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Effective parameterization of phase-field models of fission gas bubble growth

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Changing the World's Energy Future

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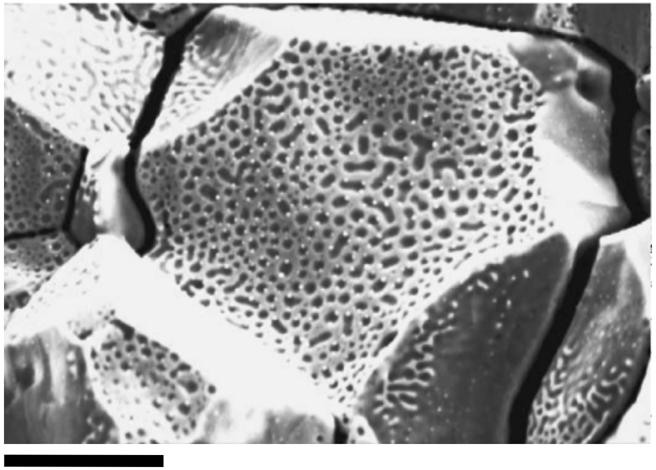
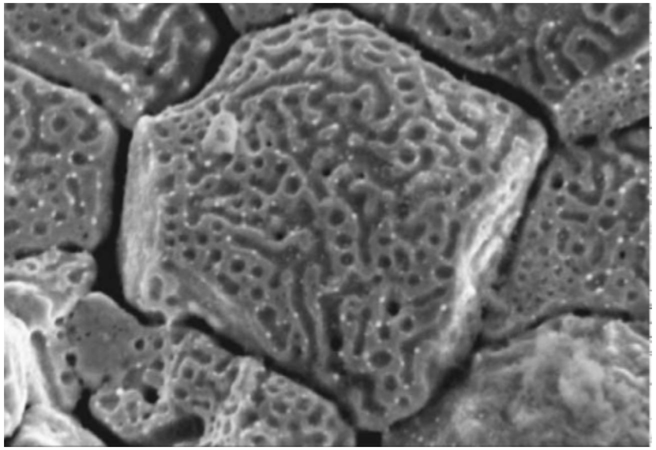
Fission Gas Evolution and Release: Background





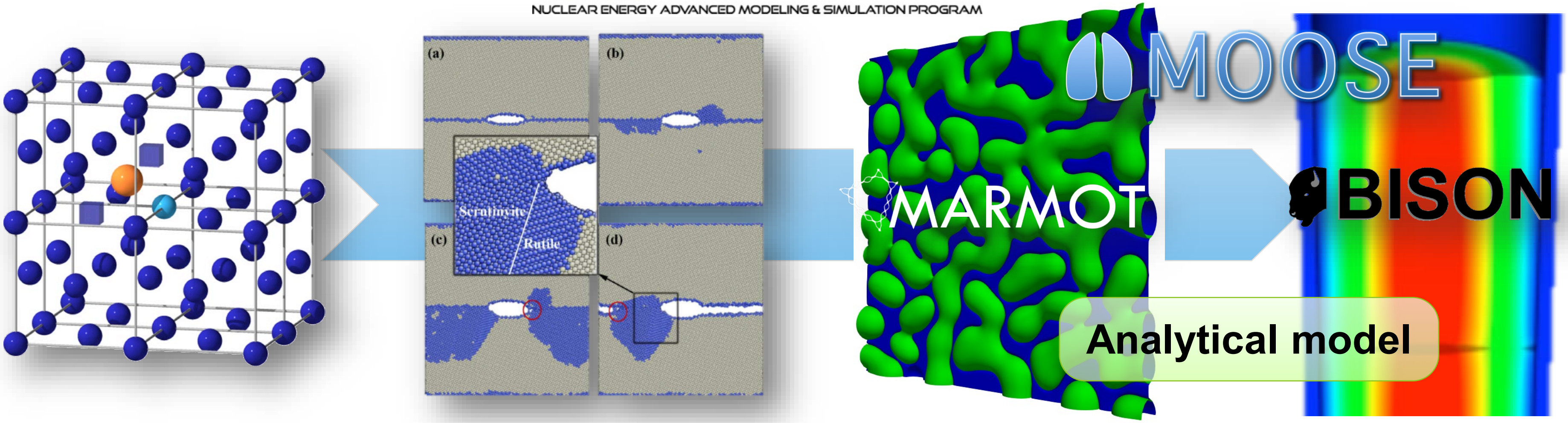
Intergranular Bubbles in UO_2
R.J. White, J. Nuc. Mater., 325, 61-77 (2004)

- Fission products produced in fuel matrix
- Bubbles of low-solubility fission products (Xe, Kr) nucleate
- Intragranular bubbles: 1-5 nm
  - Size limited by re-solution
- Intergranular bubbles: >50 nm
  - Bubbles grow and percolate on grain faces
  - When percolated faces connect to free surface, fission gas is released
  - Microstructure has a strong influence on fission gas release
- Consequences of fission gas release:
  - Reduced thermal conductivity of fuel-cladding gap
  - Increase in plenum pressure: mechanical properties of cladding

Phase-field modeling of fission gas bubble microstructural evolution

- Fundamental scientific understanding
- Inform engineering-scale nuclear fuel performance codes

NEAMS
NUCLEAR ENERGY ADVANCED MODELING & SIMULATION PROGRAM



nanometers
First Principles
- Identify critical bulk mechanisms
- Determine bulk properties

100's of nanometers
Molecular Dynamics
- Identify interfacial mechanisms
- Determine interfacial properties

microns
Mesoscale
- Predict microstructure evolution
- Determine impact on properties

millimeters and up
Engineering Scale
- Use analytical theory
- Predict fuel performance

Phase-field modeling evolution equations: defect species

- Most complete picture:
  - Vacancies
  - $\frac{\partial c_v}{\partial t} = \nabla \cdot M_v \nabla \mu_v + S_v - K_{iv} c_i c_v - K_{vs} c_v$

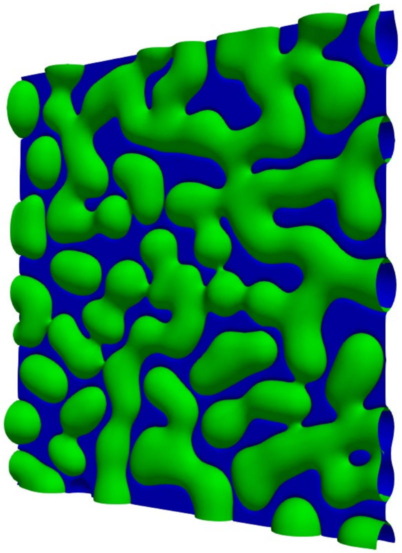
    Diffusional transport | Source | Recomb-ination | Sink
  - Interstitials
  - $\frac{\partial c_i}{\partial t} = \nabla \cdot M_i \nabla \mu_i + S_i - K_{iv} c_i c_v - K_{is} c_i$
  - Gases
  - $\frac{\partial c_g}{\partial t} = \nabla \cdot M_g \nabla \mu_g + S_g$
- Interstitials can significantly increase computational time due to much more rapid diffusion

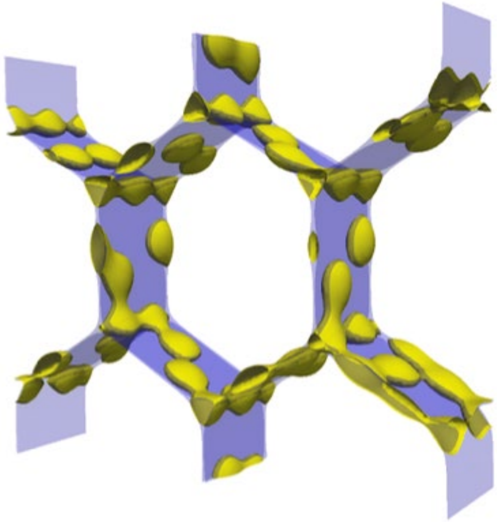
Phase-field modeling evolution equations: effective vacancy production

- Due to preferential absorption of interstitials at dislocations and faster diffusion, normally interstitial concentration is much lower than vacancy concentration and there is a net excess of vacancies
- Several past models have included vacancies with a net vacancy production rate
- Source-Only (SO)
  - $\frac{\partial c_v}{\partial t} = \nabla \cdot M_v \nabla \mu_v + S_v$
  - S_v: an effective vacancy source, but what is the right value?
- Source + Sink (SS)
  - $\frac{\partial c_v}{\partial t} = \nabla \cdot M_v \nabla \mu_v + S_v - K_v c_v$
  - Can use a physical value of S_v and K_v is an effective vacancy sink
  - Maintain steady-state vacancy concentration $c_v^{ss} = S_v / K_v$ in bulk far from bubbles

Overview

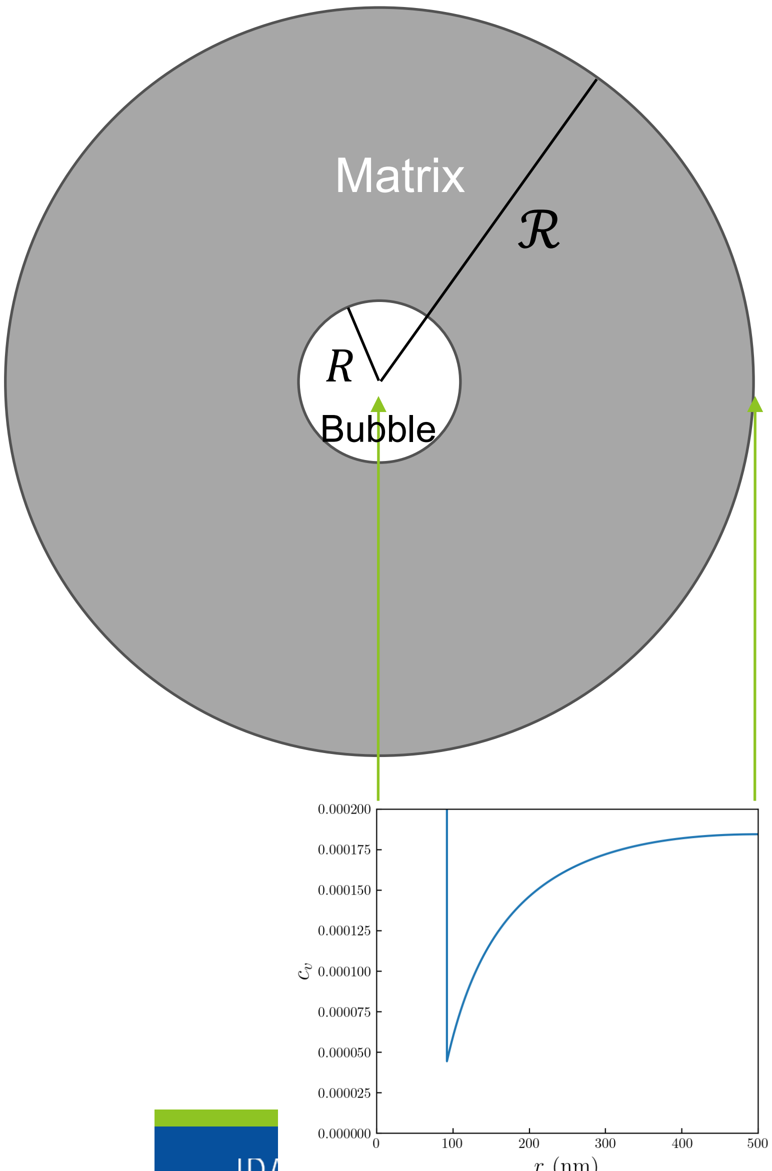
- Goal: compare SO and SS phase-field models to analytical solutions
    - Simplified geometry
    - Determine advantages/disadvantages
- Determine parameters for phase-field modeling by comparing to analytical model that includes vacancies and interstitials in a simplified geometry
    - “Chemical stress” model





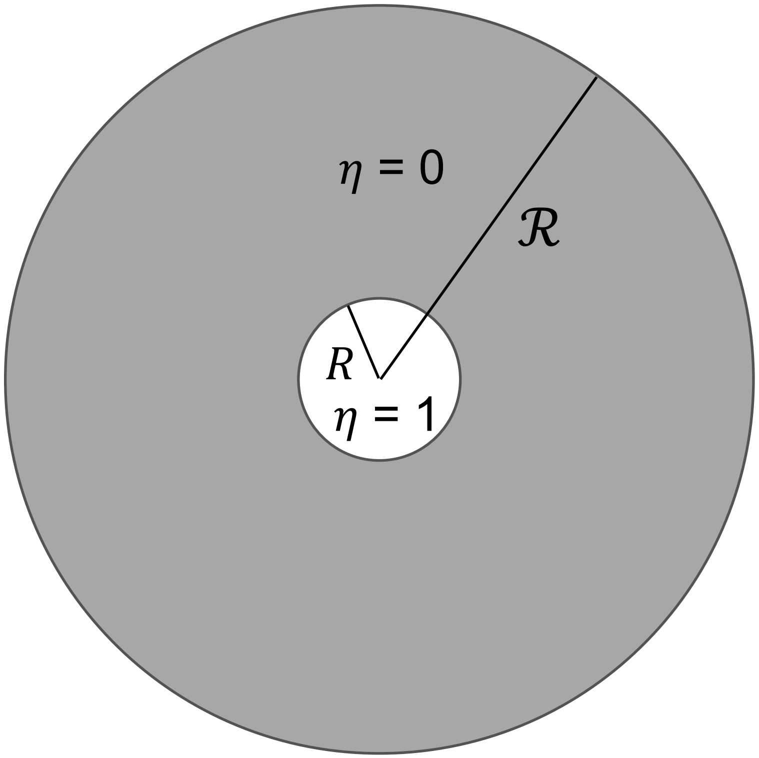
Analytical solution for Source-only (SO) approach

- For simplicity consider vacancies only in spherical domain (void)
- Spherical coordinates:
  - $\frac{\partial c_v}{\partial t} = \frac{D_v}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial c_v}{\partial r}\right) + S_v$
- Known analytical solution for quasi-steady state case:
  - $\frac{\partial c_v}{\partial t} \approx 0 \approx \frac{D_v}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial c_v}{\partial r}\right) + S_v$
  - Boundary conditions: Gibbs-Thomson condition at bubble-matrix interface, 0 gradient at domain boundary
- Analytical solution
  - $c_v(r) \approx c_{vR} + \frac{S_v \mathcal{R}^3}{3D_v R}\left(1 - \frac{R}{r}\right)$
- Take derivative to find flux, growth rate:
  - $\frac{dR}{dt} = \frac{S_v \mathcal{R}^3}{3R^2}$



Phase-field model: Essential features

- Single order parameter η to represent void (bubble) and fuel matrix phase
- Track vacancies
  - SO and SS approaches
- Chemical energy contribution
- Solid-bubble interfacial energy
  - Kim-Kim-Suzuki (KKS) approach to remove bulk energy contribution to interfacial energy



Phase-field model: Free energy functional

- $F = \int_V \left[f_{chem} + W g(\eta) + \frac{\kappa}{2} |\nabla \eta|^2 \right] dV$

- f_{chem} = bulk chemical free energy density.
  - $f_{chem} = [1 - h(\eta)] f_{chem}^{m}(c_v^m) + h(\eta) f_{chem}^{b}(c_v^b)$
- $h(\eta)$ is a smooth interpolation function.
- Chemical free energy of the matrix, bubble phases: Parabolic approximations
  - $f_{chem}^{m} = \frac{k_v}{2} (c_v^m - c_v^{m,min})^2$
  - $f_{chem}^{b} = \frac{k_v}{2} (c_v^b - c_v^{b,min})^2$

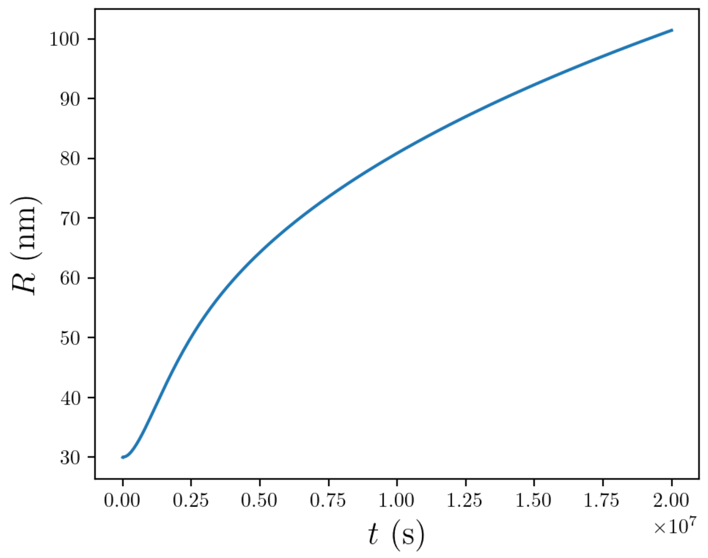
Evolution equations

- Allen-Cahn for order parameter:
  - $\frac{\partial \eta}{\partial t} = L\left[\frac{dh}{d\eta}\left[\left(f_{chem}^{m} - f_{chem}^{b}\right) - \mu_v\left(c_v^m - c_v^b\right)\right] - W\frac{dg}{d\eta} + \kappa \nabla^2 \eta\right]$
- Cahn-Hilliard for vacancies, SO approach:
  - $\frac{\partial c_v}{\partial t} = \nabla \cdot M_v \nabla \mu_v + S_v$
- Cahn-Hilliard for vacancies, SS approach:
  - $\frac{\partial c_v}{\partial t} = \nabla \cdot M_v \nabla \mu_v + S_v - K_v c_v$
- Mobilities are a function of defect diffusivities:

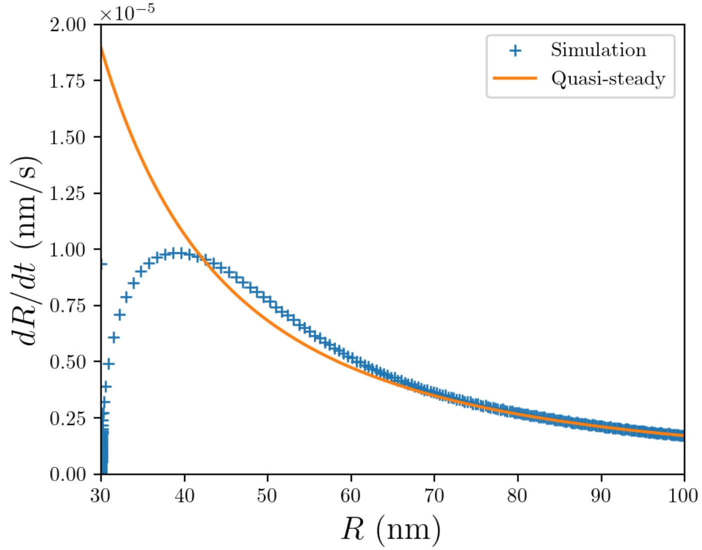
$$M_v = \frac{D_g(\phi)}{f_{c_v c_v}} = \frac{h D_v^b + (1-h) D_v^m}{f_{c_v c_v}}$$

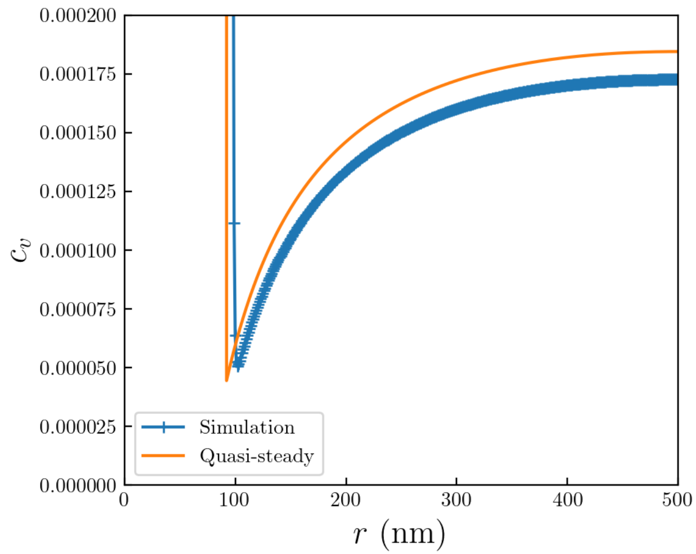
- + KKS system constraints

Source-only model simulations and comparison to quasi-steady state analytical solution

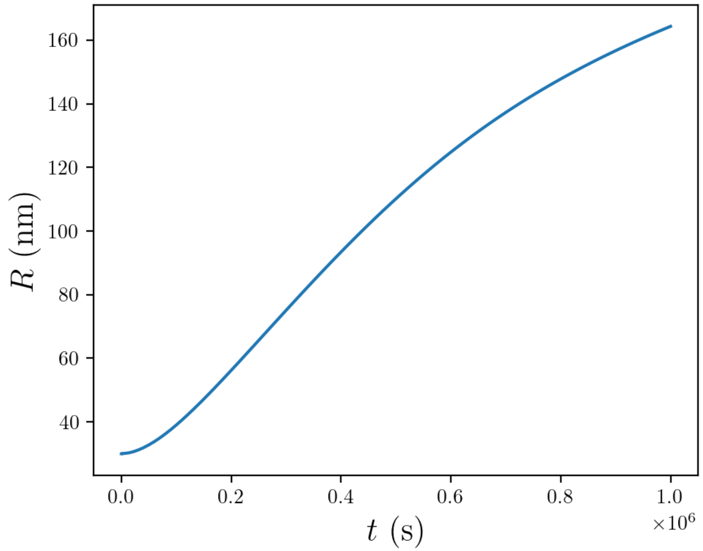


- Vacancy diffusivity based on UO_2 at 700 K
- $S_v = 4.09 \times 10^{-10}$ s^{-1} (5x gas atom production rate)
- Good match between analytical solution and phase-field simulation after c_v reaches quasi-steady state

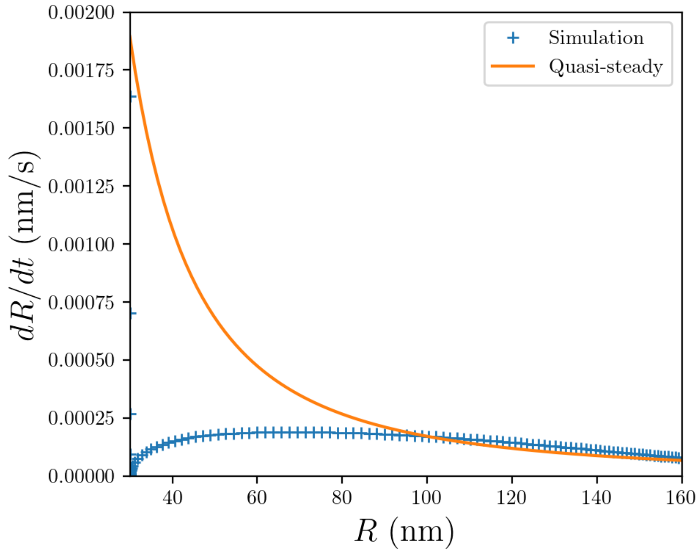


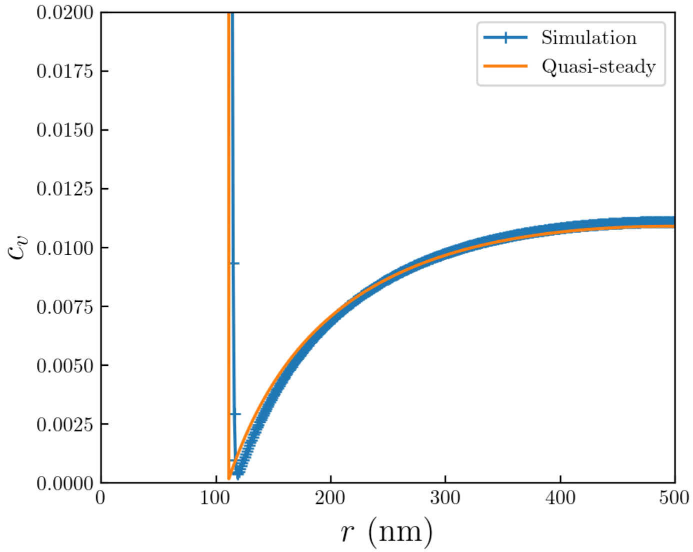


Source-only model simulations: effect of increasing S_v



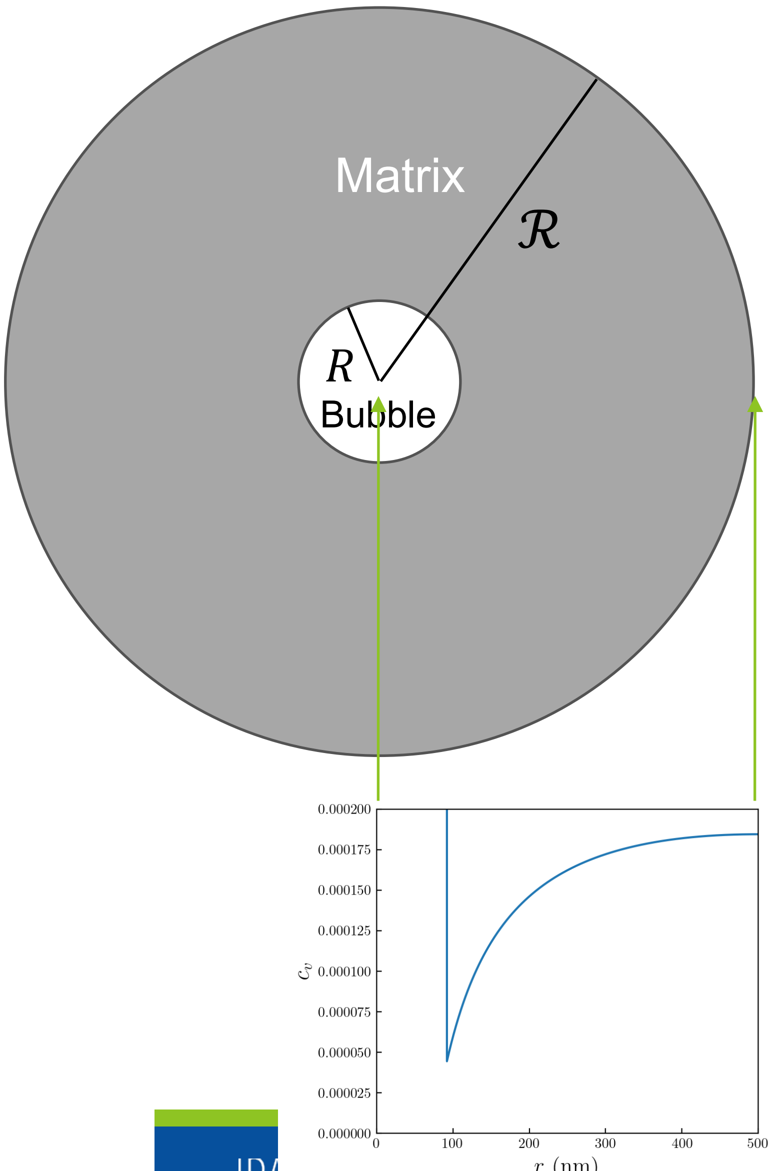
- Increase $S_v = 4.09 \times 10^{-8}$ s^{-1} (500x gas atom production rate)
- Growth rate increases significantly
- Agreement with analytical solution is only reasonable after bubble grows significantly (dR/dt becomes much smaller)





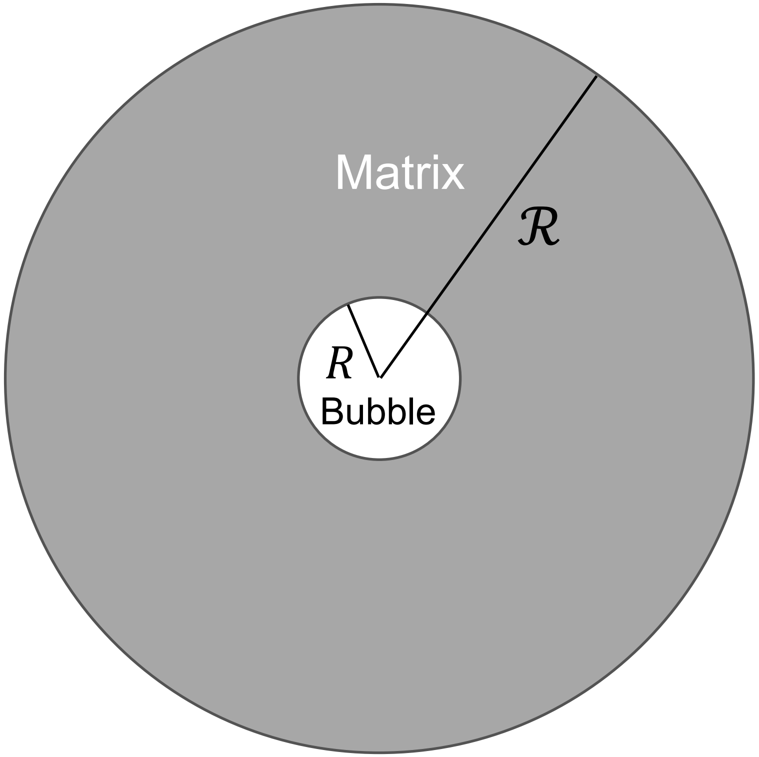
Analytical solution for Source+Sink (SS) approach

- Spherical coordinates, quasi-steady state:
  - $\frac{\partial c_v}{\partial t} \approx 0 \approx \frac{D_v}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial c_v}{\partial r}\right) + S_v - K_v c_v$
- Analytical solution not previously studied. Solve by splitting into homogeneous and particular solutions.
  - Homogeneous part: Modified spherical Bessel equation equation with n=0
  - Solutions: modified spherical Bessel functions of the first and second kinds
  - Boundary conditions: Gibbs-Thomson condition at bubble-matrix interface, 0 gradient at domain boundary
- Analytical solution
  - $c_v = \left(c_{vR} - \frac{S_v}{K_v}\right)\frac{\lambda R}{e^{-\lambda R}}\frac{e^{-\lambda r}}{\lambda r} + \frac{S_v}{K_v}$, where$\lambda = \sqrt{S_v/K_v}$
- Take derivative to find flux, growth rate:
  - $\frac{dR}{dt} = D_v(c_v^{SS} - c_{vR})\left(\sqrt{\frac{K_v}{D_v}} + \frac{1}{R}\right)$



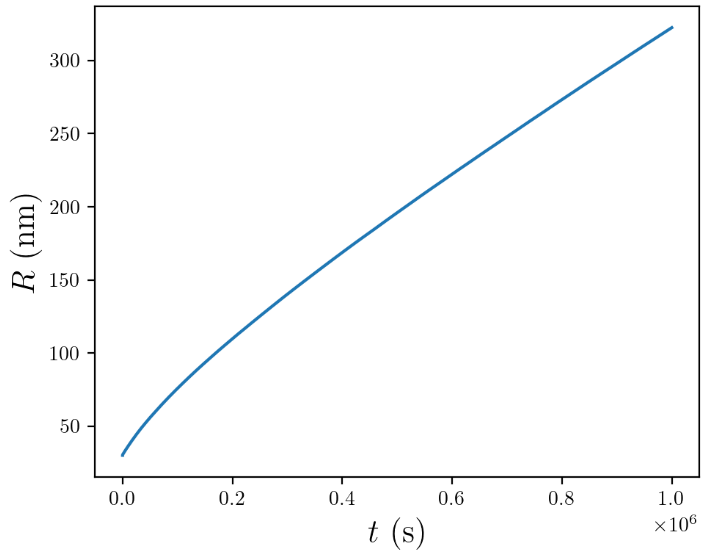
"Chemical Stress" model accounts for bubble growth due to both vacancies and interstitials

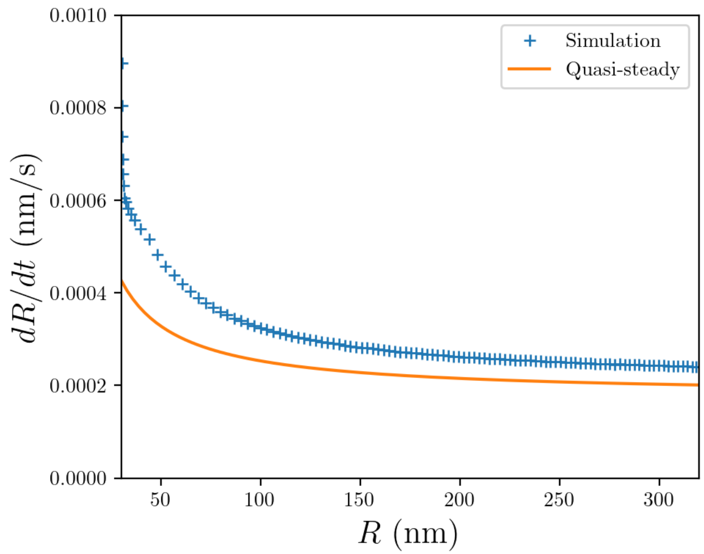
- "Produces bubble growth like a mechanical force"
- Growth due to vacancies and interstitials:
  - $\frac{dR}{dt} = \frac{1}{R}\left[D_v(c_v^{ss} - c_{vR}) + D_i(c_i^{ss} - c_{iR})\right]$
- Far from bubbles,
  - $Z_v D_v(c_v^{ss} - c_{vR}) = Z_i D_i(c_i^{ss} - c_{iR})$
- Combining results in
  - $\frac{dR}{dt} = \frac{D_v}{R}(c_v^{ss} - c_{vR})\left(1 - \frac{Z_v}{Z_i}\right)$
- Compare to SS analytical solution
  - $\frac{dR}{dt} = D_v(c_v^{ss} - c_{vR})\left(\sqrt{\frac{K_v}{D_v}} + \frac{1}{R}\right)$

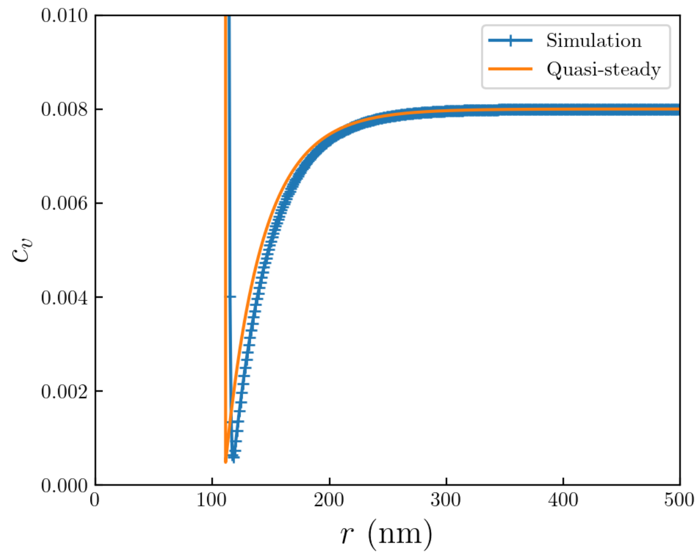


Source+sink model simulations and comparison to quasi-steady state analytical solution

- $S_v = 4.09 \times 10^{-6}$ s^{-1} (10^4 x fission rate, physically reasonable)
- Expect $c_v^{ss} \approx 8 \times 10^{-3}$ based on cluster dynamics calculations
  - Set $K_v = S_v / c_v^{ss} = 1.02 \times 10^{-3}$
- Reasonably agreement between phase-field model and analytical solution





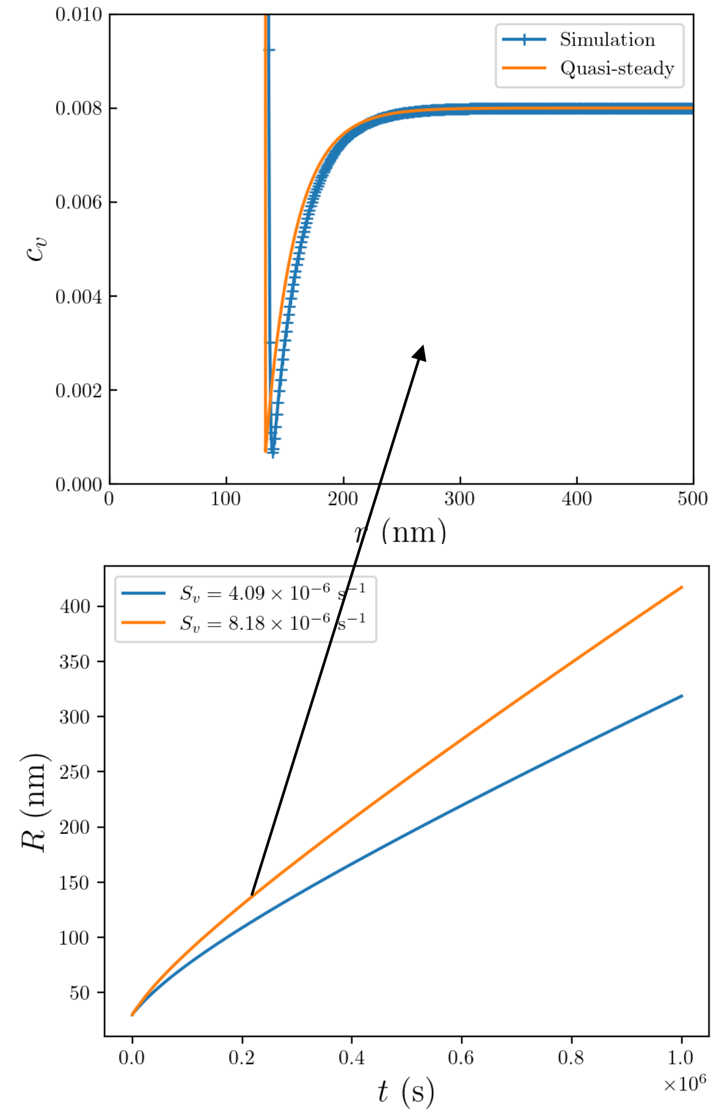


Growth rate in source+sink model has undesirable variation with parameter choices

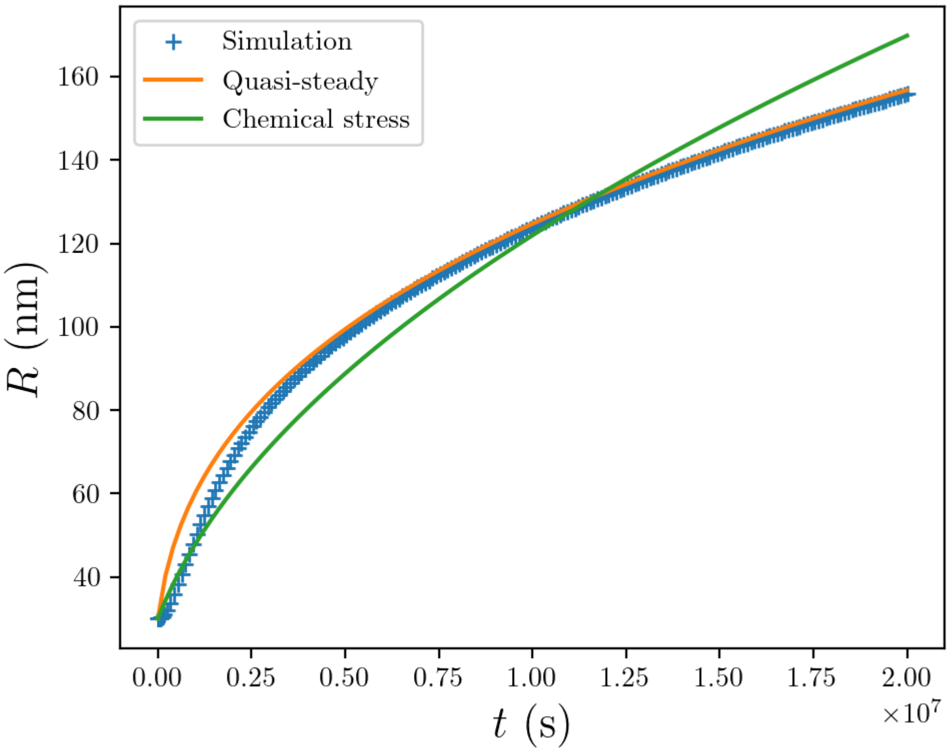
- From intuition and chemical stress model
  - $\frac{dR}{dt} = \frac{D_v}{R}(c_v^{SS} - c_{vR})\left(1 - \frac{Z_v}{Z_i}\right)$
- Expect that in the SS model, as long as c_v^{SS} remains constant, the growth rate should remain the same.
- Test by doubling S_v and K_v
  - Growth rate increases significantly!
- Insight provided by analytical solution to SS growth rate
  - $\frac{dR}{dt} = D_v(c_v^{SS} - c_{vR})\left(\sqrt{\frac{K_v}{D_v}} + \frac{1}{R}\right)$

$\parallel$
S_v/K_v

- **More difficult to parameterize SS model to match vacancy+interstitial model compared to SO, where** $\frac{dR}{dt} = \frac{S_v \mathcal{R}^3}{3R^2}$



Parameterizing Source-Only model to match vacancy+interstitial model



- Source-only analytical solution:
  - $R = [S_v \mathcal{R}^3 t + R_0^3]^{1/3}$
- Chemical stress model:
  - $R = \left[2D_v\left(1 - \frac{Z_v}{Z_i}\right) c_v^{ss} t + R_0^2\right]^{1/2}$
- Fit to determine S_v that gives best match between them
- Phase-field simulation parameterized with this value matches vacancy+interstitial model reasonably well
  - $S_v = 1.53 \times 10^{-9}$ s^{-1}, approximately 16x greater than Xe production rate

Conclusions

- Compared source-only and source+sink phase-field models to analytical solution
  - Developed new analytical solution for the source+sink case
- Analytical models predict phase-field model behavior reasonably well when bubble growth rate is small
  - Provide qualitative and quantitative insight into model behavior
- Source+sink model has significant disadvantages in parameterization
- Developed strategy to determine effective vacancy source term for source-only model
  - For conditions considered, vacancy source term approx. 16x larger than gas source term. Past work has used values in the range 1-20x gas source term