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FISSION GAS RELEASE FROM OXIDE FUELS AT HIGH BURNUPS

(AWBA Development Program)

FEBRUARY 1981

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BETTIS ATOMIC POWER LABORATORY
WEST MIFFLIN, PENNSYLVANIA

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(AWBA DEVELOPMENT PROGRAM)

C. C. Dollins

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FOREWORD

The Shippingport Atomic Power Station located in Shippingport, Pennsylvania was the first large-scale, central-station nuclear power plant in the United States and the first plant of such size in the world operated solely to produce electric power. This program was started in 1953 to confirm the practical application of nuclear power for large-scale electric power generation. It has provided much of the technology being used for design and operation of the commercial, central-station nuclear power plants now in use.

Subsequent to development and successful operation of the Pressurized Water Reactor in the Atomic Energy Commission (now Department of Energy, DOE) owned reactor plant at the Shippingport Atomic Power Station, the Atomic Energy Commission in 1965 undertook a research and development program to design and build a Light Water Breeder Reactor core for operation in the Shippingport Station.

The objective of the Light Water Breeder Reactor (LWBR) program has been to develop a technology that would significantly improve the utilization of the nation's nuclear fuel resources employing the well-established water reactor technology. To achieve this objective, work has been directed toward analysis, design, component tests, and fabrication of a water-cooled, thorium oxide-uranium oxide fuel cycle breeder reactor for installation and operation at the Shippingport Station. The LWBR core started operation in the Shippingport Station in the Fall of 1977 and is expected to be operated for about 4 to 5 years or more. At the end of this period, the core will be removed and the spent fuel shipped to the Naval Reactors Expended Core Facility for a detailed examination to verify core performance including an evaluation of breeding characteristics.

In 1976, with fabrication of the Shippingport LWBR core nearing completion, the Energy Research and Development Administration, now DOE, established the Advanced Water Breeder Application (AWBA) program to develop and disseminate technical information which would assist U.S. industry in evaluating the LWBR concept for commercial-scale application. The program is exploring some of the problems that would be faced by industry in adapting technology confirmed in the LWBR program. Information being developed includes concepts for commercial-scale prebreeder cores which would produce uranium-233 for light water breeder cores while producing electric power, improvements for breeder cores based on the technology developed to fabricate and operate the Shippingport LWBR core, and other information and technology to aid in evaluating commercial-scale application of the LWBR concept.

All three development programs (Pressurized Water Reactor, Light Water Breeder Reactor, and Advanced Water Breeder Applications) are under the technical direction of the Office of the Deputy Assistant Secretary for Naval Reactors of DOE. They have the goal of developing practical improvements in the utilization of nuclear fuel resources for generation of electrical energy using water-cooled nuclear reactors.

Technical information developed under the Shippingport, LWBR, and AWBA programs has been and will continue to be published in technical memoranda, one of which is this present report.

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ABSTRACT

Fission Gas Release from Oxide Fuels at High Burnups

(AWBA Development Program)

C. C. Dollins

The steady state gas release, swelling and densification model previously developed for oxide fuels has been modified to accommodate the slow transients in temperature, temperature gradient, fission rate and pressure that are encountered in normal reactor operation. The gas release predictions made by the model were then compared to gas release data on LMFBR-EBR-II fuels obtained by Dutt and Baker and reported by Meyer, Beyer, and Voglewede. Good agreement between the model and the data was found. A comparison between the model and three other sets of gas release data is also shown, again with good agreement.

Fission Gas Release from Oxide Fuels at High Burnups
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Introduction

When fission occurs, inert gas atoms are produced. In the design of reactors containing rod-type fuel elements it is necessary to know how much of this gas is retained in the fuel and how much escapes to the fuel rod plenum and the gap between the fuel and cladding. Gas which escapes from the fuel increases the temperature at which the fuel operates due to a decrease in thermal conductivity across the fuel-clad gap and increases the gas pressure inside the rod. These effects can potentially result in excessive clad strains during normal operation or unacceptable flow blockage due to clad rupture or clad ballooning in the case of a LOCA. Thus, if the amount of gas that escapes from the fuel is seriously underestimated, melted fuel and clad ruptures can potentially occur, resulting in unacceptable contamination in the primary coolant. If, on the other hand the amount of gas escaping the fuel is overestimated, then the fuel may not be used to its safe full potential.

The steady state swelling and gas release model recently developed⁽¹⁾ has been modified to account for changes in temperature, temperature gradient, pressure, and fission rate with time. These were program changes and do not involve changes to the theory.

The procedure to include the effect of changes in the variables during the fuel history is a straightforward modification of the previous procedure which did not permit such changes. Both programs operate on the basis of the effect of an increment in time.

As an example the calculation of the migration of gas out of a grain of material involves the solution of a modified form of Fick's second law. There is a spatial distribution of mesh points radially distributed within the grain. The concentration of gas at each mesh point is calculated at each time step. If the temperature is changed then the gas atom diffusion coefficient is changed and the calculation proceeds as in the previous time step. A change in fission rate results in a change in the diffusion coefficient as explained in Reference (1). A change in pressure would also result in an instantaneous change in the diffusion coefficient as stated in Reference (1).

Calculations using the modified model have been compared to measurements of fission gas release for LMFBR rods whose data were obtained by Dutt and Baker⁽²⁾ and reported by Meyer et al⁽³⁾. The rods contained UO_2 -25 w/o PuO_2 fuel with densities from 93% to 97% TD. The grain size was not quoted. The temperature and fission rate histories for the rods for which comparisons could be made were obtained from Meyer et al⁽⁴⁾.

Theory

The gas release model previously described⁽¹⁾ has been modified to accommodate changes in temperature, pressure, fission rate and temperature gradient with burnup.

The original model assumed that fission gas migrates to the grain faces either as isolated atoms or in bubbles. From the grain faces the gas was assumed to migrate to the intersection between three grains and from there to grain corners (the intersection of four grains). It was also assumed that the gas remained at the corners until the swelling due to gas at the corners and along the grain boundary edges reaches 5% of the total volume at which time tunnels form venting the gas to the surface. It was assumed that once the tunnels formed they remained throughout life.

Calculations using the model of Reference (1) indicate that very little gas remains at the grain boundary edges; consequently, in the modified version it is assumed that once the gas leaves the grain faces it goes directly to the grain corners.

The modified version also allows for the tunnels to pinch off once the gas escapes and for the resulting pores which formed from the tunnels to dissolve. This change is necessary to account for the case where the fuel is taken from a high temperature (where it is easy for the tunnels to form) to a low temperature (where they would normally not form). Both models predict the same gas release under steady state conditions, as expected. This provides a cross check on the two calculation procedures.

Data Comparisons

Comparisons have been made between predictions made with the modified model and the Dutt and Baker data. The density, temperature, fission rate and burnups for the Dutt and Baker data used in the analysis are shown in Table I. A grain size of 10 microns was assumed. Both the measured and predicted gas releases are given in the Table. Figure 1 is a plot of the predicted gas release versus the observed gas release for the Dutt and Baker data. The model is in generally good agreement with the data; however, in some cases too little gas is predicted to escape. Comparisons were also made between gas release predictions obtained by the model and gas release measurements made by Zimmermann⁽⁵⁾ on $\text{UO}_2\text{-PuO}_2$ fuels. Zimmermann divided his specimens into three groups depending on the final average fuel temperature. Zimmermann's specimens cooled during operation. Maximum temperatures for Zimmermann's specimens were obtained from the quoted power levels and conductivity data reported by Babcock and Wilcox⁽⁶⁾. The conductivity was modified for the inclusion of PuO_2 in the UO_2 and for the low density (88%) of the Zimmermann

specimens using techniques described by Olander⁽⁷⁾. The temperatures calculated using the maximum power conditions were averaged with the cooler end of life temperatures given by Zimmerman. A grain size of $10\mu\text{m}$ was assumed for Zimmermann's specimens. The predictions (solid curve) for the specimens with end of life mean temperature of 1000°C are shown in Figure 2 along with Zimmermann's measurements. The 1250 and 1500°C curves are shown in Figures 3 and 4 respectively. Comparisons between the gas release predicted by the Bettis unmodified model, which gives the same gas release as the modified model for fuel operated under constant conditions, and data obtained by Lewis⁽⁸⁾ and Hilbert et al⁽⁹⁾ were made in the previous work⁽¹⁾. Figure 5 is a plot of the predicted gas release versus observed gas release for the three sets of data from References (5), (8) and (9). Lewis' fuel was UO_2 with an initial density above 96% TD. No grain size was given. The data of Hilbert et al were obtained on UO_2 with densities of 90 or 97% TD. The grain sizes varied from 10 to $50\mu\text{m}$.

Discussion

Figure 6 is a plot of gas release versus burnup for burnups greater than 20,000 MWd/t. The solid curves are the predictions for the author's model⁽¹⁾. The curves were obtained at fixed temperatures. The bottom curve is for 1100°C and the top curve for 1500°C . The intermediate curves are for various specific temperatures between 1100 and 1500°C .

The Bettis model recognizes that at low temperatures less gas is released even at high burnups as shown by the 1000°C volume average temperature results of Zimmermann which has about 70% gas release at burnups as high as 114,000 MWd/t, Figure 2. A 1000°C volume average temperature specimen has a large portion of the fuel operating at much higher temperatures. The gas release results in Figure 6 assume that the entire specimen is at one temperature.

The Dutt and Baker data had an end of life average surface temperature of about 820°C . Commercial LWR cores have end of life surface temperatures about 300°C cooler since water is used as a coolant rather than liquid metal. The fact that so much of the fuel in LWR's near the pellet surface is at a much cooler temperature than in LMFBR's (Dutt and Baker) means that a good portion of the fuel would have gas release below the bottom curve of Figure 6. This will tend to keep the overall gas release in LWR's lower than for LMFBR fuel. For example, the Bettis model predicts 64% release from UO_2 operated to a burnup of 100,000 MWd/t at a surface temperature of 538°C (1000°F) and a center temperature of 2204°C (4000°F). Knowledge of gas release at high burnups can have a big impact on designs with higher burnups, for example the annuli of EPRI dual region fuel pellets⁽¹⁰⁾.

Conclusions

The Bettis model for gas release agrees well with data obtained by Hilbert et al and Lewis at low burnups and with the data of Dutt and Baker obtained at high burnups and high temperatures. The model also agrees well with the low temperature and high burnup data obtained by Zimmermann. The model will be compared to other data as they become available.

References

- (1) C. C. Dollins and F. A. Nichols, "Swelling and Gas Release in UO_2 at Low and Intermediate Temperatures", J. Nucl. Mater., 66, 143 (1977).
- (2) D. S. Dutt and R. B. Baker, "SIEX A Correlated Code for the Prediction of Liquid Metal Fast Breeder Reactor (LMFBR) Fuel Thermal Performance", HDL-TME 74-55, 1975.
- (3) R. O. Meyer, C. E. Beyer, and J. C. Voglewede, "Fission Gas Release from Fuel at High Burnup", Nuclear Safety, 19, 699 (1978).
- (4) R. O. Meyer, C. E. Beyer, and J. C. Voglewede, "Fission Gas Release From Fuel at High Burnup," NUREG-0418, March 1978.
- (5) H. Zimmermann, "Fission Gas Behavior in Oxide Fuel Elements of Fast Breeder Reactors", Nucl. Tech. 28, 127 (1976).
- (6) "Babcock and Wilcox Standard Nuclear Steam System (B-SAR-205, Vol 1)", February 27, 1976, DOCKET-STN-50-561.
- (7) D. R. Olander, "Fundamental Aspects of Nuclear Reactor Fuel Elements", ERDA, 1976.
- (8) W. B. Lewis, "Engineering for the Fission Gas in UO_2 Fuel", Nucl. Appl. 2, 171 (1966).
- (9) R. F. Hilbert, V. W. Storhok, W. Chubb, and D. L. Keller, "Mechanisms of Swelling and Gas Release in Uranium Dioxide", J. Nucl. Mater., 38, 26 (1971).
- (10) A. Roberts and H. Ocken, "Improving Nuclear Fuel Reliability", EPRI Journal, Vol. 3, No. 8, p. 75 (1978).

TABLE I

Parameters Used in Making Comparisons
with the Dutt and Baker Data

Specimen Number	Density % TD	Fission Rate f/cm ³ sec (x10 ¹³)	Burnup f/cm ³ (x10 ²⁰)	Temperature (°C)		Measured Gas Release %	Predicted Gas Release %
				Surface	Center		
PNL 3-20	96.2	2.7	6.6	670	1000	39.1	24.4
			8.8	790	1180		
			12	930	1360		
PNL 3-23	95.9	2.8	6.7	666	962	1.8	1.1
			7.3	694	1016		
PNL 4-1	94.6	5.1	1.9	770	1700	62.2	55.8
			3.9	735	1640		
			5.8	750	1645		
			7.7	790	1700		
			11.2	845	1760		
PNL 4-20	93.6	4.3	1.9	760	1450	63.8	41.9
			5.7	725	1390		
			7.6	780	1460		
			9.5	850	1550		
			11.	880	1570		
PNL 4-26	97.	4.8	1.9	770	1700	64.5	56.2
			5.7	750	1680		
			7.6	790	1720		
			9.5	830	1765		
			11.	840	1770		
PNL 4-34	95.3	4.6	1.8	815	1705	65.4	66.5
			5.5	815	1705		
			7.4	855	1745		
			9.2	940	1835		
			10.7	965	1855		
PNL 7-10	95.2	5.	6.6	850	1710	30.1	51.3

TABLE I (Continued)

Specimen Number	Density % TD	Fission Rate f/cm ³ sec (x10 ¹³)	Burnup f/cm ³ (x10 ²⁰)	Temperature (°C)		Measured Gas Release %	Predicted Gas Release %
				Surface	Center		
PNL 8-11	93.5	6.9	2.5	830	1960	92.2	70.6
			7.6	770	1900		
			10.2	870	2120		
			12.7	870	2110		
			15	870	2090		
PNL 8-25	93.8	6.8	2.6	810	1980	94.3	67.6
			7.7	790	1920		
			10.2	850	2030		
			12.8	850	2020		
			14.8	850	2010		
PNL 8-28	93.9	6.5	2.5	770	1980	62.6	61.8
			5.	740	1920		
			7.5	800	1990		
			14.6	820	1370		
PNL 8-37	93.6	6.6	7.4	770	1910	65.2	61.9
			14.3	820	1890		
PNL 8-38	N.A.	6.7	7.4	770	1900	53.6	55.9
PNL 9-30	N.A.	3.	3	760	1380	53.8	56.6
			6	710	1320		
			9.1	760	1400		
			12.7	930	1640		
PNL 10-23	N.A.	4.8	2.3	860	1840	67.5	65.1
			6.8	850	1790		
			13.1	890	1840		
PNL 10-63	N.A.	4.9	13.6	840	1825	71.5	62

TABLE I (Continued)

Specimen Number	Density % TD	Fission Rate $f/cm^3 \text{ sec}$ ($\times 10^{13}$)	Burnup f/cm^3 ($\times 10^{20}$)	Temperature ($^{\circ}\text{C}$)		Measured Gas Release %	Predicted Gas Release %
				Surface	Center		
PNL 17-6	N.A.	5.4	0.9	965	1860	58.1	64.5
			5.1	900	1770		
			8.4	880	1950		
PNL 17A-5	N.A.	6.7	0.25	1000	2190	32.6	49.4
			0.5	930	2110		
			0.75	870	2030		
			0.9	820	1960

FIGURE 1

Comparisons between the gas release predictions of the Bettis model and the measured results of Dutt and Baker.

FIGURE 2. COMPARISON BETWEEN THE MODEL PREDICTIONS, AND ZIMMERMAN'S DATA WITH END OF LIFE MEAN TEMPERATURE OF 1000°C

FIGURE 3. COMPARISON BETWEEN THE MODEL PREDICTIONS, AND ZIMMERMAN'S DATA WITH END OF LIFE MEAN TEMPERATURE OF 1250°C

FIGURE 4 COMPARISON BETWEEN THE MODEL PREDICTIONS, AND ZIMMERMAN'S DATA WITH END OF LIFE MEAN TEMPERATURE OF 1500°C

Figure 5

Comparison between the gas release predictions of the Bettis model and the measured results from References (5), (8), and (9).

FIGURE 6 PREDICTED GAS RELEASE VERSUS BURNUP FOR FUEL OPERATING AT CONSTANT TEMPERATURE ACCORDING TO THE BETTIS MODEL THE TEMPERATURES ARE SHOWN