# Crank-Nicholson Adams-Bashforth IMEX Scheme for Radiation Damage in Type 316 Stainless Steel

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# Abstract

The behavior of materials subject to large radiation fluxes are considered. The generation, diffusion, recombination, and sinking of atomic point defects are simulated for Type 316 Stainless Steel. Several numerical approaches were taken in an attempt to simulate this behavior. An implicit-explicit (IMEX) Crank-Nicholson Adams-Bashforth method (CNAB) is implemented in Python to solve the point defect balance equations for both vacancies and interstitial defects. Challenges in solving reaction diffusion problems with very small diffusion coefficients were identified. Accurate models for modeling and simulation of radiation damage in nuclear fuel materials and structural components are necessary for the evaluation of novel accident tolerant fuel concepts and performance of safety critical components used in nuclear reactors.

# Background - Radiation Damage

The effects of radiation damage can have a significant effect on the behavior of materials, especially relating to thermo-mechanical creep, material strength, embrittlement, and changes to many key thermal properties relevant to heat transfer performance. For this reason, it is desirable to have a detailed knowledge of the distribution of vacancies and interstitial impurities (both known as point defects) in materials subject to high radiation environments.

Radiation damage generally occurs due to the removal of atoms from their proper location in the crystal lattice structure. This results in the formation of point defects, line dislocations, and edge dislocations. The motion and interaction of these defects and dislocations has significant impact on material properties, which can increase or weaken material strength and ductility, affect creep behavior in high temperature environments, and reduce heat transfer effectiveness. This project focuses on point defects, or singular defects in the crystal lattice structure. In reality, these point defects tend to cluster together, but this phenomena is neglected in this project, and defects are considered as only individual particles subject to generation, destruction, and Fickian diffusion.

The two types of point defects are vacancies and interstitials. Vacancies refer to empty sites in the lattice structure which would normally be occupied by an atom. An interstitial defect refers to an atom in between crystal lattice sites, in a space where there would usually be no atom. Because the number of atoms within a material is conserved, vacancies and interstitials must be produced in pairs. The atom which occupies an interstitial location used to occupy a lattice site which has now become a vacancy. The interstitials and vacancies may then "jump" to different locations in the material due to stochastic thermodynamic behavior. This is often described simply by a diffusion process. While production of defect pairs may occur outside a high radiation environment, this effect can be neglected, as it is significantly smaller than the number of defect pairs produced when the material is subjected to significant amounts of radiation (Motta and Olander, 2017).

Defects are formed when an atom in the crystal lattice undergoes a collision (or electric field interaction for charged particles) with a particle of incident radiation, and sufficient energy is transferred to knock the atom out of its position. This initially removed atom, called the primary knock-on atom (PKA), goes on to remove other atoms from their sites in a displacement cascade. Some of these formed pairs rapidly recombine, but many displacements can result from a single incident particle-PKA interaction (Motta and Olander, 2017). The

results of this process are challenging to predict, but the Kinchin-Pease model provides a simplified approach to determining the number of displacements per atom (DPA) per second (Kinchin and Pease, 1955). The DPA measure is commonly used to evaluate cumulative radiation damage and long term effects on material properties. The Kinchin-Pease model will be discussed in greater detail in the next section.

This work is particularly applicable to evaluation of novel nuclear fuel materials and material behavior of structural components subject to large amounts of radiation.

# Model Description

While more detailed multiscale, atomistic and mesoscale models exist, they are highly complicated and require much greater computational power than is available for this project. We will use a simpler model to gain an intuition of the behavior of point defects in a sample material.

From Motta and Olander (2017), the change in concentration of both vacancies and interstitial point defects can be described as below:

$$\frac{\partial C_v}{\partial t} = D_v \nabla^2 C_v + k - K_{IV} C_i C_v - k_v^2 D_v C_v$$

$$\frac{\partial C_i}{\partial t} = \underbrace{D_i \nabla^2 C_i}_{\text{Diffusion}} \overbrace{+k}^{\text{Generation Rate}} \underbrace{-K_{IV} C_i C_v}_{\text{Recombination Rate}} \overbrace{-k_i^2 D_i C_i}^{\text{Sink Rate}}$$

## Diffusion

Fick's law provides a reasonable description of the diffusion of point defects through a material.

$$(\frac{\partial C}{\partial t})_{diff} = D \nabla^2 C$$

In this case, the challenge is in the determination of appropriate diffusion coefficients. Adding to the complexity, vacancies and interstitials diffuse via different mechanisms, each with a

different threshold energy – vacancies diffuse by being filled by atoms and leaving a new space, while interstitials diffuse by swapping of two atoms (Motta and Olander, 2017). Because of this difference, interstitials will have a smaller diffusion coefficient than vacancies.

We can calculate the diffusion coefficients for the Face-Centered Cubic (FCC) crystal lattice via the below equations (Derivation details may be found in Ch. 5 of Motta and Olander (2017)):

$$D_v = a_0^2 \nu_v exp(\frac{-E_{mv}}{k_B T})$$

$$D_i = a_0^2 \nu_i exp(\frac{-E_{mi}}{k_B T})$$

Where $a_0$ is the lattice parameter (distance between nearest neighbors in lattice structure), $E_m$ is the threshold energy for the jump between sites, $k_B$ is the Boltzmann constant, T is temperature, and $\nu$ is vibrational frequency of the diffusing atom (See Motta and Olander (2017) Ch. 5 for details).

Once these values for diffusion coefficients are calculated, computation of the diffusion term for the point defect balance equation is relatively simple.

## Generation

Since we focus on point defect production due to radiation, the generation of defect pairs due to other processes are minimal (Motta and Olander, 2017). The defect generation rate $k$ must be a function of the radiation flux, incident energy, crystal lattice structure, atomistic configuration, and threshold energy for particle ejection from a lattice site. Rather than simulating the effects of the incident radiation-PKA collision and the resulting displacement cascade, a simplified model is used to approximate the final damage state. The Kinchin-Pease (K-P) model (Kinchin and Pease, 1955) estimates the total number of displacements resulting from a single PKA removed from its lattice site due to an interaction with incident radiation. The model was derived by assuming all particle interactions can be modeled as elastic collisions between atoms. If the PKA and successive displaced atoms have sufficient kinetic energy, the next chain of atoms are also ejected from their lattice sites. This assumption results in a simple, yet reasonable approximation of the microstructural state of a material following an initial PKA ejection, except at higher energies.

The K-P model is as follows, where $\nu$ is the number of displacements resulting from a single PKA, E is the energy of the PKA, and $E_d$ is the threshold energy needed to eject the next atom in the displacement cascade:

$$\nu_{KP}(E) = \frac{E_{PKA}}{2E_d}$$

To relate the flux to number of PKAs produced, we calculate the reaction rate. For this project, it is assumed that the incident radiation is comprised of mono-energetic 1 MeV neutrons, and the material irradiated is Type 316 Stainless Steel. Then, the number of PKAs produced is found via:

$$N_{PKA} = \phi_n \sigma_{disp}^{316}(E_n)$$

Where $\phi_n$ is the neutron flux in $cm^{-2}sec^{-1}$, $\sigma_{disp}^{316}$ is the microscopic displacement cross section (probability a neutron interacts with an atom and ejects that atom from the lattice site) in $cm^2$, or barns.

Finally, to determine the generation rate in DPA/sec, we combine the above equations (Motta and Olander, 2017):

$$k = \frac{\phi_n \sigma_{disp}^{316}(E_n)}{4E_d} \Lambda E_n$$

where $\Lambda$ is the energy transfer parameter, added to account for the portion of the energy transferred from the incident neutron to the PKA. As a reminder, this generation rate provides the number of displacements per total number of atoms in the material per second. For this reason, we expect a generation rate $<<$ 1 DPA/sec.

## Recombination

Since point defects involve atoms missing from and between the proper crystal lattice sites, the presence of these vacancies and interstitials are not energetically favorable. For this reason, any vacancies and interstitials within a certain distance will tend to combine or annihilate each other. Therefore, the number of recombinations per second which occur must be dependent on the number of neighbors in the lattice structure, the size of the region in which recombination is energetically favorable, and the concentrations of both vacancies and interstitials (Motta and Olander, 2017).

The equation modeling the recombination rate is:

$$R = K_{IV} C_i C_v$$

$K_{IV}$ is known as the recombination constant, and is defined:

$$K_{IV} = \frac{Z_{IV}(D_i + D_v)}{\lambda^2}$$

Where $D_i$ and $D_v$ are the diffusion coefficients for interstitials and vacancies respectively, $\lambda$ is the jump distance for atoms to move between lattice sites, and $Z_{IV}$ is the recombination number, describing the number of favorable lattice positions, depending on the material and crystal structure. Since the recombination number is situationally dependent and challenging to determine, a reasonable number for iron is selected for this code, rather than directly calculating this parameter based on material properties.

Of note is the fact that both the vacancy and interstitial concentration are present in this term. This will present a numerical challenge in solving the system due to coupling of the two systems and the non-linearity of the term.

## Sinks

While a realistic model of radiation effects in materials must include sinks, a detailed implementation of sink behavior is not present in this code. Generally, point defects are absorbed into line and plane dislocations, spherical cavities, and grain boundaries (Motta and Olander, 2017). Effectively, the point defect (of either type) that collides with these structures are removed from the system. These structures are very spatially dependent, and implementation into this project would require defining locations and dimensions of these throughout the domain. At this time, this feature has not been added. However, if desired, a constant sink strength can be defined by the user which is uniform across the entire domain.

The sink rate equation for vacancies is shown below (the equation for interstitials is equivalent).

$$S = k_v^2 D_v C_v$$

$k_v^2$ is the total sink strength for the point defect, in $cm^{-2}$. Sink strengths for various sink types and geometries may be found in Ch 13.5 of Motta and Olander (2017).

# Numerical Approach

Initially, a simple forward time, centered space (FTCS) method was attempted. Since the time step for this method is based on forward Euler, extremely small time steps were required for stability, and the results were of questionable accuracy. For this reason the Adams-Bashforth 3 step explicit method was then attempted. Satisfactory results were achieved for the diffusion and generation terms, but when the nonlinear defect recombination was considered, the scheme rapidly became unstable. This is likely due to the instability of the diffusion term, especially near domain boundaries where the concentration gradients were large. It was then decided to use the Crank-Nicholson Adams-Bashforth (CNAB) IMEX scheme (Ascher et al., 1995) to improve stability, with hopes that this would assist in stability when including the non-linear recombination term.

The CNAB scheme in 2D is shown here:

$$\frac{C_{ij}^{n+1} - C_{ij}^{n}}{\Delta t} = \frac{D}{2(\Delta x)^2}[(C_{i+1,j}^{n+1} - 2C_{ij}^{n+1} + C_{i-1,j}^{n+1}) + (C_{i,j+1}^{n+1} - 2C_{ij}^{n+1} + C_{i,j-1}^{n+1})] + \frac{D}{2(\Delta x)^2}[(C_{i+1,j}^{n} - 2C_{ij}^{n} + C_{i-1,j}^{n}) + (C_{i,j+1}^{n} - 2C_{ij}^{n} + C_{i,j-1}^{n})] + \frac{3}{2}F(C_{ij}^{n}) - \frac{1}{2}F(C_{ij}^{n-1})$$

Where $F = \frac{dC}{dt}$, excluding the diffusion term. The CNAB method applies the implicit Crank-Nicholson term to the diffusion term of the equation and the explicit 2-step Adams-Bashforth method is used for the generation, recombination (non-linear), and sink terms. This results in unconditional stability for the diffusion term while also calculating the other terms with a faster rate of convergence than a single step method. Consequentially, the implicit portion of the method will increase the computational cost per time step, and the implementation is complicated by using a multistep explicit method (Ascher et al., 1995).

Rearranging, and letting $\frac{D\Delta t}{2(\Delta x)^2} = \alpha$ the scheme becomes:

$$(1-4\alpha)C_{ij}^{n+1} + \alpha(C_{i-1,j}^{n+1} + C_{i+1,j}^{n+1} + C_{i,j-1}^{n+1} + C_{i,j+1}^{n+1}) = (1-4\alpha)C_{ij}^{n} + \alpha(C_{i-1,j}^{n} + C_{i+1,j}^{n} + C_{i,j+1}^{n} + C_{i,j-1}^{n}) + \frac{3\Delta t}{2}F(C_{ij}^{n}) - \frac{\Delta t}{2}F(C_{ij}^{n-1})$$

Since all the terms on the right side of the equation are known when calculating at time $n+1$, the system can be placed into matrix form and solved using an efficient linear system solver. The resulting linear system can be seen below.

$$\begin{bmatrix} (1-4\alpha) & \alpha & 0 & ... & \alpha & 0 & ... & ... & 0 \\ \alpha & (1-4\alpha) & \alpha & 0 & ... & \alpha & 0 & ... & 0 \\ 0 & \alpha & (1-4\alpha) & \alpha & 0 & ... & \alpha & 0 & 0 \\ \vdots & ... & \ddots & \ddots & \ddots & 0 & ... & \ddots & \vdots \\ 0 & ... & 0 & \ddots & \ddots & \ddots & 0 & ... & \alpha \\ \alpha & 0 & ... & 0 & \ddots & \ddots & \ddots & ... & 0 \\ 0 & \ddots & 0 & ... & 0 & \ddots & \ddots & \alpha & 0 \\ 0 & ... & \alpha & 0 & ... & ... & \alpha & (1-4\alpha) & \alpha \\ 0 & 0 & ... & \alpha & 0 & ... & 0 & \alpha & (1-4\alpha) \end{bmatrix} \cdot \begin{bmatrix} C_{11}^{n+1} \\ C_{12}^{n+1} \\ \vdots \\ C_{1,k-1}^{n+1} \\ C_{21}^{n+1} \\ \vdots \\ C_{m-1,1}^{n+1} \\ \vdots \\ C_{m-1,k-1}^{n+1} \end{bmatrix}$$

$$= \begin{bmatrix} (1-4\alpha)C_{11}^n + \alpha(C_{21}^n + C_{12}^n) + \frac{3\Delta t}{2}F(C_{11}^n) - \frac{\Delta t}{2}F(C_{11}^{n-1}) \\ (1-4\alpha)C_{12}^n + \alpha(C_{11}^n + C_{22}^n + C_{13}^n) + \frac{3\Delta t}{2}F(C_{12}^n) - \frac{\Delta t}{2}F(C_{12}^{n-1}) \\ \vdots \\ (1-4\alpha)C_{1,k-1}^n + \alpha(C_{2,k-1}^n + C_{1,k-2}^n) + \frac{3\Delta t}{2}F(C_{1,k-1}^n) - \frac{\Delta t}{2}F(C_{1,k-1}^{n-1}) \\ (1-4\alpha)C_{21}^n + \alpha(C_{11}^n + C_{31}^n + C_{22}^n) + \frac{3\Delta t}{2}F(C_{21}^n) - \frac{\Delta t}{2}F(C_{21}^{n-1}) \\ \vdots \\ (1-4\alpha)C_{m-1,1}^n + \alpha(C_{m-2,1}^n + C_{m-1,2}^n) + \frac{3\Delta t}{2}F(C_{m-1,1}^n) - \frac{\Delta t}{2}F(C_{m-1,1}^{n-1}) \\ (1-4\alpha)C_{m-1,k-1}^n + \alpha(C_{m-2,k-1}^n + C_{m-1,k-2}^n) + \frac{3\Delta t}{2}F(C_{m-1,k-1}^n) - \frac{\Delta t}{2}F(C_{m-1,k-1}^{n-1}) \end{bmatrix}$$

The matrix is potentially very large and sparse, so an iterative solver is an appropriate choice. Since the matrix is symmetric, the MINRES method is used. The MINRES function in the Python SciPy (Virtanen and SciPy 1.0 Contributors, 2020) package is applied to solve the linear system.

# Results

Significantly faster convergence is seen using the CNAB IMEX scheme (for non-diffusion terms) than FTCS. Additionally, the stability concerns seen when using solely an explicit method are reduced due to the unconditional stability of the Crank-Nicholson portion of the method applied to the diffusion term. That being said, stability (and maximum time step) is still greatly limited due to the very large generation and recombination terms.

Unfortunately, for the stable case, the graphic is not very interesting. This is due to the material properties of the stainless steel and the extremely slow diffusion rate of vacancies and interstitial point defects. This results in an $\alpha \approx 0$. Numerically, this results in a

matrix very close to the identity matrix, so practically no diffusion is visible for reasonable simulation times. In realistic cases, equilibrium is reached on the order of $10^5$ seconds (Motta and Olander, 2017). With the stability concerns, the code would need to be run for an extended period of time to see diffusion occuring. When the diffusion coefficients are artificially increased, the method became rapidly unstable. Clearly this code is unsatisfactory for this application. Concentration values, however are within expectations for the 600K temperature used and the given radiation flux, but further validation would be beneficial.

Overall, improvement is seen with the CNAB method over the FTCS in terms of convergence and stability. Use of an iterative linear system solver reduces the computational cost of an implicit method. However, this project shows how challenging stiff problems may be to solve numerically. While permissive step sizes for stability are much larger for CNAB than FTCS, they are still too small to be practical. The diffusion effect is not even visible. A stability analysis is necessary to identify the best method for moving forward. The Modified CNAB method (MCNAB) may be an appropriate choice since the rapidly oscillating instabilities seen at larger time steps for the CNAB method would be damped by the additional numerical dissipation (Ascher et al., 1995), but it is anticipated that the small diffusion rate will continue to be an issue.

# Conclusion

The behavior of Type 316 Stainless Steel subject to large radiation fluxes was considered. The generation, diffusion, recombination, and sinking of atomic point defects are considered, and a Python code was developed to solve the point defect balance equations for both vacancy and interstitial defect concentrations. Several numerical methods were implemented in Python for this problem, with varying degrees of success. A FTCS method was acceptable when recombination was neglected and very small time step sizes were permissible. Convergence rate was improved when using multistep Adams-Bashford 3 method, but the method failed to converge when considering recombination. An implicit-explicit (IMEX) Crank-Nicholson Adams-Bashforth method (CNAB) was finally used, but small time step sizes were still required, making simulation of material behavior over longer time scales impractical. In fact, with the CNAB method, diffusion was not even visible on reasonable time scales. A more sophisticated numerical method would likely have better performance. The simple point defect balance model was shown to be inadequate for detailed analysis of these types of problems, both due to oversimplifications in material behavior and computational cost for

simulating longer irradiation times. Higher fidelity models accounting for realistic material behavior should be used when simulating radiation effects. However, this model is effective for estimating concentration values and building intuition about radiation processes within matter.

# Appendix A - Github Link to Code

https://github.com/mason-fox0/PointDefects

# Appendix B - Code Dependencies

The following softwares and python libraries are required to run the code.

- Python 3.0 or greater
- math (https://docs.python.org/3/library/math.html)
- numpy (https://numpy.org)
- scipy (https://scipy.org)
- matplotlib (https://matplotlib.org)

See README.md file for more info.

# References

Ascher U. M., Ruuth S. J., and Wetton B. T. R. (1995). "Implicit-Explicit Methods for Time-Dependent Partial Differential Equations". *SIAM Journal on Numerical Analysis* 32.3, pp. 797–823. URL: http://www.jstor.org/stable/2158449.

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