Winter Internship Project Report on

Numerical Estimation of Fission Gas Release Fraction from a Nuclear Fuel Element

By

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Abstract

The fission gas release in a cylindrical fuel rod is numerically estimated by various methods and the results are analyzed. This report considers a cylindrical fuel rod, 1.1 cm in diameter, at the beginning of its life-cycle, whose outer-surface temperature is fixed at 675 K. The rod is filled with Uranium Dioxide (UO_2) matrix. The radial temperature profile of the cross-section is obtained for various linear heat ratings, by doing a thermal analysis for solid cylindrical fuel pins as discussed in [TK11]. The fission gas release is then modelled with an empirical dependence on temperature as outlined in [Not70]. Two more diffusion-based models, [NH78; WT83], are discussed which consider the effect of temperature, heat rating, grain size and burn-up on the release of fission gas. These effects are highlighted by various sensitivity studies.

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1 Introduction

Fission gas release in fuel rods during nuclear fission is a critical process, and can influence several key factors such as the heat conductivity outside the fuel. It also has bearing on the swelling and cracking of fuel rods over use. The significant effect that the release of fission gases has on the functioning of several key reactor components and processes, makes it imperative for precise models to be developed which can analyze this behaviour under various environmental conditions.

However, such models often face a trade-off between their scope and their complexity. Highly accurate models, which account for the myriad contributing factors to the release of fission gas, tend towards bewildering complexity. On the other hand, empirical and pseudo-empirical models which utilize only a few key contributing factors to analyze this behaviour, often lose out on the nuanced effect that other unaccounted factors can have. This trade-off will become apparent in later sections as the report progresses to various models.

This report analyzes a hypothetical fuel rod composed entirely of Uranium Dioxide (UO_2) with a diameter of 1.1 cm and the outer surface fixed at 675 K. As the axis is significantly longer than the radial dimension, the axial effects of thermal conductivity are ignored and the radial temperature profile is determined for any given linear heat rating.

An empirical model, which estimates the fraction of fission gas released based solely on the temperature of the region, is then implemented. It is completely independent of burn-up and grain size.

Two diffusion models (Notley and Hastings, White and Tucker) are then implemented to account for the effect of burn-up and grain size and observe the nuances in this behaviour. The results of these models are compared and their relative susceptibility to change in factors such as burn-up, heat rating and grain size is analyzed.

2 Methodology

2.1 Phase I - Temperature Profiling

The circular cross-section of the rod is split into 100 concentric rings of 0.005 cm thickness. The smallest central circle is of 0.05 cm in radius. The rings are numbered in the radially outward direction (100th ring is the outermost surface). Thus, the general relation for the radius (in cm) of the nth ring is given by

$$r_n = \frac{n}{200} + \frac{1}{20} = (0.005n + 0.05) \text{ cm} \tag{1}$$

Consider a element power of $q_1 = 300W/cm$, from this we can calculate the volumetric heat flux

$$q_3 = \frac{q_1}{\text{Area of cross-section}} = \frac{300 \text{ W/cm}}{\pi 0.55^2 \text{ cm}^2} = 315.68 \text{ W/cm}^3 \tag{2}$$

This is a measure of the heat power generated per unit volume of fuel. The thermal conductivity of the UO_2 matrix can be determined based on the temperature of the region by the empirical relation

$$k = \frac{38.24}{129.4 + T} + 6.1256 \times 10^{-13}\, T^3 \tag{3}$$

where k is the thermal conductivity of UO_2 in W/cm K and T is the local UO_2 temperature in K.

We now analyze the heat conduction equation of the cylindrical fuel pellet. Symmetry allows us to ignore azimuthal temperature gradients, and for fuel pellets whose length-to-diameter ratio exceeds 10, we can ignore axial heat transfer within the fuel in comparison to the radial heat transfer for the bulk of the pellet length.

With these declarations, the steady state heat conduction equation is a one-dimensional equation in the radial direction

$$\frac{1}{r}\frac{d}{dr}\left(kr\frac{dT}{dr}\right) + q_3 = 0 \tag{4}$$

This can be integrated and written as

$$kr\frac{dT}{dr} + q_3\frac{r^2}{2} + C_1 = 0 \tag{5}$$

where C_1 is a constant of integration. For symmetrical pellets, the radial heat flux (q_2) at the centre-line ($r = 0$) will be 0.

$$q_2|_{r=0} = -k\frac{dT}{dr}|_{r=0} = 0 \tag{6}$$

Substituting Equation (6) in Equation (5), we get $C_1 = 0$, this gives us the relation

$$dT = -\frac{q_3}{2k} r dr \tag{7}$$

As we have considered 100 rings in a 1.1 cm diameter cross-section, the thickness of each ring is very small, allowing us to assume that each individual ring has a uniform temperature and there is a discrete step as you go from one ring to another. Note the negative sign, this implies that the temperature decreases in the radial direction i.e., the center-line temperature is the maximum temperature in the fuel pellet.

Our hypothetical system is constrained to have a fixed outermost surface temperature of 675 K. This is the temperature of the 100^{th} ring. We can now iterate inwards, calculating the thermal conductivity for each ring and thus, the increased temperature of the inner ring. After 100 such iterations, we arrive at the center-most solid circle in the cross-section (of radius 0.05 cm). Thus, we have found the temperature at each of the 100 rings, and have a numerical estimation of the radial temperature distribution of the fuel pellet. This method can easily be extended to a higher number of iterations for increased accuracy and continuity.

The radial temperature profile of the cross-section of the fuel pellet can be seen as below.

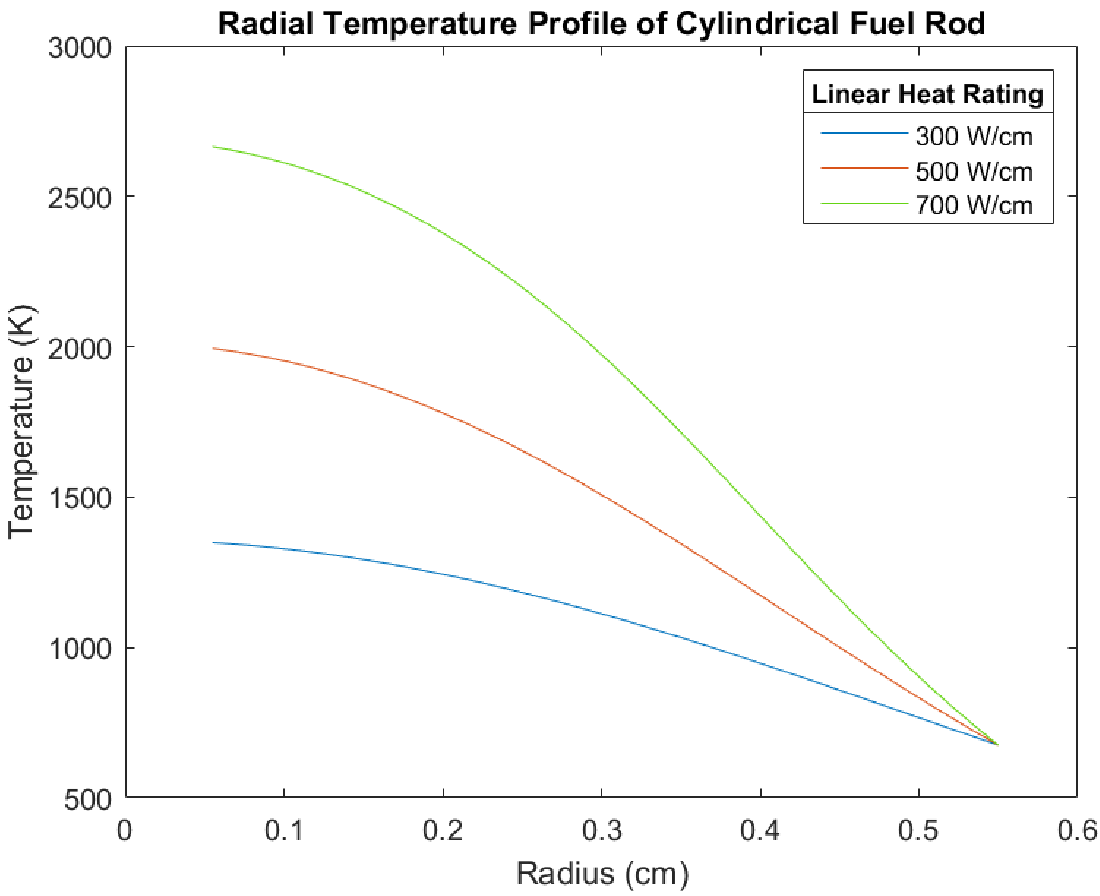

Figure 1: Radial Temperature Distribution of Fuel Pellet at Element Powers of 300 W/cm, 500 W/cm and 700 W/cm.

Observe the temperature increase and plateau as we move closer to the centre-line. The centre-line temperature is the maximum temperature attained in the system. Higher element powers result in higher temperatures. The centre-line temperatures observed at the various element powers are:

- Element power of $q_1 = 300$ W/cm - Centre-line temperature of $T_c = 1350.5$ K
- Element power of $q_1 = 500$ W/cm - Centre-line temperature of $T_c = 1997.5$ K
- Element power of $q_1 = 700$ W/cm - Centre-line temperature of $T_c = 2668.7$ K

2.2 Phase II - Empirical Model

We now move forward to the next phase, applying an empirical model to predict the fraction of fission gas released based on the temperature of the fuel. Such a formulation is given in [**Notley**] as:

$$\begin{aligned}
f &= 0.05 & T &< 1673K \\
f &= 0.10 & 1773 > T &> 1673K \\
f &= 0.20 & 1873 > T &> 1773K \\
f &= 0.40 & 1973 > T &> 1873K \\
f &= 0.60 & 2073 > T &> 1973K \\
f &= 0.80 & 2273 > T &> 2073K \\
f &= 0.98 & T &> 2273K
\end{aligned}$$

Our system has a range of temperatures spread out over a circular cross-section. Each ring (n^{th}) has a uniform temperature (T_n), and thus, a uniform value for the fraction of fission gas released (f_n). In order to find an estimate of this fraction for the entire fuel pellet, we do an areal average over the cross-section.

$$f_{avg} = \frac{\sum_{n=1}^{n=100} f_n \times \pi(r_n - r_{n-1})^2}{\pi r_{100}^2} \tag{8}$$

The average fraction of fission gas released by the fuel rod for the three element powers discussed in Section 2.1 are:

- Element power of $q_1 = 300$ W/cm - FGR fraction of $f_{avg} = 0.0500$
- Element power of $q_1 = 500$ W/cm - FGR fraction of $f_{avg} = 0.0919$
- Element power of $q_1 = 700$ W/cm - FGR fraction of $f_{avg} = 0.3166$

We now consider 10,000 data points between 300 W/cm and 700 W/cm, and plot the dependence of fission gas release fraction on the element power.

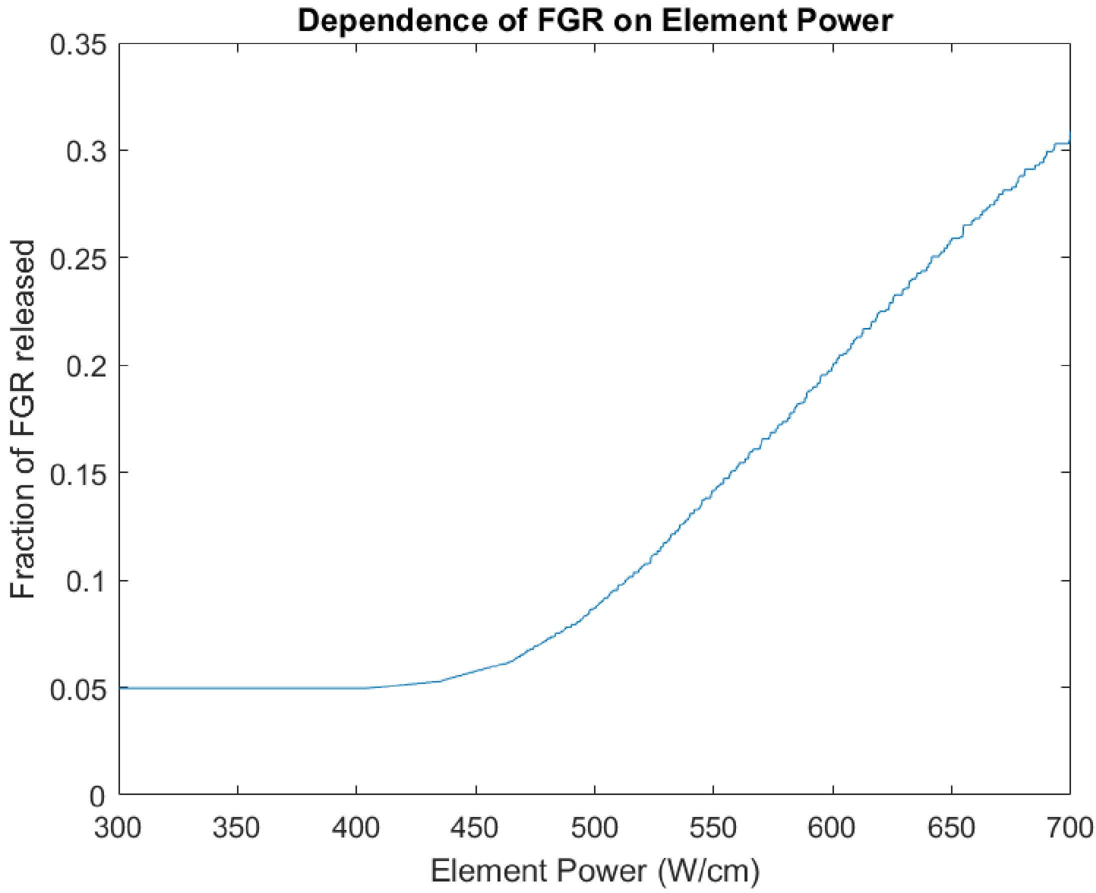

Figure 2: Fraction of FGR as a function of Element Power over a range of 10,000 values.

As this model depends exclusively on the temperature, which in turn is determined by the element power q_1, factors such as burn-up and grain-size are inconsequential to the value obtained. This calls for more comprehensive models that can account for the variety of factors involved in the release of fission gases, and analyze their effect. A few such examples, modelling the diffusion of fission gases through the fuel material after their generation, are discussed in the following subsection.

2.3 Phase III - Diffusion Models

2.3.1 Notley and Hastings

Notley and Hastings designed a microstructure-dependent model to analyze the fission product gas release. Their primary goal was to create a physically sound model considering the processes occurring at every stage of the process. This report will draw attention to their approach to modelling the diffusion of fission gas after generation and attempt to understand the role of factors such as element power, burn-up and grain size on the amount of fission gas released.

Findlay had measured the diffusional release of Krypton-85 in reactors from specimens of a known surface-to-volume ratio. A similar manner of measurement would allow the calculation of release from a sphere regardless of the manner of gaseous migration (atomically or intragranular bubbles). Findlay obtained a relation for a diffusion coefficient, D, dependent on the absolute temperature, T, given by:

$$D = 7.8 \times 10^{-2} \exp\left(-\frac{288 \text{ kJ mole}^{-1}}{R(T/1000)}\right) \text{ m}^2\text{s}^{-1} \tag{9}$$

In the system being considered in this report, we are analyzing a fuel pellet from the very beginning of its usage. This implies that there is no inventory of "old" gas, and all the gas released in the subsequent processes was generated in the time-frame being considered.

Let the grain radius of the sphere by denoted by a, and the time interval by t. The model considers two cases: the short time approximation and the long time approximation.

- If $t < \frac{a^2}{\pi^2 D}$,

$$f = 4\left(\frac{Dt}{a^2\pi}\right)^{\frac{1}{2}} - \frac{3}{2}\frac{Dt}{a^2} \tag{10}$$

- If $t \geq \frac{a^2}{\pi^2 D}$,

$$f = 1 - \frac{a^2}{15Dt} + \frac{6a^2}{\pi^4 Dt}\exp\left(\frac{-\pi^2 Dt}{a^2}\right) \tag{11}$$

where f is the fractional release of fission gas.

The complete life-time of the fuel rod at a constant burn-up (B) and element power (q_1) can be determined in the following manner. Note that the Uranium metal density in UO_2 is $\rho = 9.67 \times 10^3 \text{kg/m}^3$, and the volumetric heat flux is as given by Equation (2).

$$t_{\text{life}} = \frac{B\rho}{q_3} \tag{12}$$

Substituting t_{life} in place of t in Equation (10) and Equation (11), will give you the fractional fission gas released for a particular burn-up, grain radius, element power and temperature. The temperature profile of the radial cross-section has already been discussed for a given element power rating in Section 2.1. Following the same areal average estimation discussed in Equation (8), we obtain f_{avg} for the entire fuel pellet. This result is dependent on three key factors: burn-up, grain radius and element power.

We shall now explore its sensitivity to element power at a few values of grain radius and burn-up. For three values of burn-ups 50, 200 and 1000 MW h/kg U, the dependence of the fraction of fission gas released on element power, ranging from 300 W/cm to 700 W/cm, is plotted below.

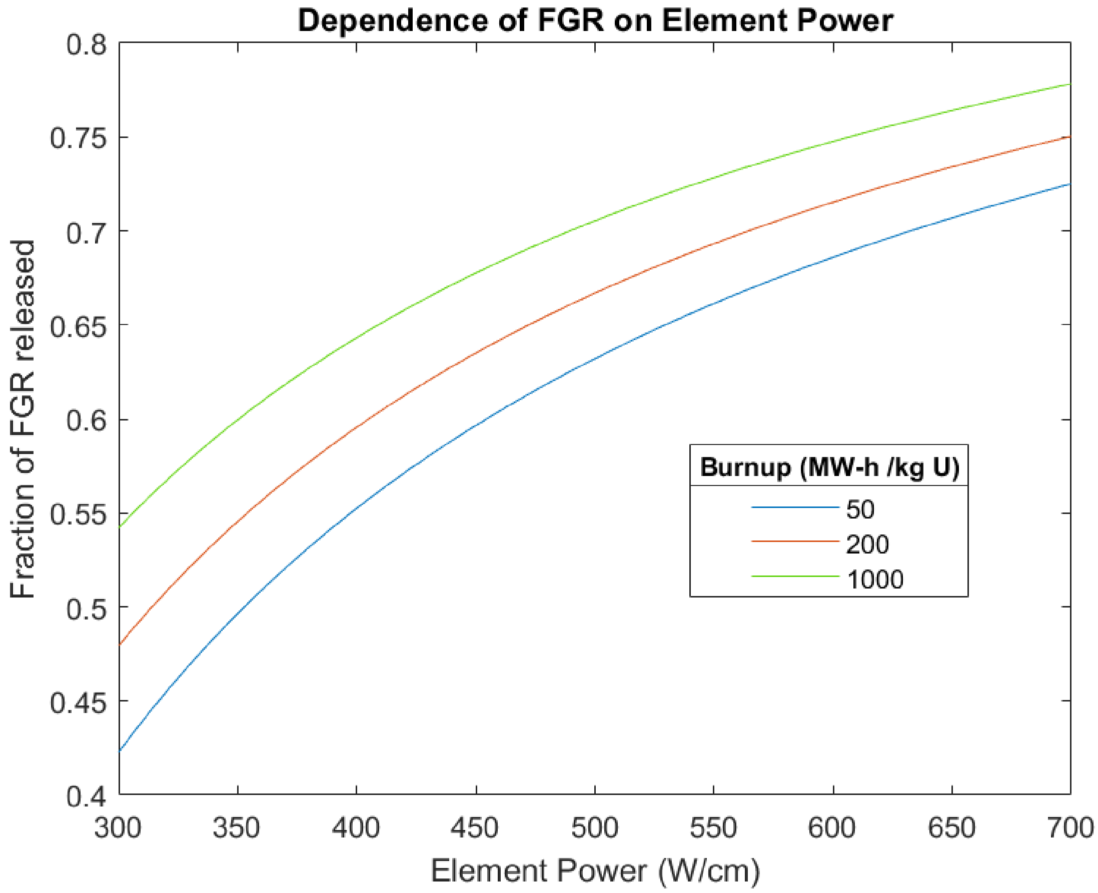

Figure 3: Notley and Hastings Model of FGR Fraction as a function of Element Power at 100 micron grain radius.

2.3.2 White and Tucker

The release of fission gases from the irradiated fuel is widely understood to be controlled by atomic diffusion to the fuel grain boundaries. This approach was pioneered by Booth, and all subsequent diffusion-based interpretations have been built around that approach. We treat the fission gas release from UO_2 as a diffusion process to the surface of a spherical grain. While, the grains in the fuel matrix may not be spherical, this approach allows a startling simplicity into the model, and it can be tailored to specific grain geometries as required.

This idealised spherical grain approach assumes that fission gas is generated uniformly at a rate (say G) throughout a grain of radius a. Thus, the spherical diffusion equation which should be consistent throughout the sphere is:

$$\frac{\partial c}{\partial t} = D\nabla^2 c + G \tag{13}$$

The constraints imposed on this differential equation (by symmetry and boundary conditions) are

- $c = 0$ at $r = a, 0 \leq t$ and $0 \leq r \leq a, t = 0$
- $\frac{\partial c}{\partial r} = 0$ at $r = 0$

For brevity in notation, we adopt the following substitution $\omega = \frac{Dt}{a^2}$. We can integrate the concentration profile thus defined up to a time t to obtain the fractional release f.

$$f = 1 - \frac{6}{\omega} \sum_{n=1}^{\infty} \frac{1 - \exp\left(-\pi^2 n^2 \omega\right)}{(n\pi)^4} \tag{14}$$

As this report is conducting a numerical estimation, we sum the exponential part up to 100 terms to get a reasonably close approximation. This model also has a short time approximation similar to Notley and Hastings. If $\omega < \frac{1}{\pi^2}$,

- If $f < 57\%$,
$$f = 4 \left(\frac{\omega}{\pi}\right)^{\frac{1}{2}} - \frac{3}{2}\omega \tag{15}$$
- If $f \geq 57\%$,
$$f = 1 - \frac{0.0662}{\omega} \left[1 - 0.93 \exp\left(-\pi^2 \omega\right)\right] \tag{16}$$

The equations Equation (14), Equation (15) and Equation (16) model the diffusional release of stable gases from a sphere of UO_2 matrix, when the initial concentration of fission gas inside the grain is zero.

For the same environment as analyzed for the Notley model, we plot the dependence of the fraction of fission gas released on element power ranging from 300 W/cm to 700 W/cm, for three values of burn-ups 50, 200 and 1000 MW h/kg U below.

To observe the sensitivity of fission gas release to burn-up, we plot the fraction of fission gas released to a range of burn-up values from 50 MW h/kg U to 1000 MW h/kg U. These are plotted for three element powers: 300, 500 and 700 W/cm, and a grain radius of 100 micron and 10 micron. Note that the values are significantly higher for 10 micron, implying that smaller grain sizes increase the fraction of fission gas released.

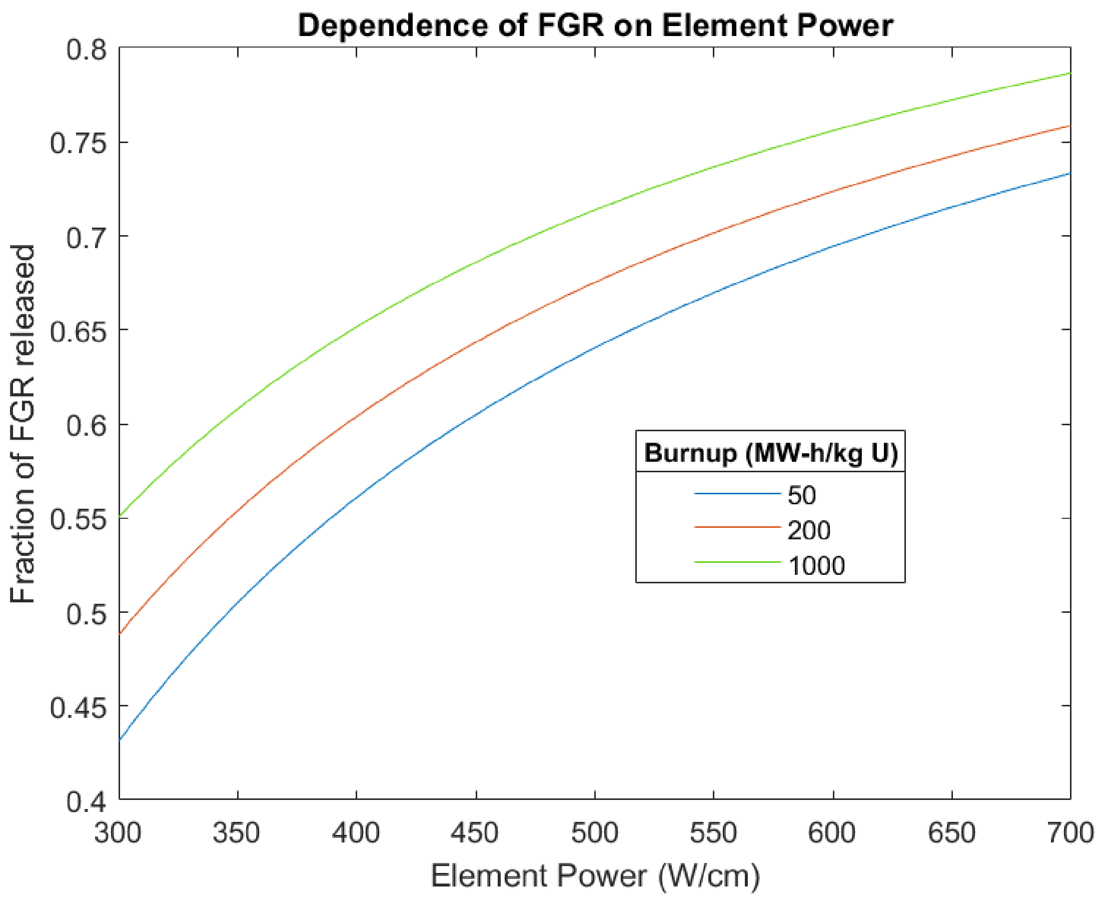

Figure 4: White and Tucker Model of FGR Fraction as a function of Element Power at 100 micron grain radius.

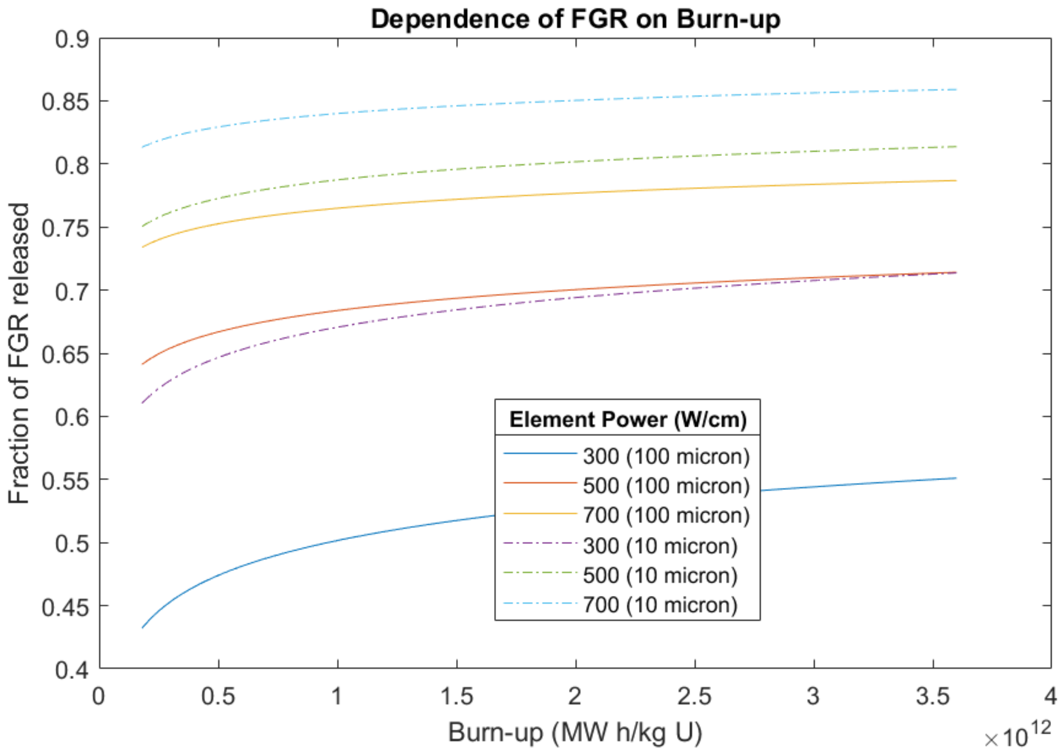

Figure 5: White and Tucker Model of FGR Fraction as a function of Burn-up at 100 micron and 10 micron grain radius.

3 Summary

This report has numerically estimated the temperature profile of the fuel pellet and thus, the release of fission gases from a nuclear fuel element. While it has dealt with the basic scenario of uniform power history and considered the hypothetical fixed temperature constraint of 675 K at the outer surface, the model is a sound starting point to be extended to particular cases as required. The release of fission gas has been empirically plotted against the temperature, and shows a strong dependence on it.

The diffusion model plots highlighted the role of grain size and burn-up to the release of fission gas. We can conclude that the temperature (influenced by the element power) is an influencing factor, and a higher fraction is released at higher temperatures i.e., higher element powers. A higher fraction is also released at higher burn-up values. A point to note however, is that increasing the grain size can reduce the fraction of fission gases released.

The nature of the plots also serve as indicators of relative sensitivity, as there is a large fluctuation over a change in the element power, whereas the fission gas release tends to plateau after crossing a threshold burn-up. This implies that element power is a primary factor of influence, and burn-up is a secondary factor of influence.

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