Fission Gas Release Comparison for PWR Duplex Fuel Pellets

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Abstract

Current research has identified Duplex pellets as an effective strategy for decreasing the centerline temperature of the fuel. However, before this design can be widely utilized several areas, including fission gas release, need to be better characterized. Therefore, this paper seeks to ascertain the relative difference in fission gas release between a standard solid UO_2 reference design and a duplex design with Zircaloy and UO_2. Although the central area in the duplex doesn't produce any fission gas, it is not well known if the higher temperatures in the outer annulus (when compared to the equivalent area in the solid design) will cause overall more or less fission gas release. Interestingly, this study concluded that the duplex fuel actually exhibits a 4.4% higher overall fission gas release when compared to the solid fuel. Due to the very approximate treatment of several important input variables it was concluded that while the duplex design does have lower average temperatures, the fission gas release is comparable to that of the solid fuel design.

Introduction

One of the difficulties facing nuclear fuel designers today is ensuring that the design stays below the melting point of the fuel or 2840°C in the case of UO_2. Recent work has shown that a significant thermal operating benefit can be obtained from the incorporation of annular fuel as shown in Figure 1 below.

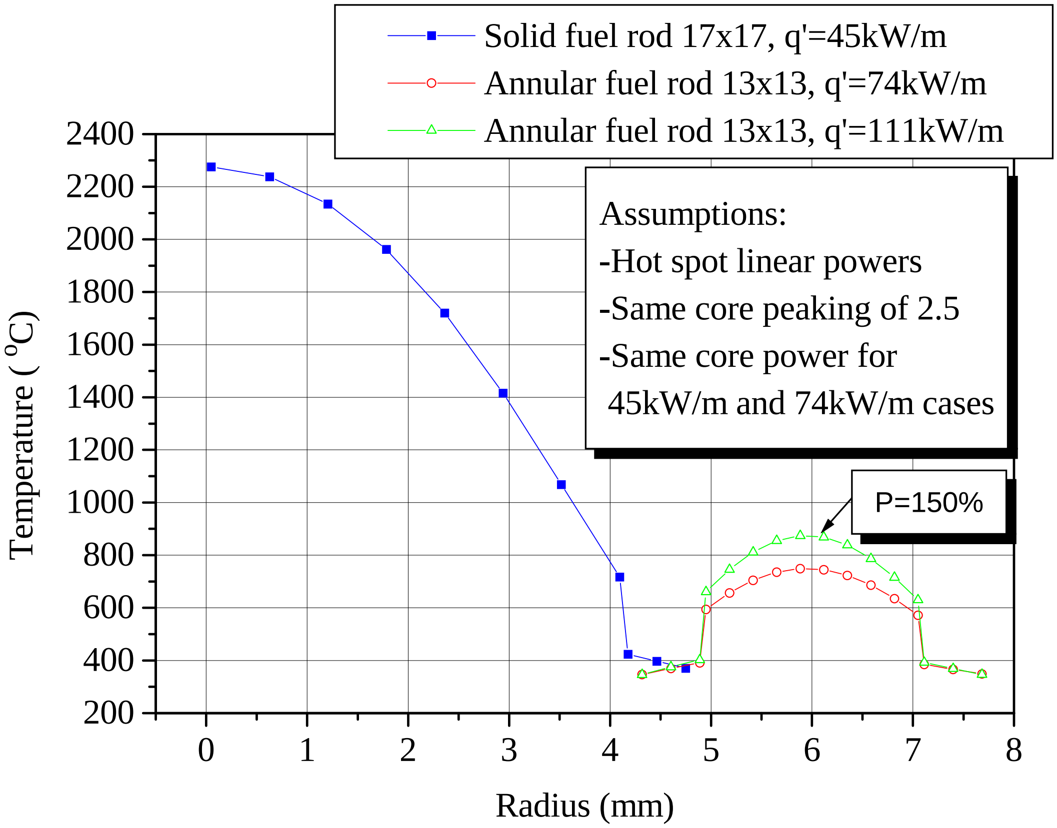

Figure 1: Solid/Annular Radial Fuel Temperature Profile (Kazimi)

These annular pellets obtain this additional thermal margin due to the unique internally and externally cooled geometry of the individual fuel pins shown in Figure 2 below.

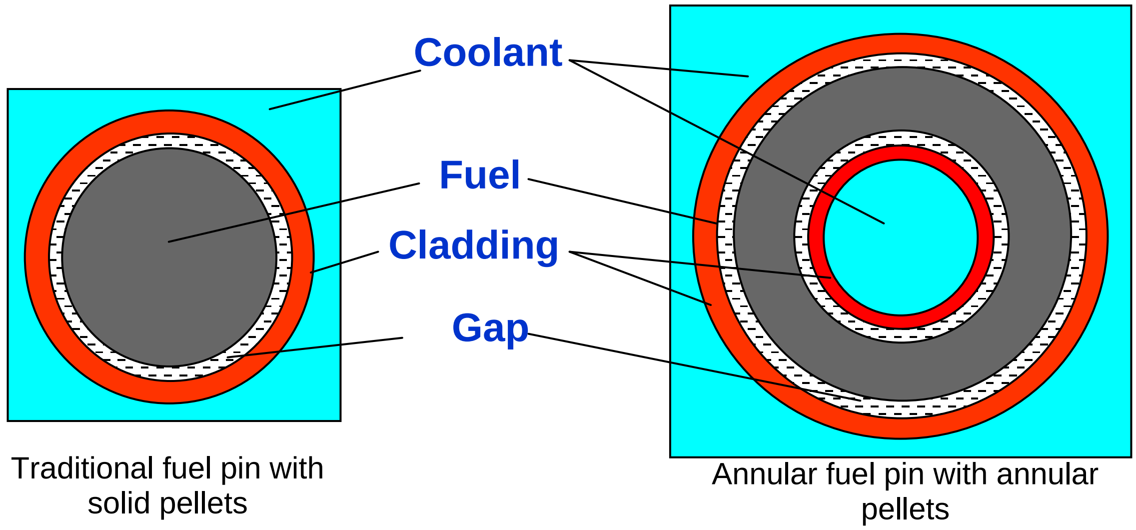

Figure 2: Annular Fuel Pin Geometry (Kazimi)

Besides the increased surface area for heat transfer, the annular pellets also benefit from not having fuel in the center of the pin which effectively spreads out the heat source. Due to fabricability concerns, additional schemes such as Duplex pellets have also been proposed in order to decrease the centerline temperature. Duplex pellets differ from annular fuel in that the inner hole of the annular pellet is filled with a solid material such as ZrO_2 or Zircaloy-4 as shown in Figure 3.

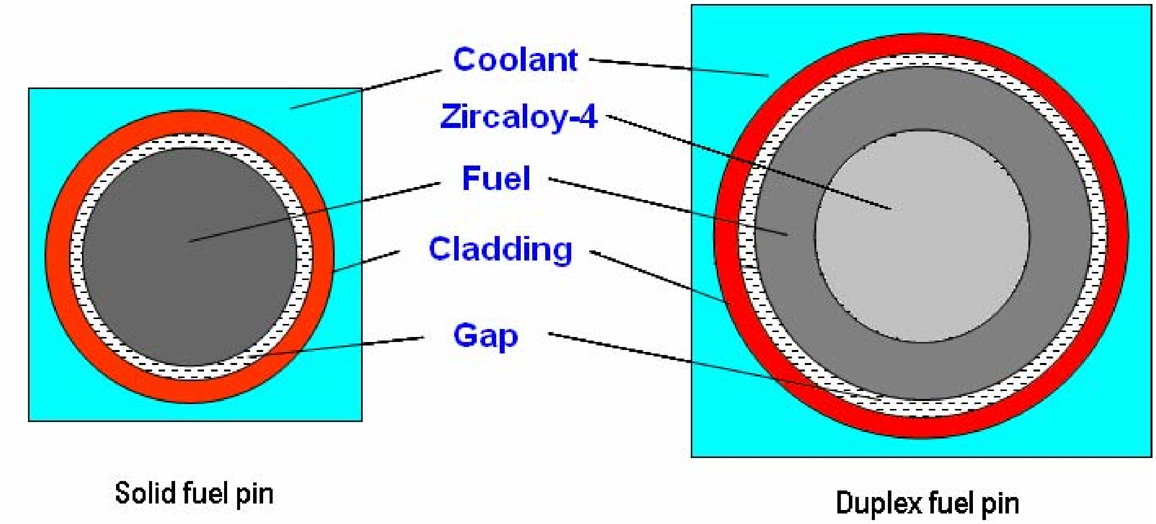

Figure 3: Duplex Fuel Pin Geometry

Like nearly all ideas within the nuclear field, the duplex pellet concept is not new. Duplex pellets were originally proposed during the late 1970's light water reactor breeder program in hopes of increasing the breeding potential of the fissile material. However, its use has started to recently resurrect due to increased interest in proliferation resistance (Shwageraus) and waste management (Bays). Shwageraus utilized this geometry as one possible avenue in order to help develop a thorium-based fuel cycle for light water reactors which reduces the plutonium generation rate and enhance the proliferation resistance of the spent fuel. However, due to the significantly large thermal resistance path from the UO_2 in the center to the ThO_2 on the periphery, it was found that the design led to higher than acceptable centerline temperatures. Bays, on the other hand, took advantage of this characteristic by putting spent fuel in a zirconium metal matrix on the periphery with thoria/zirconium CERMET in the center. This design allowed for a deeper burn of the transuranic waste because of the higher burnup "rim effect" on the periphery from the high thermal flux coming in from the moderator. Additionally, since the

CERMET shares a common metallic phase with the outer pellet, these two can be co-extruded together with the zircaloy cladding in a heated die extrusion process.

However, in order to further develop this duplex fuel pin design, there are several areas, including fission gas release, which need to be better characterized. Therefore, this paper seeks to ascertain the relative difference in fission gas release between a standard solid UO_2 reference design and a duplex design with Zircaloy and UO_2. Although the central area in the duplex doesn't produce any fission gas, it is not well known if the higher temperatures in the outer annulus (when compared to the equivalent area in the solid design) will cause overall more or less fission gas release.

Method

For purposes of this study, an equivalently sized solid UO_2 reference pellet will be compared with a UO_2/Zircaloy duplex pellet. In order to make a constant comparison, the linear heat generation rate for the two designs will be matched. This necessitates that the UO_2 in the outer ring of the duplex pellet will have to be enriched above that of the 5 weight % UO_2 in the solid pellet. The particular calculations showing this determination follow in the next section.

The next step will be to determine the temperature profiles for the two respective designs. For simplicity in the calculation, the thermal conductivity of the UO_2 will be assumed constant. This will allow the establishment of average temperature values for both the inner and outer zones. These temperature averaged zones will then be fed into a fission gas release correlation based on Booth type diffusion with grain boundary gas accumulation and resolution (Weisman).

In order to perform the fission gas estimation, this correlation also requires several other input parameters including, discharge burnup of the fuel, total in-core residence time and plutonium content. A CASMO simulation of both the solid reference fuel case as well as the duplex fuel case allowed these parameters to be determined. The pellet design parameters used to construct the CASMO input deck are shown below in Table 1.

	Solid Pellet	Duplex Pellet
Dco (mm)	9.52	9.52
Dci (mm)	8.37	8.37
Dfo (mm)	8.25	8.25
Dfi (mm)	-	2.74

Table 1: Fuel Pin Design Parameters

The solid reference fuel was ran with 5 weight percent enrichment however in order to fairly compare the duplex fuel, the total number of U^{235} atoms needs to be equivalent on a per unit length basis. Therefore the following calculation was performed to determine the enrichment of the duplex fuel.

$$\frac{10.4gU}{cm^3} * \frac{1molUO_2}{270gUO_2} * \frac{1molU}{1molUO_2} * \frac{238gU}{1molU} = 9.1674\frac{gU}{cm^3}$$

$$9.1674\frac{gU}{cm^3} * 0.05 = 0.45837\frac{gU^{235}}{cm^3}$$

The area ratio between the duplex and solid fuel is given by

$$AR = \frac{\pi(4.125mm)^2}{\pi((4.125mm)^2 - (1.370mm)^2)} = 1.10834$$

Thus multiplying the U^{235} atom density by the area ratio gives

$$0.45837 \frac{gU^{235}}{cm^3} * 1.10834 = 0.50803 \frac{gU^{235}}{cm^3}$$

Dividing this by the total original U atom density gives the new enrichment

$$\frac{0.50803 \frac{gU^{235}}{cm^3}}{9.1674 \frac{gU^{235}}{cm^3}} = 5.54\%$$

The density was determined by

$$\begin{aligned} \rho_{duplex} &= (\frac{1}{AR})(\rho_{UO_2}) + (1 - \frac{1}{AR})(\rho_{Zircaloy}) \\ &= (0.90225)(10.4 \frac{g}{cm^3}) + (0.09775)(6.44 \frac{g}{cm^3}) = 10.01 \frac{g}{cm^3} \end{aligned}$$

Figure 4 shows a plot of the Eigenvalue versus burn-up of the two designs. The simulation assumed a 3 batch refueling scheme with 3% leakage. The calculated discharge burn-ups are indicated on the graph by the arrows from the x-axis as well as in Table 2.

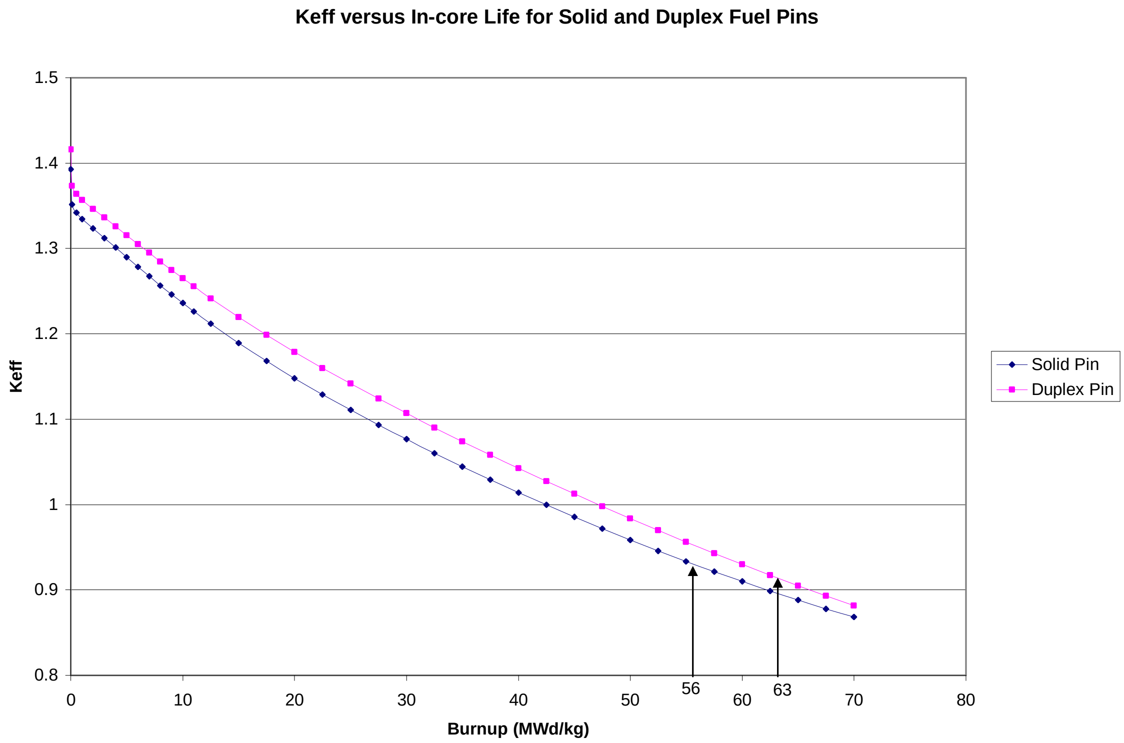

Figure 4: Keff versus In-Core Life for Solid and Duplex Fuel Pins

	Solid Fuel	Duplex Fuel
Discharge BU (MWd/kg)	56.25	63.75
Discharge BU (MWs/kg)	4860000	5508000
Specific Power (W/g)	0.0343346	0.0395727
EFPD	1638.289073	1610.95907
EFPs	141548175.9	139186863.7

Table 2: CASMO Calculated Discharge Parameters

Calculations

First the linear power ratings will be matched between the two designs.

$$q'_{solid} = q'_{duplex}$$

$$q'_{solid} = \pi(R_{fo}^2 - R_{fi}^2)q'''_{duplex}$$

$$q'''_{duplex} = 1.1228*10^6 \frac{W}{m^3}$$

Then the volumetric heat generation rate for the solid fuel is

$$q'''_{solid} = \frac{q'_{solid}}{\pi R_{fo}^2} = \frac{53.4\frac{kW}{m}}{\pi(0.0040456m)^2} = 1.03855*10^6 \frac{W}{m^3}$$

In order to determine the temperature distribution in the fuel pins we start with the general heat conduction equation as below

$$-\nabla * q'' + q''' = 0$$

which at steady state and assuming we are only considering one dimension in the radial direction, the heat conduction equation reduces to

$$\frac{1}{r}\frac{d}{dr}\left(kr\frac{dT}{dr}\right) + q''' = 0$$

$$k\frac{dT}{dr} + q'''\frac{r}{2} + \frac{C_1}{r} = 0$$

Now we write the general heat flux condition knowing that for the duplex pellet no heat flux exists at R_{fi} and for the solid pellet $R_{fi}=0$

$$q''\Big|_{r=R_{fi}} = -k\frac{dT}{dr}\Big|_{r=R_{fi}} = 0 \Rightarrow C_1 = -\frac{q'''R_{fi}^2}{2}$$

which gives

$$\int_{T}^{T\max} kdT = \frac{q'''}{4}\left[r^2 - R_{fi}^2\right] + C_1 \ln\left(\frac{r}{R_{fi}}\right)$$

Since $R_{fi}=C_1=0$ for the solid fuel, the above expression simplifies to

$$\int_{T}^{T\max} kdT = \frac{q''' r^2}{4}$$

Then evaluating the expression at $r=R_{fo}$ we find

$$\int_{Tfo}^{T\max} kdT = \frac{q'}{4\pi}$$

Plugging the in numbers (assuming that T_{fo}=400°C) gives

$$\left(3\frac{W}{m°C}\right)\left(T_{\max} - 400°C\right) = \frac{\left(53.4*10^3 \frac{W}{m}\right)}{4\pi} \Rightarrow T_{\max} = 1816.48°C$$

However, for the duplex pellet we substitute in the expression for C_1 and solve

$$\int_{T}^{T\max} kdT = \frac{q'''}{4}(r^2 - R_{fi}^2) - \frac{q''' R_{fi}^2}{2}\ln\left(\frac{r}{R_{fi}}\right)$$

$$\int_{T}^{T\max} kdT = \frac{q''' r^2}{4}\left(\left(1 - \left(\frac{R_{fi}}{r}\right)^2\right) - \left(\frac{R_{fi}}{r}\right)^2 \ln\left(\frac{r}{R_{fi}}\right)^2\right)$$

Then evaluating the expression $r=R_{fo}$ and substituting $q''' R_{fo}^2 = \frac{q'}{\pi\left(1 - \left(\frac{R_{fi}}{R_{fo}}\right)^2\right)}$ we find

$$\int_{Tfo}^{T\max} kdT = \frac{q'}{4\pi}\left(1 - \left(\frac{\ln\left(\frac{R_{fo}}{R_{fi}}\right)}{\left(\frac{R_{fo}}{R_{fi}}\right)^2 - 1}\right)\right)$$

Plugging in the numbers (assuming that T_{fo}=400°C) gives

$$\left(3\frac{W}{m^\circ C}\right)(T_{\max}-400^\circ C)=\frac{\left(53.4*10^3\frac{W}{m}\right)}{4\pi}\left(1-\left(\frac{\ln\left(\frac{4.125mm}{1.37mm}\right)}{\left(\frac{4.125mm}{1.37mm}\right)^2-1}\right)\right) \Rightarrow T_{\max}=1603.11^\circ C$$

Figure 5 below shows the plotted fuel pin temperature profile and the breakdown between Zone 1 and 2. The temperature averaged values for Zones 1 and 2 are shown in Table 3.

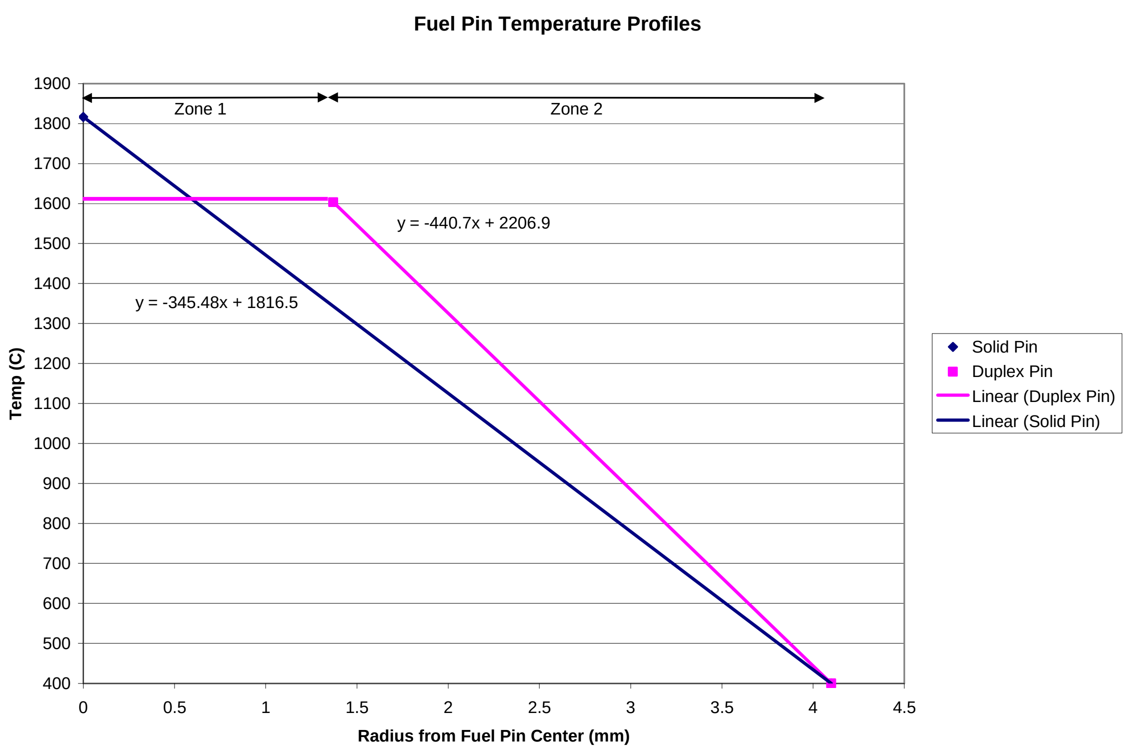

Figure 5: Fuel Pin Temperature Profiles

	Solid Pellet	Duplex Pellet
Avg Temp Zone 1 (°C)	1698.17	1603.11
Avg Temp Zone 2 (°C)	989.94	1001.52

Table 3: Zone Averaged Temperatures

This zone averaged temperature profile for both the solid and duplex fuel is depicted below in Figure 6.

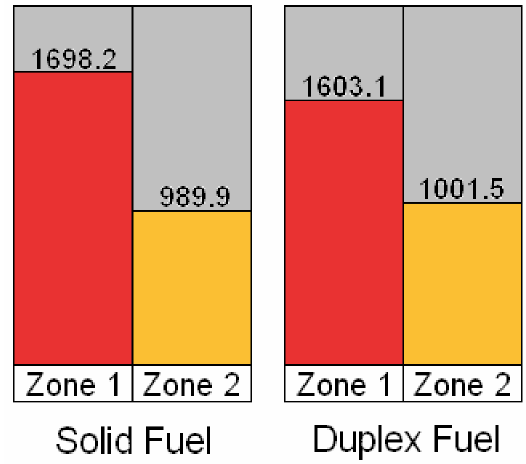

Figure 6: Zone Averaged Temperature Profiles

Now that the relevant data has been determined, we can start to calculate the fission gas release. Provided b=burn-up in MWs/kg and t=cycle length in s, we calculate the high burn-up factor as

$$bunkel = \frac{1.25*10^{14}}{\left(\left(\frac{b}{86.4}\right)^{3}\right)}$$

The fuel grain fission gas release is calculated by

$$ak1 = 9*10^{7}\left(e^{\frac{-45289.86}{ftemp}}\right) + 0.0005$$

$$ak2 = 0.00005*ak1*\left(1 - \frac{1}{\left(e^{\left(\frac{ftemp-1900}{40}\right)+1}\right) + e^{(-bunkel)}}\right)$$

$$f = (1-(1-ak1))*\left(\frac{\left(1-e^{-ak2*t}\right)}{ak2*t}\right)$$

$$fma = f * fmgpr + cc2*\left(1-e^{-ak2*t}\right)$$

where ftemp is the temperature, fmgpr is the input fission gas produced (moles), and cc2 is the specified concentration of grain trapped fission gas. Now assuming that gbbgi is the specified concentration of grain boundary trapped fission gas, the fission gas not released from the grain boundary can be calculated as

$$gbbg = gbbgi + fma$$

Assuming that comp is the specified plutonium content, fdens is the specified fuel density in kg/m^3 and fgrn is the specified fuel grain size in microns, the grain boundary fission gas release is given by

$$roth = 11.45*\frac{comp}{100}*\left(1-\frac{comp}{100}\right)*10.96$$

$$fp = 1-0.001*\frac{fdens}{roth}$$

$$p = \frac{1}{\left(1+\left(\frac{1}{fp}-1\right)*\frac{10^5}{\left(fgrn^3\right)}\right)}$$

$$fbr = \left(1-erff\,(p)\right)+e^{-bunkel}$$

where the error function is approximated by

$$
\begin{aligned}
erff(x) &= 1 - 0.348024 * \frac{1}{(1 + 0.47047 * x)} \\
&+ 0.0958798 * \left(\frac{1}{1 + 0.47047 * x} \right)^2 \\
&- 0.7478556 * \left(\frac{1}{1 + 0.47047 * x} \right)^3 \\
&+ 0.000025
\end{aligned}
$$

Finally by defining cc1 as the output concentration of grain trapped fission gas, gbgout as the output concentration of grain boundary trapped fission gas and fmgrot as cumulative fission gas released we can calculate

$$
\begin{aligned}
fmaa &= fbr * gbbg \\
cc1 &= cc2 + fmgpr - fma \\
gbgout &= gbbg - fmaa \\
fmgrot &= fmgr + fmaa
\end{aligned}
$$

Results

The calculations showed that the outer fueled annulus did indeed exhibit a significantly higher fission gas release that the equivalent region for the reference solid pellet. Since the fuel is being compared on a per unit length basis, the fission gas release results are organized into area weighted ratios as shown below in Table 4.

Solid Pellet	
Weighted Fission Gas Release Zone 1	3.23%
Weighted Fission Gas Release Zone 2	21.06%
Duplex Pellet	
Weighted Fission Gas Release Zone 1	0.00%
Weighted Fission Gas Release Zone 2	25.37%

Table 4: Weighted Fission Gas Release Ratios

Thus the final fission gas release ratios for the solid reference and duplex fuel can be easily calculated and are given below in Table 5.

Zone 2 Fission Gas Release Ratio	Total Fission Gas Release Ratio
1.2046	1.0443

Table 5: Final Fission Gas Release Ratios

As shown above, the 20.4% higher fission gas release in the duplex fuel designs outer annulus more than compensates for the larger fission gas release which occurs in Zone 1 of the solid reference fuel. Overall, the 4.4% higher fission gas release for the duplex fuel is not significantly larger than that of the solid reference. However, this result is surprising in that it goes against the conventional wisdom of a lower centerline temperature always yielding a lower cumulative fission gas release.

Conclusions

Since this was a scoping study, the treatment of several variables was fairly rough. Aside from treating the thermal conductivity as non constant, the analysis could also be further refined by considering a larger number of temperature averaged cylinder zones. The 4.4% difference in fission gas release determined in this scoping study is likely to be overshadowed by the uncertainties in the analysis. Thus it would be safe to conclude that while the duplex design does have lower average temperatures, the fission gas release is comparable to that of the solid fuel design.

References

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