Comparison of Fission Gas Swelling Models for Amorphous U_3Si_2 & Crystalline UO_2

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ABSTRACT:

Theoretical models are used in support of the I²S-LWR (Integral Inherently Safe LWR) project for a direct comparison of fuel swelling and fission gas bubble formation between U_3Si_2 and UO_2 fuels. Swelling in uranium silicide is evaluated by analyzing the bubble coarsening process' effects on the bubble population and size distribution. The uranium dioxide is examined with two models: one which calculates the swelling behavior with a fixed grain radius and a second which incorporates grain growth into the model. These two models use the diffusion of gas atoms to the grain boundaries to calculate the swelling. The different mechanisms controlling the swelling of the fuels are introduced including the "knee point" caused by the amorphous state for the U_3Si_2 . The U_3Si_2 model shows that, for the burnup expected for the I²S reactor, swelling in U_3Si_2 will not reach the "knee point" with accelerated swelling. The results from the fixed grain UO_2 model and U_3Si_2 model are used to compare the swelling of each fuel at the same fission rate.

1. Introduction

Uranium silicide (U_3Si_2) is being examined for the I²S-LWR project as a possible alternative to conventional fuel in a nuclear reactor[1]. There are potentially a multitude of benefits to using U_3Si_2 as fuel over UO_2 . These include high uranium density, increased thermal conductivity, a longer potential fuel cycle, and potentially favorable swelling characteristics. These characteristics are important to I²S-LWR as they lead to U_3Si_2 being an accident tolerant fuel that reacts favorably when compared to UO_2 in a loss of coolant accident (LOCA) scenario.

Theoretical models more readily allow for a direct comparison between fuels. More specifically, fuel swelling and fission gas bubble formation for U_3Si_2 and UO_2 . Separate models are implemented to evaluate the fission gas bubble formation and swelling behavior of U_3Si_2 and UO_2 due to different mechanisms for bubble formation. Separate models are necessary due to ceramic versus intermetallic composition and methods of bubble formation.

If swelling is too severe then contact between the fuel and cladding may occur. When this occurs there may be a fuel-cladding chemical interaction (FCCI) or a fuel-cladding mechanical interaction (FCMI.) Both of which may lead to cladding failure.

Historically U_3Si_2 has been used in RERTR testing as an aluminum dispersed plate fuel which differs from tradition pellet fuels in that they are high fission density and low temperature fuels. As a result, much of the data for U_3Si_2 is at higher fission densities and lower temperatures than conventional fuels. Originally it was thought that U_3Si_2 behaved similarly to UO_2 while being irradiated in that the fuel stayed in a crystalline state and models were built around this assumption. This was assumed because after undergoing irradiation, U_3Si_2 undergoes swelling at an unacceptably high rate, which has been attributed to amorphization. Although U_3Si_2 did not undergo this extreme swelling behavior and was assumed to be crystalline like UO_2 since it has relatively small bubbles through high burnup[2]. When models for swelling did not match experimental results, further investigation by irradiation with krypton ions showed that the fuel underwent amorphization fairly rapidly and maintained an amorphous state until high doses[3]. While U_3Si_2 undergoes amorphization under irradiation, Birtcher et al. has shown it only stays in the amorphous state up to the temperature of 250°C when under irradiation with

1.5 MeV Kr ions [4]. Although the fuel may still have regions that are amorphous while under irradiation within a reactor.

The uranium silicide model, developed by Dr. J. Rest[2], with the fuel in an amorphous state was used. Since uranium silicide rapidly becomes amorphous under irradiation, the distribution of bubbles and their effects on the material differ from those of uranium dioxide. U_3Si_2 stays amorphous until a high dose because of the higher number of the stronger Si-Si bonds in U_3Si_2 relative to U_3Si . [5] These bonds also increase with burnup as U is depleted. Normally under an amorphous state the diffusivity of the bubbles are much higher and when combined with the plastic flow rate of the fuel lead to excessive swelling. In the case of U_3Si_2 , the Si-Si bonds overcome these effects. In U_3Si there is a dramatic increase in free volume, while U_3Si_2 contracts upon amorphization reducing free volume. [6]

In the amorphous state, bubble nucleation sites are distributed uniformly and take on the free volume that is created when U_3Si_2 contracts upon amorphization.

Another beneficial factor affecting the swelling behavior of U_3Si_2 is that the solubility of the fission gas atoms increases with fuel burn-up. The mechanism for this is an increase in Si-Si bonds relative to the weaker U-Si bonds as the uranium in the fuel is spent reducing the U to Si fraction. Due to the increase in the stronger Si-Si bonds the diffusivity for the fission gas atom decreases trapping more gas atoms in solution. [6]

Initially all bubbles are assumed to be distributed uniformly. Bubbles grow as gas atoms diffuse to the bubbles. As the bubbles begin to grow their distribution has a peak of smaller bubble and a tail of larger bubbles. As they continue to grow larger, bubbles begin to coalesce. Eventually, bubbles in the tail reach a size where they, on average, coalesce with one other bubble. This point is known as the knee point and is where swelling accelerates due to the increase in bubble coalescence. The number of bubbles then decreases and the distribution shifts to larger bubbles as they continue to grow and repeat the coalescence process.

In UO_2 the nucleation sites are at vacancies and the bubbles are concentrated at grain boundaries. Vacancies are created due to irradiation and gas diffusion is dependent on vacancy migration. The gas diffusion is dependent on three parts; intrinsic diffusion at high temperatures, vacancy assisted diffusion dependent on both temperature & fission rate, and irradiation enhanced athermal diffusion [7, 8]. In addition to intragranular bubbles, there are intergranular bubbles at the grain boundaries. Over time as intragranular bubbles and gas atoms accumulate on the grain boundaries, the intergranular bubbles grow and cover the grain faces. Eventually they interconnect along the grain boundaries, which can lead to fission gas release when the interconnection reaches the surface. For UO_2 , one model calculates the swelling behavior with a fixed grain radius [8] while the second incorporates grain growth into the model [8-12]. In the grain growth model swelling happens similarly, but as the grain faces sweep across intragranular bubbles they are accumulated into the grain face.

2. Theory & Methods

2.1. Theory & Methods – U_3Si_2

The U_3Si_2 amorphous model was developed from a previously published model [2]. The following table list the inputs used to produce the swelling data for U_3Si_2 unless otherwise stated (varying temperature and fission rate.)

Table 1: U_3Si_2 Swelling Model Variables and Constants

Symbol	Description	Units	Value
Temp	Temperature	K	473
kb	Boltzmann Constant	(m ² *kg)/(s ² *K)	1.3806E-23

R _g	Radius of diffusing gas atom	m	0.216E-9
$\dot{f}$	Fission Rate	m ⁻³ s ⁻¹	1.7E20
f ₀		m ⁻³ s ⁻¹	FissR/2
n ₀	Exp(-Theta/Temp)	kg/(m*s)	2e7*f ₀ *0.1
b ₀	Initial gas atom resolution rate	m ³	2E-23
β _e		m ^{3/2} s ^{1/2}	5.6E-12
β _v	Van der Waals Constant	m ³	(5.16E-5)/(6.023E23)
α		m ⁻³ s ⁻¹	5E-10*f ₀
f _n		kg m ⁻⁶ s ⁻³	4*α*n ₀ *R _g /.1
β	Gas atoms produce per fission		0.25
γ		J m ⁻²	0.7

First t_{knee} is solved analytically before running the Matlab model to calculate the swelling at the knee with the appropriate equations and then again for after the knee point. $I(r_i)$ is the average number of bubbles a bubble of radius r_i intersect with and r_i is the smallest bubble in the tail of the bubble size distribution. To calculate t_{knee} , $I(r_i)$ is set to 1 for Eq. 1 below and then solved for t_{knee} . This is when the average number of other bubbles a bubble of radius r_i intersect with is one other bubble. Or that all bubbles in the tail of the bubble distribution participate in bubble coalescence (since r_i is the smallest bubble in the tail.)

Table 2: U_3Si_2 Swelling Model Equations

Variable	Description
$R_{ds} = \beta_v \cdot \chi \cdot \beta \cdot \dot{f} \cdot t$	ratio of the average diameter of a bubble to the average inter-bubble spacing
$\chi = \frac{4\pi \cdot f_n \cdot D_o \cdot \beta_e^2 \cdot t}{b_o \cdot \beta}$	Fraction of fission gas generated that exists in bubbles
$k = \frac{b_o}{2 \cdot \beta \cdot D_g \cdot \beta_v \cdot t}$	
$D_g = \frac{k_b \cdot Temp}{6\pi \cdot R_g \cdot n}$	Diffusivity of gas-atom
$D_o = \frac{k_b \cdot Temp}{6\pi \cdot R_g \cdot n_o}$	Trap free diffusion
$n = n_o / \dot{f}$	viscosity
$Bub_a = \frac{2\beta_v \cdot \chi \cdot \beta \cdot \dot{f} \cdot t}{\pi^{3/2}}$	
$Bub_b = k^{5/2} \cdot r \cdot \exp[-k \cdot r^2]$	
$Bub_D = Bub_a \cdot Bub_b$	Bubble Density

$$I(R_i) = \frac{Rds^3}{6\sqrt{\pi}} \left(4 \sqrt{\frac{\chi b_o R_i^2}{Rds^3 D_n}} \left(3 + \frac{\chi b_o R_i^2}{Rds^3 D_n} \right) + \sqrt{\pi} \left(3 + \frac{6\chi b_o R_i^2}{Rds^3 D_n} \right) \right) \quad Eq. 1$$

Volume swelling before and after the knee is calculated with the following equations:

Table 3: U₃Si₂ Volume Swelling Equations

Variable	Description
$R_k = \sqrt{\frac{50}{k} + \frac{196\pi k_b \cdot Temp}{2\gamma \cdot \beta_v \cdot k^{3/2}}}$	r knee
$R_p = 1/\sqrt{2k}$	radius at peak of bubble size distribution
$R_i = 3.33R_p$	radius at I=1, smallest bubble within the tail region
$R_t = 5R_p$	radius at center of tail
$N_1 = 3.6\chi \cdot \beta_v \cdot \beta \cdot t \cdot \dot{f} \cdot (k/\pi)^{3/2}$	bell shaped distribution of small diameter bubbles
$N_2 = 2 \cdot 10^{-3} \chi \cdot \beta_v \cdot \beta \cdot t \cdot \dot{f} \cdot (k/\pi)^{3/2}$	tail shaped distribution of large diameter bubbles
$m(r) = \frac{4\pi r^3}{3\beta_v}$	gas atoms "m" in bubble population
$mrp = \frac{4\pi R_p^3}{3\beta_v}$	bubble distribution peak population
$mrt = \frac{4\pi R_t^3}{3\beta_v}$	bubble distribution tail population
$dN1 = N_1/2$	
$m_o = mrt + \frac{mrp \cdot dN1}{N_2}$	
$\phi = 8 \cdot 10^{-8}$	s ⁻¹ , experimentally determined, time constant
$\chi_k = \frac{4\pi \cdot f_n \cdot D_o \cdot \beta_e^2 \cdot t_k}{b_o \cdot \beta}$	chi at knee
$\beta_s = 1.75 \cdot 10^{-29}$	m ³ , fractional swelling due to solid fission products per unit fission density

Fractional volume change before the knee point:

$$Vsk = \frac{4\pi}{3} \left(\frac{R_p^3 \cdot N_1}{2} + R_k^3 \cdot N_2 \right) + \beta_s \cdot \dot{f} \cdot t_k \quad Eq. 2$$

Fractional volume change after the knee point:

$$Vsa = \beta_s \cdot \dot{f} \cdot t + \frac{2\pi R_p^3 \cdot N_1}{3} (1 + \chi/\chi_k) \left(\frac{3k_b \cdot Temp}{8\pi\gamma} \left(m_o + \frac{\beta \cdot \dot{f} \cdot t \cdot \exp[\phi \cdot t] (1 - \chi/2)}{N_2} \right) \right)^{1/2} \quad Eq. 3$$

2.2. Theory & Methods – UO₂ Fixed Grain

The models used to calculate the swelling of UO₂ were prepared by the process described in Gibson's thesis[13]. In UO₂ bubbles form at vacancies and vacancy migration leads to bubble growth. The process is less iterative than U₃Si₂ in that the grains act as sinks for the bubbles & vacancies and continue to grow as the intragranular bubbles diffuse to the boundaries. Over time the intergranular bubbles grow causing the grain faces to grow into voids as the fuel swells.

After swelling into voids, the grain faces begin to interconnect. Eventually the grain face void interconnections reach the surface and allow fission gas to escape. This is known as the saturation point. After the saturation point is reached, the fission gas is allowed to escape.

Table 4: UO₂ Swelling Model Variables and Constants [13-17]

Symbol	Description	Units	Value
T	Temperature	K	
kb	Boltzmann Constant	(m ² *kg)/(s ² *K)	$1.3806 \cdot 10^{-23}$
P	External Pressure	Pa	10^6
t	Time	s	$3.6 \cdot 10^7$
ts	Time Step	s	3600
R	Grain Radius	m	$7.5 \cdot 10^{-6}$
r _{bs}	Average intergranular bubble radius at saturation	m ³	10^{-6}
cgb	Composite Gas Bubble Parameter [13]	m	$7.25 \cdot 10^{-8}$
Na	Avogadro's number		$6.023 \cdot 10^{23}$
PD	Power Density	kW m ⁻¹	35
C ₁	Constant		$7.6 \cdot 10^{-10}$
C ₂	Constant		$4.5 \cdot 10^{-35}$
C ₃	Constant		$2 \cdot 10^{-40}$
Q ₁ /kb	Activation Energy	K	35247
Q ₂ /kb	Activation Energy	K	13800
L	Fission fragment range	m	$6 \cdot 10^{-6}$
b	van der Waals constant for Xenon	m ³ mol ⁻¹	$5.16 \cdot 10^{-5}$
δ	Damage radius of fission fragment	m	10^{-9}
γ	Surface tension	N/m	$2.4 \cdot 10^6$

Table 5: Inputs for UO₂ Swelling Model[13-17]

Variable	Description
$F_m = PD \cdot 5.189 \cdot 10^{-14}$	Molar fission density
$F = F_m \cdot Na$	Fission Density
$G_p = F_m \cdot 0.3$	Gas atom production rate

$G_r = G_p \cdot 5.7 \cdot 10^{-8}$	Gas atom resolution rate [13]
$D' = C_1 \cdot e^{\frac{-Q_1}{k_b \cdot T}} + C_2 \cdot F \cdot e^{\frac{-Q_2}{k_b \cdot T}} + C_3 \cdot F$	Diffusivity in trap free media for UO ₂
$C_b^t = \frac{1.52 \cdot 10^{27}}{T} - 3.3 \cdot 10^{23}$	Total Bubble Density
$\bar{R}_b = 1.453 \cdot 10^{-10} \cdot e^{1.023 \cdot 10^{-3} \cdot T}$	Intragranular bubble radius, m
$g = 4\pi \cdot \bar{R}_b \cdot C_b^t \cdot D'$	Fission gas capture rate by intragranular gas bubbles
$vg = 3.03\pi \cdot L \cdot F \cdot (\bar{R}_b + \delta)^2$	Intragranular gas bubble resolution rate
$D = \frac{D' \cdot vg}{vg + g}$	Fission gas diffusion for UO ₂
$N_b = \frac{2}{3} R \cdot C_b$	Number density of gas bubbles
$N_{bs} = \frac{cgb \cdot P(1 + \kappa)}{k_b \cdot T(1 + v_p(1 + \kappa))}$	Number density of gas bubbles at saturation

Over time the intergranular bubble gas density increases which leads to fuel swelling. Additionally swelling is contributed to by the intragranular fission gas bubbles and the solid fission products. The swelling from the intergranular gas bubbles stops when saturation is reached or N_{bs} . The equation for gaseous swelling is:

$$\frac{\Delta V}{V} = \frac{4\pi \cdot r_b^3 \cdot \omega_\theta \cdot n_b}{3} \quad Eq. 4$$

In this equation r_b is the intergranular bubble radius in m, ω_θ is a geometric factor used to correct for the use of spherical bubbles with θ equal to intergranular bubble dihedral angle, n_b is the intergranular gas bubble density in m^{-3} . The dihedral angle is assumed constant.

$$\omega_\theta = 1 - 1.5 \cdot \cos \theta + 0.5 \cos^3 \theta \quad Eq. 5$$

The number of gas atoms per bubble, following van der Waals Law of gases is

$$N_{atom} = \frac{1}{k_b \cdot T} (V_b - b \cdot N_{atom}) \left(P_b + \frac{a \cdot N_{atom}^2}{V_b^2} \right) \quad Eq. 6$$

Where a & b are van der Waals constants for xenon, aN^2 is assumed to be negligible, and P_b is the bubble pressure. The pressure of the gas bubble is [18]

$$P_b = \frac{2\gamma}{r_b} + P \quad Eq. 7$$

Where γ is the bubble surface tension. The intergranular gas atom density and saturated density can be written as

$$C_b = \frac{4\pi}{3} r_b^3 \cdot \omega_\theta \cdot n_b \frac{P_b}{b \cdot P_b + k_b \cdot T} \quad \text{Eq. 8}$$

$$C_{bs} = \frac{3 \cdot N_{bs}}{2 \cdot Na \cdot R} \quad \text{Eq. 9}$$

Then comparing C_b to it's saturation value, C_{bs} , the intergranular bubble radius, r_b , can be seen through the ratio of it and it's saturation value.

$$\frac{C_b(t)}{C_{bs}} = x^2 \frac{x + \kappa}{1 + \kappa} \left(\frac{1 + (1 + \kappa)v_p}{1 + (1 + \kappa/x)v_p} \right) \quad \text{Eq. 10}$$

Where x is the ratio of r_b & r_{bs} , κ is the ratio of bubble surface tension to exterior pressure, and v_p is

$$v_p = \frac{b \cdot P}{k_b \cdot T} \quad \text{Eq. 11}$$

Then the volumetric swelling can be calculated, once r_b has been determined, through

$$\frac{\Delta V}{V} = \frac{k_b \cdot T [1 + (1 + \kappa/x)v_p] C_b}{P(1 + \kappa/x)} \quad \text{Eq. 12}$$

2.3. Theory & Methods – UO₂ Grain Growth

The grain growth model is similar to the fixed grain model and both reach a saturation point where gas is allowed to escape. In addition to gas causing the boundaries to become voids and swell, some of the grains grow consuming neighboring grains. Therefore, in the grain growth model, as some grains grow this causes other grains to shrink. As the grain faces sweep across the volume of the fuel, they pick up intragranular gas atoms and release them into the boundaries or voids. This then leads to a decrease in intragranular gas density.

The grain growth model used in Gibson's thesis [13] is developed from Jernkvist's and Massih's work [19]. The rate of grain growth is given as

$$\frac{dR}{dt} = \frac{k_g}{2} \left(\frac{1}{2R} - \frac{1}{D_{m,UO_2}} \right) \quad \text{Eq. 13}$$

$$D_{m,UO_2} = D_0 [H(T_g - T) + H(T - T_g) A_m e^{-Q_m/T}] \quad \text{Eq. 14}$$

$$k_{g,UO_2} = A_g e^{-Q_g/T} \quad \text{Eq. 15}$$

Where D_{m,UO_2} is the maximum grain diameter, D_0 is the initial grain diameter, T_g is 1550K, A_m is 615.59, Q_m is 9955K, A_g is $4.11 \cdot 10^{-9} \text{ m s}^{-1}$, Q_g is 32114.51K, and H is the Heaviside step function. The step function is used to restrict grain growth to temperatures above 1550K where H equals 1 otherwise it equals 0. The grain growth formula comes from Turnbull's work [12].

3. Results & Discussion

3.1. U_3Si_2 Bubble Formation and Density

Over time the bubbles in the amorphous U_3Si_2 grow and have a distribution with a bell shaped peak and tail. At this time there is a portion of the bubbles that reach sufficient size to interact and merge with nearby bubbles. This size is when the radius of the two bubbles is greater than the interbubble distance. The interbubble distance in relation to bubble radius can be seen in the following figure.

Figure 1: Interbubble Distance

In the figure below, $I(r)$ is the average number of bubbles that a given bubble of radius r intersects with. The “knee” occurs when $I(r)=1$ and the radius is equal to r_i . In the figure, this occurs when the radius is approximately equal to 14nm.

*Figure 2: $I(r)$ vs Bubble Radius
Fission Rate: $2.5E20m^{-3}s^{-1}$ at 473K for U_3Si_2*

The U_3Si_2 model exhibits the behavior where the bubbles reach radius r_i and merge at the “knee point”. The swelling accelerates at this point as the bubbles coalesce into a population of larger diameter bubbles.

Figure 3: Bubble Density ($10^{30} m^{-3}$) vs Bubble Radius ($10^{-8} m$)

After the knee point time (t_{knee}) the bubbles population decreases as bubbles coalesce and continue to grow. This continues until the distribution is again a peak and tail, but with a higher average radius and lower population.

3.2. U_3Si_2 Fission Gas Evolution

Figure 4 was generated from equations 2 and 3 (before and after t_{knee} .) From the fractional swelling figure below we can see that at higher fission rates, the swelling is decreased for a given fission density. The swelling is decreased because there is an increase in energetic particles. These energetic particles collide with and collapse some of the smaller bubbles leading to an increase of gas in solid solution for a given fission density. The time t_{knee} can be seen in the following figure where the swelling rate increases

as the bubbles merge. It is also clear that the “knee point” occurs at a higher fission density with higher fission rates as well.

Figure 4: Fractional Swelling vs Fission Density for U_3Si_2 for various fission rates compared to experimental values

Figure 5: Fission Rate vs Fission Density at Knee Comparison of Experimental and Analytical Values

$$R_{ds}^{crit} = 2 \left(\frac{3 \cdot f_n \cdot D_0 \cdot \beta_v \cdot \dot{f} \cdot t_{knee}^2}{b_0} \right)^{1/3} \quad Eq. 16$$

$$(\dot{f} t_{knee}) = \sqrt{\frac{b_0}{24 \cdot f_n \cdot D_0 \cdot \beta_v}} \frac{\sqrt{\dot{f}}}{\beta_e} (R_{ds}^{crit})^{3/2} \quad Eq. 17$$

Eq. 16 is used to find R_{ds} at t_{knee} , which is then used to solve for the fission rate and fission density at t_{knee} . Eq. 17 was used to generate Figure 5. From Figure 5 it can be seen that as the fission rate increases the fission density to reach the knee point increases. This is again because more gas atoms per fission are in solid solution due to an increased gas resolution corresponding to the increased fission rate. The gas resolution rate increases with fission rate because the fission products colliding with bubbles can cause them to collapse.

Figure 6: Fraction of Gas in Bubbles vs Fission Density at Knee

$$\chi_{knee} = \frac{4\pi \cdot f_n \cdot D_0 \cdot \beta_e^2 \cdot t_{knee}}{b_0 \cdot \beta}$$

Eq. 18

Figure 6 is generated using Eq. 18. At higher fission rates, the chi (fraction of gas in bubbles) is lower at the knee. This is because at these higher fission rates the fission gas resolution rate increases such that a lower fraction of gas is in bubbles. Yet the total amount of gas in bubbles still increases, albeit at a lesser degree than the fission rate increase.

Figure 7: Bubble Density vs Fission Density after t_{knee} for U_3Si_2 at a fission rate of $3 \cdot 10^{20} \text{ m}^{-3} \text{ s}^{-1}$

$$N(t) = N_2 \cdot e^{-\phi(t-t_{knee})}$$

Eq. 19

Eq. 19 describes the large bubble population evolution over time after t_{knee} . Eq. 19 is used to generate Figure 7. As the fission density increases the bubble density decreases. This is because as the number of fission gas atoms increase; the bubbles swell and then coalesce with one another and reduce the total number of bubbles.

3.3. UO_2 Fixed Grain Fission Gas Evolution

Figure 8: Fractional Swelling vs Time for UO_2 using fixed grain swelling model for grain size of $7.5 \cdot 10^{-6} \text{ m}$

Figure 8 shows the fractional swelling versus time for UO_2 with a fixed radius of $7.5 \cdot 10^{-6} \text{ m}$ and is generated with Eq. 12. Over time vacancies diffuse to the grain boundaries and gas atoms collect at the vacancies forming bubbles at the boundaries. These bubbles then interconnect and cover the grain face. Eventually as enough bubbles and grain faces interconnect the gas is allowed to escape to the surface and release. This is the saturation point and where gaseous swelling stops with this model.

Figure 9: rb/rbs vs Time for UO_2 at various temperatures and a $7.5(10^{-6})$ m grain radius

This plot shows the bubble radius versus the saturation bubble radius. The average intergranular bubble radius for the saturation point is “rbs” and “rb” is the average intergranular bubble radius. This ratio is the value “x” in equation Eq. 12. The time to reach the saturation point for a given fission rate is very sensitive to the temperature. When this fraction reaches a value of one, then the saturation point is achieved. The plot indicates that as the temperature increases, the time to reach saturation decreases.

Figure 10: Gas Density (mol m^{-3}) vs Time for UO_2 at various temperatures and a $7.5(10^{-6})$ m grain radius

The intergranular gas density can be seen with equations Eq. 8 & Eq. 9. At higher temperatures the gas density, the gas atoms in intergranular bubbles per unit area of grain boundary, necessary to reach saturation decreases because the gas atoms are more energetic and form larger bubbles.

Figure 11: Fractional Gas Sat. vs Time for UO_2 at various temperatures and a $7.5(10^{-6})$ m grain radius

Figure 11: Fractional Gas Sat. vs Time shows the number density of gas bubbles over the number density of gas bubbles at saturation versus time. This ratio can be calculated from the ratio of the last two equations in Table 5. This again shows how the time to reach saturation is sensitive to temperature.

3.4. UO_2 Grain Growth Fission Gas Evolution

Figure 12: Gas Density vs Time for UO_2 Grain Growth Model at 1600K

The Grain Growth model was run with an initial grain radius of $5 \cdot 10^{-6} \text{ m}$ and a final grain radius of $6.11 \cdot 10^{-6} \text{ m}$ at a temperature of 1600K. Similar to the Fixed Grain model, after the saturation point is reached the fission gas is allowed to escape. As the grain faces sweep across the volume of the fuel, they pick up intragranular gas atoms and release them. This then leads to a decrease in gas density. The average grain size increases with time as the grain faces sweep across the fuel and lead to the number of grains decreasing as seen below.

Figure 13: Grain Size vs Time for UO_2 Grain Growth Model at 1600K

Figure 14: rb/rbs vs Time for UO_2 Grain Growth Model at 1600K

As can be seen in *Figure 14*, compared to the fixed grain model, the saturation point is reached much more quickly due to the grain growth and grain boundaries sweeping across the fuel picking up intragranular gas atoms.

Figure 15: Fractional Swelling vs Time for UO₂ Grain Growth Model at 1600K

Similar to the decreases in gas density after the saturation point is reached and intragranular gas atoms escape, the swelling decreases as more gas atoms are swept over by the grain boundary decreasing the intergranular gas density.

Figure 16: Gas Release vs Time for UO₂ Grain Growth Model at 1600K

The saturation point can be seen where the initial release of has occurs. As the grains grow after the saturation point, more gas atoms are released.

4. Discussion & Conclusion

It is clear, that after the knee point, the swelling accelerates for the U₃Si₂ fuel. This should not be a problem for the I²S-LWR as the fuel should be achieving 37-46 GWd/tU [1] through its life in the reactor and the knee point should not be reached by this point. In Figure 4 the knee points occur at $2.55 \cdot 10^{27}$ fissions/m³ and $2.05 \cdot 10^{27}$ fissions/m³ for $2.6 \cdot 10^{20}$ fissions/(s m³) and $1.7 \cdot 10^{20}$ fissions/(s m³) respectively. These correspond to 85 GWd/tU and 69 GWd/tU respectively with an assumed 95% theoretical density or 10.735 gU/cm³. Therefore, at these burnups for I²S-LWR, there should be minimal risk of the knee point being reached. Additionally, due to the higher temperature of the reactor at approximately 350°C, the fuel should not be in an amorphous state[4] which is discuss further below.

During a simulation with BISON at a lower fission rate, Metzger calculated the swelling versus burnup for U₃Si₂ [20]. This was done with an empirical expression. This equation was developed from data generated by Finlay [3] from irradiated aluminum dispersed mini-plate fuel. It should also be noted that the data was for burnups above $2 \cdot 10^{27}$ fissions/m³, of the same order as this work. The equation is shown below. The BISON code expression results in 0.019 V/Vo swelling at 20 GWD/MTU for U₃Si₂ and 0.0125 V/Vo for UO₂. This corresponds to about $6.3 \cdot 10^{26}$ fissions/m³ and $5.37 \cdot 10^{26}$ for U₃Si₂ and UO₂ respectively. For the U₃Si₂, the model here results in 0.0123 V/Vo (it should be noted the swelling rates for different fission rates don't differ greatly until after the knee). The UO₂ fixed grain model results in 0.0117 V/Vo at 1500K (~875 hours).

$$\frac{V}{V_0} \% = (3.88008 \cdot Bu^2 + 0.79811 \cdot Bu) \cdot 100 \quad \text{Eq. 20}$$

Note that in Eq. 20 burnup is in units MWd/MtU. The table below shows the swelling at potential I²S-LWR burnups with both the empirical equation and the model from this paper.

Table 6: Empirical versus Model U₃Si₂ Swelling

	37 GWd/tU	46 GWd/tU
Model	2.282%	2.970%
Empirical Relation (Eq. 20)	3.484%	4.492%

The results from the fixed grain UO₂ model and U₃Si₂ model are used to compare the swelling of each fuel in Figure 17. The swelling rate for U₃Si₂ is at least comparable to UO₂. Although U₃Si₂ becomes amorphous under irradiation, it is only amorphous up to 250°C therefore the comparison had to be done at different temperatures [4], While amorphous the uniform distribution of bubbles helps to keep fission gases within the fuel for U₃Si₂, thereby preventing a pressure increase in the cladding plenum.

The model used for U₃Si₂ here calculates the swelling, but is not a fuel performance code. One way to pursue this would be to use this model in conjunction with a fuel performance code such as BISON which previously used Eq. 20 by Metzger[20], but in this case replace the empirical calculation with this model to calculate the fuel swelling. Another possibility is to use this model in conjunction with the grain subdivision model of high burnup fuel that has been developed by Marquez [21].

While the current U₃Si₂ model does give us some information on the swelling behavior of U₃Si₂, for a more ideal comparison a model to evaluate U₃Si₂ swelling at higher temperatures while crystalline will be pursued. This will allow us to examine the swelling behavior of the fuel more accurately when it is under I²S-LWR conditions at higher temperatures most of the time. Although the fuel in the I²S-LWR will still need to transition through the amorphous regime whenever taken online/offline or whenever the fuel temperature drops below 250°C. There also remains the possibility that since Birtcher et al. used 1.5 MeV Kr ions [4], there still may be amorphous regions within the fuel while under irradiation in a reactor. One possibility being pursued is adapting the UO₂ models in this paper to be used for U₃Si₂ in a crystalline state. Inputs for the model specific to U₃Si₂ versus UO₂ will first need to be obtain or approximated. Developing a model to compare fission bubble formation and swelling for crystalline vs amorphous states for a given fuel would provide more insight into the differences for each.

Figure 17: Massih Fixed Grain (1300K) vs JRest (523K)
 UO_2 vs U_3Si_2 at $1.7E20$ Fission Rate
 Swelling vs Time

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