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► **To cite this version:**

L. Verma, L. Noirot, Ph. Maugis. A new spatially resolved model for defects and fission gas bubbles interaction at the mesoscale.. E-MRS Spring meeting Strasbourg 2018, Jun 2018, Strasbourg, France. hal-02338994

HAL Id: hal-02338994

<https://hal.science/hal-02338994>

Submitted on 13 Dec 2019

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A new spatially resolved model for defects and fission gas bubbles interaction at the mesoscale

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Abstract

Fission gases, especially Xenon, produced during irradiation in a nuclear fuel, have very low solubility in the UO_2 fuel matrix and precipitate into bubbles. These gas bubbles interact with point defects of the fuel (vacancies, self-interstitials, etc.) causing significant microstructural evolution which may eventually affect the overall performance of the fuel. Spatially resolved models are developed to predict and model the microstructural change at the mesoscale. We present a new model, which focuses on modeling the interaction between point defects and xenon gas bubbles. This new model overcomes the limitation of the existing cluster dynamics models as it can account for spatialization as well as the limitation of the spatially resolved phase-field models as it can also account for very small defect clusters, even below the individual grid spacing. The modeling of the phenomena of coalescence of two bubbles in a vacancy supersaturation and the vanishing of a small bubble in the presence of a larger bubble (Ostwald ripening) prove the credibility of the new model. 2-D analysis of a case depicting the movement and growth of bubbles in a vacancy concentration gradient is presented and is in good agreement with the associated physics.

Keywords: gas bubbles, mesoscale, point defects, nuclear fuel

1. Introduction

When fission takes place in a nuclear reactor, the nuclear reactions lead to the generation of fission products, including inert gas atoms and certain

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disorder in the nuclear fuel in the form of defects such as vacancies, auto-interstitials and gas in substitution. The fission gases, Xe and Kr, which have a fission yield of about 0.3, have very low solubility in the fuel matrix and often precipitate into bubbles. The point defects interact with the Xenon bubbles and other microstructural features like as-fabricated pores and grain boundaries in the fuel. Such interactions can lead to macroscopic effects like swelling and fission gas release which have a direct impact on the thermo-mechanics of the fuel rod and must be considered in any fuel performance code [1].

The fission gas bubbles can be either small nanobubbles within the grain (intra-granular) or larger bubbles on grain boundaries (inter-granular). The mechanism of fission gas release out of the fuel can be understood as a two step process: the transfer of gas atoms from the grain to the grain boundaries, and then the coalescence and interlinkage of grain-boundary bubbles to give pathways to outside the fuel. While the latter is clearly observed at high temperatures ($T \geq 1400^{\circ}C$), it has recently been questioned for base irradiation [2]. The former step of gas atoms transfer within the grain has been a topic of debate for the past few decades [3, 4]. If the gas bubbles are considered as perfect traps for gas atoms and according to the effective diffusion theory, the gas should be trapped in the large number of intra-granular bubbles during annealing at high temperatures and should not escape the grain within the duration of annealing. However, it has been long observed that, in practice, the fission gas release from the grain to the grain boundaries may reach values higher than $\approx 60\%$ [5]. Several mechanisms have been proposed for the explanation of such behaviour in the past. For example, MacEwan et al. [6] suggested that the thermal resolution of gas atoms from bubbles, providing a source term for their migration to grain boundaries, could be a possible mechanism. This approach assumes that the inert gases are not essentially insoluble in UO_2 . Through an experiment using ion implantation of krypton into UO_2 , Evans et al.[7] concluded to the non realism of the mechanism of thermal resolution of gas atoms. However, they searched for the evidence of thermal resolution through the shrinking of the bubbles, whilst, in the case of thermal resolution, there would initially be Ostwald Ripening, and hence, possibly, growing of the bigger bubbles at the expense of the smaller ones. Only then, as the free surface is farther, the shrinking of these bubbles would follow at the profit of the free surface. So, from our point of view, thermal resolution cannot be completely excluded from the possibilities to be explored. Evans [8] further proposed that the mechanism could be explained

by the directed motion of bubbles up the concentration gradient caused by the arrival of thermal vacancies from grain boundaries. He also carried out a quantitative assessment [9] of the mechanism by using numerical calculations to simulate the phenomenon. The model used by Evans, however, is not completely mechanistic because he postulates that the bubbles situated at the front (defined by $C_v \approx 0$), undergo a local fractional swelling, ΔS , (a key parameter of his model) before the front progresses inwards the grain. This maintains the vacancy gradient long enough to allow bubbles that are in the gradient to reach the grain boundary. However, this has never been demonstrated. Another scenario could be that the vacancies diffuse inwards the grain and that the vacancy gradient becomes nil before most of the bubbles can reach the grain boundary. So, there arises a need to develop more mechanistic new models that would explicitly describe the bubble and their interaction with vacancies.

For a few years, spatially resolved models have been developed in the mesoscale to understand the microstructure in greater detail. The advantage of using the mesoscale is that material characteristics at the microstructure-level such as grain boundaries or other defects, can be represented in local detail and their interactions with atomic processes can be considered explicitly. Models based on Cluster Dynamics [10, 11] have been used to study the interactions of defects and gas bubbles. Although very efficient, these models lack space correlation of the microstructural elements. Even the spatially resolved models, such as the Phase Field Models [12, 13] have some limitations as they cannot model very small bubbles, less than the individual grid size. Moreover, a very high resolution meshing is required to simulate the continuous field evolution across the diffuse interface, which can be computationally demanding.

In this paper, we present a new spatially resolved model for the interaction between point defects and fission gas bubbles. The model intends to numerically simulate, in greater detail, the mechanism of directed gas bubble migration up a vacancy gradient, as proposed by Evans [8]. The phenomena of coalescence and vanishing of bubbles is simulated and a 2-D test for the movement and growth of bubbles is described in this paper. The successful implementation of these phenomena in the model provide us confidence to further analyze the mechanism of gas migration in a vacancy concentration gradient. The methodology adopted in the model is described in section 2. The general results for the phenomena simulated in the model are described in section 3 and some discussion and conclusions are presented in Section 4.

2. Model description

Assumptions:

- Only vacancies are considered as point defects in the model.
- Bubbles are assumed to be spherical and remain so. This assumption is justified for the intra-granular bubbles.
- No irradiation is considered in the model, i.e. it is only applicable for annealing tests.

2.1. Representation

In the model, the perfect infinite solid is modeled by a portion of it as a mono-crystal having cavities (bubbles). The limit conditions are either periodic or symmetric along the x-axis and periodic in the other direction(s). At present, no grains, grain boundaries or different solid phases are considered in the model. The model is developed to function in both 2-D and 3-D, but for the results presented in this paper, only 2-D analysis has been done for simplicity.

In the representation of the model, the solid portion, which is a rectangle in 2-D or a cuboid in 3-D, is discretized on a regular mesh. The independent variables which are sufficient to represent the model are spins and fields.

(i) Spins

The first variables, spins, are simply integer numbers allocated to the cells (pixel in 2-D/voxel in 3-D) to categorize them in the mesh. For a cell in the solid region, the spin is 2. Spin is 0 for a cell in a cavity and 1 for the cells in the interface between solid and cavity (Fig.1). A rule is that a 1-cell will always be between a 2-cell and a 0-cell i.e. a solid and cavity cannot be immediate neighbours and will always have an interface between them. The 1-cells are imagined to be partially solid and partially void. It is also imagined that a defect arriving towards a 1-cell will immediately reach the solid/void surface.

(ii) Concentration fields

The other variables, the fields, represent the concentrations and are calculated by a system of Ordinary Differential Equations (ODEs). The fields for a 2-cell are the concentration of defects, which here, are the concentration of vacancies C_v . For a 1-cell the fields are the concentration of crystalline atoms, C_a . All the concentrations are expressed

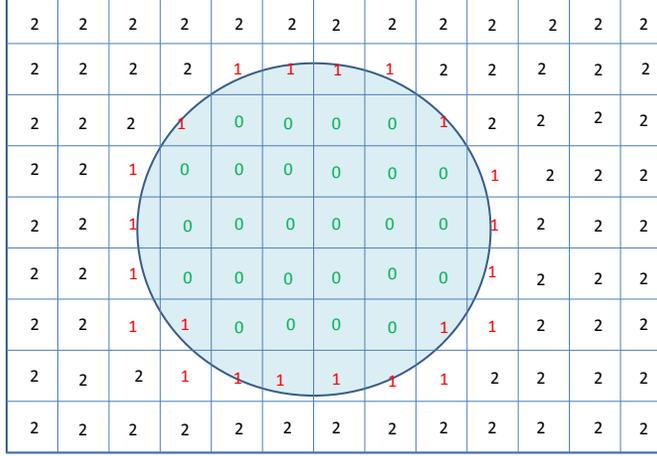


Figure 1: Representation of a regular mesh in 2-D with fuel matrix and cavity with Spins designated to each cell

in fraction of sites. C_a in particular, is the number of U atoms in the cell divided by the number of U-sites in the cell. So, C_a is assimilated to the solid fraction (r_s) in the interface cell. No fields are required for the 0-cells. The equilibrium concentration of vacancies at the considered temperature in the proximity of a surface with curvature κ and exposed to a pressure P_b are denoted as $C_v^{eq}(\kappa, P_b)$. The method to calculate the equilibrium concentrations of vacancies in the vicinity of over-pressurized bubbles has been presented by Noirot [14] and the expression for this is:

$$C_v^{eq} = \exp \left[-\frac{(\epsilon_v - s_v T)}{kT} - \frac{\Omega}{kT} (P_b - \gamma_b \kappa) \right] \quad (1)$$

where, ϵ_v is the formation energy of vacancies (more precisely, Schottky defects, with $\epsilon_v = 2.47$ eV), s_v is the excess entropy of vacancy formation, T is the annealing temperature in K, Ω is the volume of one UO_2 -site and it is equal to $40.9 \times 10^{-30} m^3$, k is Boltzmann Constant and γ_b is the surface tension at the solid-bubble interface. The excess entropy of vacancy formation, s_v , is equal to

$$s_v = k \ln \left[\left(\frac{\nu}{\nu'} \right)^\alpha \right] \quad (2)$$

where, ν is the perfect crystal vibration frequency, ν' is the vibration frequency of atoms influenced by the vacancies and α is the number of vibrational modes that are altered by the introduction of a single vacancy [15, p.59]. In this article, we have taken $s_v = 0$, but as the value of s_v is not known precisely, it can be considered as a parameter in further studies.

The 1-cells have a two-fold usage in the model. Firstly, they contain the C_a variable, which integrate every arrival or removal of U atoms with time. Secondly, the 1-cells are also used to provide a limit condition for the diffusion of vacancies in the 2-cells by imposing that, within the 1-cells, the vacancy concentration C_v is equal to the equilibrium concentration $C_v^{eq}(\kappa, P_b)$.

(iii) Bubbles

The bubbles in the model are supposed to be and remain spherical. The bubbles are entities that store specific information about each bubble in the model. Among these are the position of their center, the total volume of the 0-cells included in the bubble, the total volume of the bubble, the gas content and the list of 1-cells forming the interface. In this study, we can consider the presence of gas in the bubbles, but we do not consider gas in the solid.

2.2. Non-dimensionalization

The physical quantities used in the expressions are non-dimensionalized as follows:

- Energies by $E_{ad} = 1.6 * 10^{-19} J$, which corresponds to 1eV
- Distances by $l_{ad} = 10^{-9} m$, which corresponds to 1nm
- Time by $t_{ad} = \frac{l_{ad}^2}{D_v}$, where D_v is the vacancy diffusion coefficient

As a consequence of non-dimensionalization, the non-dimensional value of D_v is equal to 1. Concentrations of vacancies (C_v) and crystalline atoms (C_a) are already non-dimensionalized as they are expressed in fraction of sites.

The non-dimensional values are represented with a tilde in the expressions used in the paper.

2.3. General algorithm of the model

The model simulates microstructure evolution with time by taking into account the various aspects of interaction between defects and bubbles such as diffusion of defects and the update of bubble characteristics and shape. A general algorithm adopted in the model for a time step is depicted in Figure 2 and the detailed description of these aspects are presented in the following sections.

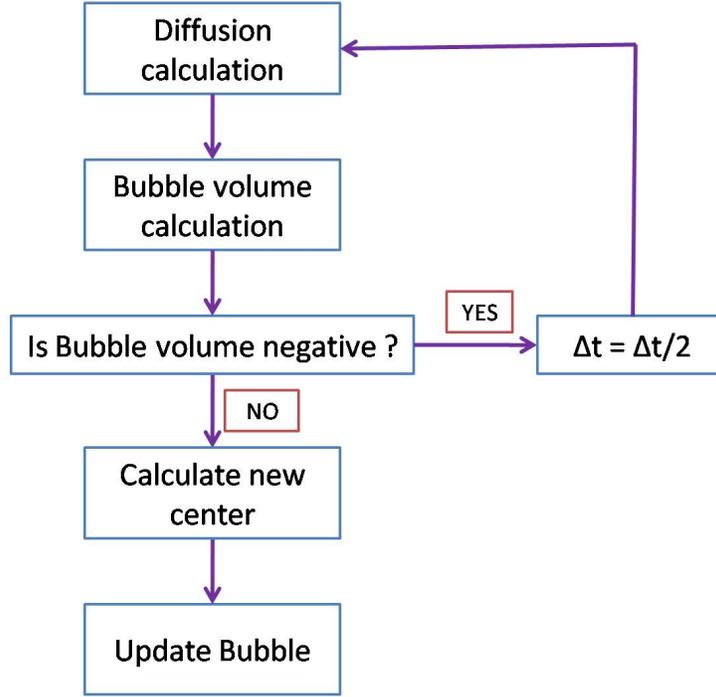


Figure 2: A general algorithm of the model

2.3.1. Diffusion of point defects and crystal atoms

(i) For the 2-cells

The diffusion of defects is governed by the Fick's law. A defect balance equation is written for each 2-cell interacting with its neighbours. So the non-dimensionalized defect balance equation is:

$$\frac{\partial C_v}{\partial \tilde{t}} = -\tilde{\nabla} \cdot \tilde{\phi}_v \quad (3)$$

$$\text{where,} \quad \tilde{\phi}_v = -\tilde{D}_v \tilde{\nabla}(C_v) \quad (4)$$

here $\tilde{\phi}_v$ represents the flux of vacancies between 2-cells.

(ii) For the 1-cells

For the 1-cells, the atom balance equation is written as:

$$\frac{\partial C_a}{\partial \tilde{t}} = -\tilde{\nabla} \cdot \tilde{\phi}_a \quad (5)$$

The flux of atoms, $\tilde{\phi}_a$, however, can occur through different phenomena. Exchanging with a 2-cell, the flux of atoms is a consequence of the flux of defects (here, only vacancies) and can be determined as:

$$\tilde{\phi}_a = -\tilde{\phi}_v \quad (6)$$

The flux of atoms could also be along the interface (i.e. exchanging with another 1-cell) due to surface diffusion. In the model, the surface diffusion is not explicitly taken into account as a flux of atoms between two 1-cells. It has, however, been incorporated in the hypothesis about the shape of the bubbles being and remaining as spheres.

These equations are then discretized in space and time. There are several spatial schemes or stencils available for space discretization depending on the number or type of neighbours being used. For the present model, only the face neighbors have been taken into consideration using a 5-point difference scheme in 2-D and 7-point difference scheme in 3-D.

The procedure used for calculating the left hand side of equation (3) or (5) is to calculate the flux of vacancies from the neighbour cells (n) to the present cell (p) and then to calculate the negative divergent of the flux. So, for the 2-cells, this procedure can be represented by the equation:

$$\frac{\partial C_v(p)}{\partial \tilde{t}} = \sum_{n \in \text{face neighbours}} \frac{\tilde{D}_v}{\tilde{h}^2} [C_v(n) - C_v(p)] \quad (7)$$

here, h is the grid size.

For a 1-cell, the procedure for the flux due to the flux of vacancies from a neighbour 2-cell, can be represented as:

$$\frac{\partial C_a(p)}{\partial \tilde{t}} = - \sum_{n \in \text{2-cell face neighbours}} \frac{\tilde{D}_v}{\tilde{h}^2} [C_v(n) - C_v^{eq}(\kappa(p), P_b(p))] \quad (8)$$

where, $\kappa(p)$ and $P_b(p)$ are the curvature and pressure of the bubble whose cell (p) is an interface cell. Once the set of space-discretized equations is written,

a set of Ordinary Differential Equations (ODE) is obtained. Explicit scheme of time discretization is used. For a solid cell p , the expression for time discretization is:

$$C_p^{t+1} = C_p^t + \sum_{n \in \text{face neighbours}} \frac{\Delta \tilde{t} \tilde{D}_v}{\tilde{h}^2} [C_n^t - C_p^t] \quad (9)$$

For an interface cell, the expression is analogous. C_v^{eq} is considered constant with time during the time step Δt in Figure 2.

2.3.2. Bubble volume update

For each bubble in the simulation, the total volume of the bubble needs to be updated after the diffusion process. Then knowing the volume of each bubble, the temperature and the equation of state of the gas, the pressure, P_b as well as the new value of C_v^{eq} is determined. At the beginning of the time step, for each bubble, the total 0-cell volume is kept into a variable $V_b^{(0-cells)}$. At the end of a time step, the total volume of the bubble, V_b , is updated by :

$$V_b = V_b^{(0-cells)} + \sum_{\text{interface cells}} (1 - C_a) * V_{cell} + \Delta a \Omega + \Delta V_{comp} \quad (10)$$

here, C_a has been calculated in the diffusion phase, V_{cell} is the volume of one cell and Ω is the volume of one site. The last two terms are for the error in number of atoms Δa and complimentary error term for balancing the volume equation ΔV_{comp} , respectively. Δa is updated each time an interface cell of the bubble is updated and its value is updated as:

$$\Delta a = \Delta a + (C_a^{new} - C_a^{old}) \frac{V_{cell}}{\Omega} \quad (11)$$

ΔV_{comp} is required to balance the volume equation when there is a transformation of a 2-cell into a 1-cell and vice versa. When a 2-cell is transformed into a 1-cell, an additional term is added to the summation over the interface cells on the right hand side of equation 10. To restore the balance of the equation, this additional term has to be removed and it is taken care by ΔV_{comp} with the expression:

$$\Delta V_{comp} = \Delta V_{comp} - (1 - C_a) * V_{cell} \quad (12)$$

Similarly, for a 1-cell to 2-cell transformation, there is a deficit of one term (from the transformed 1-cell) and thus, this term is added to ΔV_{comp} to maintain the balance. Moreover, the movement of bubble is also considered with the calculation of its new center (called Target center).

2.3.3. Re-drawing the bubble

Once the new characteristics of the bubble are updated, the list of interface cells of each bubble also has to be updated. This means that the spins of some cells may be changed and the $r_s = C_a$ (solid ratio in the interface cells) are recalculated as if the bubble was re-drawn. Δa and ΔV_{comp} have been introduced in equation 10 to respect the balance of crystal atoms without demanding a very high precision when new r_s values are calculated in the re-drawing process. If the target center of the bubble is too far from its previous center, this process of re-drawing may be done in several steps, moving the bubble center by shorter distances. Coalescence with other bubbles is also checked during the process. Once the re-drawing process is done for each bubble, a new diffusion time step may begin.

3. Results and Discussions

In order to test the capability of the model to simulate the mechanisms related to fission gas bubble behaviour, some general tests are carried out in 2-D. The simulation domain is a 64x64 cells box. The grid size is taken as 4, so the physical domain is a 256 nm x 256 nm size box. The same domain has been adopted for all the cases discussed hereafter. The bubble(s) are included in the domain box with size and number of bubbles depending on the requirements of the test case. The isothermal annealing temperature is taken as 1800°C. The following tests are carried out using the model.

3.1. Coalescence of two bubbles

Due to bubble growth and migration, coalescence between two or more bubbles can occur in the grain. Coalescence leads to the overall coarsening of the bubbles but also decreases the number density of bubbles within the grain. To simulate the the phenomenon of coalescence of two bubbles, the bubbles are modeled with the properties mentioned in Table 1.

The initial concentration of vacancies in the solid region is provided as high as $C_v^{ini} = 1.e-2$ so that coalescence can occur quickly as the two bubbles will trap vacancies and grow. The criteria for coalescence of two bubbles is

Bubble ID	Radius (nm)	Pressure (Pa)
1	12.1	28.53
2	5.9	2.85e+7

Table 1: Properties of bubbles for coalescence test

that a 1-cell of a bubble should be an immediate neighbour of a 1-cell of the other bubble. When this condition is satisfied, the two bubbles coalesce into one bigger bubble. During coalescence, the volume and content of the resulting bubble is the sum of volume and content of the two bubbles. The center of the new bubble is the barycenter (weighted by the volume) of the centers of the two coalescing bubbles. The simulation is carried out for a time of $2.e+7$ which is equivalent to about 12 s of experiment time. The situation before and after the diffusion is seen using Paraview [16] and is represented in terms of r_s , the solid fraction of a cell (Fig.3). It is very

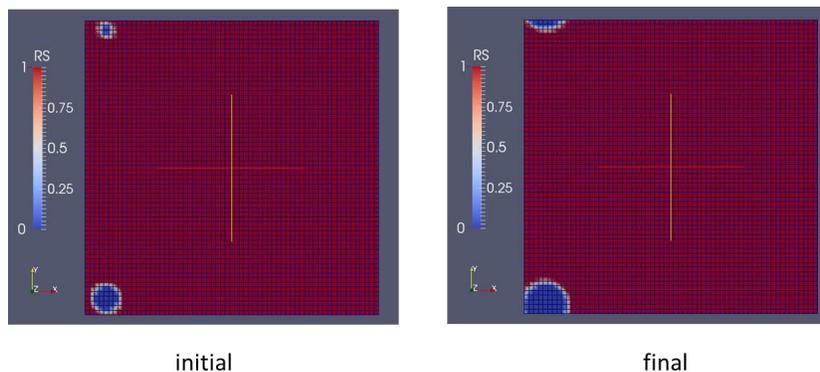


Figure 3: Coalescence of 2 bubbles, at time $t=0$ and after diffusion at $t=12s$ of experiment time

well evident from Fig.3 that coalescence occurred between the two bubbles forming a single bubble of radius 19.7 nm. The pressure within the new bubble is $2.50e+6$ Pa. The positions of the two bubbles were chosen as such to test the periodicity condition at the boundaries, which can be seen to be working perfectly.

3.2. Vanishing of a bubble in the presence of a large bubble

In the case of empty bubbles with different sizes, the smaller bubble in the vicinity of a larger bubble decreases further in size and the larger

bubble continues to grow further at the expense of the small bubble. This phenomenon is known as Ostwald Ripening. The small bubble may even vanish during the process. To simulate the phenomenon of Ostwald Ripening in the model, two bubbles are modeled with different sizes and are provided with a very large volume per atom of gas (V_{at}) as $1.e+6 \text{ nm}^3$. The bubble properties are mentioned in Table 2.

Bubble ID	Radius (nm)	Pressure (Pa)
1	20.0	28.53
2	5.0	28.53

Table 2: Properties of bubbles for vanishing test

The initial concentration of vacancies in the solid region is taken as $C_v^{ini} = 1.04e-6$, which is of the same order of magnitude as the two C_v^{eq} at the interfaces of the two bubbles. The algorithm adopted for vanishing of a bubble is that if the volume of a bubble gets less than a limit volume, the bubble is removed from the lattice and replaced by an equivalent number of vacancies. This limit volume is taken as 3% of the volume of a cell. The idea for this order of magnitude is that, in the model, each site represents a UO_2 atom, irrespective of the orientation of U atom. This site, which is a cuboid, has 26 neighbour sites. So, a vacancy needs 26 sites around it to be a vacancy and thus, the concentration of vacancies cannot exceed $\frac{1}{27} \approx 3.7\%$. The simulation is carried out for a time of $9.7e+8$ which is equivalent to 10 minutes of experiment time. As expected, the small bubble vanishes and the big bubble grows to have a new radius of 20.6 nm. The evolution of the volumes of the big bubble and the small bubble with time are depicted in Figure 4. It can

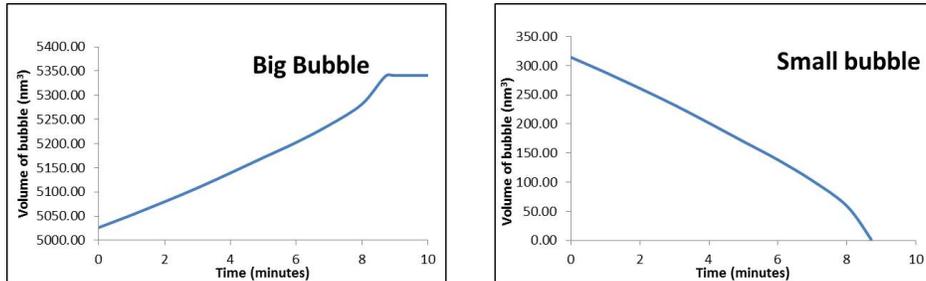


Figure 4: Evolution of volume of the small and big bubble with the experiment time

be observed through the figure that the volume of the small bubble decreases

up to the limit volume and then the bubble vanishes. The big bubble grows during this time at the expense of the small bubble. The saturation in the volume of the big bubble near the end indicates the vanishing of the small bubble at that time.

3.3. Movement and growth of bubbles

A simple 2-D case is carried out to test the ability of the model to simulate the movement and growth of bubbles in a vacancy concentration gradient. One empty bubble with a radius of 30 nm is modeled at the center of the domain. The volume per atom of gas in the bubble is provided as $1.e6 \text{ nm}^3$. Randomly generated bubbles with a random size distribution are provided with volume per atom of gas approximately equal to the volume of one U site (0.04 nm^3). These highly pressurized bubbles are randomly distributed in the domain to surround the empty big bubble. According to the mechanism proposed by Evans [8], due to the gradient of vacancy concentration between the small bubbles and the big bubble, there should be a directed motion of the small bubbles up the gradient towards the big bubble. At the same time, the smaller bubbles must also grow by trapping vacancies from the big bubble. The simulation is carried out for an experiment time of 1 hour. The initial vacancy concentration in the solid is taken as $C_v^{ini} = 2.e-6$, which is of the same order of magnitude as the C_v^{eq} at the interfaces of the big and small bubbles. The bubble evolution and migration is depicted in Figure 5 for time $t=0$ to $t=60$ min.

It can be observed in the Figure 5 that at $t=0$ the various bubbles are randomly distributed around the big bubble. As time progresses, the small pressurized bubbles trap vacancies from the bigger bubble and grow. The bigger bubble shrinks at the same time. This bubble evolution continues while the gradient of vacancy concentration lasts between the bubbles. Once the equilibrium values are reached, there is no further movement of the bubbles. It can be seen from Figure 6, that the vacancy concentration values for the big bubble and one of the small bubbles reach an equilibrium state at around 57 minutes of annealing time.

Further, to study the directed motion of bubbles up the vacancy gradient, the distance of a bubble from the center of the initially big bubble is plotted against the experiment time (Figure 7). The radius of the big bubble is also plotted to act as a reference. It is observed from Figure 7 that the bubbles further away from the big bubble move a very slight distance towards the bigger bubble. The bubbles near the big bubble, on the other hand, move

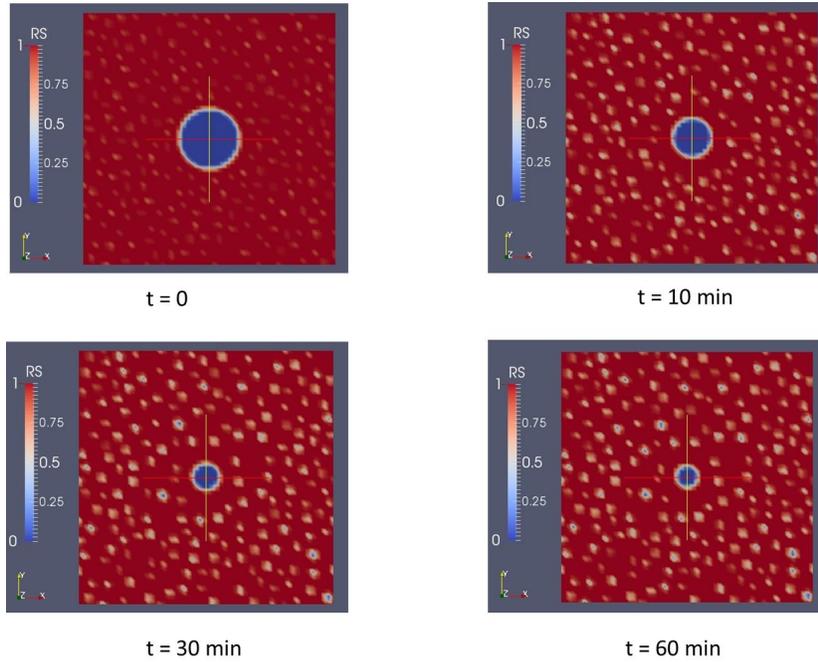


Figure 5: Movement and growth of bubbles in a vacancy gradient

much more distance towards it. For example, Bubble 5 moves a distance of 24.8 nm towards the big bubble. Some bubbles coalesce among themselves while moving towards the big bubble and thus vanish, as Bubbles 3 and 4. The kink in the curve for Bubble 5 at around 21 min is due to the coalescence of Bubble 4 with it, increasing the volume and shifting the center of Bubble 5 away from the center of the big bubble. In conclusion, it can be seen that the bubbles migrate towards the bigger bubble which has a lower value of equilibrium concentration of vacancies, thus verifying the mechanism of directed motion of bubbles in a vacancy gradient.

4. Conclusion

A new spatially resolved model to study the interaction between fission gas bubbles and point defects is presented. Motivated to study fission gas release from the grain to the grain boundary through the mechanism of directed bubble migration in a vacancy gradient, the present paper discusses the capability of this model. The phenomena of coalescence of two bubbles

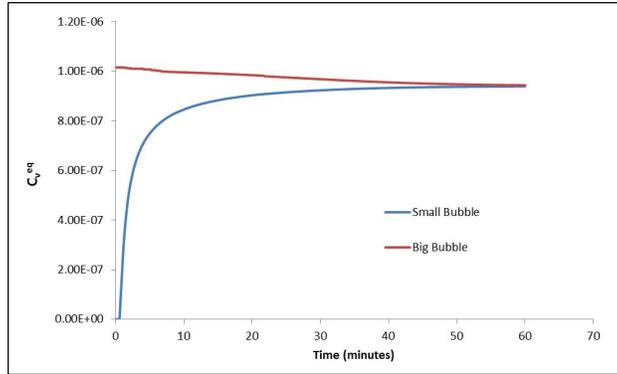


Figure 6: Equilibrium concentration of vacancies at the vicinity of bubbles with time

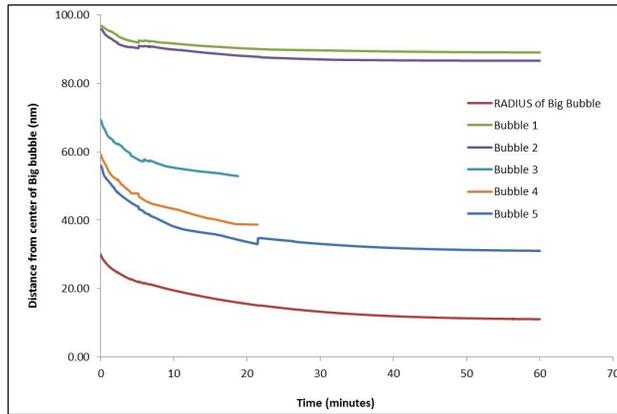


Figure 7: Directed motion of smaller bubbles towards the big bubble

and Ostwald ripening in the grain are simulated using the model. These phenomena are very well simulated by the model and show good agreement with the associated physics. To test the ability of the model to simulate bubble movement and growth in a vacancy concentration gradient, a 2-D case is analyzed. The results show the growth of small pressurized bubbles and their movement towards the large empty bubble while the vacancy gradient lasts. The results obtained prove the ability of the model to simulate the phenomena associated with bubble growth and migration. This gives us the confidence to proceed further with the model to study, in detail, gas bubble migration within the grain and its impact to the overall fission gas release.

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