Criticality studies of actinides within fissures

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CRITICALITY STUDIES OF ACTINIDES WITHIN FISSURES

by

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1997

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ABSTRACT

Criticality Studies of Actinides within Fissures

by

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The storage of nuclear waste in underground storage facilities presents numerous engineering challenges and risks. Experimental verification of engineered underground storage is impractical or prohibitively expensive, leaving scientists with few options. A 1995 report by Bowman and Venneri of the Los Alamos National Laboratory generated considerable controversy by hypothesizing that wastes composed of fissionable plutonium leached from underground storage containers could pose a nuclear criticality hazard. They proposed cases where plutonium collected in underground fractures could lead to sustained nuclear fission. In overmoderated cases, they argued that the resulting release of energy from fission could result in steam explosions, or even an underground nuclear explosion (autocriticality). Their hypothesis had severe implications for the feasibility of long-term nuclear waste storage in geologic repositories.

The Bowman and Venneri hypothesis led to the need for a study of conditions that could lead to a critical event in a geologic repository due to releases of uranium or plutonium. Information about the likely consequences of a critical event is also important in repository design. To accomplish this study a numerical simulation code, GEOCRIT,
was written to model radionuclide transport from the repository into a fracture below the repository. Once sufficient material has accumulated in the fracture and rock matrix, the neutronics portion of the code is started to simulate heat generation and fluid flow. The thermohydraulics portion of the code calculates heat generation from fission, stream functions, velocity, and pressure of the fluid in the fracture and rock matrix. The transport portion of the code incorporates numerous parameters that can be varied to simulate different radionuclide buildup in the fracture and rock matrix.

Variation of the solubility, diffusion, deposition coefficients in the program yield different accumulations of radionuclides in the fracture and rock matrix. In each case the majority of the deposition occurs in the fracture and adjacent rock matrix. The variation in accumulated radionuclides led to different neutron distributions within the fracture and rock matrix. In each case the largest neutron flux is in the fracture leading to the highest temperature also being within the fracture. The increase in temperature leads to a transient bifurcating flow within the rock matrix.

One potential risk for the geological repository is the buildup of radionuclide in the saturated rock with subsequent drying out of the rock matrix. This case is known as overmoderation, whereby the neutron flux increases as the water dries out. It has been hypothesized that certain configurations of radionuclides may be overmoderated and drying out of the water in the rock may lead to a critical event. Three cases of overmoderation were simulated with the code.

Three different levels of accumulation of radionuclides within the fracture and rock matrix, with subsequent drying out of the crack, lead to an increase in neutron flux. This increase in neutron flux indicates that the accumulated radionuclide in the saturated
fracture and rock matrix is overmoderated. In one of the overmoderated cases, the reactor reached a steady state with transient neutron behavior due to water in the surrounding rock changing from liquid to steam and back. One case shows a significant increase in neutron flux.
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<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>Area ($m^2$)</td>
</tr>
<tr>
<td>$A$</td>
<td>Atomic mass number</td>
</tr>
<tr>
<td>$A_v$</td>
<td>Avogadro’s number</td>
</tr>
<tr>
<td>$A_w$</td>
<td>Atomic weight ($u$)</td>
</tr>
<tr>
<td>$b$</td>
<td>Fracture width ($m$)</td>
</tr>
<tr>
<td>$c_0$</td>
<td>Source concentration ($kg/m^3$)</td>
</tr>
<tr>
<td>$c$</td>
<td>Concentration ($kg/m^3$)</td>
</tr>
<tr>
<td>$\dot{c}$</td>
<td>Rock fracture deposition concentration ($kg/m^2$)</td>
</tr>
<tr>
<td>$c_f$</td>
<td>Specific heat of the fluid $kJ/kgK$</td>
</tr>
<tr>
<td>$c_m$</td>
<td>Concentration in rock matrix ($kg/m^3$)</td>
</tr>
<tr>
<td>$\dot{c}_m$</td>
<td>Rock matrix deposition concentration ($kg/kg$)</td>
</tr>
<tr>
<td>$c_s$</td>
<td>Specific heat of rock matrix $kJ/kgK$</td>
</tr>
<tr>
<td>$D$</td>
<td>Neutron diffusion coefficient ($m$)</td>
</tr>
<tr>
<td>$D$</td>
<td>Mass diffusion coefficient ($m^2/s$)</td>
</tr>
<tr>
<td>$D_e$</td>
<td>Effective Diffusion Coefficient ($m^2/s$)</td>
</tr>
<tr>
<td>$g$</td>
<td>Gravity ($m/s^2$)</td>
</tr>
<tr>
<td>$I$</td>
<td>Intensity ($particles/m^2 sec$)</td>
</tr>
<tr>
<td>$I_0$</td>
<td>Initial inventory of nuclide</td>
</tr>
<tr>
<td>$K$</td>
<td>Permeability ($m^2$)</td>
</tr>
<tr>
<td>$k_d$</td>
<td>Partition coefficient ($m^3/kg$)</td>
</tr>
<tr>
<td>$k_{eff}$</td>
<td>Effective multiplication factor</td>
</tr>
<tr>
<td>$k_x$</td>
<td>Infinite multiplication factor</td>
</tr>
<tr>
<td>$k_f$</td>
<td>Thermal conductivity of the fluid $W/mK$</td>
</tr>
<tr>
<td>$k_{log}$</td>
<td>Glass log surface area rate constant $kg/m^2s$</td>
</tr>
<tr>
<td>$K_{log}$</td>
<td>Thermodynamic parameter for glass log $kg/m^3$</td>
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<tr>
<td>$k_s$</td>
<td>Thermal conductivity of rock matrix $W/mK$</td>
</tr>
<tr>
<td>$l_{log}$</td>
<td>Glass log length ($m$)</td>
</tr>
</tbody>
</table>
\( \dot{m}_{Pu} \) Mass rate of Pu available from leaching (kg/yr)

\( \dot{m}_{Pu\_trans} \) Mass rate of Pu transport away from log (kg/yr)

\( n \) Number of neutrons

\( n \) Mass flux \( \left( \frac{kg}{m^3 s} \right) \)

\( N \) Number density (atoms/m^3)

\( p \) Pressure (Pa)

\( P_{NL} \) Non-leakage probability

\( q^\prime \) Rate of heat generation \( \left( \frac{W}{m^3} \right) \)

\( Q_{log} \) Dissolved silica in water concentration \( \left( \frac{kg}{m^3} \right) \)

\( r_{log} \) Radius of glass log (m)

\( \dot{R}_{log} \) Alteration rate of glass log (kg/yr)

\( s_{log} \) Surface area of glass log (m^2)

\( S \) Neutron source (neutrons/m^2)

\( T \) Temperature (K)

\( t \) Time (s)

\( t_c \) Time at which waste container fails (yr)

\( u \) Velocity (m/s)

\( U \) Velocity of water in rock fracture (m/s)

\( v \) Velocity (m/s)

\( V \) Velocity (m/s)

\( \varphi \) Volume (m^3)

\( w \) Velocity in the z-direction (m/s)

\( wt \) Weight percent

\( Z \) Number of protons

\( \alpha_{log} \) Experimental constant for glass alteration

\( \beta \) Volume expansion coefficient \( \left( \frac{1}{K} \right) \)

\( \varepsilon \) Porosity \( (m^3/m^3) \)

\( \theta \) Angle (deg)

\( \kappa \) Lumped rock fracture deposition coefficient (m)

\( \kappa_{log} \) Glass log surface alteration constant \( \left( \frac{kg}{m^2 \ yr} \right) \)

\( \kappa_m \) Lumped rock matrix deposition coefficient (1/s)
\( \lambda \)  Decay constant \( \left( \frac{1}{s} \right) \)

\( \mu \)  Viscosity of water \( \left( \frac{kg}{ms} \right) \)

\( \nu \)  Average number of neutrons per fission

\( \rho \)  Density \( (kg/m^3) \)

\( \rho_b \)  Bulk density \( (kg/m^3) \)

\( \rho_f \)  Density of fluid \( (kg/m^3) \)

\( \rho_g \)  Density glass log \( (kg/m^3) \)

\( \rho_s \)  Density of rock matrix \( (kg/m^3) \)

\( \sigma \)  Microscopic cross section \( (b \text{ or barns} = 10^{-28} m^2/\text{nucleus}) \)

\( \sigma_a \)  Absorption microscopic cross section \( (b) \)

\( \sigma_f \)  Fission microscopic cross section \( (b) \)

\( \sigma_s \)  Scattering microscopic cross section \( (b) \)

\( \sigma_{se} \)  Elastic scattering microscopic cross section \( (b) \)

\( \sigma_{si} \)  Inelastic scattering microscopic cross section \( (b) \)

\( \sigma_t \)  Total scattering microscopic cross section \( (b) \)

\( \Sigma \)  Macroscopic cross section \( (m^2/m^3) \)

\( \Sigma_a \)  Absorption macroscopic cross section \( (m^2/m^3) \)

\( \Sigma_f \)  Fission macroscopic cross section \( (m^2/m^3) \)

\( \Sigma_s \)  Scattering macroscopic cross section \( (m^2/m^3) \)

\( \Sigma_t \)  Total macroscopic cross section \( (m^2/m^3) \)

\( \tau_c \)  Mean container failure time \( (yr) \)

\( \phi \)  Neutron flux \( \left( \frac{\text{neutrons}}{m^2s} \right) \)

\( \Phi \)  Viscous dissipation function \( \left( \frac{1}{s^2} \right) \)
ACKNOWLEDGMENTS

I would like to express my most sincere thanks to Dr. William Culbreth for his guidance and constant interest. Without his supervision, I would have not been able to complete this thesis.

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CHAPTER 1

INTRODUCTION

Geologic repositories have been proposed for the long-term storage of spent nuclear fuel. Storage in deep, stable geologic formations would allow wastes to radioactively decay to safe levels that are typical of the original ore that generated the fuel. Unfortunately, the half-lives of fissionable species of uranium, plutonium, americium, and curium will create potential criticality hazards for millions of years, long after the roughly 10,000 year lifetime of the spent fuel containers.

Once spent fuel canisters begin degrading due to the presence of water, the internal contents will be released into the near-field environment surrounding the canisters. Waste canisters will contain quantities of plutonium-239, a highly fissionable form of this element. There is a strong chance that plutonium will collect in the fissures and voids present in the rock surrounding the repository. In the presence of water, these wastes can go critical and sustain a chain reaction. In this study, the conditions that can lead to criticality will be explored. The consequences of a criticality event will also be presented based on computer models that have been validated through comparison with natural underground reactors/repositories located in Gabon, Africa.

There are no existing geological repositories for high-level nuclear waste and hence no data for the performance of the repository as a whole. Recently there has been considerable research conducted on different aspects of a potential repository to support
the need to find suitable storage sites for high-level waste. Water infiltration into the repository complicates matters since it is uncertain how the waste will behave. Unfortunately, due to the complexity and environmental concerns very little experimental data exists for how radionuclide will migrate within the repository and repository host material.

A hypothesis by Bowman and Venneri on what might happen to waste if it migrated into the repository host material was made public in the New York Times on March 5, 1995. The hypothesis stated that material may transport from the waste container and accumulate in the host material in a critical mass. The critical mass could go super-critical and lead to an uncontrolled chain reaction and possible explosion. In a series of Los Alamos National Laboratory reports in 1994 Bowman and Venneri detailed their hypothesis and gave examples of different concentrations of water and accumulated Pu-239 and the possibility that it would lead to a critical event. To simplify their models, they used a sphere of pure Pu-239. They then created cases with different radius of the sphere and water concentration. This data was used as input to the MCNP (Monte Carlo N-Particle) transport code which calculated criticality of the different scenarios. The case that most concerned them was the overmoderated case. In the overmoderated case the Pu-239 was not super-critical until the concentration of water decreased. The MCNP code calculated that the overmoderated case would become super-critical as the water concentration was reduced.

Bowman and Venneri further hypothesized that the Pu-239 may become prompt critical yielding an exponential energy generation. If the critical mass were above ground it would most likely disassemble or change shape thereby terminating the exponential
increase in energy generation. However, if the critical mass were underground the host rock could confine the mass, keeping it from disassembling or changing shape thereby terminating the critical event. The confined material could lead to an explosive event. Bowman and Venneri explored the potential explosive yield in Los Alamos National Laboratory report LA-UR-94-4022A. Potential explosive yield is beyond the scope of this work.

As one might imagine, Bowman and Venneri’s hypothesis created a stir in the nuclear and geologic scientific community. Following the release of the New York Times’ article and their papers, a series of papers and studies were commissioned to further explore the validity of the Bowman and Venneri’s hypothesis. The following is a survey of the work produced in the effort to study the Bowman and Venneri’s hypothesis.

In 1996 Myers explored mixtures of plutonium, Nevada tuff, and water for the potential systems leading to explosions. The study examined explosive characteristics of fissioning systems. The paper is primarily concerned with yields from slow fission energy release. A computer model, MRKJ, was developed to calculate the dynamic energy release of supercritical transient reactors. The code combines thermodynamics, material motions, and neutrons transport differential equations to model transient nuclear systems. The first order ordinary differential equation is solved using fourth order Runge-Kutta method. The MRKJ code uses the ONEDANT, one-dimensional discrete ordnance transport, code to calculate the neutron transport. The MRKJ code was used to study three different critical configurations of plutonium, Nevada tuff, and water. The paper concluded that “Critical configurations of plutonium, Nevada tuff, and water can exist”. They also conclude no explosion would arise for these configurations.
An analysis of bounding estimates for criticality was conducted by Rechard in 1996. As noted in the paper, detailed simulations were purposely avoided and the use of general knowledge and natural analogues were employed. The paper explores several scenarios that lead to a criticality event and assigns a probability to the event. The probability is used to determine if a particular scenario should be dismissed. The scenarios of interest are the assembly of material and the moderation of the assembly. Rechard looked at slow and fast assembly of material with low and high moderation. Bounding estimates for fast assembly of material, both moderated and unmoderated, were estimated between $10^{15}$ to $10^{20}$ fissions. According to the paper, a critical event with $10^{20}$ fissions would be a negligible event. The paper further states that “a criticality event is not easily dismissed through simple logical argument.”

Kastenberg et al. assessed scenarios leading to critical events in geologic repositories. Kastenberg explored seven events required to cause a critical event in a repository. He stated that failure of any one event occurring would nullify the possibility of a critical event taking place. The events are as follows:

1. Waste package failure
2. Removal of neutron poisons
3. Hydrological transport of fissile material
4. Accumulation of material in the host rock
5. Accumulation of material in a critical configuration
6. As the systems temperature increased there would be a positive reactivity feedback
7. The system remains super-critical until the energy being generated
becomes of concern

It was determined that none of the seven events could be ruled out. The paper assumed
transport of highly enriched uranium (HEU) in solution in groundwater and the transport
of plutonium sorbed colloids. Static neutronics calculations were conducted assuming
the material accumulated on the surfaces of multiple fractures. It was assumed that the
fractures were parallel to each other and equally spaced. A parametric study of deposited
material and water concentration was conducted. From the study it was concluded that if
sufficient fissile material were to accumulate in the host rock a critical event might occur.

In a paper submitted to waste management and a report submitted to a Japan
corporation, Ahn et al. assess the possibility of an autocatalytic criticality (see section
3.1) event occurring due to the migration of nuclear waste in an underground geological
repository. The release of the material from the repository and subsequent transport and
accumulation was studied. Mathematical models were developed to describe the release
of material from its storage in glass logs and the transport and accumulation. A spherical
shape with the same mass and surface area as the glass log is assumed for the derivation
of the equations governing the release of radionuclides. A molecular diffusion model is
derived describing the release. The release model is used as a source term for the
transport and accumulation model. The transport model is based on molecular diffusion
and advection. The model assumes that all radionuclides arriving at a location of interest
away from the original storage location accumulates there. The computer code MCNP
was then used to analyze the minimal criticality masses and water concentrations needed
to create a critical event.
Moridis et al. investigated the transport of radionuclides in the unsaturated zone of tuff located in Nevada. The paper looked at fracture and porous media flow with different permeability. Two computer codes, EOS9nT and T2R3D, were used to simulate the 3-D transport model of a potential storage location. EOS9nT and T2R3D are from the TOUGH2 (transport of unsaturated groundwater and heat) family of codes. EOS9nT simulates flow and transport of an arbitrary number of solutes or colloids. T2R3D simulates saturated or unsaturated flow of a single solute. The codes were used to generate breakthrough curves for three radionuclides (Tc-99, Np-237, and Pu-239) using intentions and continues release source terms. The codes were also used to look at mass accumulation within the host rock surrounding the storage site.

Several computer codes have been used to study criticality of a potential storage site. As stated earlier, MCNP is a Monte Carlo N-Particle transport code. The code can be used to assess the criticality of a static system. However, MCNP does not take into account fluid flow or dynamic changes in material concentrations. The TOUGH2 family of codes calculates fluid flow and heat transfer but lacks the ability to calculate criticality of a particular system.

The studies discussed above are primarily concerned with mass transport and criticality of accumulated radionuclides. Lacking in all of the scientific studies is what happens as the material builds up in the host rock. As the material builds up and starts to produce heat the question is what happens in the surrounding rock, barring an explosion. This work focuses on the transport and buildup of fissile material with subsequent heat generation and fluid flow in the surrounding host media.
This paper takes a safety factor approach instead of a probability approach. Instead of looking at the probability that an event will not happen, it looks at what will happen if an event does occur, i.e. what is the worst case scenario. The results demonstrate that accumulated actinide wastes in fissures within a repository can support neutron criticality. The overmoderated cases predicted by Bowman and Venneri were also verified, leading to the violent and periodic emission of water as steam from the fissure “reactor”. The conditions that can lead to criticality events are described along with the consequences. In this paper we analyze what may happen so that minimum safeguards can be employed.

In order to analyze the conditions that lead to criticality in a geologic repository a FORTRAN code was developed. The code simulates mass transport of radionuclides from the waste storage location into the host rock. The code tracks the buildup of radionuclides within a fracture and host rock. As the material builds up the code solves the neutron diffusion equation to calculate the heat generated by fission. The code then simulates the thermally driven fluid flow within the fracture and rock matrix. The code can be used to determine potential criticality events within the repository. Also, the code provides a tool to study how fluid flow could cause migration of radionuclides within the rock matrix.
CHAPTER 2

THEORY OF RADIONUCLIDE TRANSPORT

2.1 Waste Package Dissolution

Waste material is typically stored in cylindrical canisters. A typical container will contain a number of glass logs that have plutonium distributed uniformly in the log. An exponential failure rate will be assumed (Wilson, 1991). The exponential failure rate has a distribution function given by

\[ f_c(t) = \frac{1}{\tau_c} e^{\frac{-t}{\tau_c}} \]  

(2.1)

The probability that a container fails between time 0 and t is given by

\[ \int_0^t \frac{1}{\tau_c} e^{\frac{-t}{\tau_c}} dt_c = 1 - e^{\frac{-t}{\tau_c}} \]  

(2.2)

The mean failure time is given by

\[ \int_0^t \frac{1}{\tau_c} e^{\frac{-t}{\tau_c}} dt_c = \tau_c \]  

(2.3)

Thus \( \tau_c \) will be used throughout as the time of container failure.
It is assumed that storage is fully flooded and that there is a net flow rate through a container that has failed at time $t = \tau_c$. The dissolution rate of the log will be taken as (O’Connell, 1997)

$$\dot{R}_{\text{log}} = s_{\text{log}}(t)k_{\text{log}} \left[ 1 - \left( \frac{Q_{\text{log}}}{K_{\text{log}}} \right)^{\alpha_{\text{log}}} \right]$$

(2.4)

where $s_{\text{log}}(t)$ is surface area of the log, $k_{\text{log}}$ is a rate constant, $Q_{\text{log}}$ is the concentration of silica dissolved in the water, $K_{\text{log}}$ is a thermodynamic parameter of the glass log, and $\alpha_{\text{log}}$ is an experimentally determined constant. The surface area change is determined to be

$$s_{\text{log}} = 2\pi r_{\text{log}}(t)(r_{\text{log}}(t) + l_{\text{log}}(t))$$

(2.5)
where

\[
r_{\log}(t) = \begin{cases} 
    r_{\log} - \frac{k_{\log}}{\rho_{\log}} \left[ 1 - \left( \frac{Q_{\log}}{K_{\log}} \right)^{\alpha_{\text{aq}}} \right] & \text{for } 0 \leq t \leq \frac{r_{\log} \rho_{\log}}{k_{\log} \left[ 1 - \left( \frac{Q_{\log}}{K_{\log}} \right)^{\alpha_{\text{aq}}} \right]} \\
    0 & \text{otherwise}
\end{cases}
\]  
(2.6)

\[
l_{\log}(t) = \begin{cases} 
    l_{\log} - \frac{2k_{\log}}{\rho_{\log}} \left[ 1 - \left( \frac{Q_{\log}}{K_{\log}} \right)^{\alpha_{\text{aq}}} \right] & \text{for } 0 \leq t \leq \frac{l_{\log} \rho_{\log}}{2k_{\log} \left[ 1 - \left( \frac{Q_{\log}}{K_{\log}} \right)^{\alpha_{\text{aq}}} \right]} \\
    0 & \text{otherwise}
\end{cases}
\]  
(2.7)

Plutonium 239 has a half life of 24,110 years, as plutonium decays by alpha emission to one of its daughter products, uranium 235. The mass rate of plutonium and uranium available for transport from a glass log that is being dissolved in water is given by

\[
\dot{m}_{\text{pu}}(t) = \dot{R}_{\log} \cdot \rho_{\text{wpu}}
\]  
(2.8)

where \(\rho_{\text{wpu}}\) is the weight percent of plutonium in the glass log. The available plutonium and uranium will be dissolved into water according to the leach rates. Leach rates for plutonium vary widely from \(3.8 \times 10^{-6} \frac{g}{m^2 \text{day}}\) to \(0.38 \frac{g}{m^2 \text{day}}\) (Macfarlane, 1998) whereas leach rates for uranium have been reported as being between \(3.1 \frac{mg}{m^2 \text{day}}\) and \(8.6 \frac{mg}{m^2 \text{day}}\) (Steward, 1994).
Colloids are particles (SiO₂ for this case) in solution, ranging in size from 1nm to 1μm in radius, that tend to sorb and transport contaminants. Some of the dissolved material will be sorbed onto colloids based on the concentration of colloids and the propensity of plutonium and uranium to sorb onto the colloids. Experimental colloid concentrations range from 140 to 200 mg/L (Viswanthan, 2003) and have a 70% to 90% sorption rate in natural water from well U-20WW at the Nevada Test Site (NTS). The mass rate of the total amount of plutonium transported away from the log will be denoted as \( \dot{m}_{\text{Pu}_\text{trans}} \) and it should be noted that

\[
\dot{m}_{\text{Pu}_\text{trans}} (t) \leq \dot{m}_{\text{Pu}} (t)
\]  

(2.9)

2.2 Colloid Transport Theory

Colloids are particles that range from 1nm to 1μm in radius and are insoluble in the solution in which they are suspended (Hunter, 2001). While the concentration of plutonium and uranium dissolved in water may be low, the accumulation of these actinides onto colloids can be quite high. Colloids can transport actinides through fracture flow at very high velocities far exceeding simple diffusion. The governing equation for the advection and diffusion of suspended colloid particles is given by (Buck, 1994)

\[
\frac{\partial \bar{c}}{\partial t} = D \frac{\partial^2 \bar{c}}{\partial x^2} - U \frac{\partial \bar{c}}{\partial x} - \kappa \bar{c}
\]

(2.10)

where \( \bar{c} \) is the colloid concentration, \( D \) is the diffusion coefficient, \( U \) is the fluid velocity and \( \kappa \) is the lumped deposition coefficient. Equation (2.10) can be solved in the same manner as the governing equation for fracture and porous media flow, as described in section 2.3.
The amount of contaminant that sorbs onto a particular colloid varies by the type of colloid, the charge on the particles, the type of contaminant, and the chemistry of the solution in which it is suspended. The sorption or partition coefficient for Pu under a variety of pH levels and different types of colloids are given by Kersting, 2003 and Lu, 1998. In field and laboratory experiments it has been shown that 70% to 90% of the Pu sorbed on the colloids (Kersting, 2003, Lu, 1998, and Viswanthan, 2003).

2.3 Fracture and Porous Media Flow

Fluid flow from the waste package storage area will occur through fractures or fissures in the rock matrix. This flow will then penetrate the rock matrix through porous media flow. The model developed to study critical events in a nuclear waste repository will couple both fracture and porous media flow. The flow in a rock fracture has been idealized as fully developed laminar flow between two parallel plates as shown in Figure 2.2. This flow rate will be used in the code as limiting flow rate for water in the rock fracture. The governing equations are (Fox, 1998)

\[ 0 = \frac{\partial}{\partial t} \int_{V} \rho dV + \int_{A} \rho \bar{v} \cdot dA \]  
(2.11)

\[ 0 = \sum F = \frac{\partial}{\partial t} \int_{V} \bar{v} \rho dV + \int_{A} \bar{v} \rho \bar{v} \cdot dA \]  
(2.12)

where \( V \) is the volume. The wall shear stress is given by

\[ \tau_{xz} = \mu \frac{du}{dz} \]  
(2.13)

where \( x \) is in the direction of the fracture, \( z \) is perpendicular to the fracture, \( \mu \) is the viscosity of water, and \( u \) is the velocity in the \( x \) direction. The above equations can be solved yielding
where $b$ is the fracture width and $g$ is gravity. Taking $\frac{dp}{dx} = 0$ and integrating over area, the average velocity is given by

$$\bar{u} = \frac{\rho g}{12 \mu} b^2$$

Equation (2.15) gives the average hydrostatic flow within the fracture. This will be used as an upper bound for the fracture flow.

The governing equations for porous media flow in an isotropic homogeneous media based on conservation of mass is given by
\[
\frac{\partial}{\partial t} \left( \rho \varepsilon dxdydz + S \rho_b dxdydz \right) = \left( ndydx - \frac{\partial n}{\partial z} \frac{dz}{2} dydx + n dydz - \frac{\partial n}{\partial x} \frac{dx}{2} dydz \right) \varepsilon \\
- \left( ndydx - \frac{\partial n}{\partial z} \frac{dz}{2} dydx + n dydz - \frac{\partial n}{\partial x} \frac{dx}{2} dydz \right) \varepsilon
\]  
\tag{2.16}
\]

where \( \rho \) is the mass concentration of the solute in the fluid, \( S = k_d \rho \), \( k_d \) is the partition coefficient, \( \rho_b \) is the bulk density of the porous media, \( \varepsilon \) is the porosity of the porous media, and \( n \) is the mass flux. Equation (2.16) reduces to (Clark, 1996)

\[
\frac{\partial}{\partial t} (\rho \varepsilon + S \rho_b) + \varepsilon \nabla \cdot \vec{n} = 0
\]
\tag{2.17}

Expanding the first term and rearranging terms equation (2.17) yields

\[
\frac{\partial \rho}{\partial t} + \left( \frac{1}{1 + \frac{k_d \rho_b}{\varepsilon}} \right) \varepsilon \nabla \cdot \vec{n} = 0
\]
\tag{2.18}

Letting \( R = 1 + \frac{k_d \rho_b}{\varepsilon} \), the retardation factor, and noting that \( \vec{n} = -D \nabla \rho \) equation (2.18) can be rewritten as

\[
\frac{\partial c}{\partial t} = \frac{D}{R} \nabla^2 c
\]

where \( D \) is the diffusion coefficient and \( c \) is the molar concentration.

The next step is to combine the fracture and porous media flow into a set of coupled differential equations for the idealized one dimensional fracture flow. Using lumped parameters and conservation of mass, the governing equation is given by (Abdel-Salam, 1994)
\[ \frac{\partial c(t,x)}{\partial t} + \frac{2}{b} \frac{\partial \hat{c}(t,x)}{\partial t} = D \frac{\partial^2 c(t,x)}{\partial x^2} - U \frac{\partial c(t,x)}{\partial x} + \frac{2 \varepsilon D_c}{b} \frac{\partial \hat{c}_m}{\partial z} \bigg|_{z=b/2} \]  

(2.19)

where \( c \) is the mass concentration in the fracture, \( \hat{c} \) is the mass concentration deposited on the fracture surface, and \( c_m \) is the mass concentration in the porous media. The deposition onto the fracture surface will be modeled using a lumped irreversible deposition coefficient. Thus, the second term on the left hand side of equation (2.19) can be expressed as

\[ \frac{\partial \hat{c}(t,x)}{\partial t} = \frac{\kappa U}{b} c(t,x) \]  

(2.20)

where \( \kappa \) is the lumped deposition coefficient. The last term on the right hand side of equation (2.19) corresponds to diffusive mass flux into the rock matrix from the fracture. Assuming the rock matrix is fully saturated, the concentration in the rock matrix is given by

\[ \frac{\partial c_m(t,x,z)}{\partial t} + \frac{1}{\varepsilon} \frac{\partial \hat{c}_m(t,x,z)}{\partial t} = D_c \frac{\partial^2 c_m(t,x,z)}{\partial z^2} \]  

(2.21)

where \( \hat{c}_m \) is the concentration deposited in the rock matrix. The deposition onto the rock matrix will be modeled using a lumped irreversible deposition coefficient. Thus, the second term on the left hand side of equation (2.21) can be expressed as

\[ \frac{\partial \hat{c}_m(t,x,z)}{\partial t} = \frac{\kappa_m e}{\rho_b} c_m(t,x,z) \]  

(2.22)

where \( \kappa_m \) is the lumped deposition coefficient.

For the case of constant inlet concentration the analytical solution has been derived (Abdel-Salam, 1994) using Laplace transforms. The analytical solution for concentration in the rock fracture with diffusion into the rock matrix is given by
2.4 Heat Transfer in Porous Media

Heat transfer through saturated porous media includes both heat transfer and fluid flow (Bejan, 1995). The governing energy equation for homogeneous, fully saturated porous media will be presented in two parts. First, the three-dimensional incompressible continuity equation can be expressed as

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{V}) = 0
\]  

(2.24)

where \( \vec{V} \) is the volume averaged velocity vector. The governing equation for the solid phase is given by
\[ \rho \cdot c_s \frac{\partial T}{\partial t} = k_s \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right) + q''_s \] (2.25)

where \( \rho \) is the density of the solid, \( c_s \) is the specific heat of the solid, \( k_s \) is the thermal conductivity of the solid, \( T \) is the temperature, and \( q''_s \) is the internal heat generation.

The energy equation for the fluid is given by

\[ \rho_f c_{pf} \left( \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} \right) = k_f \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right) + \mu \Phi \] (2.26)

where \( \rho_f \) is the density of the fluid, \( c_{pf} \) is the specific heat of the fluid, \( k_f \) is the thermal conductivity of the fluid, \( \mu \) is the viscosity of the fluid, and \( \Phi \) is the viscous dissipation function. The combined thermal conductivity for porous media taking into account the porosity, \( \varepsilon \), is given by

\[ k_a = \varepsilon k_f + (1-\varepsilon) k_s \] (2.27)

and is typically determined experimentally. The combined energy equation can be written as

\[ \left( \rho_f c_{pf} + (1-\varepsilon) \rho_s c_s \right) \frac{\partial T}{\partial t} + \rho_f c_{pf} \nabla \cdot \nabla T = k_a \nabla^2 T + \mu \Phi + q''_s \] (2.28)

By introducing the stream function \( \psi \) for irrotational flow, defined as \( u = \frac{\partial \psi}{\partial y} \) and \( v = -\frac{\partial \psi}{\partial x} \), which satisfies the steady state continuity equation

\[ \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = \frac{\partial^2 \psi}{\partial x \partial y} - \frac{\partial^2 \psi}{\partial y \partial x} = 0 \] (2.29)

Equation (2.28) can be rewritten using the stream function as follows (Oosthuizen, 1999)
\[
\left( \rho_f c_p + (1 - \varepsilon) \rho_s c_s \right) \frac{\partial T}{\partial t} + \rho_f c_p \left( \frac{\partial \psi}{\partial y} \frac{\partial T}{\partial \chi} - \frac{\partial \psi}{\partial \chi} \frac{\partial T}{\partial y} \right) = k_a \nabla^2 T + \mu \Phi + q_s \nabla
\] (2.30)

The pressures can be calculated using Darcy’s Law as follows

\[ u = -\frac{K}{\mu} \frac{\partial p}{\partial \chi} \] (2.31)

\[ v = -\frac{K}{\mu} \frac{\partial p}{\partial \gamma} \] (2.32)

where \( K \) is the permeability. The momentum equation is given by

\[ \rho \frac{D\bar{V}}{Dt} = -\nabla p + \mu \nabla^2 \bar{V} \] (2.33)

where \( D \) represents the total derivative and \( p \) is the pressure. The momentum equation can be rewritten using the Boussinesq approximation to account for buoyancy effects

\[ u \frac{\partial u}{\partial \chi} + v \frac{\partial u}{\partial \gamma} = -\frac{1}{\rho} \frac{\partial p}{\partial \chi} + \frac{\mu}{\rho} \left( \frac{\partial^2 u}{\partial \chi^2} + \frac{\partial^2 u}{\partial \gamma^2} \right) + \beta g (T - T_0) \] (2.34)

where \( \beta = -\frac{1}{\rho} \left( \frac{\partial p}{\partial T} \right)_p \) is the volume expansion coefficient at constant pressure (Bejan, 1995). Combining the Darcy flow model with the Boussinesq approximation the momentum equations in the x and y direction are given by

\[ \frac{\mu u}{K} = -\frac{\partial p}{\partial \chi} + \beta g \rho_f (T - T_0) \cos \theta \] (2.35)

\[ \frac{\mu v}{K} = -\frac{\partial p}{\partial \gamma} + \beta g \rho_f (T - T_0) \sin \theta \] (2.36)

where \( g \) is the gravitational acceleration and \( \theta \) is the inclination angle of the system.
CHAPTER 3

CRITICALITY IN THE REPOSITORY ENVIRONMENT

3.1 Nuclear Theory

The atom is comprised of three basic components, the electron, neutron, and proton. Each atom is designated by its makeup of neutrons (n) and protons (Z), also known as its atomic number. Together these represent the atom mass number, A.

\[ A = n + Z \]  \hspace{1cm} (3.1)

Atoms with differing numbers of neutrons are known as isotopes. An example of an isotopes would be those of natural uranium (Foster, 1983) where the isotopes and their abundances are listed below

\[ ^{234}_{92}U \]  \hspace{1cm} 0.006%

\[ ^{235}_{92}U \]  \hspace{1cm} 0.714%

\[ ^{238}_{92}U \]  \hspace{1cm} 99.28%

The number density (N) gives the number of atoms in 1 m³ of a substance (Shultis, 2002)

\[ N = \frac{\rho A_v}{A_w} \]  \hspace{1cm} (3.2)

Here \( \rho \) (kg/m³) is the mass density and \( A_v \) is Avogadro’s number.

The probability that a neutron – nuclear interaction will occur is expressed in terms of the nuclear cross section. The microscopic cross section (\( \sigma \)) can be considered the target area of the atom that is seen by the neutron in units of target area per atom. In
general, cross sections depend on target composition, particle energy, and particle type. The neutron – nuclear interactions described by cross sections are either absorption or scattering interactions. Absorption and scattering cross sections can be expressed as follows

\[ \sigma_a = \sigma_f + \sigma_c \]  

(3.3)

\[ \sigma_s = \sigma_{se} + \sigma_{si} \]  

(3.4)

where \( \sigma_f \) is the fission and \( \sigma_c \) is the radiative capture cross section. Also, \( \sigma_{se} \) is the elastic and \( \sigma_{si} \) is the inelastic scattering cross section. The total cross section (\( \sigma_T \)) is the sum of all of the cross sections.

\[ \sigma_t = \sigma_a + \sigma_s \]  

(3.5)

Microscopic absorption cross sections are temperature dependent and decrease rapidly as temperature rises. Tables of absorption cross sections are compiled for room temperature or thermal neutrons corresponding to 293K. The absorption cross section for most materials in the thermal region decreases as the inverse square root of the kinetic energy and absolute temperature. Thus the absorption cross section at the average neutron velocity can be adjusted for temperature as follows

\[ \sigma'_a = \sigma_a \sqrt{\frac{\pi}{2}} \sqrt{\frac{293}{T}} \]  

(3.6)

where \( \sigma'_{a_{290}} \) is the absorption cross section at 293 K. The “thermal” cross section can be found in tables in most nuclear engineering books. The rate of these interactions are given by

\[ \dot{R} = \sigma IN \]  

(3.7)
where $I$ (neutrons/m$^2$ s) is the neutron beam intensity and $N$ (nuclei/m$^3$) is the density of target atoms. On a volume basis the cross sections are denoted as macroscopic cross sections $\Sigma$ (m$^2$/m$^3$) and are represented as

$$\Sigma = \sigma N$$  \hspace{1cm} (3.8)

The macroscopic cross sections for scattering and absorption can be added to give the total cross section.

Reactor power levels are a function of many factors. The neutron flux is one of the major components, and is given by

$$\phi = vN$$  \hspace{1cm} (3.9)

where $N$ is the neutron density (neutrons/m$^3$) and $v$ is the velocity of the neutrons within the reactor. The infinite multiplication factor, $k_\infty$, gives an indication of reactor criticality. The infinite multiplication factor is the ratio of the number of neutrons, $n'$, at the current time to the number of neutrons, $n$, at a previous time in an infinitely large reactor.

$$k_\infty = \frac{n'}{n}$$  \hspace{1cm} (3.10)

The infinite multiplication factor describes the criticality of a reactor. Neutrons “leak” from the outer area of reactors of finite size. The non-leakage probability, $P_{NL}$, accounts for the neutron leakage from the reactor. The effective multiplication factor is given by

$$k_{eff} = k_\infty P_{NL}$$  \hspace{1cm} (3.11)

An effective multiplication factor of 1 indicates that a reactor is operating at a stable critical condition. A value greater than 1 would indicate that the reactor is super critical. Autocatalytic criticality is a nuclear chain reaction that increases over time due to some
mechanism within the reactor that provides positive reactivity (Kastenberg, 1997). Reactor reactivity is given by

\[ \rho = \frac{k_{\text{eff}} - 1}{k_{\text{eff}}} \]  

(3.12)

and is a measure of the deviation of the reactor from \( k_{\text{eff}} = 1 \).

3.2 Bowman and Venneri Hypothesis

The Bowman and Venneri paper is primarily concerned with the possible criticality of weapons grade plutonium (w-Pu) that may reach a critical configuration after being leached from its original storage location. The focus is concentrated on a spherical geometry of Pu, water, and SiO\(_2\) mixture surrounded by a SiO\(_2\) reflector. SiO\(_2\) is used as a reasonable representation of different types of rock and sandstone, see Table 3.1 (Bowman, 1994).

<table>
<thead>
<tr>
<th>Compound</th>
<th>West Granite</th>
<th>Sandstone</th>
<th>Nevada Alluvium</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO(_2)</td>
<td>73.9</td>
<td>78.3</td>
<td>71.6</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td>14.9</td>
<td>4.8</td>
<td>12.1</td>
</tr>
<tr>
<td>H(_2)O</td>
<td>0.0</td>
<td>0.0</td>
<td>4.0</td>
</tr>
<tr>
<td>K(_2)O</td>
<td>4.5</td>
<td>1.3</td>
<td>3.5</td>
</tr>
<tr>
<td>CaO</td>
<td>3.3</td>
<td>5.5</td>
<td>2.4</td>
</tr>
<tr>
<td>MgO</td>
<td>0.0</td>
<td>1.2</td>
<td>0.0</td>
</tr>
<tr>
<td>FeO</td>
<td>2.0</td>
<td>1.4</td>
<td>0.0</td>
</tr>
<tr>
<td>CO(_2)</td>
<td>0.0</td>
<td>5.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>

Criticality curves were produced for spheres of radii of 25, 50, 100, and 200 cm and different concentrations of Pu, water, and SiO\(_2\) using the MCNP code. These curves were used to study possible feedback mechanisms and their outcomes. It is assumed that
the initial accumulation of fissile material is sub-critical, and over time changes in the amount or configuration of fissile material and concentrations of SiO₂ and H₂O cause the material to possibly go critical. Once the material is in a critical configuration the positive and negative feedback of the system are explored. Six cases of possible feedback were studied, including: migration of TFM to wet rock (with positive and negative feedback), reduction of moisture content of TFM deposits in rock (with positive feedback), and small volume systems (with positive feedback). The concerning case is the overmoderated case, where the TFM is of sufficient concentration such that as the rock moisture content decreases the neutron flux increases.

3.3 Analytic Slab Reactor Analysis

The neutron population within a reactor determines the rate of fission and the subsequent generation of heat. Neutron cross sections are used to determine the interaction rates between these particles and the atoms of uranium, plutonium, and other materials within the reactor. The transient behavior can be modeled through the time dependent neutron transport equation. The one speed, time dependent transport equation is given by

\[ \frac{1}{\nu} \frac{\partial \phi}{\partial t} - D \nabla^2 \phi + \Sigma_a \phi = S \]  

(3.13)

where

\[ S = \nu \Sigma_f \phi \]  

(3.14)

and \( D \) is the diffusion coefficient and \( \nu \) is the average number of neutrons released per fission. The first term in the equation represents the time rate of change in neutron population. The second term accounts for the diffusion of neutrons from regions of high neutron density to low density. The third term represents the rate of neutron absorption.
by materials within the reactor, and the source term $S$ represents the rate of production of neutrons through nuclear fission. By solving equation (3.13) throughout the reactor, the transient production of neutrons and heat from fission can be modeled. The neutron transport equation is difficult or impossible to solve and finite differences are often applied in its solution of reactor geometries.

The steady state form for equation (3.14) is given by

$$D \nabla^2 \phi - \Sigma_e \phi + S = 0$$

(3.15)

This equation can be solved for simple geometries and the eigenfuctions of this equation represent the steady neutron flux distributions that are possible while the eigenvalues represent the different modes of steady performance. The first eigenvalue, or fundamental solution, represents the long term steady solution for a stable nuclear reactor. By solving equation (3.15) for a particular reactor and given boundary conditions, the geometry of the reactor that will lead to a “critical” or steady state reactor, can be determined.

Although the computer code developed for this work was based on the unsteady neutron transport equation, solutions to equation (3.15) were valuable in determining the validity of the code. A simple reactor geometry analogous to the fracture reactor is a one-dimensional slab reactor show in Figure 3.1.
The governing neutron transport equation (3.15) may be rewritten as

\[ \nabla^2 \phi + B^2 \phi = 0 \]  

(3.16)

where \( B \), “buckling”, is a function of neutron cross sections, neutron escape probabilities, and neutron production rates by fission. The fundamental eigenvalue for the reactor shown in Figure 3.1 is given by the characteristic equation

\[ \cos(Bx) = 0 \]  

(3.17)

producing the first eigenvalue.
\[ B = \frac{\pi}{x_0} \]  

with the corresponding eigenfunction shown in Figure 3.1.

A more relevant case is shown in Figure 3.2, where Zone 1 represents the accumulated fissionable material in the fracture, Zone 2 represents an intermediate zone containing rock and fissionable material, and Zone 3 represents a semi-infinite layer of rock.

![Figure 3.2 Three Zone Slab Reactor](image)

The reactor as shown in Figure 3.2 has the following geometry and material properties:

Zone 1

- 0.12 % by volume \(^{239}\text{Pu}\)
- 99.88% by volume H\(_2\)O
- \(x_1\) to be determined
$^{239}$Pu and H$_2$O mixture has a density of 1.02249 g/cm$^3$

Zone 2

Rock modeled as SiO$_2$ with 30% porosity and a density of 2.2 g/cm$^3$

0.14 % by volume of the porosity of $^{239}$Pu

99.86 % by volume of the porosity of H$_2$O

$x_2 = 20$ cm

SiO$_2$, $^{239}$Pu, and H$_2$O mixture has a density of 2.50787 g/cm$^3$

Zone 3

Rock modeled as SiO$_2$ with 30% porosity and a density of 2.2 g/cm$^3$

The rock is fully saturated with H$_2$O

SiO$_2$ and H$_2$O mixture has a density of 2.5 g/cm$^3$

For the infinite slab as shown in Figure 3.2, the steady state form of equation (3.13) for Zones 1 and 2 can be rewritten as

$$-D \frac{\partial^2 \phi}{\partial x^2} + \sum_a \phi = \nu \sum_j \phi$$

(3.19)

where $\phi$ is a function of $x$. For Zone 3 the steady state diffusion equation is

$$-D \frac{\partial^2 \phi}{\partial x^2} + \sum_a \phi = 0$$

(3.20)

The boundary conditions are as follows

Zone 1

$$\frac{\partial \phi_1}{\partial x} \bigg|_{x=0} = 0$$

(3.21)

$$\phi_1(x) = \phi_2(x)$$

(3.22)

Zone 2
\( \phi_2(x_2) = \phi_3(x_2) \) \hspace{1cm} (3.23)

\[
\frac{\partial \phi_3}{\partial x}igr|_{x=x_1} = \frac{\partial \phi_4}{\partial x}igr|_{x=x_2} \tag{3.24}
\]

Zone 3

\[
\frac{\partial \phi_2}{\partial x}igr|_{x=x_2} = \frac{\partial \phi_4}{\partial x}igr|_{x=x_2} \tag{3.25}
\]

\[\lim_{x \to \infty} \phi_3 = 0 \tag{3.26}\]

Defining

\[B^2 = \frac{\nu \Sigma_f - \Sigma_u}{D} \tag{3.27}\]

and

\[L^2 = \frac{D}{\Sigma_u} \tag{3.28}\]

and noting that \( \phi \) is only a function of \( x \), equations (3.19) and (3.20) can be written as

\[
\frac{d^2 \phi}{dx^2} + B^2 \phi = 0 \tag{3.29}
\]

and

\[
\frac{d^2 \phi}{dx^2} - \frac{1}{L^2} \phi = 0 \tag{3.30}
\]

The ordinary differential equations (3.29) and (3.30) can be solved using the method of separation of variables. The solution of equation (3.29) for Zones 1 and 2 is given by

\[\phi_1(x) = A_1 \cos(B_1x) + A_2 \sin(B_1x) \tag{3.31}\]

\[\phi_2(x) = A_3 \cos(B_2x) + A_4 \sin(B_2x) \tag{3.32}\]

The solution of equation (3.30) is given by
\phi_i(x) = A_4 e^{\frac{x}{x}} + A_6 e^{\frac{x}{\xi}} \tag{3.33}

Equations (3.31) and (3.33) can be reduced by applying the boundary conditions (3.21) and (3.26) giving

\phi_1(x) = A_1 \cos(B_1 x) \tag{3.34}

\phi_2(x) = A_5 e^{\frac{x}{\eta}} \tag{3.35}

Simultaneously solving the remaining boundary conditions applied to (3.32), (3.34), and (3.35) for the constants of integration \( A_3 \), \( A_4 \), and \( A_5 \) gives

\begin{align*}
A_3 &= -\frac{-A_1 B_2 D_2 D_2 LCos(B_2 x_1) Sin(B_2 x_1) - A_1 B_2 D_2 Sin(B_2 x_1) Sin(B_2 x_1) - B_2 D_2 LCos(B_2 x_1) Sin(B_2 x_1) + D_2 Sin(B_2 x_1) Sin(B_2 x_1)}{B_2 D_2 \left[ D_2 Cos(B_2 x_1) Cos(B_2 x_1) + B_2 D_2 LCos(B_2 x_1) Sin(B_2 x_1) - B_2 D_2 LCos(B_2 x_1) Sin(B_2 x_1) + D_2 Sin(B_2 x_1) Sin(B_2 x_1) \right]} \tag{3.36}

A_4 &= \frac{A_1 B_2 D_2 Sin(B_2 x_1) \left( D_2 Cos(B_2 x_1) - B_2 D_2 LCos(B_2 x_1) \right)}{B_2 D_2 \left[ D_2 Cos(B_2 x_1) Cos(B_2 x_1) + B_2 D_2 LCos(B_2 x_1) Sin(B_2 x_1) - B_2 D_2 LCos(B_2 x_1) Sin(B_2 x_1) + D_2 Sin(B_2 x_1) Sin(B_2 x_1) \right]} \tag{3.37}

A_5 &= -\frac{A_1 B_2 D_2 \xi \frac{\xi}{\xi} LSin(B_2 x_1) \left( Cos(B_2 x_1)^2 + Sin(B_2 x_1)^2 \right)}{-D_2 Cos(B_2 x_1) Cos(B_2 x_1) - B_2 D_2 LCos(B_2 x_1) Sin(B_2 x_1) + B_2 D_2 LCos(B_2 x_1) Sin(B_2 x_1) - D_2 Sin(B_2 x_1) Sin(B_2 x_1)} \tag{3.38}
\end{align*}

Substituting (3.36) through (3.38) into (3.22) yields

\begin{align*}
0 &= A_1 \left[ Cos(B_1 x_1) \right. \\
&\left. + B_1 D_2 Sin(B_1 x_1) \left( -B_2 D_2 LCos(B_2 \left( x_1 - x_2 \right)) + D_1 Sin(B_2 \left( x_1 - x_2 \right)) \right) \right]
\nonumber
\end{align*}

\begin{align*}
&\left. + B_1 D_2 \left[ D_2 Cos(B_2 \left( x_1 - x_2 \right)) + B_2 D_2 LCos(B_2 \left( x_1 - x_2 \right)) \right] \right] 
\nonumber
&\tag{3.39}
\end{align*}

To avoid the trivial solution set
\[ \begin{align*} 0 &= \cos(B_1 x_1) \\
&+ \frac{B_2 D_2 \sin(B_1 x_1)(-B_2 D_2 L \cos(B_2 (x_1 - x_2)) + D_3 \sin(B_2 (x_1 - x_2))}{B_2 D_2 \left( D_3 \cos(B_2 (x_1 - x_2)) + B_2 D_2 L \sin(B_2 (x_1 - x_2)) \right)} \quad (3.40) \end{align*} \]

From the material and geometry of the reactor the constants can be determined as

\[ B_1 = 0.124 \quad \text{1/cm} \]
\[ D_1 = 0.335 \quad \text{cm} \]
\[ B_2 = 0.0463 \quad \text{1/cm} \]
\[ D_2 = 0.947 \quad \text{cm} \]
\[ L = 11.23 \quad \text{cm} \]
\[ D_3 = 0.946 \quad \text{cm} \]

From (3.40) it can be determined that the first positive root is at \( x_1 = 2.29 \quad \text{cm} \). By arbitrarily setting \( A_1 = 1 \), the solutions of (3.32), (3.34), and (3.35) can be plotted as shown in Figure 3.3.
3.4 Time Dependent Change in Fissile Content

Fissionable plutonium, Pu-239, decays to U-235, a fissionable isotope of uranium. In the process, Pu-239 emits an energetic alpha particle (helium nucleus). The half life, $t_{1/2}$, of a material is used to determine the rate of decay

$$\frac{dN}{dt} = -\lambda N$$

(3.41)

where $N$ represents the number of nuclei at a given time, $t$. The half life for $^{239}$Pu is $t_{1/2} = 24360$ years and for $^{235}$U is $t_{1/2} = 1.7 \times 10^8$ years. Noting that $\lambda = \frac{\ln(2)}{t_{1/2}}$, the decay constants can be determined as $\lambda_{Pu} = 2.8454 \times 10^{-5}$ yr$^{-1}$ and $\lambda_{U} = 9.7626 \times 10^{-10}$ yr$^{-1}$. The time rate of change for fissile content is given by the coupled equations.
\[\frac{dN_{Pu}}{dt} = -\lambda_{Pu} N_{Pu}\] (3.42)

\[\frac{dN_{U}}{dt} = -\frac{dN_{Pu}}{dt} - \lambda_{U} N_{U}\] (3.43)

The time rate of change for $^{239}$Pu can be solved by direction integration and the time rate of change of $^{235}$U can be solved by use of an integration factor to yield

\[N_{Pu} = N_{0, Pu} e^{-\lambda_{Pu} t}\] (3.44)

\[N_{U} = \frac{\lambda_{Pu} N_{0, Pu}}{\lambda_{U} - \lambda_{Pu}} \left( e^{-\lambda_{Pu} t} - e^{-\lambda_{U} t} \right)\] (3.45)

The decay of $^{239}$Pu to $^{235}$U can be seen in Figure 3.4. Note that by 100000 years most of the $^{239}$Pu has decayed to $^{235}$U. The decay of $^{235}$U is significantly longer as seen in Figure 3.5.
Figure 3.5 Decay of U 235
CHAPTER 4

ANALYSIS TECHNIQUE

4.1 Mass Transport Model

Proposed repositories would employ several different types of containers to hold the myriad of nuclear wastes destined for a nuclear waste repository. In this study, we are concerned about wastes accumulated from the nuclear weapons program that is highly-fissionable plutonium. To contain these wastes, the plutonium is divided into a fine “frit” and dissolved into borosilicate glass similar in composition to natural obsidian. The boron in the glass tends to absorb neutrons and suppresses the ability of the plutonium to reach a critical mass. The glass “logs” are placed in specialized waste containers for long-term storage underground. Over time, these rugged glass logs will be exposed to water which will leach the silicon, boron, plutonium, and other minerals from the glass. Boron is highly mobile when dissolved in water and this important element for the suppression of nuclear criticality will be lost early in the dissolution process of the glass log. Plutonium 239 and its decay product, uranium 235, will eventually dissolve in water and be transported through advection, diffusion, or colloidal transport for collection elsewhere in the geologic formation surrounding the nuclear waste repository.

Dissolution of the glass logs are governed by equations (2.4) through (2.7). As the glass dissolves, radionuclide frit is released. Equation (2.8) is used to determine the mass rate of release of the radionuclides. The released frit is modeled as small spheres
for the purpose of leaching into the surrounding water. As the radionuclides leach into the surrounding water they are transported by the flow of water into the fracture at the bottom of the drift.

The mass transport is governed by equations (2.19) through (2.22). The concentration in the fracture is assumed to be uniformly mixed in the z direction. Thus, equation (2.19) is modeled for the 2-D case in the x direction. The fracture and rock matrix equations are coupled using a linear interpolation. Equation (2.19) is modeled using finite differences given by

\[
\frac{c_{i,j}^{l+1} - c_{i,j}^l}{\Delta t} + \frac{2\kappa U}{b^2} c_{i,j}^{l+1} = D \left( \frac{c_{i+1,j}^{l+1} - 2c_{i,j}^{l+1} + c_{i-1,j}^{l+1}}{(\Delta x)^2} - U \frac{c_{i,j}^{l+1} - c_{i-1,j}^{l+1}}{\Delta x} \right) - \frac{2\theta D_e}{b} \frac{\partial c_m}{\partial z} \bigg|_{z=b/2}
\]

(4.1)

where \( \frac{\partial c_m}{\partial z} \bigg|_{z=b/2} \) is expressed using finite differences as

\[
\frac{\partial c_m}{\partial z} \bigg|_{z=b/2} = \frac{c_{i,j}^{l+1} - c_{i,j}^l}{\Delta z} - \frac{b}{2} \left( \frac{1}{\Delta z} - \frac{1}{4} \right)
\]

(4.2)

Equation (2.21) is modeled using

\[
\frac{c_{m_{i,j}}^{l+1} - c_{m_{i,j}}^l}{\Delta t} + \kappa_m c_{m_{i,j}}^{l+1} = D_e \left( \frac{c_{m_{i+1,j}}^{l+1} - 2c_{m_{i,j}}^{l+1} + c_{m_{i-1,j}}^{l+1}}{(\Delta x)^2} \right) - \frac{D_e}{\Delta z} \left( \frac{c_{m_{i,j+1}}^{l+1} - 2c_{m_{i,j}}^{l+1} + c_{m_{i,j-1}}^{l+1}}{(\Delta z)^2} \right)
\]

(4.3)

See section 2.3 for definition of the coefficients, \( l \) is the current time step and \( l+1 \) is the future time step. The coefficient matrix is solved using Gauss-Seidel method. To save on storage the sky-line method is used to store the non-zero values of the coefficient.
matrix. When a high enough concentration of radionuclides builds up in the fracture and rock matrix to cause \( k_a \) to be greater than 1.3, the mass transport portion of the code is suspended and the combined neutron transport, fluid flow, and heat transfer portion of the code begins to run. Using a cutoff of \( k_a \geq 1.3 \) insured that enough material accumulated in the fracture and surrounding rock to create a critical reactor.

4.2 Combined Model of Neutron Transport, Fluid Flow, and Heat Transfer

The combined neutron transport, fluid flow, and heat transfer equations are solved in a quasi-steady manner. First, the neutron transport equation is solved to find the heat generated by fission at the current time step. The neutron diffusion equation (3.13), using finite differences, can be expressed

\[
\frac{1}{\nu} \frac{\phi_{i,j}^{l+1} - \phi_{i,j}^{l}}{\Delta t} = D \left( \frac{\phi_{i+1,j}^{l+1} - 2\phi_{i,j}^{l+1} + \phi_{i-1,j}^{l+1}}{(\Delta x)^2} + \frac{\phi_{i,j+1}^{l+1} - 2\phi_{i,j}^{l+1} + \phi_{i,j-1}^{l+1}}{(\Delta y)^2} \right) - \Sigma_\sigma \phi_{i,j}^{l+1} + S \tag{4.4}
\]

See section 3.3 for definitions of the coefficients, \( l \) is the current time step and \( l+1 \) is the future time step. Once the neutron flux has been solved the energy equation is solved taking into account the energy generated by fission. Using finite differences the energy equation is given by

\[
\frac{T_{i,j}^{l+1} - T_{i,j}^{l}}{\Delta t} = \frac{k_x}{\rho c_p} \left( \frac{T_{i+1,j}^{l+1} - 2T_{i,j}^{l+1} + T_{i-1,j}^{l+1}}{(\Delta x)^2} + \frac{T_{i,j+1}^{l+1} - 2T_{i,j}^{l+1} + T_{i,j-1}^{l+1}}{(\Delta y)^2} \right) \\
+ \frac{1}{\rho c_p} q - \frac{\psi_{i,j+1}^{l+1} - \psi_{i,j-1}^{l+1}}{2\Delta y} \frac{T_{i+1,j}^{l+1} - T_{i-1,j}^{l+1}}{2\Delta x} \\
+ \frac{\psi_{i+1,j}^{l+1} - \psi_{i-1,j}^{l+1}}{2\Delta x} \frac{T_{i,j+1}^{l+1} - T_{i,j-1}^{l+1}}{2\Delta y} \tag{4.5}
\]
where the coefficients are defined in section 2.4. The new temperature values are then used to compute the stream functions. Equations (2.35) and (2.36) can be combined, and using finite differences the stream function can be written as

\[
\psi_i^{t+1} - 2\psi_i^{t+1} + \psi_{i-1,j}^{t+1} + \psi_{i,j}^{t+1} - 2\psi_{i,j-1}^{t+1} + \psi_{i,j}^{t+1} = \frac{\beta g \rho_f K}{\mu} \left( \frac{T_{i,j+1}^{t+1} - T_{i,j-1}^{t+1}}{2\Delta y} \cos \theta - \frac{T_{i+1,j}^{t+1} - T_{i-1,j}^{t+1}}{2\Delta x} \sin \theta \right) \tag{4.6}
\]

where the coefficients are defined in section 2.4. The velocities and pressures are then calculated from equation (4.6). Equations (4.4) through (4.6) are solved using the successive over-relaxation (SOR) method.

4.3 Mass Transport Code Validation

The mass of plutonium in the glass log is given by equation (2.8) and is taken to be 201.5 kg. The code calculates that the radionuclide mass released from the glass log is 201.150 kg over 3.5 million years and can be seen in Figure 4.1.
The radionuclide concentration from the code is plotted along with the analytic solution for time at 10,000 years and is shown in Figure 4.2. The maximum normalized error for this case is 0.77%.
The number of nodes is increased from 400 to 160,000 to verify the numerical accuracy of the code. If too few nodes are used the solution will not represent the physical problem and too many nodes will introduce round off error or take excessive computational time. This phenomenon can be seen in Figure 4.3. Similar results are seen in Figure 4.4 as the time step is scaled by 0.5 to 50,000.
Figure 4.3 Maximum Normalized Concentration Error by Number of Nodes

Figure 4.4 Maximum Normalized Concentration Error by Time Step
4.4 Transient Neutron Flux Code Validation

Currently there is a lack of data for geological reactors. Some data exists from the Oklo reactor site in Gabon, Africa which operated 2 billion years ago. Unfortunately, the data collected reflects conditions after the reactor quit operating. The data does not show how the reactor operated or what the neutron flux levels were during operation. Experimental setups to study geological reactors would be impractical and expensive. To validate the code, numerical results for a 2-D transient neutron flux on a 1m by 1m domain will be compared with the analytical results for the same geometry. For this case, the initial condition will be a flux of 100 \( \text{(neutrons/m}^2\text{s)} \) across the geometry. The diffusion coefficient will be set equal to 0.001 \( \text{(m}^2\text{/s)} \) and \( \Delta x \) and \( \Delta y \) equal to 0.1m. The neutron flux will be set to 0 \( \text{(neutrons/m}^2\text{s)} \) on all boundaries as seen in Figure 4.5.

\[
\phi(x, y, 0) = 100 \text{ n/m}^2\text{s}
\]

\[
\phi(0, y, t) = 0 \text{ n/m}^2\text{s}
\]

\[
\phi(x, 0, t) = 0 \text{ n/m}^2\text{s}
\]

\[
\phi(x, 1, t) = 0 \text{ n/m}^2\text{s}
\]

\[
\phi(1, y, t) = 0 \text{ n/m}^2\text{s}
\]

\[
\phi(x, y, 0) = 100 \text{ n/m}^2\text{s}
\]

\[
\phi(0, y, t) = 0 \text{ n/m}^2\text{s}
\]

\[
\phi(x, 0, t) = 0 \text{ n/m}^2\text{s}
\]

\[
\phi(x, 1, t) = 0 \text{ n/m}^2\text{s}
\]

\[
\phi(1, y, t) = 0 \text{ n/m}^2\text{s}
\]

Figure 4.5 2-D Transient Neutron Flux Geometry
The analytical solution will be derived from the time dependent neutron flux equation with no source or absorption terms, given by

\[ \frac{1}{v} \frac{\partial \phi}{\partial t} - D \nabla^2 \phi = 0 \]  

(4.7)

To solve equation (4.7) a solution of the following form is assumed

\[ \phi(x, y, t) = h(t) \omega(x, y) \]  

(4.8)

Substituting (4.8) into (4.7) and setting the result equal to a constant, yields the following

\[ \frac{1}{D} \frac{h'(t)}{h(t)} = \frac{1}{\omega} \nabla^2 \omega(x, y) = -\lambda \]  

(4.9)

Equation (4.9) can now be separated into two equations as follows

\[ h'(t) = -\lambda D h(t) \]  

(4.10)

\[ \nabla^2 \omega + \lambda \omega = 0 \]  

(4.11)

The solution to (4.10) is given by

\[ h(t) = A e^{-\lambda D t} \]  

(4.12)

Equation (4.11), known as the Helmholtz equation, results in

\[ \omega(x) = A_2 \sin \left( \frac{n \pi x}{a} \right) \]  

(4.13)

\[ \omega(y) = A_3 \sin \left( \frac{n \pi y}{b} \right) \]  

(4.14)

\[ \lambda_{nm} = \left( \frac{n \pi}{a} \right)^2 + \left( \frac{m \pi}{b} \right)^2, n = 1, 2, 3 \ldots; m = 1, 2, 3 \ldots \]  

(4.15)
where \( a \) and \( b \) are the lengths in the x and y directions respectively. Substituting (4.12) through (4.15) into (4.8) yields

\[
\phi(x, y, t) = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} B_{nm} e^{\frac{(n \pi)^2 + (m \pi)^2}{x^2 y^2}} e^{-\frac{n \pi x}{a} \sin \left( \frac{m \pi y}{b} \right)}
\]  \hspace{1cm} (4.16)

The initial conditions are used to determine the coefficients of \( B_{nm} \) from the double Fourier series as follows

\[
B_{nm} = \frac{4}{ab} \int_0^a \int_0^b \phi_0 \sin \left( \frac{m \pi y}{b} \right) \sin \left( \frac{n \pi x}{a} \right) dy dx
\]  \hspace{1cm} (4.17)

Integrating (4.17) gives the 2-D transient flux as

\[
\phi(x, y, t) = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} \frac{4 \phi_0}{ab} \left( a - a \cos \left( \frac{n \pi}{a} \right) \right) \left( b - b \cos \left( \frac{m \pi}{b} \right) \right) e^{\frac{(n \pi)^2 + (m \pi)^2}{x^2 y^2}} e^{-\frac{n \pi x}{a} \sin \left( \frac{m \pi y}{b} \right)}
\]  \hspace{1cm} (4.18)

The analytic flux at \( t = 10 \) s for a 1m by 1m domain with \( \Delta x \) and \( \Delta y \) equal to 0.1m is given in Table 4.1

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<th>0.00</th>
<th>0.00</th>
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<td>51.81</td>
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</tr>
<tr>
<td>( x ) (m)</td>
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<tr>
<td>( y ) (m)</td>
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<td>50.29</td>
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<td>96.16</td>
<td>93.34</td>
<td>81.41</td>
<td>50.29</td>
</tr>
</tbody>
</table>
The 3-D plot of the analytical solution is seen in Figure 4.6.

![Analytical 2-D Neutron Flux](image)

**Figure 4.6 Analytical 2-D Neutron Flux Solution at t=10s**

The numerical results from the code for the same problems are given in Table 4.2.

<table>
<thead>
<tr>
<th>Flux (n/m^2-s)</th>
<th>0.00-10.00</th>
<th>10.00-20.00</th>
<th>20.00-30.00</th>
<th>30.00-40.00</th>
<th>40.00-50.00</th>
<th>50.00-60.00</th>
<th>60.00-70.00</th>
<th>70.00-80.00</th>
<th>80.00-90.00</th>
<th>90.00-100.00</th>
</tr>
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<tbody>
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</tr>
<tr>
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<td>46.72</td>
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<td>92.03</td>
<td>90.23</td>
<td>81.20</td>
<td>53.79</td>
<td>0.00</td>
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<td>94.96</td>
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<td>97.94</td>
<td>97.94</td>
<td>97.94</td>
<td>96.01</td>
<td>86.36</td>
<td>57.17</td>
<td>0.00</td>
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</table>
The 3-D plot of the numerical solution is seen in Figure 4.7.

The numeric and analytic solutions are in good agreement in the center of the domain and, as expected, the error increases slightly towards the boundary. The average error for the domain is 1.34%.
4.5 Steady State Heat Conduction Code Validation

The numerical results for 2-D steady state conduction on a 1m by 1m domain will be compared with the analytical results for the same geometry. For this case the boundary conditions can be seen in Figure 4.8. The step sizes will be $\Delta x$ and $\Delta y$ equal to 0.1m.

![Figure 4.8 2-D Steady State Conduction Geometry](image)

The analytical solution will be derived from the steady state conduction equation given by

$$\nabla^2 T = 0$$

(4.19)
The following transformation will be employed to simplify the solution

\[ \zeta = \frac{T - T_c}{T_h - T_c} \]

(4.20)

Equation (4.20) transforms the boundary conditions into \( \zeta(0, y) = 0, \zeta(x, 0) = 0, \zeta(a, y) = 0, \) and \( \zeta(x, b) = 1 \) where \( a \) is the length in the \( x \) direction and \( b \) is the length in the \( y \) direction. To solve equation (4.19) a solution of the following form is assumed

\[ \zeta(x, y) = X(x)Y(y) \]

(4.21)

Substituting equations (4.20) and (4.21) into equation (4.19) and setting the result equal to a constant yields

\[ -\frac{1}{X} \frac{d^2 X}{dx^2} = \frac{1}{Y} \frac{d^2 Y}{dy^2} = \lambda^2 \]

(4.22)

Equation (4.22) can now be separated into two equations as follows

\[ \frac{d^2 X}{dx^2} + \lambda^2 X = 0 \]

(4.23)

\[ \frac{d^2 Y}{dy^2} - \lambda^2 Y = 0 \]

(4.24)

The general solution to equations (4.23) and (4.24) are as follows

\[ X = A_1 \cos(\lambda x) + A_2 \sin(\lambda x) \]

(4.25)

\[ Y = A_4 e^{-\lambda y} + A_4 e^{\lambda y} \]

(4.26)

Substituting equations (4.25) and (4.26) into (4.21) give the general solution as

\[ \zeta(x, y) = \left( A_1 \cos(\lambda x) + A_2 \sin(\lambda x) \right) \left( A_4 e^{-\lambda y} + A_4 e^{\lambda y} \right) \]

(4.27)

Using the homogenous transformed boundary conditions (4.27) can be written as
\[ \zeta(x, y) = \sum_{n=1}^{\infty} A_n \sin \left( \frac{n\pi x}{a} \right) \sinh \left( \frac{n\pi y}{a} \right) \] (4.28)

Using the non-homogenous boundary condition \( A_n \) can be solved for and the steady state solution is given by

\[ \zeta(x, y) = 2 \sum_{n=1}^{\infty} \frac{(-1)^{n+1} \sin \left( \frac{n\pi x}{a} \right)}{n} \frac{\sinh \left( \frac{n\pi y}{a} \right)}{\sinh \left( \frac{n\pi b}{a} \right)} \] (4.29)

where the temperature \( T \) is obtained from (4.20).

The analytic temperature distribution for a 1m by 1m domain with \( \Delta x \) and \( \Delta y \) equal to 0.1m is given in Table 4.3.

<table>
<thead>
<tr>
<th>x</th>
<th>y</th>
<th>Temperature (K)</th>
</tr>
</thead>
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<td>293.00</td>
</tr>
<tr>
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</table>

The 3-D plot of the analytical solution is seen in Figure 4.9.
The numerical results from the code for the same problem are given in Table 4.4.

Table 4.4 Numerical Steady State Temperature (K) Distribution

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<td>293.15</td>
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</tbody>
</table>
The 3-D plot of the numerical solution is seen in Figure 4.10.

![Numerical 2-D SS Conduction Solution](image)

**Figure 4.10 2-D Steady State Temperature Distribution of the Numerical Solution**

The numerical and analytical solutions are in good agreement for the entire domain. The average error for the domain is -0.47%.

4.6 Steady State Stream Function Code Validation

The numerical results for 2-D steady state stream function on a 1m by 1m domain will be compared with the analytical results for the same geometry. For this case the boundary conditions can be seen in Figure 4.11. The step sizes will be Δx and Δy equal to 0.1m.
The analytical solution will be derived from the steady state stream equation given by
\[ \nabla^2 \psi = 0 \]  \hspace{1cm} \text{(4.30)}

The following transformation will be employed to simplify the solution
\[ \zeta = \frac{\psi - \psi_1}{\psi_2 - \psi_1} \] \hspace{1cm} \text{(4.31)}

Equation (4.31) transforms the boundary conditions into \( \zeta(0, y) = 0, \zeta(x, 0) = 0, \zeta(a, y) = 0, \) and \( \zeta(x, b) = 1 \) where \( a \) is the length in the \( x \) direction and \( b \) is the length in

---

**Figure 4.11 2-D Steady State Stream Function Geometry**

\[ \psi(x, 1) = 5.5 \times 10^{-6} \frac{m^2}{s} \]

\[ \psi(0, y) = 0 \frac{m^2}{s} \]

\[ \psi(x, 0) = 0 \frac{m^2}{s} \]

\[ \psi(1, y) = 0 \frac{m^2}{s} \]
the y direction. The derivation of the solution to equation (4.30) is the same as in section 4.5 and is given by

$$
\zeta(x, y) = \frac{2}{\pi} \sum_{n=1}^{\infty} \left(\frac{-1}{n} + 1\right) \sin \left(\frac{n \pi x}{a}\right) \frac{\sinh \left(\frac{n \pi y}{a}\right)}{\sinh \left(\frac{n \pi b}{a}\right)}
$$

(4.32)

where the stream function, \(\psi\), is obtained from (4.31).

The analytic stream function distribution for a 1m by 1m domain with \(\Delta x\) and \(\Delta y\) equal to 0.1m is given in Table 4.5.

<table>
<thead>
<tr>
<th>Table 4.5 Analytic Steady State Stream Function (m²/s) Distribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00E+00 0.00E+00 0.00E+00 0.00E+00 0.00E+00 0.00E+00 0.00E+00 0.00E+00 0.00E+00 0.00E+00</td>
</tr>
<tr>
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</tr>
<tr>
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<tr>
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</table>

The 3-D plot of the analytical solution is seen in Figure 4.12.
The numerical results from the code for the same problem are given in Table 4.6.

Table 4.6 Numerical Steady State Stream Function (m\(^2\)/s) Distribution

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The 3-D plot of the numerical solution is seen in Figure 4.13.

![Numerical 2-D SS Stream Function Solution](image)

**Figure 4.13 2-D Steady State Stream Function Distribution of the Numerical Solution**

The numerical and analytical solutions are in good agreement for the entire domain. The average normalized error for the domain is -1.28%.
CHAPTER 5

RESULTS

5.1 Conditions that Lead to Criticality in a Fissure Reactor

The geometry for the current study can be seen in Figure 2.1. The area of interest is the fracture and rock matrix below the drift. For the purposes of simulation an area of 20m wide by 10m deep centered about the fracture is considered. Typical fracture spacing in tuff ranges from 0.6m to 4.6m (Hidna, 2003), yet fractures that can support significant flow with average widths greater than 1 mm typically have larger spacing (Seol, 2005). The area is discretized using 29 nodes in the width and 19 nodes in the depth. The temperature boundary condition is set at 20 degrees Celsius. The pressure boundary condition assumes 1 atmosphere with 3 inches of head pressure at the top of the fracture. The rock matrix has a porosity of 30%.

The rock matrix is modeled using primarily SiO$_2$ and Al$_2$O$_2$, with a porosity of approximately 30% containing water, see Table 3.1. In order to get a critical event enough fissionable material must accumulate to sustain a chain reaction. Thus, to have a critical event in the rock fracture or matrix, material must migrate from the drift containing the waste package. This migration of material is dependent on the solubility of the radionuclides and the velocity of the flow of water in the rock fracture. If the solubility or flow rate is too low, material will not migrate into the rock fracture or matrix. For the purpose of this study the flow rate in the rock fracture was fixed at 1m/yr.
(Ahn, 1997) and other parameters were varied. To achieve radionuclide migration with the given flow rate, the solubility was varied from 0.239 to 0.0239 (kg/m³). The higher solubility allows for 116 kg of nuclear material to migrate in 221 thousand years, while the lower solubility migrates 130 kg of nuclear material in 2.3 million years. If the solubility or flow rate is increased more material will migrate into the fracture and rock matrix.

If the material is dispersed throughout the rock matrix then a critical mass will not be achieved. Likewise, if the fissionable material is concentrated in a narrow fracture the neutrons will escape without causing a fission event. In order to achieve critical conditions the diffusion and deposition coefficients were varied. The diffusion coefficient in the rock fracture was varied from 7.92x10⁻⁷ to 7.92x10⁻⁹ (m²/s), while the diffusion coefficient in the rock matrix was varied 3.23x10⁻¹² to 3.23x10⁻¹³ (m²/s). The deposition coefficient for the rock fracture was varied from 1.0x10⁻⁸ to 1.0x10⁻¹⁰ (m). The deposition coefficient for the rock matrix was maintained at 0.01 (1/s). These combinations of parameters allowed for different amounts of nuclides to build up on the fracture surface and in the rock matrix. Each of the combinations resulted in different criticality events.

5.2 Results of Heat Transfer, Neutron Flux, and Fluid Flow in a Critical Fissure Reactor

Four simulations were run using different parameters as described in section 5.1. The mass transport part of the code was run until $k_n > 1.3$ at which time the mass transport was turned off and the neutronics section of the code began. The time for the neutronics started when the mass transport portion of the code stopped. The neutronics were run for 30 hours of reactor time and took 5.9 days to complete. In each simulation
the Reynolds number (Re) was checked for and found to be well below 1 implying the Darcy flow model is valid (Bejan, 1995).

The first simulation was run using a solubility of 0.239 (kg/m³). The diffusion coefficient in the rock fracture was 7.92x10⁻⁷ (m²/s), while the diffusion coefficient in the rock matrix was 3.23x10⁻¹² (m²/s). The deposition coefficient for the rock fracture was 1.0x10⁻⁸ (m). The concentration of radionuclides for 3 nodes, center of fracture, fracture wall, and the first node in the rock matrix at a depth of 5m below the drift are shown in Figure 5.1 through Figure 5.3, respectively. The neutron flux distribution for time equal to 36000s, 7.22x10⁴s, and 1.08x10⁵ can be seen in Figure 5.4 through Figure 5.6, respectively. The temperature distribution for time equal to 36000s, 7.22x10⁴s, and 1.08x10⁵s is displayed in Figure 5.7 through Figure 5.9, respectively. Finally, the pressure distribution for time equal to 36000s, 7.22x10⁴s, and 1.08x10⁵ are shown in Figure 5.10 through Figure 5.12, respectively.
Figure 5.1 Run 1 Nuclide Concentration Center of Fracture Node

Figure 5.2 Run 1 Nuclide Concentration Fracture Wall Node
Figure 5.3 Run 1 Nuclide Concentration First Interior Node from Fracture

Figure 5.4 Run 1 Neutron Flux at 36100 Seconds
Figure 5.5 Run 1 Neutron Flux at 72200 Seconds

Figure 5.6 Run 1 Neutron Flux at 108000 Seconds
Figure 5.7 Run 1 Temperature at 36100 Seconds

Figure 5.8 Run 1 Temperature at 72200 Seconds
Figure 5.9 Run 1 Temperature at 108000 Seconds

Figure 5.10 Run 1 Pressure at 36100 Seconds
Figure 5.11 Run 1 Pressure at 72200 Seconds

Figure 5.12 Run 1 Pressure at 108000 Seconds
The second simulation was run using a solubility of 0.0239(kg/m³). The diffusion coefficient in the rock fracture was 7.92x10⁻⁷(m²/s), while the diffusion coefficient in the rock matrix was 3.23x10⁻¹²(m²/s). The deposition coefficient for the rock fracture was 1.0x10⁻⁸(m). The concentration of radionuclides for 3 nodes, center of fracture, fracture wall, and the first node in the rock matrix at 5m below the drift are shown in Figure 5.13 through Figure 5.15, respectively. The neutron flux distribution for time equal to 36000s, 7.22x10⁴s, and 1.08x10⁵ can be seen in Figure 5.16 through Figure 5.18, respectively. The temperature distribution for time equal to 36000s, 7.22x10⁴s, and 1.08x10⁵ are displayed in Figure 5.19 through Figure 5.21, respectively. Finally, the pressure distribution for time equal to 36000s, 7.22x10⁴s, and 1.08x10⁵ are shown in Figure 5.22 through Figure 5.24, respectively.
Figure 5.13 Run 2 Nuclide Concentration Center Node

Figure 5.14 Run 2 Nuclide Concentration Fracture Wall Node
Figure 5.15 Run 2 Nuclide Concentration First Interior Node from Fracture

Figure 5.16 Run 2 Neutron Flux at 36100 Seconds
Figure 5.17 Run 2 Neutron Flux at 72200 Seconds

Figure 5.18 Run 2 Neutron Flux at 108000 Seconds
Figure 5.19 Run 2 Temperature at 36100 Seconds

Figure 5.20 Run 2 Temperature at 72200 Seconds
Figure 5.21 Run 2 Temperature at 108000 Seconds

Figure 5.22 Run 2 Pressure at 36100 Seconds
Figure 5.23 Run 2 Pressure at 72200 Seconds

Figure 5.24 Run 2 Pressure at 108000 Seconds
The third simulation was run using a solubility of 0.239 (kg/m³). The diffusion coefficient in the rock fracture was 7.92x10⁻⁷ (m²/s), while the diffusion coefficient in the rock matrix was 3.23x10⁻¹¹ (m²/s). The deposition coefficient for the rock fracture was 1.0x10⁻⁸ (m). The concentration of radionuclides for 3 nodes, center of fracture, fracture wall, and the first node in the rock matrix at 5 m below the drift are shown in Figure 5.25 through Figure 5.27, respectively. The neutron flux distribution for time equal to 36000s, 7.22x10⁴s, and 1.08x10⁵ can be seen in Figure 5.28 through Figure 5.30, respectively. The temperature distribution for time equal to 36000s, 7.22x10⁴s, and 1.08x10⁵ are displayed in Figure 5.31 through Figure 5.33, respectively. Finally, the pressure distribution for time equal to 36000s, 7.22x10⁴s, and 1.08x10⁵ are shown in Figure 5.34 through Figure 5.36, respectively.
Figure 5.25 Run 3 Nuclide Concentration Center Node

Figure 5.26 Run 3 Nuclide Concentration Fracture Wall Node
Figure 5.27 Run 3 Nuclide Concentration First Interior Node from Fracture

Figure 5.28 Run 3 Neutron Flux at 36100 Seconds
Figure 5.29 Run 3 Neutron Flux at 72200 Seconds

Figure 5.30 Run 3 Neutron Flux at 108000 Seconds
Figure 5.31 Run 3 Temperature at 36100 Seconds

Figure 5.32 Run 3 Temperature at 72200 Seconds
Figure 5.33 Run 3 Temperature at 108000 Seconds

Figure 5.34 Run 3 Pressure at 36100 Seconds
Figure 5.35 Run 3 Pressure at 72200 Seconds

Figure 5.36 Run 3 Pressure at 108000 Seconds
The fourth simulation was run using a solubility of 0.0239 (kg/m³). The diffusion coefficient in the rock fracture was 7.92 \times 10^{-7} (m²/s), while the diffusion coefficient in the rock matrix was 3.23 \times 10^{-11} (m²/s). The deposition coefficient for the rock fracture was 1.0 \times 10^{-9} (m). The concentration of radionuclides for 3 nodes, center of fracture, fracture wall, and the first node in the rock matrix at 5m below the drift are shown in Figure 5.37 through Figure 5.39, respectively. The neutron flux distribution for time equal to 36000s, 7.22 \times 10^4 s, and 1.08 \times 10^5 can be seen in Figure 5.40 through Figure 5.42, respectively. The temperature distribution for time equal to 36000s, 7.22 \times 10^4 s, and 1.08 \times 10^5 are displayed in Figure 5.43 through Figure 5.48, respectively. Finally, the pressure distribution for time equal to 36000s, 7.22 \times 10^4 s, and 1.08 \times 10^5 are shown in Figure 5.46 through Figure 5.48, respectively.
Figure 5.37 Run 4 Nuclide Concentration Center Node

Figure 5.38 Run 4 Nuclide Concentration Fracture Wall Node
Figure 5.39 Run 4 Nuclide Concentration First Interior Node from Fracture

Figure 5.40 Run 4 Neutron Flux at 36100 Seconds
Figure 5.41 Run 4 Neutron Flux at 72200 Seconds

Figure 5.42 Run 4 Neutron Flux at 108000 Seconds
Figure 5.43 Run 4 Temperature at 36100 Seconds

Figure 5.44 Run 4 Temperature at 72200 Seconds
Figure 5.45 Run 4 Temperature at 108000 Seconds

Figure 5.46 Run 4 Pressure at 36100 Seconds
Figure 5.47 Run 4 Pressure at 72200 Seconds

Figure 5.48 Run 4 Pressure at 108000 Seconds
The four simulations conducted for this paper show slightly different accumulations of radionuclides within the fracture and rock matrix. Each of the cases led to a different neutron flux distribution within the reactor. However, for each case the highest neutron flux was within the fracture. This is to be expected since the fracture and fracture walls have the highest concentration of nuclides within the reactor. The high neutron flux in the fracture drives the temperature within the reactor. The temperature is highest in the fracture and drops to initial temperature, 293K, a short distance from the fracture due to the thermal mass associated with the rock. The elevated temperature creates high pressure within the fracture and rock matrix leading to fluid flow within the porous media. The fluid flow is transient and leads to a bifurcating high and low pressures within the rock matrix.

5.3 Results for an Overmoderated Fissure Reactor

Bowman and Venneri contended that there may be cases where material leached into the rock matrix may be overmoderated. In the case of overmoderation, the moderator, typically water, absorbs enough neutrons to prevent a criticality case. Bowman and Venneri hypothesized that as the rock matrix dries out there may be a criticality event due to overmoderation. This drying out of the rock matrix may result from a number of scenarios; chief among these are the receding of groundwater and ejection of water from the rock due to heat generated from nuclear fission. For the purpose of this study the case where water dries out in the fracture due to the groundwater receding was evaluated. To simulate the drying out, the water concentrations of the nodes in the center of the fracture were set to zero at time equal to
1800 seconds. Every 1800 seconds thereafter the water concentration of the wall nodes were set to zero starting with the top nodes, as shown in Figure 5.49.

Three simulations were run using the parameters described in section 5.1. The mass transport part of the code was run until different $k_\infty$ values were reached, at which
time the mass transport was turned off and the neutronics section of the code began. The neutronics were run for 30 hours of reactor time and took 5.9 days to complete. In each of the simulations the Reynolds number (Re) was checked for and found to be well below 1, implying that the Darcy flow model is valid (Bejan, 1995).

The first simulation was run using a $k_c$ cutoff of 0.7 and a solubility of 0.239 (kg/m$^3$). A $k_c$ cutoff of 0.7 was determined to yield a slightly overmoderated case. The diffusion coefficient in the rock fracture was 7.92x10$^{-7}$ (m$^2$/s), while the diffusion coefficient in the rock matrix was 3.23x10$^{-12}$ (m$^2$/s). The deposition coefficient for the rock fracture was 1.0x10$^{-8}$ (m). The concentration of radionuclides for 3 nodes, center of fracture, fracture wall, and the first node in the rock matrix at 1.1m below the drift are shown in Figure 5.50 though Figure 5.52, respectively. The neutron flux for the center node at 1.1 m below the drift can be seen in Figure 5.53. The temperature for the center node at 1.1 m below the drift is displayed in Figure 5.54.
Figure 5.50 Run 5a Nuclide Concentration Center Node

Figure 5.51 Run 5a Nuclide Concentration Fracture Wall Node
Figure 5.52 Run 5a Nuclide Concentration First Interior Node

Figure 5.53 Run 5a Neutron Flux
The second simulation was run using a $k_{w}$ cutoff of 0.5 and a solubility of 0.239(kg/m$^3$). The diffusion coefficient in the rock fracture was $7.92 \times 10^{-7}$(m$^2$/s), while the diffusion coefficient in the rock matrix was $3.23 \times 10^{-12}$(m$^2$/s). The deposition coefficient for the rock fracture was $1.0 \times 10^{-8}$(m). The concentration of radionuclides for 3 nodes, center of fracture, fracture wall, and the first node in the rock matrix at 1.1m below the drift are shown in Figure 5.55 though Figure 5.57, respectively. The neutron flux for the center node at 1.1 m below the drift can be seen in Figure 5.58. The temperature for the center node at 1.1 m below the drift is displayed in Figure 5.59.
Figure 5.55 Run 5b Nuclide Concentration Center Node

Figure 5.56 Run 5b Nuclide Concentration Fracture Wall Node
Figure 5.57 Run 5b Nuclide Concentration First Interior Node

Figure 5.58 Run 5b Neutron Flux
The third simulation was run using a $k_{\infty}$ cutoff of 0.8 and a solubility of 0.239(kg/m³). The diffusion coefficient in the rock fracture was $7.92 \times 10^{-7}$(m²/s), while the diffusion coefficient in the rock matrix was $3.23 \times 10^{-12}$(m²/s). The deposition coefficient for the rock fracture was $1.0 \times 10^{-8}$(m). The concentration of radionuclides for 3 nodes, center of fracture, fracture wall, and the first node in the rock matrix at 1.1m below the drift are shown in Figure 5.60 though Figure 5.62, respectively. The neutron flux for the center node at 1.1 m below the drift can be seen in Figure 5.63. The temperature for the center node at 1.1 m below the drift is displayed in Figure 5.64.
Figure 5.60 Run 5c Nuclide Concentration Center Node

Figure 5.61 Run 5c Nuclide Concentration Fracture Wall Node
Figure 5.62 Run 5c Nuclide Concentration First Interior Node

Figure 5.63 Run 5c Neutron Flux
Three cases were studied for potential overmoderation using cutoffs for $k_\infty$ of 0.5, 0.7, and 0.8. Using these cutoffs led to three different overmoderated cases. For the cutoff of 0.5, there was an increase in neutron flux as the fracture dried out. This increase in neutron flux was not significant enough to raise the temperature of the surrounding rock. In the case where the cutoff was 0.7 there was a considerable increase in neutron flux as the fracture dried out. The transients seen in Figure 5.53 and Figure 5.54 are due to the surrounding water in the rock matrix flashing to steam and then returning to liquid. Temperature transients seen later in the simulation are similar to transients predicted to have occurred in the Oklo natural reactor (Kuroda, 1990). The third case with a cutoff of 0.8 shows the neutron flux reaching the upper limits preset in the code. The preset limits in the code were designed to keep the code from trying to simulate a supercritical event.
Also, due to storage limits in memory, the water properties data are limited from 293K to 1200K. This case warrants the need to develop a code that can predict phase change in the rock and simulate supercritical events.
CHAPTER 6

CONCLUSIONS

Two codes have been developed separately and then linked to provide a mass transport and thermal hydraulic simulation program. The overall program “GEOCRIT” tracked radionuclide mass transport from the repository and then started the thermal hydraulic simulation once $k_v$ reached a predetermined cutoff value. The thermal hydraulics code calculated neutron flux to determine temperature, stream functions, pressure, and velocities of the fluid. Several parameters for the mass transport portion of the code were varied to simulate different radionuclide buildup in the fracture and rock matrix to study the effects on neutron flux, fluid flow, temperature, and reactor operations.

Due to the lack of experimental data for geological reactors the code has been validated using a number of analytic solutions. The analytic models included mass concentration in the fracture, transient neutron flux, steady state head conduction, and steady state stream function. The analytic solutions were used to validate the different portions of the code. In each case there was good agreement between the analytic solutions and the results from the code.

Due to the low solubility of the radionuclides, a slow buildup of material in the fracture and rock matrix was observed. Varying the mass transport parameters in the code changed the buildup rates on the fracture surface and rock matrix. In all cases, the
majority of the buildup occurred on the fracture surface and the rock matrix adjacent to the fracture from the top of the fracture toward the bottom of the fracture. In each case, the neutronics were started when $k_{\infty} \geq 1.3$. As seen in section 5.2, the neutron flux was the greatest in the fracture for each case. The high neutron flux in the fracture led to increased temperatures within the fracture. The increased temperature resulted in a transient bifurcating fluid flow within the rock matrix.

In each case studied in section 5.2, it was assumed that the fracture and rock matrix were saturated. Bowman and Venneri hypothesized that a rapid drying out of the rock could lead to a criticality event due to overmoderation. In their simulation the radionuclide material was in a spherical configuration. A situation where water dries out in the fracture due to the groundwater receding was evaluated in section 5.3. Depending on the amount of radionuclide buildup in the fracture and rock matrix an overmoderated condition could exist. In the first case in section 5.3, as the fracture dried out an increase in neutron flux was experienced leading to heat generation and subsequent temperature increase in the rock. In the second case, the neutron flux also increased but was insufficient to generate enough thermal energy to increase the rock temperature. The third case showed a significant increase in neutron flux and temperature.

The possibility of overmoderated cases needs further research. Depending on the buildup of material and nature of dry out, it could pose a potential criticality risk. For the fully saturated cases, it is apparent that the material builds up slowly over time resulting in a slab type reactor. Increased solubility of radionuclides and their subsequent transport and buildup in the fracture and rock matrix warrants further investigation. In addition, more accurate colloid transport and deposition models should be further investigated.
To avert the possibility of underground criticality in a geologic repository several measures can be employed. The waste can be reprocessed and burned up in new types of reactors, reducing the amount of waste stored in the repository. The remaining waste can then be transmuted, further reducing the amount of actinides that may lead to a potential criticality. Also, less waste per container spread out over a larger area would reduce the possibility of a criticality event when the waste migrates within the host rock. Selecting a repository site where the host rock has a lower porosity may reduce the possibility of enough material building up to cause a critical event.
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