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MODELING FISSION GAS RELEASE IN HIGH BURNUP $\text{ThO}_2\text{-UO}_2$ FUEL

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Introduction

Due to potential waste and economic benefits as well as enhanced proliferation resistance features of the spent fuel, there has been renewed interest in $\text{ThO}_2\text{-UO}_2$ as high burnup LWR fuel for once through cycles [1]. Several new proposals have been made in recent years including the Radkowsky Thorium Reactor (RTR) seed and blanket concept with a once-through fuel cycle utilizing thorium [2]; and a mixed thorium-uranium dioxide fuel cycle [3]. Both these concepts are devised to drive the fuel to a very high burnup exceeding 100,000 MWd/MT.

Earlier work in the LWBR project suggests that the rate of fission gas release from Th/U fuel would be expected to be less than that from UO_2 fuel [4]. Also, thoria has a higher thermal conductivity at low and moderate temperatures typical of commercial plant operation conditions. As a result, Th/U oxide fuel is expected to operate at a lower temperature and perform better than conventional uranium oxide fuel at fuel exposures near or above the current USNRC licensing limit of 62 MWd/kg. However, the previous thorium fuel investigations had three shortcomings relative to present needs: using very highly enriched U-235, using a small fraction of total uranium, and focusing on lower burnup. Therefore, the information obtained in the previous experience cannot directly be used to ascertain the performance of thorium in power reactors under present conditions.

Fission gas release is an important factor in achieving satisfactory performance of fuel to high burnup in light water reactors (LWRs). It determines the internal pressure rise within the fuel rods, and if excessive fission gas release occurs, it might lead to fuel-cladding gap reopening at high burnup. To properly calculate the fission gas release, accurate thermal models (fuel thermal conductivity, thermal expansion, radial power distribution, etc.) are necessary. The buildup of plutonium in the outer rim of LWR UO_2 pellets has been observed to create a region of high fuel burnup, fission gas buildup and high porosity at the fuel rim. A less severe rim formation is expected in $\text{ThO}_2\text{-UO}_2$ fuel due to the lesser build-up of Pu-239 (less U-238 in $\text{ThO}_2\text{-UO}_2$ fuel) and flatter distribution of U-233 (less resonance capture in Th-232) [5]. Therefore, efforts are being made to investigate the fission gas release from mixed thoria-urania

fuels with appropriate thermal conductivity, thermal expansion, and radial power distributions.

Radial Power and Burnup Distribution for LWR Thoria Fuels

A previous study shows that there is much lower power peaking near the surfaces of ThO₂-UO₂ fuel than UO₂ fuel and this difference becomes more pronounced as burnup increases [5]. With lower power peaking at the rim, a severe rim effect is not expected to occur in mixed thoria-urania fuel until much higher pellet average burnup than in urania fuel. To incorporate this difference into the fuel performance code FRAPCON-3, a new model THUPS (Thoria-Urania Power Shape), which is relatively simple, yet reasonably accurate, was developed to replace the original TUBRNP Model [6] which is dedicated to UO₂ fuel. The new model considered the following complications of ThO₂-UO₂ fuel compared to UO₂ fuel [7]:

- More isotopes of importance in ThO₂-UO₂ fuel (Th-232, U-233, U-234)
- The difference in resonance capture between U-238 and Th-232.
- The change in average capture rate and radial distribution of capture rate due to the mixture of U-238 and Th-232.
- The dependence of Th-232 and U-238 capture on the initial content of U-235 in heavy metal.

The set of equations used in THUPS model were summarized in the following:

$$\frac{dN_{232}(r)}{dt} = -\sigma_{a,232}(r, N_{235}, N_{232})N_{232}(r)\phi(r) \quad (1)$$

$$\frac{dN_{233}(r)}{dt} = -\sigma_{a,233}N_{233}(r)\phi(r) + \sigma_{c,232}(r, N_{235}, N_{232})N_{232}(r)\phi(r) \quad (2)$$

$$\frac{dN_{238}(r)}{dt} = -\sigma_{a,238}(r, N_{235}, N_{238})N_{238}(r)\phi(r) \quad (3)$$

$$\frac{dN_{239}(r)}{dt} = -\sigma_{a,239}N_{239}(r)\phi(r) + \sigma_{c,238}(r, N_{235}, N_{238})N_{238}(r)\phi(r) \quad (4)$$

$$\frac{dN_j(r)}{dt} = -\sigma_{a,j}N_j(r)\phi(r) + \sigma_{c,j-1}N_{j-1}(r)\phi(r) \quad (5)$$

Where $N(r)$ is the concentration of isotopes, σ_a the absorption cross section, σ_c the capture cross section, j represents U-234, U-235, U-236, Pu-240, Pu-241 and Pu-242, and $\phi(r)$ is the thermal neutron flux shape (approximated by the modified Bessel function). However, $\sigma_{a,232}(r, N_{235}, N_{232})$ and $\sigma_{a,238}(r, N_{235}, N_{238})$ are equivalent one group absorption cross sections including thermal and resonance absorption, and are, therefore, functions of the radius:

$$\sigma_{a,238}(r, N_{235}, N_{238}) = \bar{\sigma}_{a,238}(N_{235}, N_{238}) * f(r, N_{238}) \quad (6)$$

The shape function $f(r, N_{238})$ has a form of $1 + u_1 \exp(-u_2 (r_0 - r)u_3)$ but area averaged to 1 by a normalization factor. The coefficients u_1 , u_2 , u_3 are all functions of N_{238} . $\sigma_{a,232}(r, N_{235}, N_{232})$ has a similar form to equation (6), but with different numerical values for the constants.

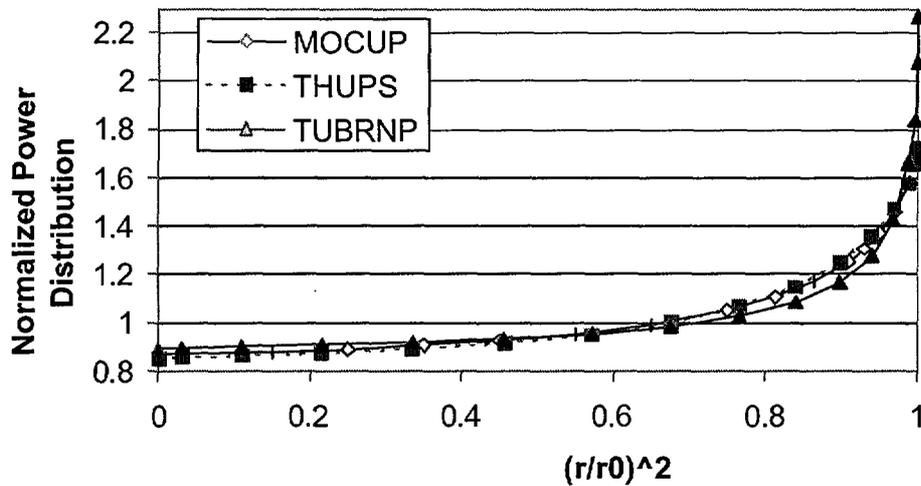


Figure 1. Comparison of power distribution from THUPS, TUBRNP and MOCUP for 65%ThO₂-35%UO₂ (19.5% U-235 enrichment) fuel at 63 MWd/kgHM

The average effective cross sections and shape functions for the Th-232 and U-238 isotopes were fitted to the reaction rates from the MOCUP monte-carlo depletion calculations [8]. The calculated radial power distributions for 65%ThO₂-35%UO₂ (U-235 enrichment 19.5%) fuel at 63 MWd/kgHM from THUPS, TUBRNP (Th-232 was replaced by U238) and MOCUP are shown in Figure 1 as an example. The THUPS model has been found to agree very well with MOCUP. The TUBRNP model, however, gives a much higher peak power near the fuel surface.

Thermal Conductivity of ThO₂-UO₂ Fuel

Three major heat transport mechanisms can be active in UO₂: (1) lattice conduction (2) radiant heat conduction, and (3) electron heat transport. Below ~2000 K, the lattice conduction dominates, and the conductivity decreases with increasing temperature. Above ~2000 K, the radiation and electronic contributions to thermal conduction become important and the conductivity increases with increasing temperature.

The radiant contribution is significant at high temperatures for an electrically non-conducting solid that is transparent to infrared radiation. Thus, for ThO₂, the radiant conduction is expected to occur at high temperatures. However, the electronic contribution will be insignificant for ThO₂ since it is principally an ionic conductor [4]. Therefore, the ThO₂ conductivity is expected to increase with increasing temperature at very high temperatures, but the magnitude of the increase for ThO₂ is expected to be less than that for UO₂.

For mixed ThO₂-UO₂ with a UO₂ content from 0% (pure ThO₂) to 30%, Belle and Berman recommended the following correlation [4]:

$$K_{Th/U}^M = \frac{1}{A(M) + B(M) \cdot T} \tag{7}$$

$$\text{where, } A(M) = \frac{1}{A_0 + A_1 \cdot M} \text{ and } B(M) = B_0 + B_1 \cdot M + B_2 \cdot M^2 \quad (8)$$

M is the mole fraction of the UO₂ in the mixed ThO₂-UO₂,

$$A_0=46.948, A_1= -112.072, B_0=1.597 \times 10^{-4}, B_1=6.736 \times 10^{-4}, B_2= -2.156 \times 10^{-3}$$

Figure 2 shows the above correlation and related thermal conductivity data for pure ThO₂. Figure 2 also shows the Belle and Berman correlations for mixed ThO₂-UO₂ along with the data (UO₂ content up to 6%) they published in their handbook. It is seen that the Belle and Berman correlations for pure ThO₂ and for mixtures of thorium and urania with high thorium content fit the data reasonably well up to about 2000 K. However, the continuous decrease in both the ThO₂ and mixed thorium-urania thermal conductivity above 2000 K is not justified. Considering the radiant contribution at high temperature which will be proportional to T³, the following component was added to equation (7):

$$\frac{(T-1340)^3}{5.43 \times 10^9} \quad (9)$$

As shown in Figure 2, the addition of the cubic term does not affect the thermal conductivity up to about 2000 K. Even though there is no data above about 2200 K, the thermal conductivity calculated using the modified correlation at the melting point of ThO₂ (3,640 K) goes through the middle of the data obtained at about 2200 K, which seems quite reasonable.

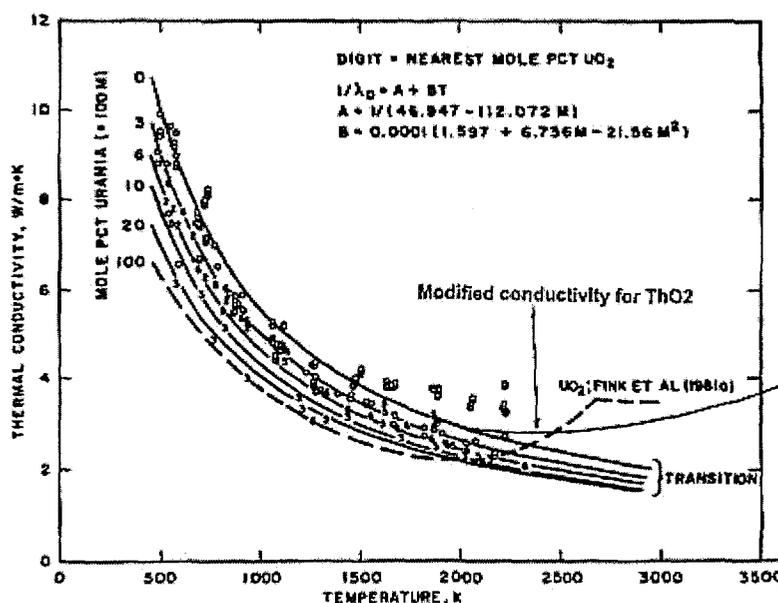


Figure 2. Thermal Conductivities, Corrected for Porosity, of ThO₂ through ThO₂-6 Mole Percent UO₂

For mixed ThO₂-UO₂ with a UO₂ content greater than 30%, the mixture conductivity can be determined by a linear interpolation between the 70%ThO₂-30%UO₂ conductivity and the pure UO₂ conductivity:

$$K_{Th/U}^M (M > 30\%) = \frac{K_U - K_{Th/U}^{0.3}}{0.7} \cdot [M - 0.3] + K_{Th/U}^{0.3} \quad (10)$$

Where, $K_{Th/U}^{0.3}$ is the thermal conductivity of 30%UO₂-70%ThO₂,
M is the UO₂ mole fraction in mixed ThO₂-UO₂.

The above thermal conductivity equations are for fresh ThO₂-UO₂ fuel at theoretical density. The correlations for burnup, porosity and irradiation effect [9] were kept the same as in FRAPCON3.

Rim Porosity

The accelerated fission gas release in high temperature fuel rods is considered due to the activated thermal release from the intermediate fuel region [10]. The high porosity and burnup in the rim region may assist this process by decreasing the thermal conductivity in the rim region, thus increasing the fuel temperature in the inner and intermediate regions. The porosity has been reported between 10-30% in the high burnup rim zone. Less constrained fuel tends to have higher rim porosity. For a typical PWR fuel the porosity reaches about 15% and does not increase for a wide range of higher burnup (up to 83 MWd/kgU). Further increase of the burnup to 102 MWd/kgU will increase the porosity to 24% [10]. The bubble size is also significantly increased.

Usually the rim width is calculated based on an empirical formula and the average porosity is modeled to increase asymptotically to 15% at high burnup. This approach is adopted in the SIERRA code [11]. However, the rim width can vary with the fissile concentration. The average porosity dependence on the average rim burnup approach may not be appropriate in the application in thorium fuel in which the fissile concentration varies over a wide range depending on the core design.

Rim porosity was not considered in FRAPCON3 code. To model the impact of the rim porosity on the thermal performance, a model developed by Losonen was adopted [12]. This model correlates the local porosity with the local burnup within the fuel pellet. The porosity increase, ΔP , in the rim zone as a function of the burnup-up and radial position used in the calculation of the thermal conductivity is

$$\Delta p(bu) = a_c \left(\frac{bu - bu_{rin}}{\Delta bu'} \right)^2, \quad \text{at } bu \geq bu_{thr} \quad (11)$$

$$\Delta bu' = bu_{r0} - bu_{rin}$$

Where bu_{r0} is the burn-up at the pellet outer surface, bu_{rin} is the burn-up in the center of the pellet, bu is the local burnup and a_c is 0.12. Below bu_{thr} no rim porosity is assumed to form.

Other Considerations

- Density of ThO₂-UO₂ fuel

The FRAPCON-3 has been modified to reflect the density of ThO₂-UO₂ fuel in order to obtain the correct burnup, densification calculation, etc.

- Thermal Expansion of ThO₂-UO₂ Fuel

The thermal expansion of mixed ThO₂-UO₂ is smaller than that of pure UO₂. The thermal expansion correlation for ThO₂ was incorporated into the FRAPCON-3 code as recommended by Belle and Berman [4]. The ThO₂-UO₂ linear thermal expansion is determined from that of pure ThO₂ and pure UO₂ by applying Vegard's law.

- Fission Gas Production

For the thorium fuel system the production of more than twice as much Kr per ²³³U-fission results in a total of approximately 10 percent more fission gas (Xe plus Kr) production [4]. During operation, ²³³U will build up and ²³⁵U will decrease in Th-U fuel. The fission gas production varies accordingly.

Fission Gas Release Model

Fission gas release data were obtained from mixed thorium-uranium fuels of various compositions, rod geometries and irradiation conditions (power histories, coolant temperature and pressure, etc.) as part of the Light Water Breeder Reactor (LWBR) development program. Fission gas release values from 59 test rods have been reported by Bettis Atomic Power Laboratory, but sufficiently detailed information is available in the open literature for only 5 of those test rods [13,14]. Therefore, they have been the basis for our comparisons to predictions using the steady-state fuel performance code FRAPCON-3.

Table 1 lists some characteristics of the LWBR test rods. Most of these rods contained 70% to 80% ThO₂, with one rod containing 93.4% ThO₂. Two rods were irradiated at peak linear power levels comparable to those for high power rods in LWRs and three rods were irradiated at power level well above the peak powers in LWRs. All 5 rods were subject to time-varying power histories: Rods 79-442, 79-506 and 79-349 were irradiated at high power levels early in life and at low power levels late in life; while Rods 79-375 and 79-405 were irradiated at relatively constant power levels.

Rod No.	79-442	79-506	79-349	79-375	79-405
Wt% of ThO ₂	93.4	82.9	80	70	80
Fuel Density (%TD)	96	95.3	95	92	94
Pellet Diameter (in.)	0.208	0.462	0.652	0.645	0.651
Cladding Outer Diameter (in.)	0.255	0.527	0.717	0.717	0.717
Fuel Clad Diametric Gap (mils)	7.1	6.9	4	12	6
Peak Linear Power (kW/ft)	10.35	14.48	22.45	19.34	22.17
Peak Burnup (MWd/MTHM)	45300	21100	8070	9990	11300
Axial Peak/Average Ratio	1.43	1.41	1.33	1.33	1.33
Measured Fission Gas Release (%)	0.5	2.0	1.9	8.4	15.0

Table 1. Characteristics of 5 LWBR test rods used in FRAPCON-3 calculations

There are two fission gas release models in FRAPCON-3: the ANS 5.4 model and the Massih model. The ANS 5.4 model [15] is currently the industry standard for the calculation of release of various fission gases. The model utilizes a mathematical formula for time-varying power histories derived from diffusion theory (one stage gas diffusion from the fuel matrix to the rod void volume). The diffusion coefficient for the ANS 5.4 model was determined as a function of fuel temperature and burnup based on steady-state fission gas release data from 45 fuel rods irradiated to low burnup (<19,000 MWd/MTM) and 19 fuel rods irradiated to high burnup (<60,000 MWd/MTM).

The Massih model [9,16] is a two-stage model: diffusion of fission gas from inside the grains to their boundaries and the release from grain boundaries to the rod void volume. The diffusion stage is described by the Booth diffusion model, which is a solution of the gas diffusion equation at constant power and temperature. A saturation criterion is imposed for release of gas from the grain boundary to the void volume. The diffusion and saturation parameters were determined based on data from 10 steady-state cases and 10 cases with power ramps at the end of life.

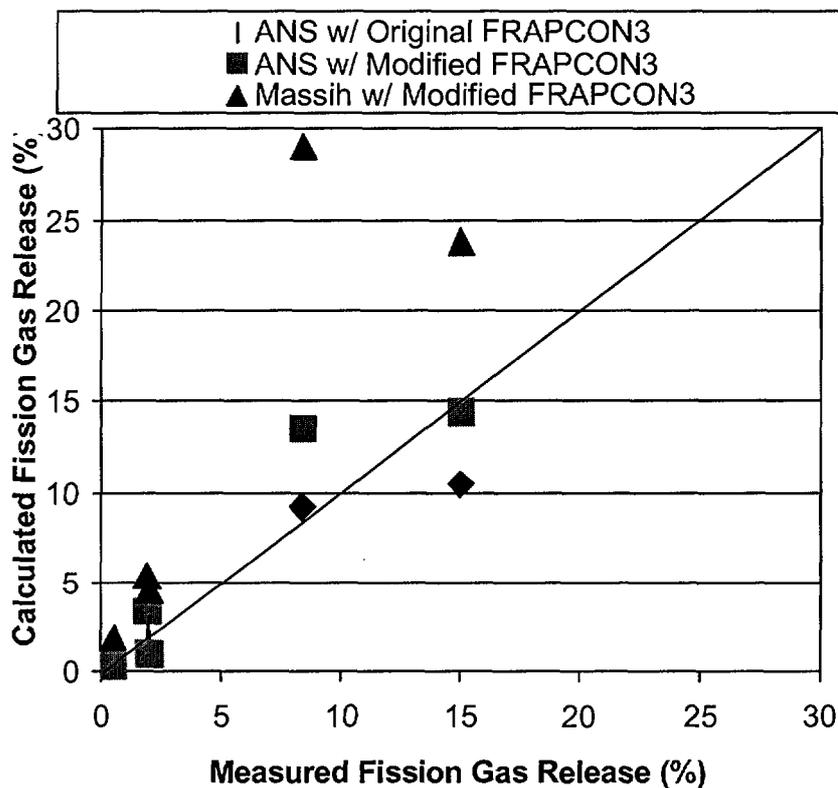


Figure 3. Fission gas release predictions vs. measurements

Figure 3 shows the effects of the modifications in FRAPCON-3 on the fission gas release predictions using the ANS 5.4 and Massih model. It is seen that fission gas release predictions using the ANS 5.4 model in modified FRAPCON-3 code are in reasonable agreement with measured values. Figure 3 also shows that the Massih model over predicts fission gas releases for all 5 test rods. None of these 5 test rods was subject to power ramps at the end of life.

Burnup Dependence of Fission Gas Release

Fission gas release increases with burnup and is accelerated at high burnup (around 45 MWd/kgU) in UO₂ fuel. The rim effect is considered directly or indirectly related to this phenomenon. The microstructure change in the rim region may lead to additional fission gas release. Also, the increased porosity and burnup in the rim region will reduce the thermal conductivity of fuel in the rim region and lead to higher inner fuel temperatures and hence more fission gas release in the inner region. By using X-ray fluorescence analysis (XRF) to measure the radial distribution of xenon in a nuclear fuel the local percentage fission gas released to the rod free volume can be obtained. The concentration measured by XRF starts deviating from the generated concentration between 60 and 80 MWd/kgU [12,19]. XRF data clearly show that some fraction of the fission gas is released from fuel at high burnup due to the formation of the rim structure. However the contribution of fission gas release from rim structure is noticeable at low temperature where the thermal central release is low [20].

There is also an upturn of fission gas release in ThO₂-UO₂ fuel, but only at a much higher burnup than UO₂ fuel. The burnup level is somewhere between 70 and 80 MWd/kgHM based on the fission gas release data from fuel rods from the LWBR program, which generally operated at the low temperature and to very high burnups [4]. This is probably due to the flatter power distribution (thus less severe rim effect) and/or higher stability of ThO₂-UO₂ fuel. Detailed geometry information on these high burnup rods is not available in the open literature. Based on the reported temperature and power history, however, unreasonably high fission gas releases (50%-70% compared to the measured value 1.8%-7.9%) were obtained by using the original ANS5.4 and Massih models. The discrepancy lies in the substantially enlarged diffusion coefficient because of the burnup enhancement factor, which increases exponentially with the burnup. The FRAPCON-3 code models the burnup dependence of fission gas release by multiplying a burnup enhancement factor by the thermal diffusion coefficient and using a separate low temperature release model in which an upturn of fission gas release occurs at 40 MWd/kgU. The burnup enhancement factor is applied to the whole temperature range in the ANS5.4 model, while only applied to high temperature in the Massih model. Since these rods were operated below the dislocation and grain boundary release temperature, the thermal diffusion process should not play an important role in the fission gas release.

To correctly reflect the accelerated fission gas release, the burnup enhancement factor is applied only above the dislocation release temperature-1700°C for ThO₂-UO₂ fuel and the low temperature release model in FRAPCON-3 was replaced by the following Belle & Berman Recommendations [4]:

$$F = (100/\rho)^{10.25} (0.05b)(1 + \exp(0.3(b - 20))) \quad (12)$$

Where, F = fission gas release (percent)

$\bar{\rho}$ = initial fuel density (percent of theoretical)

b = fission density, fission units

The above correlation shows an accelerated fission gas release at 20 fission units, which corresponds to around 80 MWd/kgHM.

Discussion

Thoria fuel has a higher thermal conductivity and lower thermal expansion. The higher thermal conductivity reduces the temperature rise within the pellet. The benefit is very limited though when there is a higher content of uranium in the fuel as shown in Figure 2. The smaller thermal expansion increases the temperature rise across the gap, which is important for temperature calculation at the beginning of the life when the gap is still open. This effect is significantly reduced after the gap is closed. The modification of these two materials properties, therefore, tends to result in a relatively small net effect on fuel temperature and fission gas release for the test rods.

Fission gas yield per U-233 fission is about 10% larger than for U-235 fission, which makes thoria fuel a little disadvantageous. However, this effect tends to be very small because the initial fissile material is U-235 and U-235 will be the dominant fission source during most of the fuel life. For 65%ThO₂-35%UO₂ (19.5% enrichment) fuel, the fission gas yield at 80MWd/kgHM is roughly 3.5% more than pure UO₂.

ThO₂-UO₂ fuel is expected to have lower fission gas release than UO₂ fuel based on the experiment data from LWBR program [14]. However this advantage is not reflected in Figure 3, where the original FRAPCON-3 predicts somewhat lower fission gas release than the modified one when using the ANS5.4 model. This is due to the limitation of the original FRAPCON-3 code, which cannot handle some of the 5 rods with high U-235 concentration and large diameter listed in table 1. The Massih model, however, gives a high prediction value that is just what we expected. To properly model the lower release in thoria fuel, the gas diffusion coefficient will have to be adjusted to a lower value. Unfortunately, 5 rods is not enough to complete this process with confidence due to the complexity of the diffusion coefficient used in the Massih model. Several data points in each temperature range and at various burnups are needed to ascertain the model.

Summary and Conclusion

A preliminary fission gas release model to predict the performance of thoria fuel using the FRAPCON-3 computer code package has been formulated. The following modeling changes have been made in the code:

- Radial power/burnup distribution
- Thermal conductivity and thermal expansion
- Rim porosity and fuel density
- Diffusion coefficient of fission gas in ThO₂-UO₂ fuel and low temperature fission gas release model

Due to its lower epithermal resonance absorption, thoria fuel experiences a much flatter distribution of radial fissile products and radial power distribution during operation as compared to uranium fuel. The rim effect and its consequences in thoria fuel, therefore, are expected to occur only at relatively high burnup levels.

The enhanced conductivity is evident for ThO₂, but for a mixture the thermal

conductivity enhancement is small. The lower thermal fuel expansion tends to negate these small advantages. With the modifications above, the new version of FRAPCON-3 matched the measured fission gas release data reasonably well using the ANS 5.4 fission gas release model.

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