	6.66E-13
0.0008	0.521	1339.	323.	1.41E-07
-0.0017	0.584	1317.	362.	-2.44E-07
0.0005	0.315	1392.	195.	1.52E-07
0.0007	0.378	1379.	235.	1.65E-07
0.0002	0.559	1326.	346.	2.58E-08
0.0002	0.155	1416.	96.	1.06E-07
-0.0007	0.241	1405.	150.	-2.69E-07
0.0003	0.221	1408.	137.	1.47E-07
0.0	0.249	1404.	154.	0.0
-0.0017	0.505	1344.	313.	-2.93E-07
-0.0017	0.582	1318.	361.	-2.45E-07
-0.0007	0.582	1318.	361.	-9.81E-08

IN-REACTOR SPECIMEN NUMBER HRB-7-10. (8TH,U)02, (GGA-6155-00-020)
 T(K)=1695.- 800.*(X**2-RIN**2*(1+2LN (X/RIN)))
 RATIO, FREE VOLUME/ OXIDE VOLUME= 0.94
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 184.0
 FINAL BURNUP, PERCENT FIMA=17.84 .RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0012	0.345	1600.	553.	3.64E-07
0.0016	0.345	1600.	553.	4.66E-07
0.0038	0.495	1499.	792.	6.69E-07
0.0020	0.396	1569.	634.	4.89E-07
-0.0002	0.343	1601.	549.	-7.35E-08
0.0019	0.437	1542.	699.	4.07E-07
0.0005	0.305	1621.	488.	1.69E-07

IN-REACTOR SPECIMEN NUMBER HRB-8-10. (8TH,U)02, (GGA-6155-00-020)
 T(K)=1600.- 725.*(X**2-RIN**2*(1+2LN (X/RIN)))
 RATIO, FREE VOLUME/ OXIDE VOLUME= 0.94
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 228.0
 FINAL BURNUP, PERCENT FIMA=20.98 .RIN= 0.16CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0035	0.549	1382.	796.	4.26E-07
0.0065	0.554	1378.	803.	7.80E-07
0.0020	0.500	1418.	726.	2.82E-07
0.0023	0.406	1480.	589.	4.25E-07
0.0015	0.546	1384.	792.	1.84E-07
0.0062	0.422	1471.	611.	1.12E-06
0.0025	0.282	1542.	409.	7.39E-07
0.0025	0.356	1486.	575.	4.88E-07
0.0017	0.538	1390.	781.	2.20E-07
0.0010	0.190	1574.	276.	4.55E-07

P8 DRIVER E11-07, COMPACT 16, 5.25/1 (TH,U)C2, 701 DAYS
 $T(K)=1588.-30.(X**2-RIN**2(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.74
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 701.0
 FINAL BURNUP, PERCENT FIMA= 8.95 ,RIN= 2.22CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0014	2.255	1588.	5.	1.24E-05
0.0008	2.490	1584.	31.	1.07E-06
0.0003	2.304	1588.	10.	1.24E-06
0.0015	2.304	1588.	10.	6.22E-06
0.0013	2.362	1587.	17.	3.23E-06
0.0007	2.403	1586.	21.	1.36E-06
0.0014	2.428	1585.	24.	2.40E-06
-0.0001	2.460	1585.	28.	-1.49E-07
0.0014	2.442	1585.	26.	2.25E-06
0.0006	2.510	1583.	33.	7.47E-07
0.0	2.368	1587.	17.	0.0
0.0004	2.308	1588.	10.	1.59E-06
0.0007	2.252	1588.	4.	7.54E-06
0.0018	2.244	1588.	3.	2.58E-05
-0.0000	2.582	1580.	41.	-5.76E-12
-0.0003	2.675	1576.	51.	-2.43E-07
0.0032	2.700	1575.	53.	2.46E-06
0.0027	2.620	1579.	45.	2.47E-06
0.0026	2.637	1578.	47.	2.29E-06
0.0014	2.655	1575.	53.	1.09E-06
0.0032	2.552	1582.	38.	3.50E-06
0.0005	2.539	1582.	36.	5.68E-07
0.0019	3.416	1512.	120.	5.98E-07
0.0	2.442	1585.	26.	0.0
-0.0007	2.525	1583.	35.	-8.31E-07
0.0015	2.392	1586.	20.	3.09E-06
0.0008	2.351	1587.	15.	2.15E-06
0.0015	3.415	1512.	120.	4.72E-07
0.0042	2.561	1581.	39.	4.48E-06
0.0009	2.643	1578.	47.	7.82E-07
0.0011	2.695	1575.	53.	8.56E-07
0.0001	2.674	1576.	51.	8.12E-08
-0.0004	2.623	1579.	45.	-3.64E-07
0.0012	3.286	1527.	109.	4.25E-07
0.0013	2.777	1570.	61.	8.69E-07
0.0013	2.908	1562.	74.	7.10E-07
0.0010	2.930	1560.	76.	5.30E-07
0.0013	3.320	1523.	112.	4.46E-07
0.0008	2.854	1565.	69.	4.72E-07
0.0006	2.821	1568.	65.	3.73E-07
0.0013	2.763	1571.	60.	8.90E-07
0.0003	2.834	1567.	67.	1.83E-07
0.0006	2.932	1560.	76.	3.17E-07
0.0006	2.959	1558.	79.	3.06E-07
-0.0013	2.893	1563.	72.	-7.25E-07
0.0016	2.927	1560.	76.	8.51E-07
0.0014	2.951	1558.	78.	7.21E-07
0.0000	2.905	1562.	73.	2.09E-12
0.0038	2.959	1558.	79.	1.94E-06
0.0031	3.028	1552.	85.	1.45E-06
0.0031	3.090	1547.	91.	1.35E-06
-0.0002	3.171	1539.	98.	-7.96E-08
0.0027	3.112	1545.	93.	1.14E-06
0.0010	3.140	1542.	95.	4.11E-07
0.0003	3.285	1527.	109.	1.06E-07

Continuation of PB Driver Ell-07, Compact 16

-0.0005	3.038	1551.	86.	-2.31E-07
-0.0001	3.091	1547.	91.	-4.34E-08
-0.0001	3.032	1552.	86.	-4.65E-08
0.0008	3.195	1537.	100.	3.10E-07
0.0032	3.202	1536.	101.	1.23E-06
0.0008	3.223	1534.	103.	3.02E-07
-0.0007	3.173	1539.	98.	-2.78E-07
0.0024	3.320	1523.	112.	8.24E-07
-0.0002	3.150	1537.	100.	-7.80E-08
0.0018	3.244	1532.	105.	6.65E-07
0.0022	3.280	1528.	108.	7.84E-07
0.0004	3.255	1530.	106.	1.46E-07
0.0011	3.298	1526.	110.	3.86E-07
0.0013	3.316	1524.	111.	4.48E-07
0.0018	3.347	1520.	114.	6.03E-07
0.0010	3.344	1521.	114.	3.36E-07
-0.0000	3.416	1512.	120.	-2.70E-12

PB DRIVER E11-07, COMPACT 18, 5.25/1 (TH,U)C2, 701 DAYS
 $T(K)=1574.-29.(X**2-RIN**2(1+2LN(X/RIN)))$
 RATIO, FREE VOLUME/ OXIDE VOLUME= 1.74
 INITIAL BURNUP, PERCENT FIMA= 0.0 DAYS= 701.0
 FINAL BURNUP, PERCENT FIMA= 8.53 ,RIN= 2.22CM

Y, CM	X, CM	T, K	K/CM	KMC
0.0024	3.444	1499.	116.	7.71E-07
-0.0009	2.244	1574.	3.	-1.34E-05
0.0004	2.301	1574.	9.	1.79E-06
0.0017	2.366	1573.	16.	4.27E-06
0.0010	2.296	1574.	9.	4.77E-06
0.0012	2.242	1574.	3.	1.95E-05
0.0	2.245	1574.	3.	0.0
0.0002	2.246	1574.	3.	2.76E-06
0.0008	2.302	1574.	9.	3.54E-06
-0.0006	2.247	1574.	3.	-7.97E-06
0.0008	2.290	1574.	8.	4.13E-06
0.0008	2.248	1574.	3.	1.02E-05
0.0018	2.240	1574.	2.	3.22E-05
0.0010	3.466	1497.	117.	3.15E-07
-0.0003	2.380	1573.	18.	-6.90E-07
0.0004	2.365	1573.	16.	1.01E-06
0.0011	2.436	1571.	24.	1.89E-06
0.0005	2.493	1570.	30.	6.87E-07
0.0039	3.415	1503.	113.	1.28E-06
0.0029	2.524	1569.	33.	3.59E-06
0.0007	2.580	1567.	38.	7.38E-07
0.0007	2.653	1564.	46.	6.19E-07
0.0000	2.532	1569.	34.	1.15E-11
0.0025	2.493	1570.	30.	3.43E-06
0.0017	2.477	1570.	28.	2.47E-06
-0.0008	2.512	1569.	32.	-1.03E-06
0.0	3.393	1505.	111.	0.0
0.0020	2.654	1564.	46.	1.77E-06
0.0012	2.682	1562.	48.	9.98E-07
0.0007	2.623	1565.	43.	6.63E-07
-0.0003	2.683	1562.	49.	-2.49E-07
0.0003	2.616	1565.	42.	2.89E-07
0.0015	2.563	1568.	37.	1.66E-06
0.0013	2.743	1559.	54.	9.61E-07

Continuation of PB Driver Ell-07, Compact 18

0.0016	2.753	1559.	55.	1.16E-06
0.0004	2.800	1556.	60.	2.68E-07
0.0011	2.854	1553.	65.	6.77E-07
0.0010	3.345	1510.	107.	3.51E-07
0.0009	2.738	1560.	54.	6.71E-07
0.0031	2.795	1556.	59.	2.09E-06
0.0041	2.848	1553.	64.	2.55E-06
0.0002	2.812	1555.	61.	1.31E-07
0.0008	2.980	1544.	76.	4.14E-07
0.0026	3.053	1538.	83.	1.23E-06
0.0025	3.092	1535.	86.	1.13E-06
0.0018	3.323	1513.	106.	6.44E-07
0.0028	3.043	1539.	82.	1.34E-06
0.0024	3.022	1541.	80.	1.18E-06
0.0011	3.193	1526.	95.	4.46E-07
0.0031	3.010	1542.	79.	1.54E-06
0.0022	2.900	1550.	69.	1.27E-06
0.0025	2.933	1547.	72.	1.37E-06
0.0009	3.154	1529.	91.	3.80E-07
0.0038	3.087	1535.	86.	1.73E-06
0.0032	3.225	1523.	97.	1.26E-06
0.0025	3.091	1535.	86.	1.13E-06
0.0011	3.088	1535.	86.	5.00E-07
0.0028	3.142	1530.	90.	1.20E-06
0.0002	3.268	1518.	101.	7.54E-08
0.0027	2.969	1545.	75.	1.42E-06
0.0011	3.024	1540.	80.	5.39E-07
0.0019	3.168	1528.	93.	7.91E-07
0.0039	3.162	1529.	92.	1.63E-06
0.0016	3.352	1510.	108.	5.57E-07
0.0027	3.153	1529.	91.	1.14E-06
0.0022	3.322	1513.	106.	7.88E-07
0.0018	3.448	1499.	116.	5.76E-07
0.0015	3.470	1496.	118.	4.71E-07

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