## **High Burnup Effects Program**

# A State-of-the-Technology Assessment

June 1982

Prepared for the Sponsors of the High Burnup Effects Program



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**High Burnup Effects Program** 

# A State-of-the-Technology Assessment

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Program Manager M. D. Freshley

lune 1982

This report was prepared for the sponsors of the High Burnup Effects Program in May 1979. It represents an assessment of the state-of-the-technology related to high burnup fission gas release from urania, light water reactor fuel as it existed in the open literature at that time. This assessment formed the basis for the research currently being conducted in the HBEP.

This report was prepared for the U.S. Department of Energy, which is one of the sponsors of the work, by Pacific Northwest Laboratories of Battelle Memorial Institute.

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#### SUMMARY

As a part of Task 1 of the High Burnup Effects Program, a report titled "High Burnup Effects, A State-of-the-Technology Assessment" was prepared to provide an updated evaluation of the literature as it pertains to high burnup effects in Zircaloy-clad  ${\rm UO_2}$  fuels used in light water reactors.

Because fission gas release is one of the major concerns related to high burnup operation, this assessment was made with emphasis on obtaining well-characterized data which could possibly be used to expand the data base and aid in developing a correlation that describes the effects of fuel temperature and burnup (above 20 GWd/MTM) on fission gas release.

To provide background for the assessment, various analytical models and empirical correlations describing the fission gas release phenomenon were examined. The dates of the analytical models range from 1957 to 1978. These models were categorized into four types: the atomic diffusion models, which described fission gas release by the mechanism of gas atom diffusion; the modified atomic diffusion models, which considered temperature gradient in addition to concentration gradient as the driving force for gas atom diffusion; the bubble migration models, which introduced the concept of bubble migration to replace atomic diffusion; and the transient gas release models, which predicted fission gas behavior during transient conditions. The specific features and capabilities of each model were discussed and compared.

The dates of the empirical correlations range from 1964 to 1979. These correlations were categorized into three types: the temperature-dependent correlations; the burnup and/or linear heat generation rate-dependent correlations; and the correlations for fission gas release at low temperature and/or burnup. The data bases used in developing and presenting each correlation were discussed and compared. As a burnup-dependent correlation for fission gas release, the NRC correlation was also examined in terms of its data base, derivation, and application.

One of the major parts of the state-of-the-technology assessment of

high burnup effects was to survey the existing high burnup irradiation data, with emphasis on fission gas release behavior. An extensive literature search was made, from which a total of 91 journal articles and technical reports were reviewed. In order to select those which were most pertinent to the interest of the High Burnup Effects Program, the following criteria were used:

- Burnup levels above 20 GWd/MTM,
- Fuel type  $U0_2$  and mixed oxide only,
- Fuel form pelletized only, and,
- Fission gas release experimental data required.

Application of these criteria resulted in identification and evaluation of thirteen sources.

The data were obtained from a wide variety of fuel types and irradiation conditions. This resulted in a wide range of both fission gas release, <1 to 100%, and apparent burnup dependencies. The wide variations in fuel rod design and irradiation conditions required a division of the data in order to make meaningful comparisons and correlations.

The data sets were divided into two groups on the basis of the fuel diameters and fission rate. The first group comprised data obtained from fuels with diameters less than 7 mm and fission rates greater than  $3 \times 10^{13}$  fission/cm<sup>3</sup>-sec. The second group, with diameters greater than 7 mm and fission rates less than  $4 \times 10^{13}$  fissions/cm<sup>3</sup>-sec., are more representative of LWR fuel designs and irradiation conditions. Thus, this grouping allows comparison of the data from various fuels with temperatures, temperature gradients, and fission rates that are typical of LWR conditions.

Due to the high fission rates, the majority of the high burnup data available are from the first group of specimens. All of the data sets from this group show evidence of burnup-enhanced fission gas release. The apparent burnup dependence is sensitive to the parameter selected to make the correlation and, by using the LHGR, leads to a high apparent burnup dependence. This is especially true at low LHGR which is attributed to the thermal feedback effect. A temperature related parameter is considered to be more appropriate than the LHGR, but sufficient information

is not readily available to determine selection of the proper parameter.

The data from the second group of data sets, i.e., those data more representative of LWR irradiations, showed little direct evidence for burnup-enhanced fission gas release. Comparisons of the data sets on the basis of temperature was not possible because the temperatures or the necessary information to calculate temperatures were not provided in most of the reports. Also, meaningful correlations based on LHGR were not possible due to the wide variety of LHGRs that were reported.

The gas release data from  $\rm UO_2$  fuels irradiated at less than 350 W/cm (peak LHGRs) were compared with the predictions of GAPCON-THERMAL-2 computer code both with and without the NRC correction factor. The experimental data exhibited such a large amount of scatter that no conclusion could be made as to the form or extent of a burnup dependence. There is not sufficient information regarding the fuel, the fuel rod design, or the irradiation conditions to determine the reason for this large amount of scatter.

In summary, the current experimental data regarding fission gas release from fuels irradiated under conditions typical of light water reactors are not adequate to define the effects of burnup on fission gas release. There exists a large amount of scatter in the data and the information necessary to evaluate the data have not been reported. Consequently, additional information as well as well-characterized experimental data at higher burnups are needed before the effects of burnup on fission gas release in LWR fuels can be established.

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#### INTRODUCTION

The objective of the High Burnup Effects Program is to provide information on the high burnup (60 to 65 GWd/MTM) behavior of zircaloy-clad  $\rm UO_2$  LWR fuel with emphasis on obtaining well-characterized data and on developing a correlation that describes the effect of fuel temperature on fission gas release during irradiation.

The program is organized into three tasks - Task 1, High Burnup Effects Evaluations; Task 2, Fission Gas Sampling; and Task 3, Parameter Effects Study. Task 1, included 1) an updated evaluation of the current state-of-the-technology on high burnup effects, 2) an assessment of the relevant data reported in the literature and identification of the data needs, 3) an evaluation of existing irradiated fuel rods and data, some of which have been made available for use in this program, and 4) the development of a program plan for Tasks 2 and 3. This document presents the results of the state-of-the-technology evaluation, an assessment of the relevant data, and identification of the data needs. The evaluation of existing rods and data and the program plan are presented in the program plan document.

The advantage of extended burnup has become more attractive recently for political, economical and environmental reasons, among which the most significant one is the increased utilization of uranium resources. However, associated with extended burnup is the probability of an increased cost penalty caused by premature fuel failure. Because fission gas release is one of the major concerns related to high burnup operation, this state-of-the-technology assessment examined the various analytical models and empirical correlations describing the fission gas release phenomenon. Comparisons of the parameters considered in the analytical models and the variables used in the empirical correlations were made.

With this background information, an evaluation was made of the current pertinent experimental data on the subject of high burnup fission gas release. Data reported by individual investigators were compared and evaluated in relation to their applicability to the content and scope of the High Burnup Effects Program. These evaluations then form the bases for defining the data needs, and the selection of variables to be studied in this program.

The twenty-five participants who supported the Task 1 effort are:

Babcock-Brown Boveri Reaktor Babcock and Wilcox Belgonucleaire British Nuclear Fuels Limited Central Research Institute of Electric Power Industry Centre d'Etude de l'Energie Nucleaire Combustion Engineering, Incorporated Comitato Nazionale per l'Energie Nucleaire Department of Energy, U.S. Government **Electric Power Research Institute** Exxon Nuclear Company, Incorporated Framatome General Electric Company Hitachi, Limited Japan Atomic Energy Research Institute Kraftwerk Union Aktiengesellschaft Mitsubishi Heavy Industry, Limited Netherlands Energy Research Foundation Nuclear Fuels Industry, Limited Risø National Laboratory Studsvik Energiteknik AB Swiss Federal Institute for Reactor Research Toshiba Corporation Technical Research Center of Finland

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# BACKGROUND SUMMARY - ANALYTICAL MODELS DESCRIBING THE FISSION GAS RELEASE PHENOMENON

Early efforts to explain the fission gas release phenomenon in  $\rm UO_2$  fuels were based mainly on the assumption that fission gas atoms migrate by diffusion. Later models, some developed with the aid of experimental evidence, provide more detailed descriptions of the generation and movement of fission gas atoms or bubbles. Recent analytical models further attempt to describe the life history of a fission gas bubble from its nucleation, distribution, migration, to trapping and resolution. Temperature effects and transient conditions are also considered.

A brief summary of the various types of analytical models describing the fission gas release phenomenon is presented below. For a more extensive review, we shall refer to the 1977 report prepared by investigators at UCLA (Baldewicz).

#### ATOMIC DIFFUSION MODELS

Gas atom diffusion was first used to interpret fission gas release by Booth in 1957. In this model the  $\rm UO_2$  fuel is replaced by a number of equivalent spheres. Fission gas is assumed to be generated in the sphere, then it migrates to the surface, and diffuses out of the surface (boundary).

Speight (1969) applied the basic features in Booth's model to describe the rate-controlling process of fission gas release, with one additional mechanism: a resolution process in which fission gas atoms can be ejected from the boundary to the interior of the sphere, or captured by immobile fission gas bubbles existing in the intergranular regions (among spheres).

A later model developed by Ronchi and Matzke (1972) made similar assumptions to those of the Speight model. However, Ronchi and Matzke assumed that the sphere boundary acted only as a sink for fission gas atoms; the model did not permit ejection of fission gas atoms to induce resolution.

MacEwan and Stevens (1964) observed an apparent decrease in diffusion coefficient with increasing fission rate and exposure. They interpreted their results in terms of defect traps that immobilized the fission gas atoms. The fraction of atoms trapped was an exponential function of burnup (fiss/cm $^3$ ), while the remaining untrapped fraction of atoms diffused out of the fuel according to the Booth model.

The most recent model that considers atomic diffusion the controlling mechanism was developed by Hargreaves and Coolins (1976). They assumed that resolution of the fission gas from grain boundary bubbles generated a high concentration of fission gas atoms next to the boundaries. These atoms in turn acted as a barrier against further fission gas release through the boundary by random atomic diffusion, until some minimum burnup was reached.

#### MODIFIED ATOMIC DIFFUSION MODELS

Several investigators recognized that in order to successfully predict fission gas release behavior, not only concentration gradient but also temperature gradient should be the driving force for atomic diffusion. Yuill, Baston and McFadden (1971) considered a "Gibbs-free-energy gradient" in their model. This energy gradient is affected by both the temperature gradient and the concentration gradient used in the Booth model.

Carroll, Perez, and Sisman (1965) performed sweep-gas experiments which showed that the principal release at low temperatures (<600°C) was by a knock-out process rather than by direct recoil. Their results lead to a new area of analysis in which different models are used to describe fission gas behavior in different temperature regimes, such as the models developed by Hayns and Wood.

#### BUBBLE MIGRATION MODELS

In an irradiation experiment with  ${\rm UO}_2$  foils, Barnes and Mazey (1963) observed the apparent fission gas bubble nucleation, migration through a temperature gradient, and coalescence. This observation introduced the concept that fission gas release may be controlled by bubble behavior instead of atomic diffusion.

Using the Monte Carlo technique, Nichols and Warner (1971) developed the BUBL computer code model in which the nucleation, distribution, coalescence,

interaction with dislocations and grain boundaries, and thermal gradient migration of fission gas bubbles were considered. This model described fission gas behavior above 1200°C. In a parallel effort, Dollins and Ocken (1970) developed a similar model for lower temperatures, where fission gas behavior was assumed to be controlled by small bubble dynamics. Resolution was considered an important mechanism in the low temperature model.

More recently, Dollins and Nichols (1976) extended the range of analysis by these investigators to a new intermediate temperature of 1700°C, the columnar grain growth temperature. In this model, partial bubble destruction by fission-induced resolution is included.

A similar computer code, the GRASS model, was developed by investigators at Argonne National Laboratory (Rest, et al. 1976). According to the description of the GRASS model by Poeppel (1971), the basic features in GRASS are essentially the same as those in BUBL. However, the mechanisms of bubble nucleation, migration and coalescence are slightly different. Bubble diffusion is described by the evaporation-condensation process in addition to surface and volume diffusion.

#### TRANSIENT GAS RELEASE MODELS

The ability of the analytical models to predict fission gas behavior during transient conditions has also been investigated. Hayns and Wood (1976, 1977) developed two models to describe fission gas behavior in fast reactor fuel elements during steady state and transient conditions. Their first model accounts for the random migration, coalescence and resolution of gas atoms at low temperature. At higher temperature, their second model is used to account for biased migration, coalescence and resolution of gas bubbles under the thermal gradient driving force.

Computer code models with transient capabilities include GRASS-SST, FRAS and PFRAS. GRASS-SST (Rest, 1978) is the latest development of the GRASS code. While GRASS was originally developed for the prediction of fission gas behavior in LMFBR fuels during steady-power irradiation, GRASS-SST is designed to predict fission gas behavior in  $\rm UO_2$  fuels during both steady state and transient conditions.

Using the extrapolated power history of two irradiations in the H. B. Robinson reactor, GRASS-SST also predicted the fission gas release at high burnup (up to 8.5 at.%). Results for burnups >5 at.% and centerline temperatures <1200°C indicate that an enhanced release of fission gas occurs from fuel regions where swelling exceeds 7%, the assumed value for long-range porosity interconnection. This "breakaway" gas release occurs at higher burnups for rods with lower centerline temperatures.

FRAS was developed mainly for transient gas release and swelling in fast reactor oxide fuels (Gruber, 1974). Based on the finite difference analysis of the transient bubble distribution, this model calculates the effective bubble mobility, which in turn is used to calculate gas release at the grain boundaries by random and biased migration. A parametric representation of FRAS which requires considerably less computer time was given in the computer code PFRAS.

Other investigations of the transient gas release behavior include the temperature transient analyses by Turnbull and Tucker (1974) for  $\rm UO_2$ , the computer subroutine KURZZEIT intended for fast reactor fuels (Bogensberger and Ronchi, 1976), and the fast thermal transient analysis by Esteves (1975).

A summary of the major analytical models describing the fission gas behavior is presented in Table 1.

TABLE 1. Summary of Analytical Models for Fission Gas Release

| INVESTIGATORS  MODEL FEATURE                  | воотн           | McEWAN<br>AND<br>STEVENS | SPEIGHT         | NICHOLS<br>AND WARNER<br>(BUBL) | DOLLINS<br>AND<br>OCKEN | YUILL           | POEPPEL<br>(GRASS) | RONCHI<br>AND<br>MATZKE | GRUBER<br>(FRAS) | DOLLINS<br>AND<br>NICHOLS | HARGREAVES<br>AND<br>COLLINS | HAYNS<br>AND<br>WOOD | REST<br>(GRASS-SST) |
|---|-----------------|--------------------------|-----------------|---------------------------------|-------------------------|-----------------|--------------------|-------------------------|------------------|---------------------------|------------------------------|----------------------|---------------------|
| DATE  | 1957            | 1964                     | 1969            | 1970-71                         | 1970-73                 | 1971            | 1971               | 1972-76                 | 1974             | 1976                      | 1976                         | 1976-77              | 1978                |
| TYPE OF FUEL                                  | UO <sub>2</sub> | UO <sub>2</sub>          | U0 <sub>2</sub> | U0 <sub>2</sub>                 | U0 <sub>2</sub>         | uo <sub>2</sub> | MOX                | MOX                     | MOX              | UO <sub>2</sub>           | UO <sub>2</sub>              | MOX                  | UO <sub>2</sub>     |
| DRIVING FORCE<br>FOR DIFFUSION <sup>(a)</sup> | С               | С                        | С               | T                               | T                       | C,T             | T                  | С                       | Т                | T                         | С                            | С,Т                  | Ţ                   |
| DEFECT TRAPPING                               | NO              | YES                      | NO              | YES                             | NO                      | YES             | YES                | NO                      | NO               | NO                        | NO                           | NO                   | YES                 |
| BUBBLE NUCLEATION(b)                          |                 |                          | FSI             | FSI                             | Н                       | NO              | Н                  | H, FSI                  | FSI              | Н                         | NO                           | Н                    | Н                   |
| BUBBLE SIZE<br>DISTRIBUTION                   |                 |                          | FSI             | YES                             | FSI                     | NO              | YES                | FS1                     | YES              | NO                        | NO                           | YES                  | YES                 |
| BUBBLE MIGRATION(C)                           |                 |                          | NO              | D                               | R                       | D               | R, D               | NO                      | R, D             | R, D                      | NO                           | R, D                 | R, D                |
| BUBBLE RESOLUTION                             |                 |                          | YES             | NO                              | YES                     | NO              | YES                | NO                      | NO               | YES                       | YES                          | YES                  | YES                 |
| GAS LAW IN BUBBLES <sup>(d)</sup>             |                 |                          |                 | VDW                             | VDW                     | IDEAL           | нА                 | VDW                     | IDEAL            | VDW                       |                              | VDW                  | нА                  |
| BUBBLE DIFFUSION<br>MECHANISMS <sup>(e)</sup> |                 |                          |                 | S                               | S                       | S               | S, V, EC           |                         | S                | S                         |                              | S                    | S, V, EC            |
| TEMPERATURE RANGE (OC)                        | ANY             | ANY                      | ANY             | >1200                           | < 1200                  | ANY             | ANY                | ANY                     | ANY              | < 1700                    | ANY                          | ANY                  | ANY                 |
| TRANSIENT CAPABILITY                          | YES             | NO                       | NO              | NO                              | NO                      | NO              | YES                | NO                      | YES              | NO                        | NO                           | YES                  | YES                 |

<sup>(</sup>a) C = CONCENTRATION GRADIENT; T = TEMPERATURE GRADIENT

<sup>(</sup>b) H = HOMOGENEOUS NUCLEATION; FSI = FIXED SIZE INPUT FOR MODEL CALCULATIONS

<sup>(</sup>c) R = RANDOM; D = DIRECTIONAL

<sup>(</sup>d) HA = HARRISON'S EXTRAPOLATED EQUATION OF STATE, IN WHICH THE VAN der WAALS CONSTANT IS TEMPERATURE AND PRESSURE DEPENDENT; VDW = VAN der WAALS EQUATION OF STATE

<sup>(</sup>e) S = SURFACE DIFFUSION; V - VOLUME DIFFUSION; EC = EVAPORATION-CONDENSATION

# BACKGROUND SUMMARY - EMPIRICAL CORRELATIONS INTERPRETING THE FISSION GAS RELEASE DATA

Current analytical models describing fission gas release generally contain a number of submicroscopic parameters (fission gas resolution, fission fragment/gas bubble interaction, etc.) that are difficult to quantify. As a result, simplified empirical correlations have been developed (for use in the actual design and licensing of power reactors) to describe fission gas release as a function of various parameters that contribute to its generation and enhancement. The major contributing parameters, or independent variables in the empirical correlations, include temperature, linear heat generation rate and burnup.

In the following sections a brief summary of the various empirical correlations interpreting the fission gas release data is given. A comparison of some of the correlations can be found in the 1977 UCLA report by Baldewicz.

#### TEMPERATURE-DEPENDENT CORRELATIONS

The Hoffman and Coplin model (1964) is one of the earliest fission gas release models based entirely on experimental data. This model was developed to describe the then little known fission gas behavior at high temperature. Effects of grain growth, void migration, sintering, etc., on fission gas release from UO<sub>2</sub> fuels at volume-averaged temperatures up to 2000°C were studied. Fractional release was considered a nonlinear function of the volumetric average fuel temperature at the peak power position. No burnup dependence of fractional release was observed at burnup levels up to 11 GWd/MTU.

Another temperature-dependent correlation was developed later by Lewis (1966). In this model, data obtained from  $\mathrm{UO}_2$  fuels irradiated in reactors at Chalk River for 3 years were categorized into various fuel temperature zones between the centerline and the fuel surface. Fission gas release in each zone is proportional to the thermal-conductivity of the fuel integrated over the temperature range of the entire zone.

Notley (1970) described a computer program designed to predict the performance of  $\rm UO_2$  fuel elements irradiated in CANDU type reactors up to a burnup level of 10 GWd/MTU. In this program, experimental data on retained fission gas in  $\rm UO_2$  fuels up to 1800°C (Lewis, et al., 1964) were used as the basis to derive the temperature dependence of fission gas release, which in turn was used to predict fractional gas release as a function of time-averaged linear heat generation rate.

Cox and Homan (1970) studied the fission gas release data from mixed-oxide fuels in fast flux experiments to determine the effects of temperature, burnup, fuel fabrication form and power cycling. They suggested that, to a first approximation, gas release may be empirically represented by a function of temperature distribution. A 3-zone model was then developed, in which 98% of the fission gas was assumed to be released from fuels with centerline temperatures above 1800°C (columnar grain region), 50% from fuels between 1400 and 1800°C (equiaxed grain region) and 30% at temperatures below 1400°C.

In a review on BWR fuel design and experience, Williamson and Ditmore (1971) compared the fission gas release data given in several reports from General Electric Company to the 2-zone design model: 4% release for T <  $1650^{\circ}$ C, 100% for T  $\geq 1650^{\circ}$ C, where T is the maximum fuel rod volume-average temperature. The model predicts a greater fractional release than actual data from selected prototype fuel rods. However, for conditions of gross centermelt (maximum fuel centerline temperature around  $2800^{\circ}$ C), the predicted 60% fractional release agrees with reported data.

Recognizing the fact that imprecise temperature estimates are the major contributor to the large variance in reported gas release data, Beyer and Hann (1974) reviewed the extensive body of literature and selected a well-characterized set of in-reactor data for  $\rm UO_2$  fuels to construct an empirical correlation for fission gas release and fuel temperature. A multiple linear regression code was used to develop the Beyer and Hann model for fission gas release at high temperature (>1200°C). The resultant fractional release was the sum of fractional releases in three temperature zones: 1200 to 1400°C,

1400 to 1700°C and above 1700°C. For fuel temperatures below 1200°C, they developed a low temperature release model which was based on the Bellamy and Rich (1969) model.

The Beyer and Hann model was based on experimental data at high temperature and low burnup ( $\leq$ 18 GWd/MTM). No influence of burnup or fuel density on gas release was observed. However, the authors pointed out that at low temperature (<1200°C) and higher burnup levels (>20 GWd/MTM), experimental evidence indicated an increase in gas release with burnup and they included burnup dependence in their low temperature model.

#### BURNUP AND/OR LINEAR HEAT GENERATION RATE-DEPENDENT CORRELATIONS

In an experiment to study the high burnup effect, miniature  $\rm UO_2$  fuel pins were irradiated at low temperature in a heavy water reactor (DIDO, U.K.) to about 5% (44 GWd/MTM) burnup. Results indicate a marked increase in gas release above 3% burnup. Bellamy and Rich (1969) explained this phenomenon by the interconnection of grain boundary gas bubbles and the fracture under thermal stress of grain boundaries weakened by gas bubbles. They further interpreted the results by a linear formula : at maximum fuel centerline temperatures below 1250°C, the fractional gas release is 0.075%/1% burnup for burnup levels below 2%. However, as the burnup level increases to above about 3%, the fractional release becomes 2.0%/1% burnup.

Based on previously published data by investigators at General Electric Company and Idaho Nuclear Corporation, and some conservative assumptions, Baston, MacFadden and Yuill (1971) developed a fission product fuel model computer code (FPFM). In this model the fractional fission gas release during steady state operation is affected by fuel surface temperature, pellet diameter and linear heat generation rate. Least square fits were applied to establish the correlations between fractional release and linear heat generation rate, fuel surface temperature and thermal conductivity, and fuel radius and fractional release.

Dutt, et al. (1972), used regression techniques to best fit a set of test results from fast reactor fuels irradiated in the EBR-II reactor. In their correlated fission gas release model, the principal parameters are fuel microstructure, linear heat generation rate and burnup. The authors claimed that

the best correlation was obtained by assigning 100% fission gas release to all restructured (columnar and equiaxed grain growth) fuel, and describing the fractional release in the nonrestructured fuel by an exponential function of local linear heat generation rate and local burnup.

In a different parametric approach to interpret fission gas release data, Johnson and Hofman (1974) attempted to provide a simple alternative that used readily available design parameters (smeared density, fuel weight, cladding dimensions, peak linear power, etc.) to calculate fission gas release based on a 2-zone release model. A pseudo-isotherm of 1475°K (value empirically determined) was used as the dividing line between the "cold" and "hot" zones. In the "hot" zone, the amount of gas released is proportional to the amount produced. In the "cold" zone, however, the amount released is reduced by fuel smear density and temperature. The effect of these two factors on gas release is represented by Arrhenius functions.

Roberts, et al., (1977) studied the fuel modeling and performance of high burnup  ${\rm UO}_2$  and MOX fuel rods irradiated in various PWRs. Their fission gas release model was derived from experimental data at burnup levels up to 50 GWd/MTU. Fractional gas release is represented by F =  ${\rm k}_1$  +  ${\rm k}_2{\rm B}^2$ , where B is the fuel burnup and  ${\rm k}_1$ ,  ${\rm k}_2$  are empirical constants related to fuel temperature.

#### THE NRC CORRELATION

In view of the conclusion that "a high burnup enhancement of fission gas release has been recognized in the range of LWR fuel burnups," and that no existing model has described gas releases adequately at high burnup, the NRC developed a correction method to account for the high burnup effect in  $\rm UO_2$  fuels.

The Dutt and Baker correlation (1975), based on high burnup fission gas release data from LMFBR fuels irradiated in the EBR-II reactor, was used to derive the NRC correction function. By separating the variables (linear heat generation rate and burnup) in the Dutt and Baker correlation such that the fractional release was correlated with only the local burnup, a new function was derived. In this function, the burnup dependent fractional release is the sum of the fractional release at 20 GWd/MTU and the remaining fraction multiplied

by an enhancement factor. This enhancement factor is an exponential function of burnup above 20 GWd/MTU. Nonlinear regression techniques were used on Dutt and Baker's data to calculate the coefficients in the exponential function.

The fractional release at 20 GWd/MTU is temperature dependent. Although low temperature data from various sources have indicated very low releases ( $\leq 0.2\%$ ) at burnups as high as 20 GWd/MTU, for safety reasons the NRC requires that a conservative value of 1% for fractional release at low temperature and 20 GWd/MTU must be used by reactor vendors in licensing analyses.

A comparison of the fractional release predicted by the NRC correlation with measured values from high burnup  $\rm UO_2$  fuel rods irradiated in various LWRs indicates that, in general, the NRC correlation overpredicts fission gas release at high burnup. While there are pros and cons on using mixed-oxide data to predict  $\rm UO_2$  behavior (Baldewicz, 1977; ANS, 1977), the consensus is that more complete and high quality data are needed to describe fission gas behavior in  $\rm UO_2$  fuels at high burnup.

#### FISSION GAS RELEASE AT LOW TEMPERATURE AND/OR BURNUP

Empirical correlations developed for fission gas release at low temperature or burnup generally show no temperature and burnup dependence. Based on low temperature (<600°C) irradiation data from UO $_2$  fuels, Szuta (1975) developed a mathematical model in which the fractional release rate was a first order function of fission rate and decay constant. Friskney, et al. (1977) measured the fission gas release from monocrystalline UO $_2$  irradiated up to 1.64 GWd/MTM and used regression analysis to interpret fractional gas release as a trigonometric function of diffusion coefficients and decay constants of the fission gas isotopes and their immediate precursors. In a more recent article, Friskney and Turnbull (1979) reported that they had further extended the experiment to polycrystalline, "small" (20 to 40  $\mu$ m) and "large" (100 to 250  $\mu$ m) grain UO $_2$  fuels. The empirical Correlation developed for gas release in monocrystalline UO $_2$  was used to predict gas release in polycrystalline UO $_2$ . Very good agreement between measured and calculated results was obtained.

Fission gas release at low burnup ( $\leq$ 15 GWd/MTU) from UO $_2$  irradiated in an advanced gas cooled reactor has also been investigated recently (1979) by

Greatley and Hargreaves (1979) at Windscale, England. In their experiment a small capsule containing 3  $\rm UO_2$  pellets (3.5%  $^{235}\rm U$ ) was irradiated to 15 GWd/MTU with fuel centerline temperatures below 1170°C. Based on the measured fission gas release, the authors derived a correlation in which the fractional release was a first order function of the decay constant of the fission gas, total volume of the fuel, and the volume of fuel contributing to the knock-out release mechanism (outer zone of fuel).

A summary of the various empirical correlations for fission gas release is presented in Table 2.

TABLE 2. Empirical Correlations for Fission Gas Release (FGR)

| AUTHOR                                      | HOFFMAN<br>AND<br>COPLIN  | LEWIS                      | BELLAMY<br>AND<br>RICH   | NOTLEY                                       | COX<br>AND<br>HOMAN                                 | WILLIAMSON<br>AND<br>DITMORE                        | BASTON,<br>MacFADDEN<br>AND YUILL            | DUTT,<br>et. al.   | JOHNSON<br>AND<br>HOFMAN   | BEYER<br>AND<br>HANN                                | NRC   | ROBERTS                                       | FRISKNEY<br>AND<br>TURNBULL  | GREATLEY<br>AND<br>HARGREAVES   |
|---|---|----------------------------|--|--|---|---|--|--|--|---|---|---|--|---|
| DATE  | 1964  | 1966                       | 1969   | 1970   | 1970  | 1971  | 1971   | 1972   | 1974   | 1974  | 1975  | 1977  | 1977, 79   | 1979  |
| REACTOR TYPE                                | LWR   | HWR                        | HWR  | HWR  | FBR   | BWR   | LWR  | FBR  | FBR  | LWR   | FBR   | PWR   | HWR  | AGR   |
| FUEL TYPE                                   | uo <sub>2</sub>   | uo <sub>2</sub>            | u0 <sub>2</sub>  | u0 <sub>2</sub>                              | WOX   | uo <sub>2</sub>                                     | UO <sub>2</sub>                              | MOX  | MOX  | uo <sub>2</sub>                                     | MOX   | uo <sub>2</sub> , mox                         | UO <sub>2</sub>  | uo <sub>2</sub>   |
| INDEPENDENT (a)                             | VAT   | T                          | BU   | T, LHGR                                      | T   | ĭ   | LHGR. T <sub>s</sub> , PD                    | LHGR, BU   | LHGR, T <sub>I</sub> , T <sub>C</sub> ,  | T   | BU  | BU  | λ. D   | ۸, P <sub>K</sub> . V <sub>K</sub>  |
| METHOD OF<br>DETERMINING<br>CORRELATION (b) | BE  | со                         | BE   | ВЕ   | BE  | со  | со   | BE   | BE   | BE  | со  | ВЕ  | BE   | BE  |
| METHOD OF<br>PRESENTING<br>CORRELATION      | PLOT  | 3-ZONE<br>MODEL            | EQUATION   | PLOT   | 3-ZONE<br>MODEL                                     | 2-ZONE<br>MODEL                                     | PLOT   | EQUATION   | EQUATION   | 3-ZONE<br>MODEL                                     | EQUATION  | EQUATION                                      | EQUATION   | EQUATION  |
| DATA BASE<br>BURNUP RANGE,<br>GWd/MTM       | 0, 1-11.0   | ≤ 10                       | 7.5-44.0   | < 10   | 30-50   | ≤ 72,6  | ≤ 36. 5                                      | 8, 4-51.3  |  | 0.4-18.3  | 1.5-63  | ÷ 50  | ≤ 6.4  | -15   |
| REMARKS                                     | VAT (≤ 1400 <sup>O</sup> C)<br>IS MEASURED<br>AT PEAK<br>LIFETIME POWER | WEIGHTED SUM<br>OF FGRs IN | FGR 1S A<br>LINEAR<br>FUNCTION OF<br>BU AT<br>T <sub>Q</sub> < 1250 °C | FGR IS<br>DESCRIBED BY<br>A COMPUTER<br>CODE | FGR IS THE<br>WEIGHTED SUM<br>OF FGRS IN 3<br>ZONES | FGR IS THE<br>WEIGHTED SUM<br>OF FGRS IN 2<br>ZONES | FGR IS<br>DESCRIBED BY<br>A COMPUTER<br>CODE | FGR IS AN EXPONENTIAL FUNCTION OF LHGR AND BU IN NONRESTRUCTURED FUEL REGION | FGR IS<br>CALCULATED<br>IN 2 ZONES<br>SEPARATED BY<br>THE 1475 °K<br>PSEUDO<br>I SOTHERM | FGR IS THE<br>WEIGHTED SUM<br>OF FGRS IN<br>3 ZONES | FGR IS AN<br>EXPONENTIAL<br>FUNCTION OF<br>BU AT LEVELS<br>> 20 GWd/MTU | FGR 15 A<br>SECOND ORDER<br>FUNCTION<br>OF BU | FGR IS A TRIGONOMETRIC FUNCTION OF A AND D AT LOW BU AND T   < 1550 °C | FGR IS A<br>LINEAR<br>FUNCTION OF<br>A, P <sub>K</sub> AND V <sub>K</sub> |

<sup>(</sup>a) VAT = VOLUME-AVERAGE FUEL TEMPERATURE; T = LOCAL FUEL TEMPERATURE; BU = BURNUP; LHGR = LINEAR HEAT GENERATION RATE;

\$\bar{t}\_{\text{q}}\$ = FUEL SUBFACE TEMPERATURE; PD = PELLET DENSITY; T, = IRRADIATION TEMPERATURE; F\_c = CLADDING ID TEMPERATURE;

\$\bar{t}\_{\text{q}}\$ = FUEL SMEAR DENSITY; \(\text{d} = DECAY CONSTANT; \) D = DIFFUSION COEFFICIENT; P\_K = PROBABILITY OF FISSION GAS KNOCK-OUT;

\$V\_K = VOLUME FRACTION OF FUEL CONTRIBUTING TO KNOCK-OUT MECHANISM.

<sup>(</sup>b) BE = BEST ESTIMATE; CO = CONSERVATIVE OVERPREDICTION

#### OTHER PARAMETERS AFFECTING FISSION GAS RELEASE

The effect of grain size on the diffusion coefficient of  $^{133}$ Xe was studied by Klima, et. al. (1972). These investigators measured the fission gas release from irradiated (4x10 $^{-6}$  atom % burnup) and annealed (1200- $1600^{\circ}$ C) UO $_2$  pellets. They found that the diffusion coefficient of Xe increased substantially with increasing grain size: a change in grain size from 10  $\mu$ m to  $100~\mu$ m increased the diffusion coefficient by 4 orders of magnitude. In an attempt to study the effect of grain size on swelling and gas release properties of UO $_2$ , Turnbull (1973) irradiated specimens of UO $_2$  at  $1750^{\circ}$ C to a burnup level of 4 GWd/MTM and found that significant reductions (to about 1/2 the original values) in fission gas release and dimensional changes were caused by increasing the grain size of the specimens from 7 to 40  $\mu$ m. These reductions, in turn, led to a significant improvement in fuel performance. In a later paper, Turnbull and Friskney (1978) reported that not only grain size, but also grain size distribution could affect the rate of swelling and fission gas release of UO $_2$  fuels.

Evidence has shown that small amounts of additives, such as  ${\rm Ti0}_2$ ,  ${\rm Y}_2{\rm O}_3$ ,  ${\rm La}_2{\rm O}_3$  and  ${\rm N}_{\rm b2}{\rm O}_5$  will improve the sinterability and increase the grain size of sintered UO<sub>2</sub> pellets (Sowman and Ploetz, 1956; Ainscough, et. al., 1974). These additives have little effect on fission gas release at low burnups and gas concentrations. (a) However, at higher burnups ( ${\scriptstyle \geq 8}$ x10<sup>-4</sup> atom %) and gas concentrations ( ${\scriptstyle \geq 10}$ 16 ions/cm<sup>2</sup>), they retard the fission gas release (Matzke, 1966).

The fabrication of burnable poison oxide fuels (Littlechild, et. al., 1973) and the behavior of  $\rm U0_2\text{-}Gd_20_3$  fuel (Wada, et. al, 1973) have also been studied, based on the consideration that burnable poison fuel enables a more constant core reactivity with time. The benefit of adding burnable poisons to oxide fuels comes from the fact that these poisons have high neutron capture cross sections; as the overall fissile reactivity drops

<sup>(</sup>a) Xenon ions were injected into the fue] lattice by ion bombardment at doses between  $8 \times 10^2$  ions/cm<sup>2</sup> and  $2 \times 10^{16}$  ions/cm<sup>2</sup>.

with increasing burnup, they can absorb neutrons and revert to radionuclides with lower capture cross sections, thus balancing the decrease in reactivity due to burnup.

# CURRENT EXPERIMENTAL DATA ON FISSION GAS RELEASE AT HIGH BURNUPS

#### DATA SELECTION CRITERIA

One of the major parts of the state-of-the-technology assessment of high burnup effects was to survey the existing high burnup irradiation data, with emphasis on fission gas release behavior. An extensive literature search was made, from which a total of 91 journal articles and technical reports were reviewed. In order to select those which were most pertinent to the interests of the High Burnup Effects Program, the following criteria were used:

- Burnup levels above 20 GWd/MTM,
- Fuel Type UO2 and mixed-oxide only,
- Fuel Form pelletized form only, and
- Fission Gas Release experimental data required.

As a result of applying these criteria, 13 sources of data were identified in the literature. Information obtained from these data sources is summarized in Table 3, and a brief abstract of each publication is given in Appendix III. In the following sections, the data reported in the individual experiments are evaluated, compared, and discussed in relation to their applicability to the contents and scope of the High Burnup Effects Program.

Table 3 presents a wide range of fuel types and experimental conditions which leads predictably to a wide range of fission gas release (<1% to 100%). There are differences in the detailed information regarding the fuel design and irradiation conditions that are reported as well as the manner in which the data are presented. These differences lead to large uncertainties and make detailed comparisons of the various results difficult to interpret.

The high burnup gas release data are plotted in Figure 1. The individual data points from the reports of Dutt and Baker, and Zimmermann (1975,77) have been omitted for clarity and their data are represented by a range of values. The lower boundary for the Zimmermann data corresponds to specimens having a calculated end-of-life average temperature of 1000°C while the upper boundary corresponds to 1500°C for the calculated end-of-life temperatures. For the Dutt and Baker correlation, the upper and lower bound-

TABLE 3. Experimental Data on High Burnup (>20 GWd/MTM) Fission Gas Release

| INVESTIGATORS                            | AND RICH                                   | SMALLEY                             | BAROCH<br>AND RIGDON                    | SMALLEY  | ZIMMERMANN                                  | DUTT  | HERING AND<br>MANZEL | ROBERTS  | CARLSEN                  | ZIMMERMANN   | BOUFFIOUX AND<br>DeMEULEMEESTER   | PATI, BESSETTE<br>AND CORSETTI  |  |
|--|--|-------------------------------------|---|--|---|---|----------------------|--|--------------------------|--|---|---------------------------------|--|
| PONSORING<br>ORGANIZATION                | AERE<br>HARWELL                            | WESTINGHOUSE                        | BABCOCK<br>AND WILCOX                   | WESTINGHOUSE   | KARLSRUHE<br>NUCLEAR<br>RESEARCH CENTER     | HEDL  | KWU                  | WESTINGHOUSE   | RISØ                     | KARLSRUHE<br>NUCLEAR<br>RESEARCH CENTER  | BELGO<br>NUCLEA IRE   | COMBUSTION<br>ENGINEERING       | EPRI (a)   |
| DATE                                     | 1969                                       | 1971                                | 1973                                    | 1974   | 1975  | 1975, 77  | 1977                 | 1977   | 1978                     | 1978   | 1978, 79  | 1979                            | 1979   |
| EACTOR NAMES                             | DIDO                                       | SAXTON                              | BAW PWR                                 | SAXTON   | FR2, BR2, DFR<br>AND RAPSODIE               | EBR 11  | OBRIGHEIM            | ZORITA   | DR3                      | FR2  | 8R3   | CALVERT<br>CLIFFS I             | (P)  |
| EACTOR TYPE                              | HWR  | PWR                                 | PWR                                     | PWR  | HWR, FBR, LWR                               | FBR   | PWR                  | PWR  | H₩R                      | HWR  | PWR   | PWR                             | LWR  |
| LIMBER OF RODS<br>XAMINED                | 26   | 18                                  | 28                                      | 15   | 159   | 77  | 35                   | 50   | 2                        |  | 18  | 6                               | *-   |
| ELLET TYPE                               | uo <sub>2</sub>                            | MOX                                 | UO <sub>2</sub>                         | мох  | UO <sub>2</sub> (32 RODS)<br>MOX (127 RODS) | MOX   | UO <sub>2</sub>      | <sup>UO</sup> 2                                      | uo <sub>2</sub>          | UO <sub>2</sub>  | uo <sub>2</sub>   | UO <sub>2</sub>                 | no <sup>5</sup>                                  |
| //O Pu                                   | 0  | 6.6                                 | 0                                       | 6,6  | 0-35  | 25  | 0                    | 0  | 0                        | 0  | 0   | 0                               | 0  |
| ELLET DENSITY<br>6 TD)                   | 95 AND 98                                  | 94-96                               | 93.5                                    | 94-96  | UO <sub>2</sub> : 88-93<br>MOX: 84-95       | 90-96   |                      | >92.5  |                          | 96.7-98.2  |   | 93-95                           |  |
| NRICHMENT<br>J. 235 <sub>U)</sub>        | •••  |                                     |   |  | UO <sub>7</sub> < 12<br>MOX: 0,7-93.0       | 30-75   |                      |  | 2,28                     | 15 AND 20  |   | 2.33-2.82                       |  |
| LADDING TYPE                             | ss   | ss                                  | ZIRCALOY                                | SS   | SS ·  | SS  | ZIRCALOY             | ZIRCALOY   | ZIRCALOY                 | TZM  | ZIRCALOY  | ZIRCALOY                        |  |
| AP SIZE (mm)                             | 6x10 <sup>-3</sup><br>-12x10 <sup>-3</sup> | Q17-Q21                             | Q1-Q2                                   | Q 17-Q21   |   | 0.15-0,20   |                      | 0.14-0.24  | 0.24                     |  |   | 0.21                            |  |
| RESSURIZED OR NOT                        | NO   | NO                                  |   | NO   |   |   | вотн                 | вотн   | NO.                      |  |   | YES                             | вотн   |
| UEL TEMPERATURE<br>PARAMETERS CONSIDERED | CENTERLINE                                 | CENTERLINE                          | SURFACE AND<br>CENTERLINE               |  | EOL VOLUME<br>AVERAGE                       |   |                      |  | CENTERLINE               | EOL VOLUME<br>AVERAGE  |   |                                 |  |
| NETHOD OF<br>ETERMINATION                | CALCULATED                                 | CALCULATED                          | CALCULATED                              |  |   |   |                      |  | CALCULATED               | MEASURED AND CALCULATED  |   |                                 | •  |
| EMPERATURE RANGE ( <sup>Q</sup> C)       | 1000-1700                                  | ≤2200                               | SUR: 700-<br>1050<br>CTR: 1800-<br>2800 |  | 1000-1500                                   |   |                      |  | 1500-2150                | 980-1750   |   |                                 |  |
| URNUP RANGE<br>GWIAMTMI                  | 8 - 44                                     | 15 - 21                             | 8 - 65                                  | 25 - 40  | 7 - 114                                     | 2 - 63  | 11 - 40              | ≤65  | ≤38                      | 9 - 87   | ≤48   | 18 - 29                         | ≤ 30   |
| EAK LHGR (w/cm)                          |  | 300-560                             | 460-705                                 | 370-650  | UO <sub>Z</sub> : 350-690<br>MOX: 450-700   | 140-450   | 250-470              | <460   | 550 & 600                | ***  | 275-440   | <360                            |  |
| VE LHGR (w/cm)                           |  | 180-250                             | ***                                     | 200-300  | 150-570                                     |   | 140-360              | 120-300  |                          |  |   | 180-220                         | •  |
| S POWER HISTORY<br>VAILABLE              | NO   | YES                                 | NO                                      | YES  | YES   | YES   | YES                  | NO   | YES                      | NO   | YES   | YES                             | NO   |
| GR RANGE (%)                             | 0.01-10                                    | 4-32                                | 10-88                                   | 26-37  | 10-95                                       | 1,8-100   | 0.2-55.5             | 3-27   | 36.6 -48.6               | 10-90  | 1-13  | <1                              | 1-27   |
| THER PARAMETERS<br>ON SIDERED            | SWELLING,<br>GRAIN GROWTH                  | CORROSION AND<br>TENSILE PROPERTIES | SWELLING                                | CORROSION AND<br>TENSILE PROPERTIES                    | RETAINED FG                                 | GRAIN SIZE  |                      | SWELLING, CLADDING<br>CREEP AND CORROSION            |                          | SWELLING,<br>Cs RELEASE  | RELATIVE<br>POWER   | CLADDING CREEP,<br>GRAIN GROWTH | **-  |
| ONCLUSION                                | FGR INCREASES<br>WITH BU                   |                                     | FGR INCREASES<br>WITH Bu                | FGR INCREASES DUE<br>TO HIGHER BU AND<br>POWER HISTORY | FGR INCREASES<br>WITH BU AT<br>CONSTANT T   | FGR IS AN<br>INCREASING<br>FUNCTION OF<br>BU AND LHGR |                      | FGR HAS STRONG BU<br>AND POWER HISTORY<br>DEPENDENCE | FGR INCREASES<br>WITH BU | FGR IS DETERMINED<br>PRIMARILY BY<br>IRRADIATION<br>TEMPERATURE AT<br>CONSTANT<br>TEMPERATURE, FGR | FGR INCREASES WITH RELATIVE POWER LEVEL. NRC CORRELATION OVEREST IMATES FGR IN LWR. | NO BU<br>ENHANCEMENT<br>OF FGR  | FGR IS DETERMINED PRIMARILY TEMPERATUI BURNUP HA |

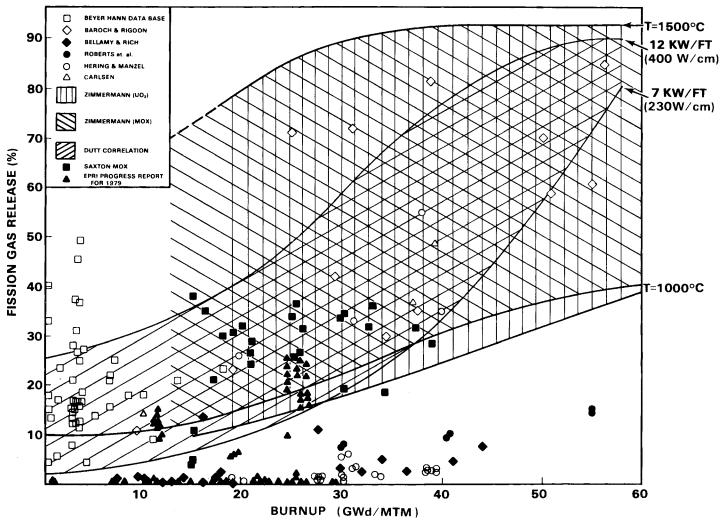


FIGURE 1. A Graphic Presentation of Existing Experimental Data on High Burnup Fission Gas Release.

aries correspond to LHGRs of 365 to 215 W/cm (12 and 7 kW/ft), respectively. The low burnup data base used in the Beyer-Hann gas release correlation is also included in Figure 1 to complete the burnup range from zero to 60 GWd/MTM.

Figure 1 shows wide variations in the fractional fission gas release and, for a given set of data, wide variations in the apparent burnup dependence are also evident. Fuel temperature is the most important factor in determining fission gas release and is primarily responsible for the wide range of gas release values. However, other factors such as 1) temperature gradients, 2) fission rate, 3) flux depression, 4) fuel type, 5) fuel microstructure 6) power history and 7) burnup could also influence gas release and their effects need to be considered when evaluating gas release at high burnups.

In the following sections, the specific experimental data will be evaluated and their potential application to establishing the burnup dependence of fission gas release in LWR fuels will be discussed.

#### EVALUATION OF DATA

### Bellamy and Rich (1969)

The data of Bellamy and Rich were among the first to suggest that fission gas release fraction increases with increasing burnup. These data are especially significant with regards to commercial LWR operations because the data were obtained from  $\rm UO_2$  fuels and operated at low temperatures,  $\sim 1200\,^{\circ}\rm C$ . However, the specimens were small, 4 to 6 mm in diameter, and were irradiated at high fission rates, 6 to 8 x  $\rm 10^{13}$  fissions/cm $^3$ -sec. Both of these factors are atypical of LWR fuels and care should be exercised in directly using these data because the effect of these factors on gas release has not been clearly established.

It should also be noted that the increased gas release fractions observed beyond 20 GWd/MTM may be related to power history rather than burnup per se. This possibility is based on examining the calculated centerline temperatures for each of the nine reactor cycles. In general, fuel temperatures decrease with increasing burnup until the last reactor cycle where temperature increases

of 200 to 400°C were calculated. Since all of the low temperature specimens with burnups greater than 20 GWd/MTM were irradiated in the ninth reactor cycle, the increased release fraction could be associated with the increased power levels and fuel temperatures either through increased diffusional release or by other processes such as fuel cracking.

#### Smalley (1971 and 1974)

These reports present fission gas release data from mixed-oxide fuels irradiated in the Saxton (PWR) reactor. The fuel rods were irradiated at various power levels to burnups ranging from 9 to 37 GWd/MTM. The fuel dimensions and the average fission rates were typical of a PWR reactor fuel. However, the local fission rate within the  $PuO_2$  particles was much higher and could influence gas release. Consequently, caution should be exercised in directly using these data to predict gas release from  $UO_2$  fuels.

Gas release data were obtained from both the Core II and Core III cycles. The power levels in Core III were generally higher than in Core II and the rods in Core III were subjected to load-following cycles throughout much of this irradiation period. The measured gas release ranged from 4 to 38% and was directly related to the instantaneous peak pellet LHGR as shown in Figure 2. Comparison of the data from Core II and Core III shows no indication of an effect from either burnup or power cycling on fission gas release. A similar plot using time-averaged LHGRs shows the same general trend but with greater scatter in the data.

### Baroch and Rigdon (1973)

These data represent fission gas release from UO $_2$  fuels operating at estimated LHGRs of 400 to 790 W/cm (13 to 26 kW/ft). The measured fission gas release varied from 10 to 88% and appeared to increase with increasing burnup. However, a large amount of scatter exists in the data which is most likely associated with differences in the fuel temperatures. There appears to be large uncertainties in the estimated LHGRs and the calculated temperatures as evidenced by unexpected fuel melting in four of the fuel rods. The LHGRs used in these experiments were not typical of normal LWR operating conditions, and when combined with large uncertainties in the power level, they make these data of little value in determining the fission gas release from operating LWRs.

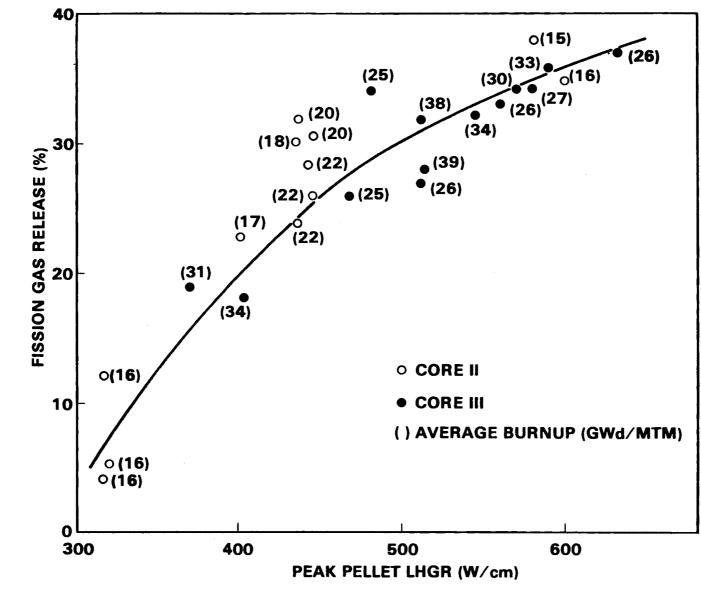


FIGURE 2. The Relationship Between Fission Gas Release and Peak Pellet LHGR for the Saxton Mixed-Oxide Data.

### Zimmermann (1975)

This paper provides a large amount of gas release data from several individual experiments. Data are presented from  $\rm UO_2$  and a variety of mixed-oxide fuels with the latter having been irradiated in either fast or thermal reactors. The specimens were all approximately 5 mm in diameter and were irradiated at high fission rates, 6 to 11 x  $\rm 10^{13}$  fissions/cm<sup>3</sup>-sec.

The gas release data are presented as a function of burnup and a reasonable correlation exists between fractional gas release and the calculated average temperature at the end of the irradiation. The release fraction increased with increasing temperature and also increased with burnup. The burnup range where the gas release showed the most rapid change decreased with increasing temperature. Also, in contrast to the NRC high burnup correlation, the rate of increase in gas release with burnup, i.e., the apparent burnup dependence, decreased with decreasing temperature.

There is a large amount of scatter in the gas release data below 60 GWd/MTM. This is probably associated with the wide range of fuel types and irradiation conditions used. The individual gas release data points were not identified which precludes a detailed evaluation of the data. However, measurements were also made on the retained gas and these data suggest greater gas retention, i.e., lower release, in the UO $_2$  fuels than the mixed-oxides for equivalent temperatures and burnups. The difference between the two fuel types increases with decreasing temperature and thereby indicates that caution should be exercised when comparing low temperature gas release from UO $_2$  and mixed-oxide fuels.

### Dutt and Baker (1975)

This paper presented a fission gas release correlation based on results obtained from release measurements made on 77 fuel pins irradiated in the EBR-II reactor. The mixed-oxide fuels had small diameters, approximately 5 mm, and were irradiated at high fission rates, 3 to 7 x  $10^{13}$  fissions/cm<sup>3</sup>-sec. These factors, as well as the decreased flux depression, are not typical of LWR irradiations. More importantly, higher fuel surface temperatures yield much higher volume-averaged temperatures, hence correlation against LHGR is very misleading. Therefore, caution is required in directly using these data.

The fission gas release correlation shows an exponential dependence on the local power level and burnup. For a constant LHGR, the gas release fraction increases with burnup and the maximum rate of increase, i.e., the apparent burnup dependence, increases with decreasing LHGR. This latter observation is in contrast to the Zimmermann (1975, 1978) data where the apparent burnup dependence decreased with decreasing volume-averaged temperature at the end of the irradiation. The difference between the two apparent burnup dependencies is most likely associated with the relationship between fuel temperature and gas release in the following manner: at constant power level, the release of fission gas decreases the heat conductance across the fuel-cladding gap and thereby increases the fuel temperatures. This, in turn, results in a greater fraction of the gas being released and the cycle continues until the gap conductivity stabilizes. This type of a temperature feedback would result in a larger apparent burnup dependence at constant LHGR, which is in accordance with the difference between the two correlations.

#### Hering and Manzel (1977)

Gas release data from 23 "standard" fuel rods, 4 high power experimental rods, and 8 fuel rods from a cycling experiment are presented. The rods were all irradiated in the Obrigheim pressurized water reactor or under normal operating conditions and, thereby provide valuable information regarding fission gas release from  $\rm UO_2$  fuels.

The standard rods were irradiated at low heat ratings (<300 W/cm peak) to burnups from 11 to 40 GWd/MTM. The gas release from these rods ranged from 0.21 to 6.5% and the highest releases occurred at approximately 30 GWd/MTM. No definite relationships between gas release fraction and burnup, LHGR, or prepressurization of the fuel rods could be established. Thus, based on the limited information available, it appears that the observed range of gas release is due to either processing variables or to differences in the detailed power/temperature histories of the individual rods.

The four high power rods contained experimental fuel with small grain size and high porosity. This microstructure is conducive to densification which may explain the high gas releases (33 to 55%) that were reported.

The eight rods from the cycling experiment were all irradiated to approximately 30 GWd/MTM and received a total of 880 cycles. The gas release ranged from 0.83 to 6.9%. This is the same range of gas release as that observed for the standard rods at 30 GWd/MTM and indicates cycling did not significantly influence fission gas release. However, in this experiment the two nonpressurized rods showed the highest gas release and reflect the "thermal feedback effect" between gas release and fuel temperatures.

The large power gradients along the axis of the cycled rods would accentuate the thermal feedback effect and would explain why the effects of prepressurization were more pronounced in these rods than the standard rods.

## Roberts, et al.(1977)

Gas release data from the Zorita Research and Development programs were discussed in this paper. The authors propose that power history and burnup are the predominant factors in determining fission gas release. However, specific data were given for only 6 of the 50 fuel rods examined, and neither the power levels nor fuel design parameters were given. Consequently, it is not possible to evaluate the data due to the lack of information.

# <u>Carlsen (1978)</u>

This paper presents fission gas release data from two well-characterized BWR type fuel rods irradiated to approximately 38 GWd/MTM. The rods were irradiated at high power levels (400 to 600 W/cm), and the measured gas release values were 37 and 49%, respectively.

The Danish fuel performance code WAFER-2 underpredicted the measured release fractions unless the NRC high burnup correlation was included. This was taken as evidence for enhanced fission-gas release at high burnup. In contrast, Hastings and Notley (1979) have reported that the fuel performance code ELESIM correctly predicted the measured gas release in these fuel rods without the addition of a correction factor. However, it should be noted that the ELESIM code considers microstructure evolution and thereby contains an inherent burnup dependence.

#### Zimmermann (1978)

Fission gas release data from  $\rm UO_2$  fuel irradiated under nearly isothermal conditions are presented in this paper. The specimen diameters were less than 5 mm and the fission rates ranged from 3 to 14 x  $10^{13}$  fissions/cm $^3$ -sec. Both of these factors, as well as the isothermal irradiation condition, are not typical of light water reactor fuels and caution should be exercised in directly using these results.

Gas release is reported to increase with both temperature and burnup. The general shape of the constant temperature release curves are similar to those reported previously for the mixed-oxide irradiations (Zimmermann, 1975). The isothermal release curves are shifted downward in accordance with the expected difference between isothermal and volume-averaged temperatures in specimens that have large temperature gradients. The enhanced release at high burnup was attributed to a saturation of the fuel matrix; and measurements of the fission gas ratained in the fuel tended to support this view.

The individual data points for fission gas release were not provided. Thus, it is not possible to evaluate the degree of scatter in the data or the quantity of data on which the correlations are based.

The initial grain size was reported to have no significant effect on fission gas release in specimens irradiated to about 80 GWd/MTM at mean temperatures of 1480K. The measured gas release was approximately 87% in specimens that had initial grain size of either 7  $\mu m$  or 40  $\mu m$ . The formation of subgrains in the large-grained material was proposed to explain the similar release fractions. However, no definite evidence for subgrain formation was given.

The release fraction of  $^{137}$ Cs as a function of burnup and temperature was also presented. There is a large amount of scatter in the data, but at higher temperatures there is a definite increase with increasing burnup. In comparison to the fission gas release data, the release of cesium is delayed to higher burnups and at low temperatures (1250K) the release fractions are considerably lower.

#### Bouffioux and DeMeulemeester (1978 and 1979)

These papers present fission gas release data from the Belgonucleaire data base. The data base includes a wide range of LHGRs and burnups for both  $\rm UO_2$  and mixed-oxide fuels. The  $\rm UO_2$  data base should be directly applicable to LWR irradiations but the experimental details were not given. This precludes a definite conclusion as to the applicability of these data.

The combined data base shows a wide range of gas release values for both low and high burnups. This wide range is primarily associated with differences in LHGR and the fuel temperatures. The authors compare the gas release from two high burnup  $\rm UO_2$  fuel rods with the predictions from the COMETHE fuel performance code and obtain good agreement without the NRC correction factor. However, because the code considers the evolving microstructure, it contains an inherent burnup dependence and the addition of the correction factor may be unwarranted.

## Pati, et al. (1979)

Fission gas release data from six prepressurized  $\rm UO_2$  fuel rods are presented. The data include both stable and unstable fuels that were irradiated in the Calvert Cliffs 1 reactor to peak burnups of 18 to 29 GWd/MTM. The peak LHGRs were less than 350 W/cm and the measured release fractions were all less than 1%.

The data do not show any evidence of burnup enhancement. Zimmermann (1975,78) has reported that the burnup range where enhanced fission gas release occurs increases with decreasing temperatures. Thus, the lack of a burnup effect in the data presented by Pati, et al., may be related to their low temperatures and relatively low burnups.

## EPRI Progress Report (1979)

This report presents a plot of fission gas release as a function of burnup (to 30 GWd/MTM) for both PWR and BWR type fuel rods. The fuel rods were irradiated in various commercial reactors under normal operating conditions and thereby provide gas release data for typical LWR fuels. The PWR data contain both prepressurized and nonpressurized rods while the BWR rods were all nonpressurized.

The measured gas release was less than 1% for all of the prepressurized rods, whereas the nonpressurized rods showed a wide range of gas release, <1 to 27%. This difference was attributed to the improved thermal stability of rods containing the higher helium contents. Power history, fuel stability, and gap size were considered to be the important factors contributing to the wide range of gas release in the nonpressurized rods. These parameters could all influence gas release but because the details of the fuel rod designs or the power histories were not given, it is not possible to evaluate these data until the final report is published.

#### DISCUSSION OF HIGH BURNUP GAS RELEASE DATA

As stated previously, fission gas release most likely depends on many factors including:

- fuel temperature
- temperature gradients in the fuel
- fission rate
- flux depression across the fuel radius
- fuel type
- fuel microstructure
- power/temperature history
- burnup

Temperature is the dominant factor and at high temperatures is expected to overpower the effects of the other factors. That is, for temperature conditions where the majority of the gas is being released, the contributions, if any, from the other factors will be relatively insignificant. However, the temperatures in LWR fuels under normal operating conditions are relatively low and therefore, the other factors could have significant effects on fission gas release.

Many of the above factors are interrelated which makes identification and evaluation of their individual effects difficult. In addition, constant conditions are normally not maintained during the lifetime of the fuel which can lead to different interpretations as to the influence of burnup on fission gas release.

For clarification, burnup enhancement as used in this discussion, is defined as follows: The fission gas release fraction increases with increasing burnup during irradiation at a constant fuel temperature. This definition was chosen because it does not require selecting a specific gas release model for comparing with the experimental data in order to establish burnup enhancement. From diffusion theory, as well as the low temperature knock-out mechanism, one would expect the release fraction to increase with burnup because of the higher concentration of fission products within the fuel. Therefore, the important consideration is not the existence of a burnup enhancement but its magnitude and form as a function of burnup.

The importance of the functional form of the burnup dependence is illustrated in Figure 3. Both curves A and B adequately represent the data point, but large differences may exist in the predicted gas release at high burnup. The shapes of the curves were not intended to represent a specific gas release mechanism. However, the shape of curve A represents a process that continuously increases with burnup while curve B could represent a process that saturates with increasing burnup. There are other curves of various shapes which could pass through the single data point however, the shape of the proper curve would be expected to depend on the fuel design and irradiation conditions.

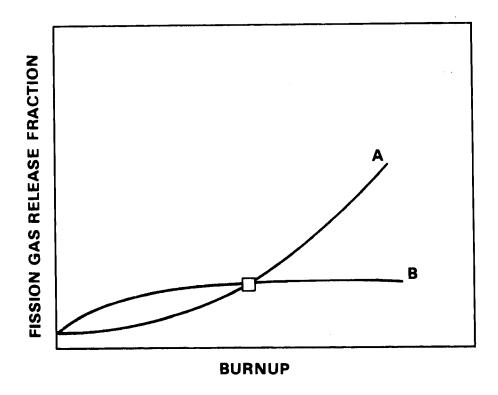


FIGURE 3. A Schematic Illustration Showing Different Types of Burnup Dependencies of Fission Gas Release.

Increasing the number of data points and their burnup range will allow the proper curve shape to be defined as long as the irradiation conditions remain constant. However, because constant conditions are rarely maintained during irradiation, the best approach to establishing the proper burnup dependence is to compare the results from similar irradiation conditions and evaluate parameters that can relate these results to other irradiation conditions.

The high burnup gas release data that have been reviewed represent a wide range of fuel types, fuel designs, and irradiation conditions. The data can be broadly placed into two groups. The first group is comprised of specimens that have diameters less than 7 mm and fission rates greater than  $3 \times 10^{13}$  fissions/cm<sup>3</sup>-sec. The second group, with diameters greater than 7 mm and fission rates less than  $4 \times 10^{13}$  fissions/cm<sup>3</sup>-sec, are more representative of LWR fuel designs and irradiation conditions. Thus, this grouping allows the data from specimens with temperatures, temperature gradients, and fission rates that were typical of LWR conditions to be compared, separately.

Due to the high fission rates, the majority of the high burnup data are from the first group. The data of Zimmermann (1975,78) and the Dutt and Baker (1975) correlation provide the most graphic evidence for enhanced fission gas release at high burnup. However, as shown in Figure 1, there are large differences in the apparent burnup dependences which are most likely associated with the particular parameters chosen to develop the correlation. Dutt and Baker used the LHGR which, as discussed previously, could lead to a higher apparent burnup dependence because of thermal feedback effects.

Because gas release is directly related to temperature, a correlation based on temperature seems more appropriate than on LHGR. However, the end of life volume-averaged temperature may not be the proper parameter because it does not account for temperature history effects. For example, a higher release fraction is expected from a fuel rod whose temperature continuously decreased with burnup compared with one irradiated continuously at the same end-of-life temperature. The temperature or power histories were not given for any of the data reported by Zimmerman (1975, 1978) or Dutt and Baker (1975) and thereby precludes any evaluation of the effects of power histories or identification of other temperature related parameters that could provide a more meaningful correlation.

The data of Bellamy and Rich (1969) are also included in the first group of data. In these experiments, the temperatures were reduced by minimizing

the fuel-cladding gap and using small specimens,  $\sim 4$  mm diameter. The low temperature data show a gradual increase in gas release fraction with burnup to 20 GWd/MTM and a more rapid increase in gas release may be associated with the temperature increase that occurred during the final reactor cycle.

For specimens with centerline temperatures <1200°C, the measured release fractions are less than one-half of those reported by Zimmermann (1978) for isothermal irradiations. The temperature profiles were not given by Bellamy and Rich but the large difference in gas release suggests that temperatures remain an important parameter at temperatures less than 1000°C. Thus, the increased release fraction observed following the final reactor cycle could be directly related to the increase in fuel temperature.

In summary, all of the experimental data from small diameter fuel pins (<7 mm dia) show evidence of burnup-enhanced fission gas release. The apparent burnup dependence is sensitive to the parameter selected to make the correlation. Using the LHGR as the parameter leads to a high burnup dependence, especially at low heat ratings. For most of the data, the LHGR and temperature histories or the specific details of the fuel rod design were not available. Consequently, a large effort would be required to obtain the necessary data in order to conduct further analysis. In view of the atypical nature of these data, this amount of effort is considered unwarranted.

The second group of data are more applicable to LWR irradiations. The fuel diameters and fission rates are typical of operating reactors and with the exception of the Saxton data, all contain UO<sub>2</sub> fuels. The fuel rods were irradiated under a variety of conditions. When this is combined with the differences in the fuel rod design and fuel microstructures, a wide range (<1 to 90%) of gas release fractions results. Because the data represent fuels with different microstructural characteristics irradiated under different conditions, it is best to first look for evidence of burnup enhancement within a given set of data and then compare the data sets and try to establish the important parameters.

Within a specific data set, only the data of Baroch and Rigdon (1973)

and Roberts, et al (1977) show evidence of burnup-enhanced fission gas release. The Baroch and Rigdon data were obtained from rods irradiated at high LHGRs and there is a large uncertainty associated with the estimated values. The data suggest a burnup-enhanced gas release but due to the large uncertainties in the calculated LHGRs and temperatures, a definite relationship between gas release and burnup would not be meaningful.

The specific data reported by Roberts, et al, show an increase in fission gas release fraction with increasing burnup. However, the power histories, power levels, or any of the details regarding the fuel, or the fuel rod design are not available. Consequently these data are of little value in establishing the effect of burnup on fission gas release.

None of the other individual data sets in the second group show a strong burnup enhancement. In some cases, there are insufficient data to obtain a correlation - while in others, the scatter in the data prevents a relation-ship from being identified. Comparing the data from different sets leads to additional difficulties which will not be addressed in this discussion.

In order to combine the data sets, it is necessary to have a common parameter on which to base the comparison. The importance of temperature on gas release suggests that this parameter would provide the most meaningful comparison. However, only one third of the data sets in this group report temperatures and of the remaining six only one gives the dimensions of the fuel-cladding gap. Consequently, with the available information, it is not possible to compare the data on the basis of temperature.

The peak LHGR is the parameter most often reported to describe the data. However, there is no consistency in the form of the reported value. In some cases, an instantaneous peak value is given, whereas the time-averaged peak value is given in others. And finally, some of the reports give only peak power values that appear to be derived from design criteria and not operating experience. Consequently, meaningful comparisons between the data sets are not possible.

In summary, the data from fuels with diameters greater than 7 mm show a wide range of fission gas release fractions. There is insufficient speci-

fic information regarding the fuel rod designs or the irradiation conditions to make meaningful comparisons between the data. Therefore, the available data are not sufficient to establish either the magnitude or the form of the burnup dependence on gas release in fuels irradiated under LWR conditions.

#### EVALUATION OF DATA NEEDS

The release of fission gases from the fuel during irradiation is an important consideration in determining fuel performance and can significantly influence fuel rod design, especially at high burnup. The NRC recently developed a correction factor to account for the apparent burnup enhancement and has required that this factor be included in calculations of fission gas release for burnups above 20 GWd/MTM. The correction factor is based on the Dutt and Baker (1975) correlation which, as discussed previously, indicates a strong burnup dependence.

The effect of the NRC correction factor on the calculated fission gas release surfaces for typical PWR and BWR type fuel rods is illustrated in figures 4 and 5, respectively. The surfaces were constructed from gas release fractions that were calculated with the GAPCON THERMAL 2 (Beyer, et al, 1975) computer code. The fuel rod parameters for the two rods are given in Table 4 and a complete listing of the input is given in Appendix IV.

TABLE 4. Fuel Rod Parameters Used for Calculating the Fission Gas Release Surfaces

| Parameter             | BWR Rod | PWR Rod |
|-----------------------|---------|---------|
| Fuel-cladding gap, mm | 0.23    | 0.16    |
| Initial Pressure, MPa | 0.1     | 2.7     |
| Plenum Volume, %      | 14.7    | 9.2     |
| Axial Profile         | cosine  | cosine  |
| Fuel Enrichment, %    | 3       | 3       |
| Fuel Density, %TD     | 95      | 95      |

The upper surfaces, in figures 4 and 5, represent the calculated fission gas release fractions when the NRC correction factor is applied. At 60 GWd/MTM the release fraction ranges from approximately 40 to 80% for both the PWR

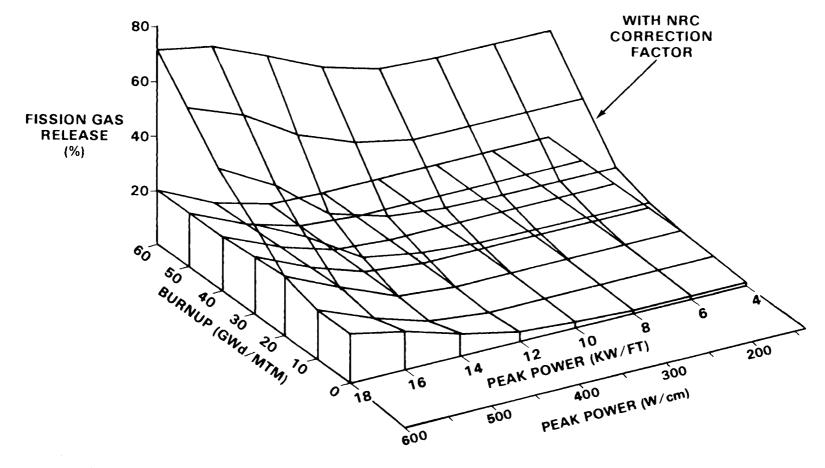


FIGURE 4. Fission Gas Release as a Function of Burnup and Peak Power with and without the NRC Correction Factor for a Prepressurized PWR Fuel Rod.

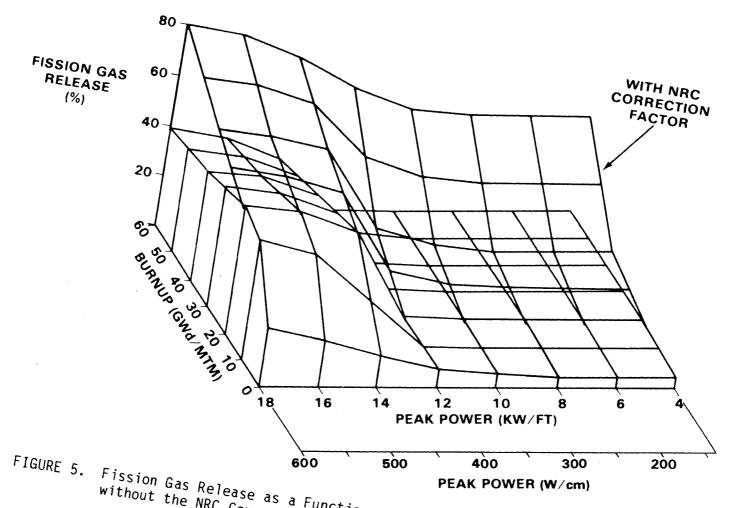


FIGURE 5. Fission Gas Release as a Function of Burnup and Peak Power with and without the NRC Correction Factor for a Nonpressurized BWR Fuel Rod.

and BWR cases. The PWR surface without the correction factor is relatively flat below 350 W/cm and increases gradually with power thereafter. The BWR surface is also relatively flat at low LHGR levels but has a large hump above 350 W/cm. The difference between the two surfaces at the high LHGR levels is due to the larger fuel-cladding gap and the lower helium content in the BWR-type fuel rod. Both of these factors enhance the thermal feedback effect and thereby drive the temperatures and gas release fractions upward.

The fission gas release surfaces are derived from calculations and therefore depend on the specific input and models used. Thus, a wide range of surfaces can be produced by either changing the input parameters or by using different models. Consequently, experimental data are needed to define the fission gas release surface at high burnups in the fuel temperature operating regimes of light water reactors. This allows fission gas release correlations to be developed that are based on experimental data and thereby removes the current reliance on models or calculations.

It is not necessary to establish the entire gas release surface and efforts should be concentrated in those temperature regimes that are most representative with LWR fuels. Some of the existing data are from fuels that were irradiated under these conditions and could be used if more detailed information could be obtained. The location of the existing data in terms of the time-averaged peak LHGR and burnup is shown in Figure 6. The peak LHGR is used in this case because fuel temperature data are not available. To convert from volume-averaged LHGR to peak LHGR, an axial peak-to-average ratio at 1.3 was assumed for the data of Smalley (1971, 1974) and a 1.1 peak-to-average was assumed for the Herring and Manzel (1977) data. The data from the Zorita program or those given in the EPRI progress report are not included because the necessary LHGR data are not available.

The solid line represents a typical peak pellet design envelope for a high burnup irradiation and thereby defines the upper region of interest. As can be seen, the majority of the existing data fall within the region of interest. The amount of data diminishes with increasing burnup until only one data point exists beyond 40 GWd/MTM.

The use of data derived from mixed-oxide fuels to predict gas release

in  $\mathrm{UO}_2$  has been criticized and arguments both for and against have been presented. However, there is insufficient evidence to prove either point of view and thus the data, if used, should be used with caution. It should also be noted that the time-averaged peak LHGR may not represent the proper position for the data on figure 6. This is especially true for the Saxton data where large power variations in LHGR occurred. If the instantaneous peak LHGR is used, all of the Saxton Core III data are shifted above the region of interest.

If the Saxton data are excluded, the LHGRs for the remaining fuel rods within the region of interest are less than 350 W/cm and the rods are all PWR-types. The calculated gas release surface for the PWR type fuel rod was relatively flat below 350 W/cm (Figure 4) which provides a basis for comparing the experimental data with the calculated surfaces. This comparison is illustrated in Figure 7 where a peak value of 330 W/cm was used in the calculation. As can be seen, a considerable amount of scatter exists in the data and precludes identification of a specific surface. Part of this scatter might be removed if evaluations were made on the basis of temperature rather than peak power. However, other factors such as fuel microstructure and stability, power history, or manufacturing tolerances could also contribute to the existing data scatter. Additional detailed information is required to more fully utilize these data.

The scatter in the existing information illustrates the need for using well-qualified data for producing the experimental fission gas release surfaces. The primary surfaces should be derived from data obtained with fairly constant power histories and from fuels with stable microstructures. Prepressurization should also be considered because some studies have indicated less data scatter from prepressurized rods. When these surfaces have been established, the effects of variable power history can be quantified by comparing these data with the constant power surfaces.

The release of fission products other than the noble gases can also influence fuel performance by 1) chemical reactions with the fuel and cladding, 2) mechanical interaction between the fuel and cladding, and 3) altering the heat conductance across the fuel-cladding gap. This is

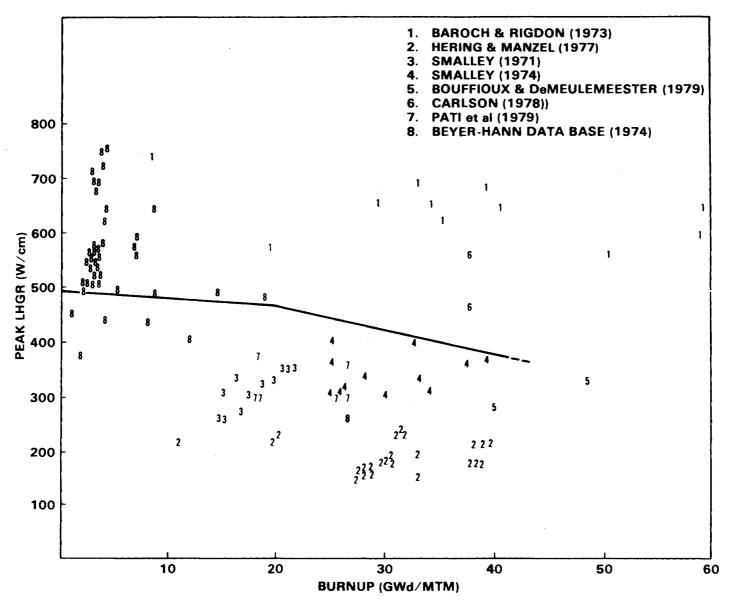


FIGURE 6. The Location of the Published Fission Gas Release Data with Respect to Time-Averaged Peak Pellet LHGR and Burnup.

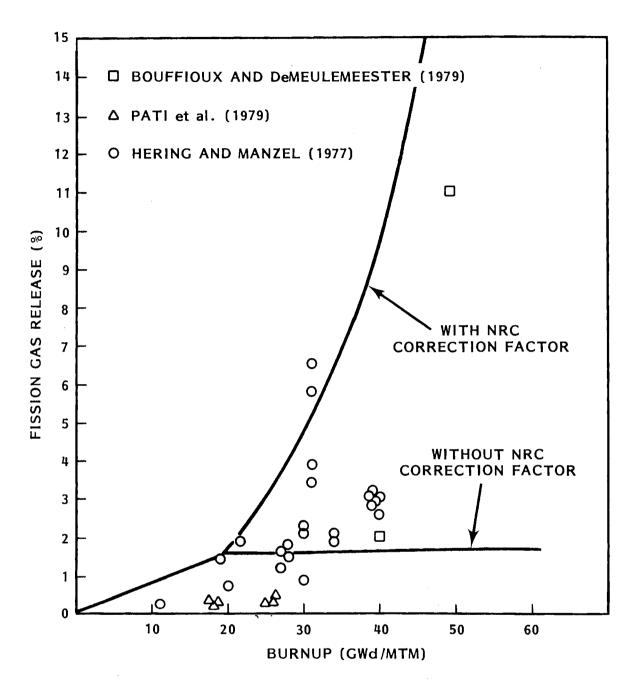


FIGURE 7. Comparison of Published Data with GT-2 Predictions for a Prepressurized PWR Type Fuel Rod with and without the NRC Correction Factor.

(Peak LHGR <350 W/cm)

especially true at high burnups due to the increased accumulation of fission products in the fuel-cladding gap. Zimmermann (1978) has reported that burnup-enhanced release of cesium occured during isothermal irradiation of  ${\rm UO}_2$ . If all fission products experience enhanced release at high burnup, their effects on fuel performance could be significant. Therefore, information is needed regarding the release of fission products other than the noble gases and their effects on fuel performance during high burnup irradiations.

In conclusion, the high burnup (>20 GWd/MTM) fission gas release data from thirteen sources have been examined and evaluated. The majority of these data were obtained under irradiation conditions that are not typical of current light water reactor fuels and are therefore considered to be of limited value. Even for those data that appear to be directly applicable, the presently available information regarding the fuel rod design or the detailed irradiation conditions is insufficient to allow a complete evaluation to be made. Therefore, until additional information is obtained, the high burnup gas release data that exists in the open literature can not be used to establish the effects of burnup on gas release in LWR fuels in a meaningful way.

#### REFERENCES

- ANS (American Nuclear Society) Working Group 5.4. 1977. Status Report: Fission Product Release from UO<sub>2</sub> Fuel. Report No. N218.
- Bailey, W. E., et al. 1969. "Effect of Temperature and Burnup on Fission Gas Release in Mixed Oxide Fuel." Ceramic Nuclear Fuels, Special Publication No. 2:195-210, American Ceramic Society, Columbus, Ohio.
- Baldewicz, W. L. 1977. State-of-the-Art of Fission Gas Release from LWR Fuels. UCLA-ENG-7740, University of California, Los Angeles.
- Barnes, R. S. and D. J. Mazey. 1963. "The Migration and Coalescense of Inert Gas Bubbles in Metals." Proc. of Roy. Soc. A275:47.
- Baroch, C. J., and M. A. Rigdon. 1973. "Irradiation Behaviour of UO<sub>2</sub> at Burnups From 10 to 80 GWd/tonne U." In <u>Proceedings of the International</u> Conference on Nuclear Fuel Performance, paper 58.
- Baston, V. F., T. H. MacFadden and W. A. Yuill. 1971. Analytical Method for Calculating Steady-State Fission Gas Release--Fission Product Fuel Model (FPFM) Code. ANCR-1010, Aerojet Nuclear Co., Idaho Falls, Idaho.
- Bellamy, R. G., and J. B. Rich. 1969. "Grain-Boundary Gas Release and Swelling in High Burnup Uranium Dioxide." <u>J. Nucl. Mat.</u> 33:64-76.
- Beyer, C. E., and C. R. Hann. 1974. <u>Prediction of Fission Gas Release from UO<sub>2</sub> Fuel</u>. BNWL-1875, Pacific Northwest Laboratory, Richland, Washington.
- Beyer, C. E., et al. 1975. <u>User's Guide for Gapcon-Thermal-2: A Computer Program for Calculating the Thermal Behavior of an Oxide Fuel Rod.</u>
  BNWL-1897, Pacific Northwest Laboratory, Richland, Washington.
- Beyer, C. E., and R. O. Meyer. 1976. "Semiempirical Model for Radioactive Fission Gas Release from  $\rm UO_2$ ." ANS Trans. 23:172.
- Bogensberger, H. G., and C. Ronchi. 1976. "Effects Due to Fission Gas During Unprotected Overpower Transients in a Liquid Metal Fast Breeder Reactor." Nucl. Tech. 29:73-85.
- Booth, A. H. 1957. A Method of Calculating Fission Gas Diffusion from UO<sub>2</sub>
  Fuel and its Application to the X-2-f Loop Test. AECL No. 496, Chalk River,
  Ontario.
- Bouffioux, P., and E. De Meulemeester. 1978. "Benchmarking Results of the COMETHE Code." <u>COMETHE Publication</u> at the IAEA Specialist Meeting on Fuel Element Performance, Report BN7803-03, Belgonucleaire, Belgium.

- Bouffioux, P., and E. De Meulemeester. 1979. "Prediction of Fission Gas Release at High Burnup." Paper presented at the ANS Topical Meeting on LWR Fuel Performance, Portland, Oregon.
- Buescher, B. J., and R. O. Meyer. 1973. "Thermal-Gradient Migration of Helium Bubbles in UO2." <u>J. Nucl. Mat.</u> 48:143.
- Carlsen, H. 1978. "Fission Gas Release in LWR Fuel Rods Exhibiting Very High Burnup." Paper presented at the IAEA Specialists' Meeting on Fuel Element Performance Computer Modeling, Blackpool, U.K.
- Carroll, R. M., R. B. Perez, and O. Sisman. 1965. "Release of Fission Gas During Fissioning of UO<sub>2</sub>." <u>J. Am. Cer. Soc</u>. 48:55.
- Core Performance Branch, U. S. Nuclear Regulatory Commission. 1975. <u>The Role of Fission Gas Release in Reactor Licensing</u>. NUREG-75/077. Washington, D.C.
- Cornell, R. M., M. V. Speight and B. C. Master. 1969. "The Role of Bubbles in Fission Gas Release from Uranium Dioxide." J. Nucl. Mat. 30(1&2):170.
- Cox, C. M., and F. J. Homan. 1970. "Performance Analysis of a Mixed-Oxide LMFBR Fuel Pin." Nucl. Appl. and Tech. 9(3):317-325.
- Dias, J. W., D. Okrent and R. C. Erdmann. 1974. "Fission Gas Behavior in Fast Reactor Fuel under Transient Power Conditions." <u>Nucl. Tech.</u> 24(1): 20-31.
- Dias, J. W., and R. B. Poeppel. 1973. <u>Transient Swelling Studies with the GRASS Code</u>. ANL-7992, Argonne National Laboratory, Hinsdale, Illinois.
- Dollins, C. C. 1973. "Fission Gas Swelling and Long-Range Migration at Low Temperatures." J. Nucl. Mat. 49(1):10-20.
- Dollins, C. C. 1976. "On Fission Gas Swelling below  $\sim 1,200^{\circ}$ C." <u>J. Nucl. Mat.</u> 60(1):107-110.
- Dollins, C. C., and H. Ocken. 1970. "A Fission Gas Swelling Model Incorporating Re-Solution Effects." <u>Nucl. Appl. and Tech.</u> 9:141.
- Dollins, C. C., and F. A. Nichols. 1976. <u>Swelling and Gas Release in UO, at Low and Intermediate Temperatures</u>. WAPD-T-2654, Bettis Atomic Power Laboratory, West Mifflin, Pennsylvania.
- Dutt, D. S., and R. B. Baker. 1975. <u>SIEX, A Correlated Code for the Prediction of LMFBR Fuel Thermal Performance</u>. <u>HEDL-TME 74-55</u>, Hanford Engineering Development Laboratory, Richland, Washington.

- Dutt, D. S., D. C. Bullington, R. B. Baker and L. A. Pember. 1972. "A Correlated Fission Gas Release Model for Fast Reactor Fuels." ANS Trans. 15:198.
- Esteves, R., et al. 1975. "Elementary Model for Nonequilibrium Fission Gas Behavior in Fast Transient." ANS Trans. 21:180.
- Esteves, R. G. 1975. <u>Fission Gas Behavior during Fast Thermal Transients</u>. Ph.D. Dissertation in Engineering, University of California, Los Angeles.
- Friskney, C. A., and J. A. Turnbull. 1979. "The Characteristics of Fission Gas Release from Uranium During Irradiation." J. Nucl. Mat. 70:184-198.
- Friskney, C. A., J. A. Turnbull, F. A. Johnson, A. J. Walter and J. R. Findlay. 1977. "The Characteristics of Fission Gas Release from Monocrystalline Uranium Dioxide During Irradiation." J. Nucl. Mat. 68:186-192.
- Greatley, J., and R. Hargreaves. 1979. "The Measured Emission of Fission-Product Gases from Operating UO<sub>2</sub> Fuel." J. Nucl. Mat. 79:235-245.
- Gruber, E. E. 1967. "Calculated Size Distributions for Gas Bubble Migration and Coalescence in Solids." J. Appl. Phys. 38:243.
- Gruber, E. E. 1973. "Migration and Coalescence of Fission Gas Bubbles in Oxide Fuels by Surface Diffusion." In <u>Proceedings of the International Conference on Physical Metallurgy of Reactor Fuel Elements</u>. CEGB, Berkeley, U.K. September.
- Gruber, E. E. 1974. <u>Calculation of Transient Fission-Gas Release from Oxide</u> Fuels. ANL-8143, Argonne National Laboratory, Hinsdale, Illinois.
- Gruber, E. E. 1976. "A Generalized Parametric Model for Transient Fission-Gas Release and Swelling in Oxide Fuels." ANS Trans. 24:279.
- Hargreaves, R., and D. A. Collins. 1976. "A Quantitative Model for Fission Gas Release and Swelling in Irradiated Uranium Dioxide." J. Br. Nucl. Energy Soc. 15:311.
- Hayns, M. R., and R. Bullough. 1975. "The Nucleation and Growth of Fission Gas Bubbles." In <u>Proceedings of Symposium on Thermodynamics of Nuclear Materials</u> 1974. SM-190/15, October 20-25, IAEA, Vienna, Austria.
- Hayns, M. R., and M. H. Wood. 1976. <u>Factors Influencing Fission Gas Release</u> and Swelling in Nuclear Fuels. AERE-R 8153, Harwell, U.K. February.
- Hayns, M. R., and M. H. Wood. 1976. "On the Rate Theory Model for Fission Gas Behavior in Nuclear Fuels." J. Nucl. Mat. 59:293-302.

- Hayns, M. R. and M. H. Wood. 1977. "Models of Fission Gas Behavior in Fast Reactor Fuels under Steady State and Transient Conditions." J. Nucl. Mat. 67:155-170.
- Hering, W., and R. Manzel. 1978. <u>Measured Fission Gas Release Data from PWR Fuel Rods</u>. NUREG-0418, Appendix C, Nuclear Regulatory Commission, Washington, DC
- Hoffman, J. P., and D. H. Coplin. 1964. The Release of Fission Gases from Uranium Dioxide Pellet Fuel Operated at High Temperatures. GEAP-4596, General Electric Co., San Jose, California.
- Johnson, D. L., and G. L. Hofman. 1974. "A Parametric Approach to the Release of Fission Gas from U, Pu-0<sub>2</sub> Irradiated in EBR-II." ANS Trans. 19:138.
- Klima, J., M. Podest and V. Vins. 1974. "Contribution to the Studies of Fission Gas Released from Irradiated Uranium Oxide Pellets." In <u>Proceedings of the Behavior and Chemical State of Irradiated Ceramic Fuels Conference</u>, pp. 221-224. IAEA, Vienna, Austria.
- Lewis, W. B. 1966. "Engineering for the Fission Gas in UO<sub>2</sub> Fuel." Nucl. Appl. and Tech. 2(2):171.
- Lewis, W. B., J. R. MacEwan, W. H. Stevens and R. G. Hart. 1964. "Fission-Gas Behavior in UO, Fuel." In <u>Proceedings of the Eleventh International Conference on Peaceful Uses of Atomic Energy</u>. IAEA:405, Vienna, Austria.
- Littlechild, J. E., G. G. Butler and G. W. Lester. 1973. "Production of Burnable Poison Oxide Fue." In <u>Proceedings of the International Conference on Nuclear Fuel Performance</u>, p. 65. British Nuclear Energy Society, London.
- MacEwan, J. R., and W. H. Stevens. 1964. "Xenon Diffusion in UO<sub>2</sub>: Some Complicating Factors." <u>J. Nucl. Mat.</u> 11(1):77-93.
- Matzke, H. 1966. On the Effect of TiO<sub>2</sub> Additions on Defect Structure, Sintering and Gas Release of UO<sub>2</sub>.° AECL-2585, Atomic Energy of Canada Limited, Chalk River, Ontario, Canada.
- Meyer, R. O. 1976. "Fission Gas Release at High Burnups." Presentation to the ACRS, U.S. Nuclear Regulatory Commission, October 15.
- Meyer, R. O., C. E. Beyer and J. C. Voglewede. 1978. <u>Fission Gas Release</u> from Fuel at High Burnup. NUREG-0418, Core Performance Branch. U.S. Nuclear Regulatory Commission.
- Michels, L. C., and R. B. Poeppel. 1972. "Influence of Grain Boundaries on Fission Gas Release in Mixed-Oxide Fuels." ANS Trans. 15:199.

- Nelson, R. S. 1969. "The Stability of Gas Bubbles in an Irradiation Environ-ment." J. Nucl. Mat. 31(2):153.
- Nichols, F. A. 1969. "Kinetics of Diffusional Motion of Pores in Solids." J. Nucl. Mat. 30(1&2):143.
- Nichols, F. A., and H. R. Warner. 1971. "Swelling and Gas-Release Models for Oxide Fuel Rods." In Proceedings of Conference on Fast Reactor Fuel Element Technology, p. 267, ANS, Hinsdale, Illinois.
- Notley, M. J. F. 1970. "A Computer Program to Predict the Performance of UO<sub>2</sub> Fuel Elements Irradiated at High Power Outputs to a Burnup of 10,000 MWd/MTU." Nucl. Appl. and <u>Tech.</u> 9:195.
- Notley, M. J. F., and I. J. Hastings. 1978. Fuel Modelling; ELESIM Code Calculations of Stable Fission Product Gas Releases. PR-FM- 45P, Sec. 2.2, Chalk River Nuclear Laboratory, Chalk River, Ontario.
- Pati, S. R., D. E. Bessette and L. V. Corsetti. 1979. "Fission Gas Release and Dimensional Changes of Test Fuel Rods Containing Densifying and Nondensifying Fuel." Paper presented at the ANS Topical Meeting on LWR Fuel Performance. Portland, Oregon.
- Poeppel, R. B. 1971. "An Advanced Gas Release and Swelling Subroutine." In Proceedings of Conference on Fast Reactor Fuel Element Technology, pp. 311-326, ANS, Hinsdale, Illinois.
- Rest, J. 1975. "SST: A Computer Code to Predict Fuel Response and Fission Product Release from Light-Water Reactor Fuels during Steady-State and Transient Conditions." ANS Trans. 22:462.
- Rest, J. 1978. <u>GRASS-SST: A Comprehensive, Mechanistic Model for the Prediction of Fission-gas Behavior in UO<sub>2</sub>-base Fuels during Steady-state and <u>Transient Conditions</u>. <u>NUREG/CR0202</u>, Argonne National Laboratory, Argonne, Illinois.</u>
- Rest, J., M. G. Seitz, L. R. Kelman and S. M. Gehl. 1976. "Development and Experimental Verification of SST: A Steady-State and Transient Fuel Response and Fission-Product Release Code." Paper presented at the OECD-CNSI meeting on the Behavior of Water Reactor Fuel Elements under Accident Conditions. Norway.
- Roberts, E., et al. 1977. "Fuel Modeling and Performance of High Burnup Fuel Rods." <u>Water Reactor Fuel Performance</u>, pp. 133-146. ANS Topical Meeting, St. Charles, Illinois.
- Roberts, E., M. G. Balfor, G. W. Hopkins and W. R. Smalley. 1977. "Fuel Modeling and Performance of High Burnup Fuel Rods." <u>Water Reactor Fuel Performance</u>, pp. 133-146. ANS Topical Meeting, St. Charles, Illinois. (An abstract of this article is given in Appendix III.)

- Roberts, J. T. A., et al. 1979. <u>LWR Fuel Performance Program: Progress in 1978</u>. EPRI-NP-1024-SR, Electric Power Research Institute, Palo Alto, California.
- Ronchi, C., and H. J. Matzke. 1972. <u>Calculations on the In-Pile Behaviour of Fission Gas in Oxide Fuels</u>. An Extended Parametric Study. <u>EUR-4877e</u>, European Atomic Energy Community, Ispra, Italy.
- Smalley, W. R. 1971. Saxton Core II Fuel Performance Evaluation, Part I: Materials. WCAP-3385-56, Part I.
- Smalley, W. R. 1974. <u>Evaluation of Saxton Core III Fuel Materials Performance</u>. WCAP-3385-57.
- Speight, M. V. 1969. "A Calculation of the Migration of Fission Gas in Material Exhibiting Precipitation and Re-Solution of Gas Atoms under Irradiation." Nucl. Sci. and Engng. 37(2):180.
- Stahl, D., and T. J. Patrician. 1974. <u>Fission Gas Behavior During a Mild Over-Power Transient</u>. ANL-8069, Argonne National Laboratory, Argonne, Illinois.
- Szuta, M. 1975. "Fission Gas Release from UO<sub>2</sub> Fuel During Low-Temperature Irradiation." J. Nucl. Mat. 58:278-284.
- Tucker, M. O., and J. A. Turnbull. 1975. "The Morphology of Interlinked Porosity in Nuclear Fuels." Proc. Roy. Soc. Lond. A. 343:299.
- Turnbull, J. A. 1974. "The Effect of Grain Size on the Swelling and Gas Release Properties of UO<sub>2</sub> during Irradiation." <u>J. Nucl. Mat.</u> 50:62.
- Turnbull, J. A., and C. A. Friskney. 1975. "The Release of Fission Products from Nuclear Fuel During Irradiation by Both Lattice and Grain Boundary Diffusion." J. Nucl. Mat. 58(1):31.
- Turnbull, J. A., and C. A. Friskney. 1978. "Relation between Microstructure and the Release of Unstable Fission Products During High Temperature Irradiation of UO2." J. Nucl. Mat. 71:238-248.
- Turnbull, J. A., and M. O. Tucker. 1974. "The Release of Fission Gas from Nuclear Fuels during Temperature Transients." J. Nucl. Mat. 50(1):53.
- Turnbull, J. A., and M. O. Tucker. 1974. "The Release of Unstable Fission Products during Variable Reactor Operating Histories." J. Nucl. Mat. 50(1):47.
- Turnbull, J. A., and M. O. Tucker. 1974. "Swelling in UO<sub>2</sub> under Conditions of Gas Release." Phil. Mag. 30:47.

- Wada, T., K. Noro and K. Tsukui, 1973. "Behavior of U02-Gi203 Fuel." In The Proceedings of the International Conference on Nuclear Fuel Performance, paper 63. British Nuclear Energy Society, London.
- Wang, W. L. 1976. "Relaxation Times for Nonequilibrium Fission-Gas Bubbles." ANS Trans. 24:280.
- Warner, J. R., and F. A. Nichols. 1970. "A Statistical Fuel Swelling and Fission Gas Model." Nucl. Appl. and Tech. 9:148-166.
- Williamson, H. E., and D. C. Ditmore. 1971. "Current BWR Fuel Design and Experience." Reactor Tech. 14(1).
- Wood, M. H., and M. R. Hayns. 1975. Modelling Fission Gas Release and Swelling in Fast Reactor Fuel Pins. AERE-R 8012, Harwell, U.K.
- Wood, M. H., and M. R. Hayns. July 1976. A Concise Review of Harwell Modelling of Fission Gas Behavior. AERE-R-8372. Harwell, U.K.
- Yuill, W. A., V. F. Baston and J. H. McFadden. 1971. <u>An Analytical Model</u>
  <u>Describing the Behavior of Fission Products in Operating Fuel Pins</u>. IN-1467,
  Idaho Nuclear Corporation.
- Zimmermann, H. 1975. "Fission Gas Behavior in Oxide Fuel Elements of Fast Breeder Reactors." Nucl. Tech. 28:127-133.
- Zimmermann, H. 1978. "Investigations on Swelling and Fission Gas Behavior in  ${\rm UO_2}$ ." J. Nucl. Mat. 75:154-161.

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# APPENDIX I

ABSTRACTS OF REFERENCES RELATED

TO ANALYTICAL MODELS FOR FGR

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- Baldewicz, W. L. 1977. State-of-the-Art of Fission Gas Release from LWR Fuels. UCLA-ENG-7740, University of California, Los Angeles.
- A review is made on the various models and some experimental data describing the fission gas behavior in solid oxide fuel. The dependence of fission gas on fuel burnup in LWRs is discussed in terms of current data from fast breeder and LWR fuels, empirical correlations and analytical models.

Various analytical models used to calculate the amount of fission gas released in the irradiated fuel are compared. Individual models were discussed in the following publications:

- Bogensberger, H. G., and C. Ronchi. 1976. "Effects Due to Fission Gas During Unprotected Overpower Transients in a Liquid Metal Fast Breeder Reactor." Nucl. Tech. 29:73-85.
- Booth, A. H. 1957. A Method of Calculating Fission Gas Diffusion from UO<sub>2</sub> Fuel and its Application to the X-2-f Loop Test.

  AECL No. 496, Chalk River, Ontario.
- Buescher, B. J., and R. O. Meyer. 1973. "Thermal-Gradient Migration of Helium Bubbles in UO<sub>2</sub>." J. Nucl. Mat. 48:143.
- Cornell, R. M., M. V. Speight and B. C. Master. 1969. "The Role of Bubbles in Fission Gas Release from Uranium Dioxide." J. Nucl. Mat. 30(1&2):170.
- Dias, J. W., D. Okrent and R. C. Erdmann. 1974. "Fission Gas Behavior in Fast Reactor Fuel under Transient Power Conditions." Nucl. Tech. 24(1):20-31.
- Dias, J. W., and R. B. Poeppel. 1973. <u>Transient Swelling Studies</u> with the GRASS Code. ANL-7992, Argonne National Laboratory, Hinsdale, Illinois.
- Dollins, C. C. 1973. "Fission Gas Swelling and Long-Range Migration at Low Temperatures." J. Nucl. Mat. 49(1):10-20.
- Dollins, C. C. 1976. "On Fission Gas Swelling below  $\sim 1,200^{\circ}$ C." J. Nucl. Mat. 60(1):107-110.
- Dollins, C. C., and H. Ocken. 1970. "A Fission Gas Swelling Model Incorporating Re-Solution Effects." <u>Nucl. Appl. and Tech.</u> 9:141.
- Dollins, C. C., and F. A. Nichols. 1976. <u>Swelling and Gas</u>
  Release in UO<sub>2</sub> at Low and Intermediate Temperatures. WAPD-T-2654,
  Bettis Atomic Power Laboratory, West Mifflin, Pennsylvania.

- Esteves, R., et al. 1975. "Elementary Model for Nonequilibrium Fission Gas Behavior in Fast Transient." ANS Trans. 21:180.
- Esteves, R. G. 1975. <u>Fission Gas Behavior during Fast Thermal Transients</u>. Ph.D. Dissertation in Engineering, University of California, Los Angeles.
- Gruber, E. E. 1976. "A Generalized Parametric Model for Transient Fission-Gas Release and Swelling in Oxide Fuels." ANS Trans. 24:279.
- Gruber, E. E. 1967. "Calculated Size Distributions for Gas Bubble Migration and Coalescence in Solids." J. Appl. Phys. 38:243.
- Gruber, E. E. 1974. <u>Calculation of Transient Fission-Gas Release</u> from Oxide Fuels. ANL-8143, Argonne National Laboratory, Hinsdale, Illinois.
- Gruber, E. E. 1973. "Migration and Coalescence of Fission Gas Bubbles in Oxide Fuels by Surface Diffusion." In <u>Proceedings of</u> the International Conference on Physical Metallurgy of Reactor Fuel Elements. CEGB, Berkeley, U.K. September.
- Hargreaves, R., and D. A. Collins. 1976. "A Quantitative Model for Fission Gas Release and Swelling in Irradiated Uranium Dioxide." J. Br. Nucl. Energy Soc. 15:311.
- Hayns, M. R., and R. Bullough. 1975. "The Nucleation and Growth of Fission Gas Bubbles." In <u>Proceedings of Symposium on Thermodynamics of Nuclear Materials</u> 1974. SM-190/15, October 20-25, IAEA, Vienna, Austria.
- Hayns, M. R., and M. H. Wood. 1976. <u>Factors Influencing Fission Gas Release and Swelling in Nuclear Fuels</u>. AERE-R 8153, Harwell, U. K. February.
- Hayns, M. R., and M. H. Wood. 1976. "On the Rate Theory Model for Fission Gas Behavior in Nuclear Fuels." J. Nucl. Mat. 59:293-302.
- Michels, L. C., and R. B. Poeppel. 1972. "Influence of Grain Boundaries on Fission Gas Release in Mixed-Oxide Fuels." ANS Trans. 15:199.
- Nelson, R. S. 1969. "The Stability of Gas Bubbles in an Irradiation Environment." J. Nucl. Mat. 31(2):153.

- Nichols, F. A. 1969. "Kinetics of Diffusional Motion of Pores in Solids." J. Nucl. Mat. 30(1&2):143.
- Nichols, F. A., and H. R. Warner. 1971. "Swelling and Gas-Release Models for Oxide Fuel Rods." In Proceedings of Conference on Fast Reactor Fuel Element Technology, p. 267, ANS, Hinsdale, Illinois.
- Poeppel, R. B. 1971. "An Advanced Gas Release and Swelling Subroutine." In Proceedings of Conference on Fast Reactor Fuel Element Technology, pp. 311-326, ANS, Hinsdale, Illinois.
- Rest, J. 1975. "SST: A Computer Code to Predict Fuel Response and Fission Product Release from Light-Water Reactor Fuels during Steady-State and Transient Conditions." ANS Trans. 22:462.
- Rest, J., M. G. Seitz, L. R. Kelman and S. M. Gehl. 1976.
  "Development and Experimental Verification of SST: A SteadyState and Transient Fuel Response and Fission-Product Release
  Code." Paper presented at the OECD-CNSI meeting on the Behavior
  of Water Reactor Fuel Elements under Accident Conditions.
  Norway.
- Ronchi, C., and H. J. Matzke. 1972. <u>Calculations on the In-Pile</u>
  Behaviour of Fission Gas in Oxide Fuels. An Extended Parametric
  Study. EUR-4877e, European Atomic Energy Community, Ispra,
  Italy.
- Speight, M. V. 1969. "A Calculation of the Migration of Fission Gas in Material Exhibiting Precipitation and Re-Solution of Gas Atoms under Irradiation." Nucl. Sci. and Engng. 37(2):180.
- Stahl, D., and T. J. Patrician. 1974. <u>Fission Gas Behavior</u>
  <u>During a Mild Over-Power Transient</u>. <u>ANL-8069</u>, Argonne National Laboratory, Argonne, Illinois.
- Tucker, M. O., and J. A. Turnbull. 1975. "The Morphology of Interlinked Porosity in Nuclear Fuels." Proc. Roy. Soc. Lond. A. 343:299.
- Turnbull, J. A. 1974. "The Effect of Grain Size on the Swelling and Gas Release Properties of UO<sub>2</sub> during Irradiation." J. Nucl. Mat. 50:62.
- Turnbull, J. A., and C. A. Friskney. 1975. "The Release of Fission Products from Nuclear Fuel During Irradiation by Both Lattice and Grain Boundary Diffusion." J. Nucl. Mat. 58(1):31.

- Turnbull, J. A., and M. O. Tucker. 1974. "The Release of Fission Gas from Nuclear Fuels during Temperature Transients."
  J. Nucl. Mat. 50(1):53.
- Turnbull, J. A., and M. O. Tucker. 1974. "The Release of Unstable Fission Products during Variable Reactor Operating Histories." J. Nucl. Mat. 50(1):47.
- Turnbull, J. A., and M. O. Tucker. 1974. "Swelling in UO<sub>2</sub> under Conditions of Gas Release." Phil. Mag. 30:47.
- Wang, W. L. 1976. "Relaxation Times for Nonequilibrium Fission-Gas Bubbles." ANS Trans. 24:280.
- Warner, J. R., and F. A. Nichols. 1970. "A Statistical Fuel Swelling and Fission Gas Model." <u>Nucl. Appl. and Tech.</u> 9:148-166.
- Wood, M. H., and M. R. Hayns. 1975. Modelling Fission Gas Release and Swelling in Fast Reactor Fuel Pins. AERE-R 8012, Harwell, U.K. July.
- Core Performance Branch, U. S. Nuclear Regulatory Commission. 1975. The Role of Fission Gas Release in Reactor Licensing. NUREG-75/077.

The release of fission gases from oxide pellets to the fuel rod internal voidage (gap) is reviewed with regard to the required safety analysis in reactor licensing. Significant analyzed effects are described, prominent gas release models are reviewed, and various methods used in the licensing process are summarized. The report thus serves as a guide to the literature including company reports and government documents up to 1974. The state of the art of gas release analysis is also discussed, with emphasis on evaluating the current methods used in reactor licensing.

Hayns, M. R. and M. H. Wood. 1977. "Models of Fission Gas Behavior in Fast Reactor Fuels under Steady State and Transient Conditions." J. Nucl. Mat. 67:155-170.

Two models are used to discuss gas release and swelling in a fast reactor oxide fuel element during steady state and transient conditions. The first model is based on the random motion and coalescence of gas atoms and bubbles within fuel grains and on the re-solution of gas atoms from bubbles. This model is used to describe fission gas behavior at low temperatures. At higher temperatures and/or under transient conditions, the second model, based on the biased migration, coalescence and re-solution of gas bubbles under a thermal gradient driving force, is more feasible and should be used instead of the first model.

In-pile and out-of-pile experimental data were obtained from test fuels irradiated at low burnups (<3.2%) under both steady power history and tran-

transient conditions. Comparison between experimental and calculated results shows that using different models for different temperature regimes can appropriately describe the physical mechanisms of fission gas release in fast reactor fuels.

MacEwan, J. R., and W. H. Stevens. 1964. "Xenon Diffusion in  $\rm UO_2$ : Some Complicating Factors." J. Nucl. Mat.  $\rm 11(1):77-93$ .

Sintered and singlecrystal powder UO<sub>2</sub> specimens containing 1.9 w/o  $^{235}$ U were irradiated (average neutron flux 9 x  $^{10^{12}}$  neutrons/cm²/sec) and annealed (1400°C, 3 to 7 hr) to study the diffusion of fission xenon. Results indicate that increasing the irradiation exposure above  $^{10^{15}}$  fissions/cm³ reduces the apparent diffusion coefficient.

A possible explanation of this phenomenon is that an appreciable fraction of the fission xenon is immobilized on lattice traps after irradiation exposure has exceeded  $10^{15}$  fissions/cm $^3$ . These lattice traps include the original closed porosities in the specimens and irradiation induced vacancy clusters.

The fraction F of trapped xenon atoms is calculated by the formula F = A[1-exp(-2n)], where n is the exposure, A and 2 are constants. Nontrapped xenon atoms are assumed to move by atomic diffusion.

Rest, J. 1978. GRASS-SST: A Comprehensive, Mechanistic Model for the Prediction of Fission-gas Behavior in UO<sub>2</sub>-base Fuels during Steady-state and Transient Conditions. NUREG/CR-0202, Argonne National Laboratory, Argonne, Illinois.

The Steady-State and Transient Gas Release and Swelling Subroutine (GRASS-SST) is based on the GRASS code first reported by Poeppel. While GRASS was originally developed for the prediction of fission-gas behavior in LMFBR fuels during steady-power irradiations, GRASS-SST is designed to predict fission gas behavior in UO,-based fuels during both steady state and transient conditions. This most current version of the GRASS code has evolved through comparisons of code predictions with the fission-gas releases and physical phenomena that occur during LWR operation, and during transient Direct Electrical Heating (DEH) tests on irradiated LMFBR and LWR fuel.

GRASS-SST calculations include the effects of production of gas from fissioning uranium atoms, bubble nucleation, bubble diffusion, bubble migration, bubble coalescence, re-solution, temperature and temperature gradients, interlinked porosity, and fission-gas interaction with structural defects based on both the distribution of fission-gas within the fuel and the amount of fission-gas released from the fuel. Swelling and total fission-gas release as a function of time for steady-state and transient conditions are also calculated. Fission gas released from the fuel reaches the fuel surface by successively diffusing from the grains to grain boundaries and then to the grain edges, where the gas is released through a network of interconnected tunnels of fission-gas and fabricated porosity.

# Wood, M. H., and M. R. Hayns. July 1976. A Concise Review of Harwell Modelling of Fission Gas Behavior. AERE-R-8372.

A review is made on the recent Harwell studies of fission gas behavior in nuclear fuels. Following extensive investigations with the rather sophisticated rate theory models, the authors established the important physical mechanisms of fission gas release and swelling under a wide range of conditions.

It was concluded that two different models are required to explain the gas release and swelling over the range of conditions to be expected in normally operating fast reactor oxide fuel, each model being dominant in different regions. A temperature of  $\sim 1500\,^{\circ}\text{C}$  was found to define the dividing line between the two regions. In the cooler parts of the fuel, the gas behavior is dominated by the random diffusion of both single gas atoms and small gas bubbles, and it is necessary to allow for gas atom re-solution and the spatial variation of the concentrations within the grain. In the higher temperature region, it is necessary to model the directional motion of gas bubbles under the influence of both high temperatures and temperature gradients. Experimental observations of gas release and swelling explained by these models are in turn used as a guide to derive more simplified and economical models.

# Yuill, W. A., V. F. Baston and J. H. McFadden. 1971. An Analytical Model Describing the Behavior of Fission Products in Operating Fuel Pins. IN-1467, Idaho Nuclear Corporation.

An analytical model was developed to describe the release of fission products to the fuel-cladding gap and the distribution of fission products in the fuel during reactor operation. This model was derived from the postulates that 1) the driving force of fission product migration is the Gibbs free energy gradient, and 2) this energy gradient is affected by both temperature and concentration gradients.

Noble gases in the fuel lattice are considered defects that can migrate toward the hot central region of the fuel pin. Gases that are released after reaching the central void or a fissure in the fuel can migrate along the cracks toward the surface of the fuel, or reenter the fuel as a defect.

Predicted release of noble gases compare well with results obtained from in-pile reactor experiments. The authors conclude that models including both a concentration and a thermal gradient as the driving force give a more complete description of fission product release than models considering concentration gradient alone.

# APPENDIX II

# ABSTRACTS OF REFERENCES RELATED TO EMPIRICAL CORRELATIONS FOR FGR



ANS (American Nuclear Society) Working Group 5.4. 1977. Status Report: Fission Product Release from UO<sub>2</sub> Fuel. Report No. N218.

In an effort to produce a standard analytical model to predict the release of radioactive volatile and gaseous fission products from oxide fuel pellets to the internal fuel rod void space, the ANS Working Group 5.4, established in 1974, has empirically fitted the Booth model to a selected set of thermal reactor high temperature data. An enhancement of release at high burnup is believed to occur. Therefore, the Working Group establishes a mathematical formulation to account for the time and burnup dependent diffusion parameters as well as a variable power history.

Arguments for and against using mixed-oxide data to predict UO, outcome are presented. Questions regarding diffusion parameters for non-noble gases, burnup dependence and low temperature release have not been resolved. It is concluded that additional experimental information is needed for the analysis of fission gas behavior.

Baldewicz, W. L. 1977. State-of-the-Art of Fission Gas Release from LWR Fuels. UCLA-ENG-7740, University of California, Los Angeles, California.

A review is made on the various models and some experimental data describing the fission gas behavior in solid oxide fuel. The dependence of fission gas on fuel burnup in LWRs is discussed in terms of current data from fast breeder and LWR fuels, empirical correlations and analytical models.

A number of empirical correlations used to predict the amount of fissional gas released in the irradiated fuel are quantitatively compared. These correlations are discussed in the following references:

- Bailey, W. E., et al. 1969. "Effect of Temperature and Burnup on Fission Gas Release in Mixed Oxide Fuel." <u>Ceramic Nuclear Fuels</u>, Special Publication No. 2:195-210, American Ceramic Society, Columbus, Ohio.
- Baston, V. F., T. H. MacFadden and W. A. Yuill. 1971. Analytical Method for Calculating Steady-State Fission Gas Release--Fission Product Fuel Model (FPFM) Code. ANCR-1010, Aerojet Nuclear Co., Idaho Falls, Idaho.
- Beyer, C. E., and C. R. Hann. 1974. <u>Prediction of Fission Gas</u>
  <u>Release from UO, Fuel</u>. BNWL-1875, Pacific Northwest Laboratory,
  <u>Richland</u>, Washington.
- Beyer, C. E., and R. O. Meyer. 1976. "Semiempirical Model for Radioactive Fission Gas Release from UO<sub>2</sub>." ANS Trans. 23:172.
- Core Performance Branch, U. S. Nuclear Regulatory Commission. 1975. The Role of Fission Gas Release in Reactor Licensing. NUREG-75/77, Washington, DC.

- Cox, C. M., and F. J. Homan. 1970. "Performance Analysis of a Mixed-Oxide LMFBR Fuel Pin." Nucl. Appl. and Tech. 9(3):317-325.
- Dutt, D. S., D. C. Bullington, R. B. Baker and L. A. Pember. 1972. "A Correlated Fission Gas Release Model for Fast Reactor Fuels." ANS Trans. 15:198.
- Hoffman, J. P., and D. H. Coplin. 1964. The Release of Fission Gases from Uranium Dioxide Pellet Fuel Operated at High Temperatures. GEAP-4596, General Electric Co., San Jose, California.
- Johnson, D. L., and G. L. Hofman. 1974. "A Parametric Approach to the Release of Fission Gas from U, Pu-O<sub>2</sub> Irradiated in EBR-II." ANS Trans. 19:138.
- Lewis, W. B. 1966. "Engineering for the Fission Gas in UO<sub>2</sub> Fuel." Nucl. Appl. and Tech. 2(2):171.
- Meyer, R. O. 1976. "Fission Gas Release at High Burnups." Presentation to the ACRS, U.S. Nuclear Regulatory Commission, October 15.
- Notley, M. J. F. 1970. "A Computer Program to Predict the Performance of UO<sub>2</sub> Fuel Elements Irradiated at High Power Outputs to a Burnup of 10,000 MWd/MTU." Nucl. Appl. and Tech. 9:195.
- Williamson, H. E., and D. C. Ditmore. 1971. "Current BWR Fuel Design and Experience." Reactor Tech. 14(1).
- Bellamy, R. G., and J. B. Rich. 1969. "Grain-Boundary Gas Release and Swelling in High Burnup Uranium Dioxide." J. Nucl. Mat. 33:64-76. (An abstract of this article is given in Appendix III.)
- Friskney, C. A., and J. A. Turnbull. 1979. "The Characteristics of Fission Gas Release from Uranium During Irradiation." J. Nucl. Mat. 70:184-198.

Samples of polycrystalline, small (20 to 40 mm) and large (100 to 250 mm) grained UO, (1.46%  $^{235}$ U) in spherical form (diameter 1.2 mm) were irradiated in the DIDO heavy water reactor for a total of 19 reactor cycles (28 days/cycle) to a cumulative burnup of 6407 MWd/MTU.

Following the same practice as that reported in the previous paper (Friskney, et al., 1977), release rates for the Xe and Kr isotopes were measured in the temperature range 700 to 1550°C. Experimental results indicated that the dependence of fractional gas release on decay constant and diffusion coefficient for small and large grains and polycrystalline UO, were similar to that for monocrystalline UO. Therefore, the same empirical correlation developed in the previous paper was applicable to both mono- and polycrystalline UO.

Friskney, C. A., J. A. Turnbull, F. A. Johnson, A. J. Walter and J. R. Findlay. 1977. "The Characteristics of Fission Gas Release from Monocrystalline Uranium Dioxide During Irradiation." J. Nucl. Mat. 68:186-192.

Single crystals of natural composition (0.72%  $^{235}$ U) stoichiometric UO, in the form of 2.5 mm diameter right cylinders were irradiated in the DIDO heavy water reactor at Harwell for a period of 8 reactor cycles (28 days/cycle) to a cumulative burnup of 1640 MWd/MTU. The temperature range during the experiment was 700 to 1550°C.

Release rates for the rare gases <sup>85m</sup>Kr, <sup>87</sup>Kr, <sup>88</sup>Kr, <sup>133</sup>Xe, <sup>135</sup>Xe and <sup>138</sup>Xe were measured and analyzed in terms of diffusion of the rare gases and their halogen precursors. The diffusion coefficients for xenon and iodine were found to be similar whilst krypton also had a similar mobility at ~1200°C but otherwise diffused more slowly.

Using the least square technique to analyze the results, an empirical correlation was derived, in which the fractional release of a rare gas isotope was a trigonometric function of diffusion coefficients and decay constants of the rare gas isotope and its immediate halogen precursor.

Greatley, J., and R. Hargreaves. 1979. "The Measured Emission of Fission-Product Gases from Operating UO<sub>2</sub> Fuel." <u>J. Nucl. Mat.</u> 79:235-245.

A small stainless steel capsule containing 3 UO, pellets (3.5% <sup>235</sup>U) was irradiated in the Windscale Advanced Gas Cooled Reactor. During the 2-year experiment, the burnup level was below 15,000 MWd/MTU, and the centerline temperature of the fuel was below 1170°C.

Isotope concentration in the purge gas released during irradiation was measured in three ways: continuous measurement by single channel analyzers, direct measurement by the pulse height analyzer, and gas sampling to confirm the results obtained by direct measurement.

Results indicated that, in the ranges of temperature and burnup during this experiment, the steady-state fission gas release was principally due to the knock-out mechanism occurring at the fuel surface. Fractional release showed a strong dependence on isotopic half-life (or decay constant), which is in good agreement with theory.

- Lewis, W. B., J. R. MacEwan, W. H. Stevens and R. G. Hart. 1964. "Fission-Gas Behavior in UO, Fuel." In Proceedings of the Eleventh International Conference on Peaceful Uses of Atomic Energy. IAEA:405, Vienna, Austria.
- Meyer, R. O., C. E. Beyer and J. C. Voglewede. 1978. Fission Gas Release from Fuel at High Burnup. NUREG-0418, Core Performance Branch. U.S. Nuclear Regulatory Commission, Washington, DC.

The approach used to derive the correction function is to separate the variables (burnup and linear heat generation rate) in the Dutt and Baker correlation, and use the burnup dependence alone for LWR fuels at burnup levels above 20,000 MWd/MTM.

Because the high burnup fission gas release data used to develop the Dutt and Baker correlation were obtained from LMFBR fuels, it was assumed that LWR and LMFBR fuels having the same release fraction at 20,000 MWd/MTM, provided their respective temperatures remained the same. Under this assumption, an arbitrary enhancement factor, which was an exponential function of burnup above 20,000 MWd/MTM, was derived from the Dutt and Baker correlation using nonlinear regression techniques.

Related information, which includes some previously unpublished data, is also summarized to help provide guidance for the analysis of high burnup gas release in licensing situations.

Roberts, E., M. G. Balfor, G. W. Hopkins and W. R. Smalley. 1977. "Fuel Modeling and Performance of High Burnup Fuel Rods." <u>Water Reactor Fuel Performance</u>, pp. 133-146. ANS Topical Meeting, St. Charles, Illinois. (An abstract of this article is given in Appendix III.)

Szuta, M. 1975. "Fission Gas Release from UO<sub>2</sub> Fuel During Low-Temperature Irradiation." J. Nucl. Mat. 58:278-284.

Fission gas release from UO2 fuel during low temperature (<600°C) irradiation is interpreted mathematically in terms of a defect trap model and the knock-out process. By assuming that gases dissolved in the fuel and trapped in bubbles during irradiation are in an "intermediate" (excited) state, and that fission gas release rate is proportional to the knock-out rate and concentration of gas trapped in bubbles, this model can predict the relative proportion of isotopes in the steady-state fission gas release.

The relationship between fission rate and fission gas release rate is also examined by the model calculations. Depending on the fission rate interval considered, the model can predict regimes in which the fission gas release rate is a first or second order function of fission rate.

The proposed model is also used to interpret the experimental results obtained previously by Carroll and by the author.

#### APPENDIX III

# ABSTRACTS OF REFERENCES RELATED TO EXPERIMENTAL DATA ON HIGH BURNUP AND FISSION GAS RELEASE



Baroch, C. J., and M. A. Rigdon. 1973. "Irradiation Behaviour of UO<sub>2</sub> at Burnups From 10 to 80 GWd/tonne U." In <u>Proceedings of the International</u> Conference on Nuclear Fuel Performance, paper 58.

Thirty-three UO2 fuel rods were irradiated in a Babcock & Wilcox PWR to burnups up to 80 GWd/tonne U in 3 years to determine their irradiation performance at high burnup. These test rods (6 to 11.5 in. long, Zirc-4 cladding OD 0.385 to 0.395 in.) were designed to maintain a cladding surface temperature of 650°F and operate at 18 (powder) and 21.5 kW/ft (powder and pellets).

Postirradiation examination includes the analysis of fuel swelling, burnup and fission gas release. For burnup up to 65 GWd/tonne U, the average swelling rates are 1.10% increase in volume per  $10^{20}$  fissions/cc for pellet rods and 0.88% for powder rods. The percentage of fission gas release tends to increase linearly with burnup, ranging from about 10% at 8 GWd/tonne to 88% at 65 GWd/tonne.

While powder fuel experiences a much greater amount of restructuring than pellet fuel, there was no significant difference in fission gas release rates between them.

Bellamy, R. G., and J. B. Rich. 1969. "Grain-Boundary Gas Release and Swelling in High Burnup Uranium Dioxide." J. Nucl. Mat. 33:64-76.

Stainless steel clad UO2 pins were irradiated to burnups ranging from 0.8 to 5.0% burnup at calculated fuel center temperatures of  $\leq 1630\,^{\circ}\text{C}$ . Results show that the fractional release of fission gas from UO2 of maximum calculated centerline temperature below 1250°C increases approximately linearly between 1% and 2.2% burnup. This release appears to be primarily a "knock-out" process. The apparent diffusion coefficient D' is essentially unaffected by burnup in this range.

As burnup increases to above 3%, D' increases rapidly at all temperatures. Fission gas release appears to increase with burnup: at centerline temperature below 1250°C, fractional release is 0.075%/1% burnup for burnup less than 2%. As burnup increases to over 3%, this factor becomes 2.0%/1% burnup.

Bouffioux, P., and E. De Meulemeester. 1978. "Benchmarking Results of the COMETHE Code." <u>COMETHE Publication</u> at the IAEA Specialist Meeting on Fuel Element Performance, Report BN7803-03, Belgonucleaire, Belgium.

Various experiments to investigate the effects of fuel density, gap conductance, burnup and power level on fuel rod behavior were performed to benchmark the results of the COMETHE Code developed in Belgonucleaire.

Results from one of the experiments, the postirradiation examinations of 18 rods irradiated in the BR3/VII reactor, indicate a high sensitivity of fission gas release to power level and burnup. A factor of 40 between the release of the lowest and highest rated rod exists when the variation of power

levels is 25% due to the power tilt through the fuel assembly. Measured fractional fission gas release increases approximately linearly with relative power levels.

To calculate the fractional fission gas release, a version COMETHE III-J was used to establish a model which considers diffusion, trapping and resolution in the matrix of the Booth's sphere. Predicted results compare well with experimental data.

Bouffioux, P., and E. De Meulemeester. 1979. "Prediction of Fission Gas Release at High Burnup." Paper presented at the ANS Topical Meeting on LWR Fuel Performance, Portland, Oregon.

A fission gas release data base has been formed in Belgonucleaire. It covers a wide range of heat ratings and burnup levels for both  ${\rm UO}_2$  and  ${\rm MOX}$  fuels.

In an attempt to compare experimental results with predictions from the NRC high burnup correlation and the code COMETHE developed by Belgonucleaire, UO2 fuel rods with linear heat generation rates of 270 W/cm to 850 W/cm and MOX fuel rods with 450 W/cm were examined. Results show that fission gas release depends strongly on linear heat generation rate, rather than burnup.

In LMFBRs, high fuel temperatures lead to pronounced gaseous bubble swelling and columnar grain growth. These two phenomena produce very high fission gas release. Using the NRC correlation (developed on the basis of LMFBR fuels) to predict fission gas release from LWR fuels would result in overestimation.

Carlsen, H. 1978. "Fission Gas Release in LWR Fuel Rods Exhibiting Very High Burnup." Paper presented at the IAEA Specialists' Meeting on Fuel Element Performance Computer Modeling, Blackpool, U.K.

Two fuel rods containing sintered UO2 pellets clad in Zirc-2 tubing were irradiated in the heavy-water test reactor DR3 at Risé up to burnup levels of 38,000 MWd/MTUO2. Postirradiation examination of the two rods indicates that the fission gas release fractions are 36.6 and 48.6%. Using a modified Beyer-Hann model incorporating the NRC high burnup correction factor, the fuel performance code WAFER-2 predicts the release fractions to be 41.5 and 56.6%, respectively. This overprediction of 15% is considered reasonable, for no similar agreement could be obtained without the burnup dependence of the release model.

Dutt, D. S., and R. B. Baker. 1975. SIEX, A Correlated Code for the Prediction of LMFBR Fuel Thermal Performance. HEDL-TME 74-55, Hanford Engineering Development Laboratory, Richland, Washington.

The SIEX computer program is a steady-state heat transfer code developed to provide thermal performance calculations for a mixed-oxide fuel element in a fast neutron environment. Fuel restructuring, fuel-cladding heat conduction and fission gas release are modeled to provide assessment of the temperatures.

Modeling emphasis has been placed on correlations related to measurable quantities (local burnup, local linear heat rate, number of full power reactor cycles, etc.) from EBR-II irradiation tests and the inclusion of these correlations in a physically based computational scheme.

The mechanisms of fission gas release are described by bubble migration to the central void and diffusion in the high fuel temperature regions, and release through grain boundaries in medium and lower temperature zones. Fission gas release data are analyzed by the FISGAS routine in which a regression analysis code REEP is used to fit the percent fission gas release an an exponential function of local burnup and local linear heat generation rate. Fission gas release (%) is plotted against burnup (MWd/kg) at 3 linear heat generation rates: 7, 10 and 12 kW/ft.

Hering, W., and R. Manzel. 1978. Measured Fission Gas Release Data from PWR Fuel Rods. NUREG-0418, Appendix C, Nuclear Regulatory Commission, Washington, DC

Fission gas release data were obtained from fuel rods irradiated in the Obrigheim PWR. The 35 fuel rods, clad in zircaloy, include standard, high-power test rods and rods of a cycling experiment. Among these fuel rods, 21 are reload pins that have been prepressurized.

Fractional fission gas releases at various linear rod powers and up to a burnup level of 40,000 MWd/MTM were presented. The burnup and fission gas release ranges for standard rods (24) are 11,000 to 40,000 MWd/MTM and 0.2 to 6.5%. The ranges for the high power rods (4) are 20,000 to 40,000 MWd/MTM and 33.2 to 55.5%, and for the cycling experiment rods (8) are 30,000 to 32,000 MWd/MTM and 0.8 and 6.9%.

Pati, S. R., D. E. Bessette and L. V. Corsetti. 1979. "Fission Gas Release and Dimensional Changes of Test Fuel Rods Containing Densifying and Nondensifying Fuel." Paper presented at the ANS Topical Meeting on LWR Fuel Performance. Portland, Oregon.

Combustion Engineering and Electric Power Research Institute conducted a research program at Calvert Cliffs (Unit 1) to evaluate the performance of test rods containing UO<sub>2</sub> fuel pellets fabricated with different microstructures and various propensity for in-reactor densification.

Sixty test rods with 93 to 95% TD and 2.33 to 2.82% enrichment have been inspected following 1 and 2 reactor operating cycles with peak rod average burnups between 18,000 and 29,500 MWd/MTU. Among the test rods, 6 were examined in a hot-cell to evaluate fission gas release behavior as a function of burnup and fuel microstructure.

Fission gas release from all 6 rods was less than 1%. There is no indication of burnup enhancement up to 29,000 MWd/MTU. Changes in porosity distribution and grain growth as related to initial grain size and fuel type were observed and explained on the basis of the differences in power and temperature history of the rods during irradiation.

Roberts, E., et al. 1977. "Fuel Modeling and Performance of High Burnup Fuel Rods." Water Reactor Fuel Performance. ANS Topical Meeting, St. Charles, Illinois.

The irradiation performance of zircaloy-clad PWR fuel rods over a wide range of power and burnup levels for both pressurized and nonpressurized rods were examined. Data obtained were used as the basis for the performance analysis and design (PAD) code.

Of particular interest to high burnup fuel performance are data obtained from Zorita and Saxton III reactors, which give a comparison of UO2 and MOX fuel rods. The fission gas model used in the code has a strong burnup dependence (second order). Experimental and calculated fission gas release results were plotted in a scattered diagram for these reactors.

Examinations of the irradiated fuel rods indicate that fuel rod growth, cladding corrosion, cladding creepdown and fuel swelling were well within acceptable limits. Measured fission gas release in Zorita fuel showed an increase with increasing burnup (30,000 to 55,000 MWd/MTU). At similar burnups, time-averaged and peak power levels, dissimilar specific power histories result in markedly different fission gas release. Prepressurization with helium has little effect on the above observation.

Roberts, J. T. A., et al. 1979. <u>LWR Fuel Performance Program: Progress in 1978</u>. EPRI-NP-1024-SR, Electric Power Research Institute, Palo Alto, California.

In order to increase commercial reactor plant availability by improving fuel rod reliability, the Electric Power Research Institute carried out an LWR Fuel Performance Program to develop a comprehensive fuel performance data base with verified predictive models and codes. Several projects that were completed during 1978 have yielded valuable information on zircaloy properties and fuel performance limits including fuel rod bow and fission gas release.

High burnup fission gas release measurements have been made under the bundle surveillance projects conducted by Combustion Engineering, Babcock & Wilcox, General Electric, and Exxon. The key results to date include 1) fission gas release from PWR fuel rods is less than 1% for burnup levels up to 30 GWd/MTM, 2) prepressurization strongly enhances the thermal stability and significantly reduces fission gas release in LWR fuel rods, and 3) two populations of fractional gas releases (>16% and <2%) exist for BWR fuels. The higher fractional release may have been caused by high fuel rod power during the first irradiation cycle, a large as-fabricated diametral gap (about 0.3 mm), and fuel densification.

Smalley, W. R. 1971. <u>Saxton Core II Fuel Performance Evaluation, Part I:</u> aterials. WCAP-3385-56, Part I.

To develop information concerning the use of Pu-enriched fuel in PWR systems, irradiation in the Saxton Plutonium Project was carried out in the Saxton reactor between 1965 and 1966.

Among the many examinations to determine the fuel performance, 18 rods (13 pellet, 5 Vipac) were punctured in a calibrated vacuum system to collect and sample fission gas. Results indicate that the measured burnup range is 9,300 to 20,700 MWd/MTM, and fission gas release range is 4 to 32% for the pellet rods. For vipac rods, the numbers are 15,300 to 20,700 MWd/MTM and 10 to 37%, respectively.

Good dimensional stability was maintained throughout the irradiation. Both pellet and vipac fuels performed equally well, with no evidence of fuel rod failures. None of the changes in dimensions, microstructure, or properties indicate basic operational limits for PuO<sub>2</sub>-UO<sub>2</sub> fuel in irradiation environments similar to those of Saxton Core II.

### Smalley, W. R. 1974. <u>Evaluation of Saxton Core III Fuel Materials Performance</u>. WCAP-3385-57.

Approximately 250 mixed-oxide fuel rods from Saxton Core II were reconstituted into 7 new loose lattice assemblies and irradiated in Core III to peak burnups up to 51,000 MWd/MTM. A sampling of these rods (1 removed from BOL, 7 from MOL and 7 from EOL) representing a wide range of peak power levels and burnups were subjected to hot-cell examinations to determine, among other things, the fission gas release and burnup.

Examinations show that all nonfailed rods were dimensionally stable. The highest fractional release of fission gas is slightly higher at the end of Core III (37%) than at the end of Core II (32%), which is consistent with the power history of the rods involved and higher burnup.

Microstructure evidence showed progressive closure of the fuel-cladding gap at high burnup and increasing fuel-cladding bonding. These phenomena, however, have no apparent adverse effect on dimensional stability or overall performance of the fuel.

## Zimmermann, H. 1975. "Fission Gas Behavior in Oxide Fuel Elements of Fast Breeder Reactors." <u>Nucl. Tech.</u> 28:127-133.

Under the fast breeder project irradiation program, a total of 159 fuel pins (80 mm long) composed of  $UO_2$ -Pu $O_2$  or  $UO_2$  pellets (4.0 to 6.25 mm diameter) were irradiated in reactors FR2, BR2, DFR and Rapsodie. These fuel pins were operated at mean linear heat generation rates of 150 to 570 W/cm to burnups of 1 to 13% of heavy atoms.

Fission gas produced is measured by the amount released, retained in pores and bubbles and retained in the matrix. Data for percentage of fission gas release at burnups up to 13.1% are presented at three volume-averaged fuel temperatures (at the end of irradiation): 1000, 1250 and 1500°C.

Experimental results indicate that a marked increase of fission gas release is seen in the burnup range of 1.5 to 7%. The higher the fuel temperature, the sooner this increase occurs.

While fission gas retained in pores continues to increase, that in the matrix reaches saturation at low burnup. Concentration of total retained gas (matrix + pore) increases with burnup to 3%, then decreases with burnup up to 6%. Above 6% it increases again to a value higher than that of the maximum (at 3% burnup) obtained previously. This effect is observed in all temperature ranges of the experiment.

Zimmermann, H. 1978. "Investigations on Swelling and Fission Gas Behavior in  $U0_2$ ." J. Nucl. Mat. 75:154-161.

Capsules containing UO2 pellets, clad in Mo alloy TZM, were irradiated at temperatures between 1000 and 2100 K to 12.6% burnup to study their swelling and fission gas behavior. The amount of fission gas was measured in three steps as that released, retained in pores and bubbles, and retained in the matrix. Swelling data were obtained by summing the fractional external volume change and the volume fraction of initial porosity that has disappeared during irradiation.

Data on fission gas release (%) as a function of burnup (%) were presented for irradiation temperatures 1250, 1500, 1750 and 2000 K. It is apparent that fission gas behavior is primarily determined by the irradiation temperature. With increasing temperature the mobility of the gas increases, resulting in higher fission gas release and swelling. Gas release and swelling also increase (at different rates) with burnup at all temperatures.

#### APPENDIX IV

INPUT VALUES FOR GAPCON-THERMAL-2

CALCULATIONS OF FISSION GAS RELEASE SURFACES

| Fuel Composition 0.0000 Weight Fraction PU02 0.0000 Weight Fraction PU239 0.0000 Weight Fraction PU240 0.0000 Weight Fraction PU241 1.0000 Weight Fraction U02 .0300 Weight Fraction U235 .9700 Weight Fraction U238   |   | FRPU02<br>FR39<br>FR40<br>FR41<br>FRU02<br>FR35<br>FR38  | 2112  |  |
|--|---|--|---|--|
| Fuel Density Restructured Fuel Density Pellet Diameter Initial Restructured Fuel Diam. Initial Center Hole Diameter Pellet-To-Clad Gap Clad Inside Diameter Clad Outside Diameter Fuel Length Sorbed Gas Content Fraction of Sorbed Gas Which is H2 Plenum Volume Coolant Temperature Axial Temperature Gradient Across Core Coolant Passage Equivalent Diameter Coolant Velocity Pressure on Clad OD Fuel Surface Roughness, Arith. Mean Clad ID Surface Roughness, Arith. Mean Diameter Of Auxiliary Basket Basket Thermal Conductivity Basket-To-Clad Heat Transfer Coeff. Fill Gas Pressure Crud Thickness Fill Gas Composition 1.00000 Mole Fraction Helium 0.00000 Mole Fraction Argon 0.00000 Mole Fraction Nitrogen 0.00000 Mole Fraction Nitrogen 0.00000 Mole Fraction Krypton 0.00000 Mole Fraction Krypton 0.00000 Mole Fraction Xenon | (Fraction TD) (Fraction TD) (Inches) (Inches) (Inches) (Inches) (Inches) (Inches) (CC/Gram)  (Cu. In.) (Deg F) (Deg F) (Inches) (Ft/Sec) (PSI) (Inch) (Inch) (Inch) (Inches) (BTU/HR-FT2-F) (Atomospheres) (Inches) | FRDEN FRSIN DFS DSIN DVOIDZ GAP DCI DCO LFUEL S XH VPLENZ TINLET (1) DTEMP DE V EXTP ROUF ROUC DBO KB HBC ATMOS CRUDTH | BWR<br>.95<br>.965<br>.410<br>0.0<br>0.00<br>0.00<br>0.419<br>0.483<br>150.0<br>0.0<br>1.83<br>550.0<br>20.0<br>0.0<br>1050.0<br>0.8 E-4<br>0.2 E-4<br>0.0<br>0.0 | PWR<br>.95<br>.965<br>.3179<br>0.0<br>0.0065<br>0.3244<br>0.374<br>144.0<br>0.0<br>0.0<br>1.677<br>550.0<br>64.0<br>0.0<br>2060.0<br>0.8 E-4<br>0.2 E-4<br>0.0<br>0.0<br>27.2<br>0.0 |

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