INFORMATION TO USERS

The most advanced technology has been used to photograph and reproduce this manuscript from the microfilm master. UMI films the text directly from the original or copy submitted. Thus, some thesis and dissertation copies are in typewriter face, while others may be from any type of computer printer.

The quality of this reproduction is dependent upon the quality of the copy submitted. Broken or indistinct print, colored or poor quality illustrations and photographs, print bleedthrough, substandard margins, and improper alignment can adversely affect reproduction.

In the unlikely event that the author did not send UMI a complete manuscript and there are missing pages, these will be noted. Also, if unauthorized copyright material had to be removed, a note will indicate the deletion.

Oversize materials (e.g., maps, drawings, charts) are reproduced by sectioning the original, beginning at the upper left-hand corner and continuing from left to right in equal sections with small overlaps. Each original is also photographed in one exposure and is included in reduced form at the back of the book. These are also available as one exposure on a standard 35mm slide or as a 17" x 23" black and white photographic print for an additional charge.

Photographs included in the original manuscript have been reproduced xerographically in this copy. Higher quality 6" x 9" black and white photographic prints are available for any photographs or illustrations appearing in this copy for an additional charge. Contact UMI directly to order.
Nuclear excursions in criticality accidents with fissile solutions

Pribyl, David James, M.S.

The University of Arizona, 1989
NUCLEAR EXCURSIONS IN CRITICALITY ACCIDENTS
WITH FISSION SOLUTIONS

by

David James Pribyl

A Thesis Submitted to the Faculty of the
DEPARTMENT OF NUCLEAR AND ENERGY ENGINEERING
In Partial Fulfillment of the Requirements
For the Degree of
MASTER OF SCIENCE
WITH A MAJOR IN NUCLEAR ENGINEERING
In the Graduate College
THE UNIVERSITY OF ARIZONA
1989
STATEMENT BY THE AUTHOR

This thesis has been submitted in partial fulfillment of requirements for an advanced degree at the University of Arizona and is deposited in the University Library to made available to borrowers under the rules of the Library.

Brief quotations from this thesis are allowable without special permission, provided that accurate acknowledgment of source is made. Requests for permission for extended quotation from or reproduction of this manuscript in whole or in part may be granted by the head of the major department or the Dean of the Graduate college when in his or her judgment the proposed use of the material is in the interests of scholarship. In all other instances, however, permission must be obtained from the author.

SIGNED: Daniel J. Pugh

APPROVAL BY THESIS DIRECTOR

This thesis has been approved on the date shown below:

D. L. Hetrick

Professor of Nuclear and Energy Engineering
To my parents
I would like to express my deep gratitude to David L. Hetrick for his overwhelming patience and continued support, and without whom this work would not be possible. I would also like to extend my appreciation to Robert L. Seale for the opportunity to study at the University of Arizona and the Nuclear and Energy Engineering Department's financial assistance. Finally, I would like to thank Norm Pruvost for his technical efforts on this project and my fellow graduate students for their valuable suggestions.
TABLE OF CONTENTS

LIST OF TABLES .......................................................... 7
LIST OF FIGURES .......................................................... 8
ABSTRACT ................................................................. 9
1. INTRODUCTION ....................................................... 10
2. EQUATION OF STATE, ENERGY, AND MOMENTUM .............. 14
   2.1 Description of Excursion Dynamics .......................... 14
   2.2 Equation of State .............................................. 16
   2.3 Gas Production ................................................. 19
   2.4 Equations of Energy and Momentum ......................... 22
3. RADIOLYTIC GAS BUBBLE GROWTH ............................... 25
   3.1 Nucleation Process ............................................ 25
   3.2 Bubble Growth ................................................. 27
4. LOS ALAMOS CRITICALITY ACCIDENT ........................... 31
5. MCNP REACTIVITY FEEDBACK MODEL ............................ 35
   5.1 Solution Geometry and MCNP Output ....................... 35
   5.2 Reactivity Feedback .......................................... 41
   5.3 Fission Dynamics and Step Input Reactivity .............. 44
TABLE OF CONTENTS  < continued >

6. CHEMISTRY OF THE SOLVENT ........................................... 47
   6.1 Physical Constants .................................................. 47
   6.2 Gas Production Data ............................................... 50
   6.3 Data Summary ..................................................... 51

7. RESULTS ................................................................. 53
   7.1 Simulation Procedure .............................................. 53
   7.2 Output Data ....................................................... 55
   7.3 Time Scale of the Accident ...................................... 64

8. CONCLUSIONS .......................................................... 66

APPENDIX A. NOMENCLATURE .............................................. 68

REFERENCES ................................................................. 72
LIST OF TABLES

4.1 Los Alamos Criticality Accident: Solvent-Treating Tank Parameters 33

5.1 MCNP Output for the Los Alamos Accident: $H_b = 4$ cm 39

5.2 Volume Coefficients of Reactivity and Relative Density
Changes for Solvent Volumes 43

5.3 Delayed Neutrons from Thermal Fission for Pu$^{239}$: Dynamics

6.1 Specific Volume of 3:1 Kerosene and Transformer Oil Mixture:

6.2 Physical Constants of Kerosene: Los Alamos Criticality Accident 52

7.1 Initial Conditions for the DARE P Simulation 54
### LIST OF FIGURES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.1</td>
<td>Solvent Treating-Tank (Paxton; 1959)</td>
<td>33</td>
</tr>
<tr>
<td>5.1</td>
<td>Cross Sectional View of Vessel Geometry and Initial Solution Configuration</td>
<td>37</td>
</tr>
<tr>
<td>5.2</td>
<td>Cross Sectional View of Simulated Supercritical Configuration</td>
<td>37</td>
</tr>
<tr>
<td>5.3</td>
<td>Cross Sectional View of Thermally Expanding Solvent for a Thickness L</td>
<td>38</td>
</tr>
<tr>
<td>5.4</td>
<td>Plot of Effective Multiplication Factor versus Solvent Volume</td>
<td>40</td>
</tr>
<tr>
<td>5.5</td>
<td>Least Squares Approximation for Multiplication versus Solvent Volume</td>
<td>42</td>
</tr>
<tr>
<td>7.1</td>
<td>Plot of Power versus Time: $\omega_m=15.37 \text{ sec}^{-1}$</td>
<td>57</td>
</tr>
<tr>
<td>7.2</td>
<td>Plot of Pulse Energy versus Time: $\omega_m=15.37 \text{ sec}^{-1}$</td>
<td>58</td>
</tr>
<tr>
<td>7.3</td>
<td>Plot of Reactivity versus Time: $\omega_m=15.37 \text{ sec}^{-1}$</td>
<td>59</td>
</tr>
<tr>
<td>7.4</td>
<td>Plot of Peak Pressure Increases for Various Momentum Dissipation Constants: $\omega_m=15.37 \text{ sec}^{-1}$</td>
<td>60</td>
</tr>
<tr>
<td>7.5</td>
<td>Plot of Peak Pressure Increase versus Time for a Momentum Dissipation Constant of $5(10)^4 \text{ sec}^{-1}$: $\omega_m=15.37 \text{ sec}^{-1}$</td>
<td>61</td>
</tr>
<tr>
<td>7.6</td>
<td>Plot of Solvent Volume versus Time: $\omega_m=15.37 \text{ sec}^{-1}$</td>
<td>62</td>
</tr>
<tr>
<td>7.7</td>
<td>Plot of Temperature versus Time: $\omega_m=15.37 \text{ sec}^{-1}$</td>
<td>63</td>
</tr>
</tbody>
</table>
An accidental criticality may occur in a solution of fissile material. Since the processing of nuclear materials in solution is prevalent throughout the fuel cycle, it would be judicious to have the capability to predict a possible hazard. In view of this concern, a computer simulation was performed of the Los Alamos accident of December 30, 1958, in which the actuation of an electric stirrer produced a sudden criticality.

A complete equation of state for a liquid containing gas bubbles was coupled with the equations of energy, momentum, and space-independent point kinetics. Multiplication calculations, implemented with the Monte Carlo Code for Neutron and Photon Transport (MCNP), were performed on thermally expanding solution geometries, to generate a reactivity feedback representation. With the knowledge of the total energy produced in the accident, the maximum reciprocal period on which the power rose was computed.
CHAPTER 1.
INTRODUCTION

In chemical processing with homogeneous fissile solutions, there exists a possibility of a sudden criticality. As evidence, eight supercritical accidents, caused by maloperation or processing difficulty, have occurred between 1955 and 1978. A description of these criticality accidents have been summarized in Stratton(1967), Paxton(1972), and Forehand(1981). Although the accidents generated some valuable information concerning nuclear excursions in fissile solutions, the total understanding of the excursion dynamics was uncertain.

The Kinetic Experiment on Water Boiler (KEWB) program was initiated in the early 1960's to study the safety characteristics and dynamic behavior of homogeneous solution reactors. The KEWB experiments demonstrated the importance of fission track nucleation of radiolytic gas bubbles as a dominant shutdown mechanism for a short-period power transient (Spiegler; 1962). The experiment not only confirmed the mechanism of the solution excursions, but provided valuable data from which mathematical simulations could be benchmarked (Dunenfeld and Stitt; 1963).

In November 1968, the French Commissariat a L'Energie Atomique launched an experimental study of solution excursions. The program, Consequences Radiologiques d'un Accident de Criticite (CRAC), simulated supercritical volumes of
enriched uranyl nitrate by pumping the solution into vertical cylinders (Lecorche and Revol; 1971 and Seale; 1971). The experiments demonstrated the dependence of solution addition rate and uranium concentration upon the size of the excursion, and also revealed the possibility of multiple bursts.

More recently, there has been a revitalization of interest in solution excursion experiments. The SHEBA (Solution High Energy Burst Assembly), a critical solution assembly, was constructed at the Los Alamos National Laboratory in 1980 (Malenfant, Forehand, and Koelling; 1981). Although not in the original experimental plan, the SHEBA has now been considered to continue the experimental research in solution excursions (Hetrick; 1989).

In association with the solution excursion experiments, mathematical simulations have been performed. A nonlinear shutdown model was first used by Hetrick and Gamble (1958). It was assumed that the dissolved gas concentration was a linear function of the energy release and the nucleation rate of bubbles was proportional to the power. This model met with moderate success.

A more successful model developed by Dunenfeld and Stitt (1963), correlated pressure data with the power data using an energy–threshold concept. The model assumed that the acceleration in the fluid was proportional to the transient pressure and that the feedback was proportional to the surface displacement (Hetrick; 1971). Although sound in premise, this method could not simulate solution excursions without a preconceived knowledge of the transient pressure. This limitation was removed by Forehand (1981). Pre-selected pressure shapes of exponential configuration were assumed. This model was successful in correlating peak power,
burst energy, and peak pressure with the maximum inverse period for the first burst in both KEWB and CRAC experiments.

The United Kingdom Atomic Energy Authority (UKAEA) has also developed a computer code (CRITEX) to simulate a sudden excursion in fissile solutions (Mather and Shaw; 1984). CRITEX computes the growth and movement of a gas void as it migrates toward the solution surface. This model has been moderately successful in predicting the power oscillations that were observed in the CRAC experiments. However, the simulation of transient pressure was ignored.

A new method in solution excursion simulations has been developed at the University of Arizona. A complete equation of state for a liquid containing bubbles of radiolytic gas has been derived by Hetrick (1987). The nuclear fission dynamics and the reactivity feedback are coupled with equations of state, energy, and momentum. This method, derived from quasi-static thermodynamic theory, has been successful in computing the maximum power and pressure for a number of excursions of various sizes for the first bursts which were observed in the KEWB and CRAC experiments.

With the revived interest in nuclear excursion experiments, computer simulations of criticality accidents are necessary. The work in this report represents the initial effort in simulating the Los Alamos Criticality Accident of December 30, 1958, in which the actuation of an electric stirrer in a processing vessel produced a sudden criticality. Using the quasi-static thermodynamic model, the equations of state, energy, and momentum were coupled with the nuclear fission dynamics. Although not sensitive to the excursion dynamics for the accident, a derivation for
radiolytic gas bubble growth was included in the text. The fission dynamics were computed from the equations of space-independent point kinetics and a unique feedback model. Multiplication calculations, implemented with the Monte Carlo Code for Neutron and Photon Transport (MCNP), were performed on thermally expanding solution geometries. From the MCNP output, a volume coefficient of reactivity was derived to compute the feedback reactivity. Knowing the total energy produced in the accidental burst, the point kinetics predicted a maximum reciprocal period for a step insertion of reactivity. The fission dynamic calculations were all implemented with the DARE P Continuous System Simulation Language developed at the University of Arizona.
CHAPTER 2.
EQUATIONS OF STATE, ENERGY, AND MOMENTUM

A sudden nuclear excursion may occur in a homogeneous solution of fissile material. The excursion dynamics are very complex, and therefore it will be prudent to first develop an understanding of the physical mechanisms occurring within the solution. After acquiring an understanding for the physical dynamics, a mathematical simulation will be explored.

2.1 Description of Excursion Dynamics

The system is a cylindrical vessel containing a homogeneous solution with a critical amount of fissile material. The solution in which fission is occurring is subject to high-energy radiation. The radiation consists primarily of fission fragments, alpha particles (heavy particle radiation), and fast electrons (light particle radiation). The slowing down of this radiation has two primary effects; the production of radiolytic gas contributing to the nucleation and growth of radiolytic gas bubbles, and the rapid deposition of energy causing a buildup of inertial pressure within the solution.
High-energy radiation will dissociate the solution into free ions along the path of the radiation. In aqueous and organic solutions, the products of radiolysis are hydrogen and/or oxygen ions. Many of these ions recombine, to exist as a gas \((H_2, O_2)\) dissolved in the solution. It has been observed (Spiegler; 1962), that radiolytic gas bubbles will nucleate along the radiation path, but will redissolve into the solution until the concentration of dissolved gas reaches a supersaturated threshold. Only after this threshold, will the microbubbles develop and grow.

The nucleation process is not unlike the nucleation phenomenon observed in bubble chambers, and will be discussed in the next chapter. The subject of radiolysis of solutions by high-energy radiation is described in great detail in (Forehand; 1981). Interested readers are encouraged to study further.

The microbubble will nucleate at an initial radius of approximately \(10^{-8}\) meters (Spiegler; 1962). Bubble growth will occur as the dissolved gas in the solution diffuses into bubbles. The growth of the gas bubbles along with the thermally expanding solution (energy deposition associated with the slowing down of radiation) will decrease the solution density. The density change will increase the neutron leakage, providing negative reactivity, terminating the sudden excursion.

Bubbles will continue to grow, depleting the amount of dissolved gas in the solution. Due to buoyancy effects, the bubbles will rise up and migrate out of the solution. The absence of the gas bubbles will provide positive reactivity, and the possibility of multiple excursions exists.

High-energy radiation also contributes to the creation of the inertial pressure within the solution. The slowing down of radiation, predominately fission fragments,
deposits kinetic energy within the solution. The rapid energy deposition occurs at a rate faster than the solution's ability to expand, creating the inertial pressure within the solution. This surge of pressure contributes to the violent disturbance of the solution which has been observed in both experimental and accidental excursions.

2.2 Equation of State

A complete equation of state for a solution containing radiolytic gas bubbles has been derived by Hetrick (1987). The differential equation of state along with the computed coefficients have been developed from quasi-static thermodynamic theory. The accuracy of this method is hence limited to the application of quasi-static thermodynamics. However, this application has warranted an interesting investigation.

The quasi-static differential representation for liquid pressure, in terms of volume and temperature, is the following equation.

\[ dP = \frac{\beta}{\kappa}dT - \frac{1}{\kappa V}dV \]  

(2-1)

where:

\[ P \equiv \text{liquid pressure} \]
\[ T \equiv \text{absolute temperature} \]
\[ V \equiv \text{system volume} \]
\[ \kappa \equiv \text{isothermal compressibility} \]
\[ \beta \equiv \text{isobaric compressibility} \]
The coefficients in the equation of state, $\kappa$ and $\beta$, are computed for a solution containing radiolytic gas bubbles. Defining the solution volume ($V$) to be the sum of the volumes of liquid ($V_l$) and gas bubbles ($V_g$), the coefficients can be written in the form

$$\kappa = -\frac{1}{V} \left( \frac{\partial V_l}{\partial P} \right)_T - \frac{1}{V} \left( \frac{\partial V_g}{\partial P} \right)_T$$

$$\beta = \frac{1}{V} \left( \frac{\partial V_l}{\partial T} \right)_P + \frac{1}{V} \left( \frac{\partial V_g}{\partial T} \right)_P.$$

The volume fraction of gas in bubbles ($f$), defined as $f = V_g / V$, is introduced into the coefficients. The gas and liquid volumes can be represented in terms of the volume fraction. Defining the isothermal and isobaric compressibilities for the liquid alone as $\kappa_o$ and $\beta_o$, respectively, the coefficients for the differential equation of state become

$$\kappa = (1 - f)\kappa_o - \frac{f}{V_g} \left( \frac{\partial V_g}{\partial P} \right)_T \quad (2-2)$$

$$\beta = (1 - f)\beta_o + \frac{f}{V_g} \left( \frac{\partial V_g}{\partial T} \right)_P. \quad (2-3)$$

To compute the derivatives in equations (2-2) and (2-3), it will be assumed that each bubble adheres to the equation of mechanical equilibrium (2-4), the ideal gas law (2-5), and that all bubbles be of uniform size.

$$P = P_i - \frac{2\sigma}{r_b} \quad (2-4)$$

$$P_i = NR_g T / V_g \quad (2-5)$$
where

\[ P_i \equiv \text{pressure within gas bubble} \]
\[ \sigma \equiv \text{liquid surface tension} \]
\[ r_b \equiv \text{bubble radius} \]
\[ N \equiv \text{mass of gas in bubbles} \]
\[ R_g \equiv \text{gas constant per unit mass} \]

It may also be assumed that the geometry of the bubble is spherically symmetric. Combining the above equations with this assumption produces

\[ PV_g = NR_gT - 2\sigma \left( \frac{4\pi n_b}{3} \right)^{1/3} V_g^{2/3} \]

where

\[ n_b \equiv \text{total number of bubbles}. \]

A differentiation may be performed on equation (2-6), and after some algebra, the following expression may be written.

\[ \left( P + \frac{4\sigma}{3r_b} \right) \frac{dV_g}{V_g} + dP = \left( P + \frac{2\sigma}{r_b} \right) \frac{dT}{T} \]

(2 - 7)

Two equations may be generated from equation (2-7), one based upon a constant temperature process \((dT = 0)\), and another derived from a constant pressure process \((dP = 0)\).

\[ \frac{1}{V_g} \left( \frac{\partial V_g}{\partial P} \right)_T = -\frac{1}{P + 4\sigma/3r_b} \quad dT = 0 \]

(2 - 8)

\[ \frac{1}{V_g} \left( \frac{\partial V_g}{\partial T} \right)_P = \frac{1}{T} \frac{P + 2\sigma/r_b}{P + 4\sigma/3r_b} \quad dP = 0 \]

(2 - 9)
Substituting equations (2-8) and (2-9) into equations (2-2) and (2-3), respectively, the computed coefficients for the differential equation of state become

\[ \kappa = \kappa_0 (1 - f) + \frac{f}{P + 4\sigma/3r_b} \]  
\[ \beta = \beta_0 (1 - f) + \frac{f(P + 2\sigma/r_b)}{T(P + 4\sigma/3r_b)}. \]

In summary, the differential equation of state (2-1) along with the computed coefficients (2-10) and (2-11), may be integrated to simulate the inertial pressure within the solution.

2.3 Gas Production

To simulate the production of radiolytic gas in bubbles, it is assumed that the amount of gas in bubbles is the excess amount of dissolved gas in solution above the supersaturated concentration. It is necessary, therefore, to compute the amount of dissolved gas produced in the solution. The amount of dissolved gas produced \( (x_E) \), which will be represented as a mass fraction (mass of gas per mass of solution), can be computed by the following equation.

\[ x_E = \frac{GE(t)}{M} \]
where;

\[ G \equiv \text{dissociation rate in mass of gas of} \]

hydrogen and/or oxygen per unit energy

\[ t \equiv \text{time} \]

\[ E(t) \equiv \text{nuclear energy in a pulse} \]

\[ M \equiv \text{mass of solution.} \]

When the concentration of dissolved gas \((x_E)\) reaches the supersaturated threshold, defined as the mass fraction \((x_T)\), stable radiolytic gas bubbles are nucleated. The amount of gas in bubbles \((x)\), represented as the excess amount of dissolved gas above threshold, can be written as

\[
x = \begin{cases} 
0 & \text{when } x_E < x_T \\
x_E - x_T & \text{when } x_E > x_T 
\end{cases} \tag{2-13}
\]

where

\[ x \equiv \text{mass fraction of gas in bubbles.} \]

The dissociation rate \((G)\) as well as the threshold conditions are known empirically for some solvents (Forehand; 1981 and Stieglitz; 1982). When the solution absorbs the energy associated with threshold, obtained from the nuclear fission dynamics (Chapter 5), equation (2-13) may be used to simulate the mass fraction of gas in bubbles.
Since the coefficients of the differential equation of state are in terms of the volume fraction of gas \( f \), it is necessary to represent the volume fraction in terms of the mass fraction \( x \). The volume fraction of gas can be written as \( f = x v_g / v \), where \( v_g \) is the specific volume of a gas, and \( v \) is the specific volume of a solution. Again, using the ideal gas law and the equation of mechanical equilibrium, the specific volume of a gas can be represented as

\[
v_g = \frac{R_g T}{(P + 2\sigma/r_b)} \quad (2-14)
\]

Using the definition of a mixture, an expression for the specific volume of the solution can be obtained.

\[
v = (1 - x) v_\ell + xv_g \quad (2-15)
\]

where

\[
v_\ell \equiv \text{specific volume of the liquid.}
\]

Combining the expression \( f = x v_g / v \) with equations (2-14) and (2-15), the volume fraction of a gas can be represented as

\[
f = \frac{x R_g T}{(1 - x)v_\ell(P + 2\sigma/r_b) + x R_g T} \quad (2-16)
\]

Upon computing equation (2-13) for the mass fraction of gas \( x \), the expression for the volume fraction may be used to evaluate the coefficients of the differential equation of state.
2.4 Equations of Energy and Momentum

The energy equation from quasi-static thermodynamics can be used to simulate the temperature change within the solution. The internal energy, based upon a dependence of temperature and volume, can be related to the difference between the nuclear energy in a pulse and the $PdV$ work performed on the system. Rearranging this energy equation yields

$$dT = \frac{1}{c_v}(dE - \frac{\beta T}{\kappa}dV)$$  \hspace{1cm} (2-17)

where

- $T \equiv$ absolute temperature
- $c_v \equiv$ heat capacity
- $E \equiv$ nuclear energy in a pulse; computed from fission dynamics
- $\beta \equiv$ isobaric compressibility
- $\kappa \equiv$ isothermal compressibility
- $V \equiv$ system volume.

Volume changes for a vertical cylinder of solution are computed from a momentum balance performed on the entire solution. The “lumped-parameter” momentum equation describes the net acceleration of the solution in the upward vertical direction, driven by a pressure difference. The momentum balance may be written in the following form.
The rate of momentum accumulation may be represented as the product of the solution mass \((M)\) and the net upward acceleration \((dv_z/dt)\), where \(v_z\) is the velocity of the solution’s center of mass in the upward \(z\) direction.

In solutions of fissile material, the important force acting on the system is the solution’s inertial pressure. The inertial pressure \((P)\), defined by the equation of state, is created by the rapid energy deposition of radiation. It is the difference between the atmospheric pressure \((P_o)\) at the top of the solution and the buildup of inertial pressure that accelerates the solution upward.

It is suggested that the pressure gradient in the vertical cylinder of solution is approximately linear (Spiegler; 1962 and Hetrick; 1989). Since the equation of state represents the pressure at the center of mass, twice the difference between the atmospheric and the inertial pressures represents the total pressure difference between the top and the bottom of the solution. Therefore, the sum of the forces acting on the system is \(2A(P - P_o)\), where \(A\) is the cross sectional area of the cylinder.

Momentum is lost by the solution as it expands against the vessel. The fissile solution expands in all directions as the radiation deposits its energy. Since the “lumped-parameter” momentum balance represents a net upward motion, the equation must compensate for the rate at which momentum is dissipated, in the
form of friction, as the solution expands against the vessel. The rate of momentum loss can be represented as the product of the solution's momentum, $Mv_z$, and a rate constant $\Lambda$, where $\Lambda$ represents the average rate of momentum dissipation. $\Lambda$ is a "lumped-parameter" that will have to be evaluated during the simulations.

From the foregoing expressions, the momentum balance may be written mathematically as

$$M \frac{dv_z}{dt} = 2A(P - P_o) - \Lambda M v_z. \quad (2-18)$$

The velocity in the momentum balance can be related to the volume ($V$) change, using the equation of continuity.

$$\frac{1}{V} \frac{dV}{dt} = \frac{\partial v_z}{\partial z}$$

A linear velocity profile will be assumed, where the velocity of the solution at the center of mass is $v_z$. Using the continuity expression and the assumed velocity profile, the momentum balance becomes

$$\frac{d^2V}{dt^2} = \frac{4A^2}{M}(P - P_o) - \Lambda \frac{dV}{dt}. \quad (2-19)$$

In summary, the energy equation (2-17) and the momentum balance (2-19) may be integrated to simulate the temperature and volume, respectively, of the solution.
CHAPTER 3.
RADIOLYTIC GAS BUBBLE GROWTH

Radiolytic gas bubbles are nucleated along a radiation path in a solution of fissile material. The nucleation process is not unlike the nucleation phenomenon observed in bubble chambers, and many of the associated theories are applicable. Once nucleated, the bubbles will grow by the diffusion of dissolved gas in solution. In this chapter, the process of nucleation and bubble growth will be explored, and a mathematical representation for bubble growth will be developed.

3.1 Nucleation Process

The nucleation of radiolytic gas bubbles in fissile solutions will be successful only if the solution medium becomes sensitive to high-energy radiation. In comparison, the sensitivity in liquid bubble chambers is achieved by superheating the liquid, while suppressing the boiling. The passage of radiation in this metastable liquid will locally excite the liquid into a phase transition, thus initiating bubble formation. Sensitivity may also be attained by supersaturating a liquid with a dissolved gas. In these gas–liquid chambers, the dissolved gas provides a strong reduction of the liquid’s surface tension, permitting the nucleation of bubbles by the energy deposition of high-energy radiation (Shutt; 1967). It is believed (by this
author) that this mechanism allows for the nucleation of gas bubbles in solutions of fissile material.

In solutions of fissile material, the supersaturated condition is achieved by the radiolysis of the solution by ionizing radiation. Prior to this supersaturated condition, it has been observed (Spiegler; 1962) that nucleated bubbles will redisolve into solution. It is concluded that prior to the supersaturated threshold, the reduction of the surface tension provided by the dissolved gas is not sufficient to permit the nucleation of stable gas bubbles. When the dissolved gas concentration has reached the supersaturated threshold, the surface tension has been minimized, and stable gas bubbles are nucleated by the deposition of energy by radiation.

The accepted theory of bubble nucleation was developed by Seitz(1958) for bubble chamber research. It was assumed that bubbles are formed by thermal spikes (localized “hot spots”) produced by the heat released in ion recombinations of unbound electrons. The electrons, produced by the interaction between the radiation and the nuclei of the solution, remain very close to the radiation path (coulomb scattering). Clusters of ions recombine near the path and generate a highly localized evolution of heat (thermal spike). The evaporation of the liquid in a concentrated volume creates a gas–vapor void. Since generally fissile solutions are below the boiling temperature, the vapor quickly condenses, leaving only a gas bubble. The gas bubble rapidly grows by the diffusion of dissolved gas in the solution.
3.2 Bubble Growth

A gas bubble in a liquid-gas state will grow by the diffusion of dissolved gas. An expression for bubble growth may therefore be derived from diffusion theory (Epstein and Plesset; 1950). It will be assumed that at the time $t_N$, a spherical bubble is nucleated at an initial radius $r_o$, and the concentration of dissolved gas is supersaturated at $c_T$. Since the growth is driven by a supersaturated concentration, it may be assumed that for all times, the concentration at the bubble surface (bubble radius; $r_b$) is at saturation $c_{sat}$ (Epstein and Plesset; 1950). Bubble growth will be spherically symmetric and therefore the diffusion process is governed by the following diffusion equation.

$$\frac{\partial c}{\partial t} = D \frac{\partial}{r^2} \left( r^2 \frac{\partial c}{\partial r} \right)$$

where

- $c(r, t) \equiv$ dissolved gas concentration
- $t \equiv$ time
- $r \equiv$ radial coordinate centered at the bubble origin
- $D \equiv$ gas diffusivity

The initial and boundary conditions are

- $c(r, t_N) = c_T$
- $c(r_b, t) = c_{sat}$
- $c(\infty, t) < \infty$. 
It will be convenient to transform the concentration into a parameter $u$, where $u = r(c - c_T)$, and the time into a parameter $\tau$, where $\tau = t - t_N$. The diffusion equation transforms into

$$\frac{\partial u}{\partial \tau} = D \frac{\partial^2 u}{\partial r^2}. \quad (3 - 1)$$

Applying the Laplace transform and the initial condition to equation (3-1) yields

$$\frac{\partial^2 \tilde{u}}{\partial r^2} - \frac{s}{D} \tilde{u} = 0 \quad (3 - 2)$$

where

$$s \equiv \text{transform parameter}$$

$$\tilde{u}(r, s) \equiv \text{Laplace transform of } u.$$

The solution to the second-order differential equation, after applying the appropriate boundary conditions becomes

$$\tilde{u} = r_b(c_{sat} - c_T)e^{-(r-r_b)\sqrt{s/D}}/s.$$

Inverting back into the original space $u$ (Churchill; 1958) and then transforming back into the original parameters $c$ and $t$, the concentration of dissolved gas outside a bubble can be written as

$$c(r, t) = c_T - \frac{r_b}{r}(c_T - c_{sat})erfc\left(\frac{r - r_b}{2\sqrt{D(t - t_N)}}\right), \quad t > t_N \quad (3 - 3)$$

It is desired to compute the concentration gradient at the bubble surface ($r = r_b$). Differentiating equation (3-3), and evaluating at $r = r_b$ produces

$$\left.\frac{\partial c}{\partial r}\right|_{r_b} = (c_T - c_{sat})\left\{\frac{1}{\sqrt{\pi D(t - t_N)}} + \frac{1}{r_b}\right\}, \quad t > t_N. \quad (3 - 4)$$
Representing the mass of gas in a bubble as \( m \), the mass rate of flow into a bubble can be written as
\[
\frac{dm}{dt} = 4\pi r_b^2 D \frac{\partial c}{\partial r}
\]
Equations (3-4) and (3-5) were derived for a stationary bubble boundary. However, it is reasonable to use this result for dynamic bubble growth. The region of diffusion surrounding a bubble is much larger than the bubble itself, therefore the concentration gradient outside a bubble is not strongly influenced by the size of the bubble (Epstein and Plesset; 1950). Bubble size is only important in regard to the surface area in which mass transfer takes place. Therefore, it is legitimate to neglect the transport term (generated from bubble motion) in the diffusion equation and use the stationary analysis to simulate bubble growth.

An expression for the growth rate of a bubble can be computed from the mass influx. The mass of the gas in a bubble can be represented as \( m = \rho_b \frac{4}{3} \pi r_b^3 \), where \( \rho_b \) is the density of the gas inside a bubble. A differentiation may be performed on the definition of the mass generating
\[
\frac{dm}{dt} = 4\pi r_b^2 \rho_b \frac{dr_b}{dt} + \frac{4}{3} \pi r_b^3 \frac{d\rho_b}{dt}.
\]
The rate at which the density changes within a bubble can be evaluated using the ideal gas law and the equation of mechanical equilibrium. The gas density \( (\rho_b) \) can be written as
\[
\rho_b = \frac{1}{R_g T} \left( P + \frac{2\sigma}{r_b} \right).
\]
where  

\[ P \equiv \text{liquid pressure} \]
\[ T \equiv \text{absolute temperature} \]
\[ \sigma \equiv \text{surface tension} \]
\[ R_g \equiv \text{gas constant per unit mass.} \]

Applying the chain rule of calculus, the density rate becomes

\[
\frac{d\rho_b}{dt} = \frac{1}{R_gT} \frac{dP}{dt} - \frac{\rho_b}{T} \frac{dT}{dt} - \frac{2\sigma}{R_g T r_b^2} \frac{dr_b}{dt} \quad (3-7)
\]

Combining equations (3-5), (3-6), and (3-7), the growth rate for a bubble can be expressed as

\[
\frac{dr_b}{dt} = \left( r_b \rho_b - 2 \frac{\sigma}{3 R_g T} \right)^{-1} \left[ r_b D \frac{\partial c}{\partial r} \bigg|_{r_b} - r_b^2 \left( \frac{1}{R_g} \frac{dP}{dt} - \rho_b \frac{dT}{dt} \right) \right] \quad (3-8)
\]

The pressure and temperature changes can be evaluated using the equations of state and energy presented in the preceding chapter. A radiolytic gas bubble in a fissile solution will nucleate at an initial radius \( r_0 \) of \( 5 \times 10^{-6} \) cm (Spiegler; 1962). With the knowledge of the concentration gradient at the bubble surface, equation (3-4), equation (3-8) may be integrated to simulate bubble growth.
CHAPTER 4.
LOS ALAMOS CRITICALITY ACCIDENT

On December 30, 1958, a nuclear-critical accident occurred in a processing vessel at the Los Alamos National Laboratory plutonium-processing plant. The vessel was located in the TBP (tri-n-butyl phosphate) solvent extraction plant, where residues from a plutonium recovery operation are concentrated and purified. In this plant, plutonium is separated by a repeated batch extraction into TBP carried in an organic solvent, and then concentrated by evaporation in an aqueous-phase extraction. The resulting solution, containing plutonium at a few grams per liter, then is fed back into an earlier recovery stage (Paxton; 1959).

The accident occurred during a plutonium inventory in which the normal flow into the TBP-solvent-extraction plant was interrupted. The operator, who was to perform the inventory to determine the plutonium content in the processing vessels, departed from prescribed procedures and handled several batches of material at one time. Due to many interconnecting transfer lines, plutonium-rich solids from two separate vessels were washed into a large vessel that contained dilute aqueous and organic solutions. Most of the aqueous phase was removed, and the remaining 151 liters of material was transferred into a large stainless-steel solvent-treating tank in which the accident occurred. In addition to the 303 liters of aqueous and organic emulsions that were present upon the 151 liter transfer, 49 liters of nitric acid was
also added. Air sparging mixed the emulsion content, extracting the plutonium into the solvent. After an estimated 2.5 minutes, the phases separated (Paxton; 1959).

Immediately prior to the accident, the initial configuration of the solution that is believed to have been contained in the 0.965 meter, 852 liter solvent-treating tank, was 160 liters of an organic solvent, resting upon 331 liters of an aqueous solution (Figure 4.1). The 21.87 centimeter thick solvent layer, containing approximately 3.27 kg of plutonium, is assumed to have been barely subcritical with a plutonium concentration of 20 gm/liter (Table 4.1). The aqueous solution is believed to have contained 40 grams of plutonium, with an additional 60 grams of plutonium solids suspended at the organic-aqueous interface (Paxton; 1959).

The operator actuated an electric stirrer located at the bottom of the solvent-treating tank. The initial action of the stirrer forced aqueous solution up along the tank walls, displacing the outer portion of the solvent layer, thickening the central portion (Paxton; 1959). With the new configuration, the system achieved supercriticality.

At this time, the operator fell from a two-step ladder, where he had been looking into a sight port on top of the tank. He then turned the electric stirrer off and then on again, and ran out of the room calling for assistance. Two other operators, working nearby, came to his aid, turned the stirrer off, and lead the injured operator away.
Table 4.1

Los Alamos Criticality Accident:
Solvent-Treating Tank Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Solvent Volume (liters)</td>
<td>160.0</td>
</tr>
<tr>
<td>Aqueous Volume (liters)</td>
<td>330.8</td>
</tr>
<tr>
<td>Tank Diameter (meters)</td>
<td>0.965</td>
</tr>
<tr>
<td>Tank Cross Sectional Area (m²)</td>
<td>0.732</td>
</tr>
<tr>
<td>Solvent Layer Height (meters)</td>
<td>0.219</td>
</tr>
<tr>
<td>Plutonium Mass/Solvent (kg)</td>
<td>3.27</td>
</tr>
<tr>
<td>Plutonium Mass/Aqueous (kg)</td>
<td>0.04</td>
</tr>
<tr>
<td>Plutonium Suspended Solids (kg)</td>
<td>0.06</td>
</tr>
</tbody>
</table>

Figure 4.1 Solvent Treating–Tank (Paxton;1959).
Although the motor was turned on a second time, radiation-detector traces indicate the existence of one nuclear burst. Radiochemical analysis estimates the total number of fissions in the single burst at $1.5 \times 10^{17}$. It was concluded that the violence of the burst along with the continual stirring prevented a recriticality. The shock of the burst, however, did displace the tank approximately 3/8 inches at its supports. The tank was not ruptured, nor did any plutonium escape (Paxton; 1959).

The entire plant was evacuated when the burst activated a radiation alarm 175 feet away, set to sound at 10 mrem/hr. The injured operator received an estimated dose of 12,000 rem ±50%, and died 36 hours later. The other two operators received estimated doses of 134 and 53 rem, and continued on regular duty (Paxton; 1959).

On January 1, 1959, the solution in the tank was vacuum-transferred into fabricated 5 and 6 inch diameter withdrawal cylinders. The transfer occurred without incident. The contents of each storage cylinder was examined by chemical analysis and by radiation surveys. The data presented, concerning the accident, was obtained from these procedures.
CHAPTER 5.

MCNP REACTIVITY FEEDBACK MODEL

The principal difficulty in simulating any nuclear excursion is in the computation of the feedback reactivity. For the Los Alamos accident of December 30, 1958, this difficulty is enhanced by the uncertainty in the solution configuration as it was altered by the motion of an electric stirrer. In this chapter, a reactivity feedback model will be developed for proposed solution geometries. An expression for the density coefficient of reactivity will be derived using the multiplication ($k$) calculations performed at the Los Alamos National Laboratory. The feedback reactivity will then be coupled with point kinetics and a reactivity step insertion approximation. The $k$ calculations for the proposed geometries were implemented with the Monte Carlo Code for Neutron and Photon Transport (MCNP), and performed by Norm Pruvost on the Cray XMP system.

5.1 Solution Geometry and MCNP Output

In the Los Alamos accident of December 30, 1958, the actuation of an electric stirrer located at the bottom of a processing vessel produced a sudden criticality. It is intuitive that the action of stirrer forced aqueous solution up along the tank walls, thickening the central portion of the plutonium-rich solvent layer. Paxton
(1959) estimated that an average increase of 0.4 inches in solvent layer thickness would account for the supercritical burst. This estimate was the initial working assumption that was later verified by the simulation. It was also assumed that the burst was terminated by the increase in neutron leakage as the solvent expanded thermally by the deposition of radiative energy.

The change in the multiplication for the accident was calculated by the Monte Carlo Code for Neutron and Photon Transport (MCNP). MCNP, developed by the Los Alamos National Laboratory, duplicates on a high-speed computer the actual histories of neutrons and/or photons as they move about a specified medium. The Monte Carlo method, in general, predicts the probability of an “event” by correlating random numbers with probability data for a given particle/target interaction. A description and user guide for MCNP has been edited by Briesmeister (1986).

The vessel geometry (MCNP input) was represented by a right circular cylinder. In association with this geometry, the initial solution configuration is depicted in Figure 5.1. The solution contents were presented in Chapter 4. The supercritical configuration was simulated by altering the surface at the organic–aqueous interface. The best surface geometry that was believed to reflect the influence of the stirrer was a paraboloid of revolution. The volume of a paraboloid of revolution is $\frac{1}{2}AH_b$, where $A$ is the cross sectional area of the tank (0.732 m$^2$) and $H_b$ is the maximum height of the paraboloid. Conserving both solvent mass and volume and also maintaining a flat top solvent surface, the Paxton estimate suggests a paraboloid height of 4 centimeters. MCNP, for this geometry (Figure 5.2), generated an effective multiplication factor of $1.0148\pm0.0045$, referenced at $0.997\pm0.0048$. 
Figure 5.1. Cross Sectional View of Vessel Geometry and Initial Solution Configuration.

Figure 5.2. Cross Sectional View of Simulated Supercritical Configuration
The termination of the excursion was simulated by the volumetric expansion of the solvent. While conserving the solvent mass, the top surface of the solvent was extended a thickness $L$ from the original surface (Figure 5.3). The height of the paraboloid at the organic-aqueous interface was maintained at 4 centimeters. For uniform density changes, MCNP computed the values for the effective multiplication factor ($k$) as well as the neutron generation time ($\ell$) for preselected values of $L$. The data for these MCNP cases are summarized in Table 5.1 and plotted in Figure 5.4. With the MCNP output, an expression for the density coefficient of reactivity may be obtained.

Figure 5.3. Cross Sectional View of Thermally Expanding Solvent for a Thickness $L$. 
Table 5.1
MCNP Output for the Los Alamos Accident:
$H_b=4.0 \text{ cm}$

<table>
<thead>
<tr>
<th>Case #</th>
<th>$L$ (cm)</th>
<th>$V$ ($m^3$)</th>
<th>$k$</th>
<th>$\ell$ ((\mu\text{sec}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>DHE 031</td>
<td>0.0</td>
<td>0.1600</td>
<td>1.0148±0.0045</td>
<td>60.66±0.61</td>
</tr>
<tr>
<td>DHE 215</td>
<td>0.5</td>
<td>0.1637</td>
<td>1.0124±0.005</td>
<td>61.98±0.60</td>
</tr>
<tr>
<td>DHE 216</td>
<td>1.0</td>
<td>0.1673</td>
<td>1.0112±0.005</td>
<td>63.10±0.57</td>
</tr>
<tr>
<td>DHE 217</td>
<td>2.0</td>
<td>0.1747</td>
<td>1.0014±0.005</td>
<td>64.44±0.59</td>
</tr>
</tbody>
</table>
Figure 5.4. Plot of Effective Multiplication Factor versus Solvent Volume.
5.2 Reactivity Feedback

The feedback reactivity ($\rho_f$), induced by a decrease in the solvent density, was represented as the product of the volume change and the volume coefficient of reactivity ($\phi_v$). The foregoing expression can be written mathematically as

$$ \partial \rho_f = \phi_v \partial V $$ \hspace{1cm} (5 – 1)

where

$\rho_f \equiv$ Feedback reactivity  
$\phi_v \equiv$ Volume coefficient of reactivity  
$V \equiv$ Solvent Volume.

An expression for the volume coefficient of reactivity was developed using the MCNP output. In simulating the volumetric expansion for the accident, MCNP provided effective multiplication factors for specified solvent volumes (Table 5.1). These values were plotted (Figure 5.4) to suggest a trend in the multiplication for the system. Because of the sizeable uncertainty on $k$ associated with the Monte Carlo techniques, a regression through the data points was generated more by intuition rather than by forcing a rigid fit. A least squares approximation for the first, second, and fourth data points (quadratic in volume) was chosen, in order to maintain a close linearity ($k$ versus $V$) between the first two data points. Linearity was preferred because density changes in this region were sufficient to terminate the accidental excursion. A least square approximation for the four points did not produce the linearity. The smooth quadratic also provided a good fit with the final data point. The quadratic regression (Figure 5.5) through the data yielded

$$ k = a_0 + a_1 V + a_2 V^2 $$ \hspace{1cm} (5 – 2)
Figure 5.5. Least Squares Approximation for MCNP Multiplication versus Solvent Volume.
where 

\[ a_o = 0.46762 \]

\[ a_1 = 7.40819 \, \frac{1}{m^3} \]

\[ a_2 = -24.91255 \, \frac{1}{m^6} \]

The definition for the volume coefficient of reactivity may be approximated by

\[ \phi_v \approx \frac{\partial k}{\partial V}. \]

Upon differentiating equation (5-2) with respect to the volume, the volume coefficient of reactivity may be expressed as

\[ \phi_v \approx a_1 + 2a_2 V. \]

(5 - 3)

For a given solvent volume, Table 5.2 presents the associated volume coefficient of reactivity and the relative density change.

Table 5.2

Volume Coefficients of Reactivity and Relative Density Changes for Solvent Volumes

<table>
<thead>
<tr>
<th>( V ) ((m^3))</th>
<th>( \phi_v ) (($/m^3))</th>
<th>Relative Density Change</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.160</td>
<td>-187.94</td>
<td>0.0</td>
</tr>
<tr>
<td>0.161</td>
<td>-204.55</td>
<td>-0.0062</td>
</tr>
<tr>
<td>0.162</td>
<td>-221.16</td>
<td>-0.0123</td>
</tr>
<tr>
<td>0.163</td>
<td>-237.76</td>
<td>-0.0184</td>
</tr>
<tr>
<td>0.164</td>
<td>-254.38</td>
<td>-0.0244</td>
</tr>
<tr>
<td>0.166</td>
<td>-287.59</td>
<td>-0.0361</td>
</tr>
<tr>
<td>0.168</td>
<td>-320.81</td>
<td>-0.0476</td>
</tr>
<tr>
<td>0.170</td>
<td>-354.03</td>
<td>-0.0588</td>
</tr>
</tbody>
</table>
5.3 Fission Dynamics and Step Input Reactivity

The simulation of the Los Alamos accident will focus upon the neutronics in the solvent, since the solvent was concentrated with the majority of the plutonium. The nuclear fission dynamics will be simulated by the equations of space-independent point kinetics.

\[
\frac{dn}{dt} = \frac{\rho - \beta_d}{\ell} n + \sum \lambda_i c_i \quad (5 - 4)
\]

\[
\frac{dc_i}{dt} = \frac{\beta_i}{\ell} n - \lambda_i c_i
\]

where

\( n \equiv \text{power} \)
\( c_i \equiv \text{precursor density} \)
\( t \equiv \text{time} \)
\( \rho \equiv \text{reactivity} \)
\( \beta_d \equiv \text{delayed neutron fraction} \quad \beta_d = \sum \beta_i \)
\( \ell \equiv \text{neutron generation time (MCNP Output)} \)
\( \lambda_i \equiv \text{decay constant for precursor density.} \)

The nuclear energy in the pulse \( E \) may be obtained by integrating the point kinetics equations.

\[
\frac{dE}{dt} = n \quad (5 - 5)
\]
With the estimated 5.0w% of Pu$^{240}$, the delayed neutron fraction for the system was approximated at 0.003, and the expression, $\frac{2\lambda}{\tau}$, was 49.46. Table 5.3 presents the delayed neutron data for Pu$^{239}$.

Table 5.3

Delayed Neutrons from Thermal Fission for Pu$^{239}$:

<table>
<thead>
<tr>
<th>Group No.</th>
<th>Decay Constant $\lambda_i$, sec$^{-1}$</th>
<th>Relative Yield $\beta_i/\beta_d$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.0128</td>
<td>0.035</td>
</tr>
<tr>
<td>2</td>
<td>0.0301</td>
<td>0.298</td>
</tr>
<tr>
<td>3</td>
<td>0.124</td>
<td>0.211</td>
</tr>
<tr>
<td>4</td>
<td>0.325</td>
<td>0.326</td>
</tr>
<tr>
<td>5</td>
<td>1.12</td>
<td>0.086</td>
</tr>
<tr>
<td>6</td>
<td>2.69</td>
<td>0.044</td>
</tr>
</tbody>
</table>

The simulation of the total reactivity of the system was based upon correlating the total energy produced in the burst with a step insertion of positive reactivity above critical. Since there was a high degree of uncertainty in the initial subcritical state of the system, the net positive reactivity induced by the distortion of the solvent (MCNP output) could not be referenced accurately. However, the total fissions produced in the burst, $1.5(10)^{17}$, was estimated by radiochemical analysis performed after the accident. Therefore, for the simulation, the appropriate amount of input reactivity was chosen to generate the 4.8 MJ of energy created in the accident. The total reactivity ($\rho$) of the system may be written as
\[ \rho = \rho_o + \rho_f \]

where

\[ \rho_o \equiv \text{Step insertion of positive reactivity above critical} \]

\[ \rho_f \equiv \text{Feedback reactivity.} \]

With the volume coefficient of reactivity defined (5-3), the point kinetics were coupled with the equations of state, energy, and momentum to compute the maximum reciprocal period \( (\omega_m) \) on which the power rose. \( \omega_m \), defined as the measure of the maximum reactivity excess above prompt critical, can be expressed as

\[ \omega_m = \frac{\rho_o - \beta d}{\ell}. \]

This \( \omega_m \) may then be used to characterize a ramp input of reactivity for future simulations. The ramp rate can not be estimated because the initial state is not known. From the step insertion simulation, an appropriate ramp rate and initial condition may be approximated.

In summary, the maximum reciprocal period and the associated excursion dynamics for the Los Alamos accident were computed by generating the known 4.8 MJ of energy with the defined MCNP reactivity feedback representation.
CHAPTER 6.
CHEMISTRY OF THE SOLVENT

The simulation of the Los Alamos criticality accident requires the determination of the physical properties of the system's solution. The majority of the solution, in which the nuclear fission dynamics occurred, was the organic solvent, Gulf BT kerosene. This chapter is devoted to computing kerosene's characteristic properties and its response to ionizing radiation. The parameters generated for kerosene will be summarized in Table 6.2.

6.1 Physical Constants

The supercriticality of the Los Alamos nuclear accident was achieved by the distortion of the organic solvent layer by an electric stirrer. The organic solvent was Gulf BT kerosene, which contained a plutonium nitrate complex of hydrolyzed 35% TBP (tri-n-butyl phosphate). Since the majority of the plutonium (3.27 kg) had been extracted into the TBP–BT solvent (160 liters) from the aqueous solution, the nuclear dynamic simulation will focus upon the properties of the TBP–BT solvent.

The complexity of the plutonium TBP–BT kerosene solution as well as the unavailability of the data would make the determination of the physical constants
virtually impossible. Therefore, since approximately $\frac{2}{3}$ of the solution was the solvent, it will be assumed that the physical constants of the solution are those of kerosene.

Although kerosene is a familiar substance, the data available to determine the isothermal and isobaric compressibilities is limited. However, volume changes for a 3:1 mixture of kerosene and transformer oil have been performed by Maslennikova (1974). The experiment generated values for the specific volume of the mixture at various temperatures and pressures (Table 6.1).

Table 6.1

Specific Volume [cm$^3$/g] of 3:1 Kerosene and Transformer Oil Mixture:

<table>
<thead>
<tr>
<th>P, bar</th>
<th>20°C</th>
<th>100°C</th>
<th>150°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.24</td>
<td>1.34</td>
<td>1.40</td>
</tr>
<tr>
<td>1000</td>
<td>1.16</td>
<td>1.21</td>
<td>1.25</td>
</tr>
<tr>
<td>1500</td>
<td>1.13</td>
<td>1.18</td>
<td>1.21</td>
</tr>
<tr>
<td>2000</td>
<td>1.11</td>
<td>1.15</td>
<td>1.18</td>
</tr>
<tr>
<td>2500</td>
<td>1.09</td>
<td>1.13</td>
<td>1.15</td>
</tr>
<tr>
<td>3000</td>
<td>1.07</td>
<td>1.11</td>
<td>1.13</td>
</tr>
<tr>
<td>3500</td>
<td>1.06</td>
<td>1.09</td>
<td>1.11</td>
</tr>
<tr>
<td>4000</td>
<td>1.04</td>
<td>1.07</td>
<td>1.09</td>
</tr>
<tr>
<td>4500</td>
<td>1.03</td>
<td>1.05</td>
<td>1.07</td>
</tr>
<tr>
<td>5000</td>
<td>1.02</td>
<td>1.04</td>
<td>1.06</td>
</tr>
</tbody>
</table>

To obtain the isothermal compressibility ($\kappa_\circ$), the specific volume data, at a temperature of 20°C, was first regressed (squared in pressure) and then differentiated. Using the definition of isothermal compressibility, $\kappa_\circ = \frac{1}{v} \frac{\partial v}{\partial P}_T$, the
differential was divided by the second-order polynomial and then the entire expression was evaluated at 1 bar. This method yielded a smooth estimate of $6.03(10)^{-4}$ 1/MPa for the isothermal compressibility.

Similarly, the isobaric compressibility ($\beta_o$) was obtained by regressing the specific volume data, linear in temperature, at a pressure of 1 bar, and then differentiating. Using the definition of isobaric compressibility, $\beta_o = \frac{1}{v} \left( \frac{\partial v}{\partial T} \right)_P$, the expression, when evaluated at 25°C, generated an estimate of $9.88(10)^{-4}$ 1/°C for the isobaric compressibility. Because of the linearity noticed between the specific volume and the temperature, the isobaric compressibility for kerosene expressed as a constant was reasonable.

The specific heat at constant pressure ($c_p$) was calculated from measured enthalpy values ($h$) for kerosene (Lenoir and Hipken; 1973). The liquid enthalpy data, at 20 psia, was first regressed (cubic in temperature) and then differentiated. The specific heat at constant pressure, defined as $\left( \frac{\partial h}{\partial T} \right)_P$, was generated from this differential. The differential, when evaluated at 24°C, yielded an estimate of $1.73(10)^{-3}$ MJ/kg-°C for the specific heat ($c_p$).

The specific heat at constant volume ($c_v$) was computed from the following familiar quasi-static thermodynamic relation.

$$c_p - c_v = \frac{vT\beta_o^2}{\kappa_o} \quad (6-1)$$

The specific volume ($v$) of kerosene at 24°C (T) is $1.247(10)^{-3}$ m$^3$/kg (Lenoir and Hipken; 1973). From equation (6-1), the specific heat at constant volume was $1.13(10)^{-3}$ MJ/kg-°C.
Finally, the surface tension of kerosene may be approximated by using the surface tension of the hydrocarbon dodecane. It was noticed by Goodger (1975), that the overall average properties of kerosene are roughly equivalent to dodecane. Therefore, $2.49 \times 10^{-2}$ kg/sec$^2$ will represent the maximum value of the surface tension for kerosene (Thermodynamic Tables Hydrocarbon; 1985).

6.2 Gas Production Data

The dissociation of a solution by the passage of high-energy ionizing radiation generates radiolytic gas along the path of the radiation. The dissociation rate ($G$), required for the simulation, can be expressed as the maximum mass of gas produced per energy absorbed by the solution. In a TBP-kerosene solution, the principal product of radiolysis is hydrogen. The dissociation rate for hydrogen in a 19% TBP-kerosene solution has been evaluated by Stieglitz (1982). It was found that 1.14 molecules of hydrogen are produced when the TBP-kerosene solution absorbs 100 eV of energy. Thus, the dissociation rate for hydrogen can be converted into $2.36 \times 10^{-4}$ kg of hydrogen per MJ.

The continual dissociation of the solution by radiation will produce a supersaturated concentration of radiolytic hydrogen gas. Bubble nucleation begins when the concentration of dissolved gas reaches the supersaturated threshold. The threshold concentration ($x_T$) observed in the KEWB experiments was $2.20 \times 10^{-5}$ kg of hydrogen per kg of solution (Spielger; 1962). There is no reason to doubt that this is a reasonable value for the threshold concentration in kerosene.
When the threshold concentration is reached, bubbles are nucleated and grow by the diffusion of dissolved gas. It has been observed, however, that the excursion dynamics for the quasi-static thermodynamic model are not sensitive to the bubble size (Hetrick; 1987). Therefore, due to the unavailability of certain bubble growth parameters required for the simulation, reasonable approximations for the following bubble growth parameters are acceptable.

The gas diffusivity ($D$) for the system may be approximated by a system of air dissolved in water. The value of the gas diffusivity for this system is $2.0(10)^{-9}$ m$^2$/sec (Epstein and Plesset; 1950). The initial radius of a bubble nucleated by a fission track was calculated by Spiegler (1962) using the KEWB data. The hydrogen threshold concentration was successfully correlated with the liquid temperature using an initial bubble radius of $5.0(10)^{-8}$ meters. Finally, the saturated concentration of dissolved gas can be approximated using the CRAC data presented by Mather and Shaw (1984). The ratio between the saturated and supersaturated concentration for the CRAC experiments generated a fraction by which the supersaturated concentration ($x_T$) may be multiplied by to predict the saturated concentration ($x_{sat}$). When the saturated concentration, derived from Henry's Law, is divided by the supersaturated threshold (observed in the CRAC experiments), a ratio of 0.436 is produced. When the supersaturated concentration is multiplied by this fraction, the saturated concentration is $9.60(10)^{-6}$ kg of hydrogen gas per kg of solution.

6.3 **Data Summary**

The foregoing physical properties of kerosene are presented in Table 6.2.
Table 6.2

Physical Constants of Kerosene: Los Alamos Criticality Accident

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$V$</td>
<td>Total solvent volume</td>
<td>$m^3$</td>
<td>0.160</td>
</tr>
<tr>
<td>$v_t$</td>
<td>Specific volume</td>
<td>$m^3/kg$</td>
<td>$1.25(10)^{-3}$</td>
</tr>
<tr>
<td>$M$</td>
<td>Total solvent mass</td>
<td>$kg$</td>
<td>128.0</td>
</tr>
<tr>
<td>$\kappa_o$</td>
<td>Isothermal compressibility</td>
<td>1/MPa</td>
<td>$6.03(10)^{-4}$</td>
</tr>
<tr>
<td>$\beta_o$</td>
<td>Isobaric compressibility</td>
<td>1/°C</td>
<td>$9.88(10)^{-4}$</td>
</tr>
<tr>
<td>$c_p$</td>
<td>Specific heat-constant pressure</td>
<td>$MJ/kg°C$</td>
<td>$1.73(10)^{-3}$</td>
</tr>
<tr>
<td>$c_v$</td>
<td>Specific heat-constant volume</td>
<td>$MJ/kg°C$</td>
<td>1.13 (10)^{-3}</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>Liquid surface tension</td>
<td>$kg/sect^2$</td>
<td>2.49(10)^{-2}</td>
</tr>
<tr>
<td>$G$</td>
<td>Radiolytic hydrogen formation constant</td>
<td>$kg/MJ$</td>
<td>2.36(10)^{-4}</td>
</tr>
<tr>
<td>$x_T$</td>
<td>Supersaturated concentration</td>
<td>$kg(gas)/kg(soln.)$</td>
<td>2.20(10)^{-5}</td>
</tr>
<tr>
<td>$x_{sat}$</td>
<td>Saturated concentration</td>
<td>$kg(gas)/kg(soln.)$</td>
<td>9.60(10)^{-6}</td>
</tr>
<tr>
<td>$R_g$</td>
<td>Hydrogen gas constant</td>
<td>$MJ/kg°C$</td>
<td>4.16(10)^{-3}</td>
</tr>
<tr>
<td>$D$</td>
<td>Gas diffusivity</td>
<td>$m^2/sec$</td>
<td>2.00(10)^{-9}</td>
</tr>
<tr>
<td>$r_o$</td>
<td>Initial bubble radius</td>
<td>$m$</td>
<td>5.0(10)^{-8}</td>
</tr>
</tbody>
</table>
CHAPTER 7.
RESULTS

The excursion dynamics for the Los Alamos accident were computed by the equations of point kinetics and quasi-static thermodynamics. In this chapter, the simulation output for the accident will be presented. Also, the time scale of the accident will be approximated.

7.1 Simulation Procedure

The fission dynamics and the equations of state, energy, and momentum were input for the fortran based DARE P Continuous System Simulation Language. DARE P, developed at the University of Arizona, is a user-oriented simulation package that numerically solves ordinary differential equations for a selected integration routine (Wait; 1983). EPISODE (Experimental Package for the Integration of Systems of Ordinary Differential Equations), developed by the Lawrence Livermore Laboratory, is one of thirteen routines offered by DARE P. EPISODE, excellent for stiff equations, performed the numerical integration on the inputed equations.

The DARE P numerical integration required the initial conditions for the point kinetics and the equations of state, energy, and momentum. The atmospheric
pressure (0.1 MPa) and the ambient temperature (20°C) were chosen for the initial state of the solvent. The volume of the solvent was initially 160 liters. The initial energy of the system was set to zero, and the initial power was chosen arbitrarily at one kilowatt. The solvent did not initially generate 1 KW of power, but this initial condition was inconsequential for the simulation. The initial power for the simulation merely translates the burst in time. Finally, all the precursor concentrations were initialized to zero. The initial conditions for the simulation are presented in Table 7.1. Other system parameters have been presented in chapters 5 and 6.

Table 7.1

Initial Conditions for the DARE P Simulation

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Equation</th>
<th>Initial Condition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure</td>
<td>Equation of State ((2 - 1))</td>
<td>0.1 MPa</td>
</tr>
<tr>
<td>Temperature</td>
<td>Equation of Energy ((2 - 17))</td>
<td>20°C</td>
</tr>
<tr>
<td>Volume</td>
<td>Momentum Equation ((2 - 19))</td>
<td>0.160 m³</td>
</tr>
<tr>
<td>Energy</td>
<td>Integrated Power ((5 - 5))</td>
<td>0.0 MJ</td>
</tr>
<tr>
<td>Power</td>
<td>Point Kinetics ((5 - 4))</td>
<td>1.0 KW</td>
</tr>
<tr>
<td>Precursor Density</td>
<td>Point Kinetics ((5 - 4))</td>
<td>0.0 KW</td>
</tr>
</tbody>
</table>

To initiate the excursion for the simulation, a step insertion of positive reactivity was inputed. 1.29 dollars of positive reactivity above critical was required to generate the total 4.8 MJ of energy produced in the accident. Converging on the 4.8 MJ of energy for the simulation, the power rose on a maximum reciprocal period of 15.37 sec\(^{-1}\). A summary of the simulation output is presented in the following section.
7.2 Output Data

The simulation of the accident generated some interesting results. To begin with, the simulation demonstrated the absence of bubble nucleation during the excursion. Unlike an aqueous solution where the dissociation of water produces both hydrogen and oxygen gas, the dissociation of kerosene yields only hydrogen. Hence, the supersaturated concentration of dissolved gas required for bubble nucleation was never achieved. It was estimated by equation (2-12), using the data in Table 6.2, that 11.9 MJ of energy must be absorbed by the solution before bubbles are nucleated. The accident generated only 4.8 MJ. Therefore, the nuclear burst was terminated solely by the thermal expansion of the solvent.

For the simulation to generate 4.8 MJ of energy, a step insertion of 1.29 dollars of positive reactivity above critical was required. The power increased on a maximum reciprocal period \((\omega_m)\) of 15.37 sec\(^{-1}\), where \((\omega_m)\) measures the maximum reactivity excess above prompt critical. The power peaked at 17.8 MW and the trace is plotted in Figure (7.1). Without the feedback reactivity associated with bubbles, a delayed neutron tail on the power trace developed. The power leveled off at approximately 1.9 MW. At the onset of the plateau, the total energy of the system was approximately 4.8 MJ (Figure 7.2). The total reactivity of the system is presented in Figure (7.3).

The pressure of the system was an artifact of the momentum dissipation constant \((A)\) in the equation of motion (2-19). Although the neutronics of the system were not sensitive to the dissipation constant, the peak pressures were incremented uniformly with the values of the dissipation constant. In the simulation of the
KEWB experiments, the peak pressures were successfully correlated with the maximum reciprocal periods for a dissipation constant of 2500 sec\(^{-1}\). It would not be prudent to assume this value for the accident, therefore Figure (7.4) presents peak pressure increases for various dissipation constants. The pressure increase for the system was represented as \(2(P - P_o)\), where \(P_o\) is the atmospheric pressure and \(P\) is the equation of state pressure. A typical pressure trace is shown in Figure (7.5).

The increase in the solvent volume is presented in Figure (7.6). The solvent expanded to 164 liters, corresponding to a relative decrease in solvent density of 2.44\%. Therefore, the linearity chosen between the first two MCNP data points for the least squares approximation was justified. The temperature rose to approximately 48°C (Figure 7.7), and therefore steam voids did not contribute any feedback reactivity.

The simulation output is summarized in the following plots.
Figure 7.1. Plot of Power versus Time: $\omega_m=15.37 \text{ sec}^{-1}$
Figure 7.2. Plot of Pulse Energy versus Time: $\omega_m=15.37 \text{ sec}^{-1}$
Figure 7.3. Plot of Reactivity versus Time: $\omega_m = 15.37 \text{ sec}^{-1}$
Figure 7.4. Plot of Peak Pressure Increases for Various Momentum Dissipation Constants: $\omega_m=15.37 \text{ sec}^{-1}$
Figure 7.5. Plot of Peak Pressure Increase versus Time for a Momentum Dissipation Constant of $5 \times 10^4 \text{ sec}^{-1} \omega_m = 15.37 \text{ sec}^{-1}$
Figure 7.6. Plot of Solvent Volume versus Time: $\omega_m = 15.37 \text{ sec}^{-1}$
Figure 7.7. Plot of Temperature versus Time: $\omega_m = 15.37 \text{ sec}^{-1}$
7.3 Time Scale of the Accident

With the maximum reciprocal period defined, the time scale of the accident was estimated. It was assumed that a source neutron from the spontaneous fission of Pu$_{240}^{240}$ sponsored a persistent fission chain for a near critical assembly. The initial neutron population ($N_o$) was approximated by the “shutdown power level,” derived from steady state point kinetics (Hetrick: 1971).

\[
N_o = \frac{q_o}{\rho_o/\beta_d} \frac{\ell}{\beta_d}
\]  \hspace{1cm} (7 - 1)

where

\[
q_o \equiv \text{effective source strength}
\]

\[
\rho_o/\beta_d \equiv \text{initial subcritical reactivity}
\]

\[
\frac{\beta_d}{\ell} = 49.46.
\]

The source strength was approximated using the half life and the number of prompt neutrons released for the spontaneous fission of Pu$_{240}^{240}$. The decay constant (1.8(10)$^{-19}$ sec$^{-1}$), obtained from the half life of Pu$_{240}^{240}$ (1.2(10)$^{11}$ years), was multiplied by the number of atoms per mass of material to generate the spontaneous fission frequency per mass of plutonium. This frequency, when multiplied by the number of prompt neutrons released in the spontaneous fission of Pu$_{240}^{240}$ (2.17 neutrons/fission) and the estimated 5w% of Pu$_{240}^{240}$ present in the vessel (0.164 kg), predicted an effective source strength of 1.63(10)$^5$ neutrons per second.

The initial subcritical state was computed by subtracting the net positive reactivity (MCNP output) from the step input reactivity of 1.29 dollars above critical (point kinetics). From the supercritical geometry (MCNP case # DHE 031),
5.85±2.16 dollars of net reactivity was induced by the distortion of the solvent. Therefore, the system was assumed to be initially 4.56 dollars below critical.

From equation (7-1), the initial neutron population was 723 neutrons. The initial power of the system was approximated by multiplying the initial neutron population by the neutron thermal velocity \(2.2(10)^5\) cm/sec and the macroscopic fission cross section \((0.037\ \text{cm}^{-1})\). Assuming that 200 MeV of energy is released per fission, the initial power was \(1.9(10)^{-4}\) watts.

The power of the system \((n)\) was governed by the following expression.

\[
n = n_0 e^{\omega_m t}
\]

where

\[
n_0 \equiv \text{initial power}
\]

\[
\omega_m \equiv \text{maximum reciprocal period}
\]

\[
t \equiv \text{time}
\]

For the step insertion approximation, the power rose with the maximum reciprocal period of 15.37 sec\(^{-1}\) to the simulation's initial condition, 1 KW. Equation (7-2), upon solving for time, represents the time it took for the system to reach 1 KW from the initial subcritical state, if a source neutron was present to initiate a persistent fission chain. Equation (7-2) generated 1.01 seconds.

From the simulation, the time for the system to reach peak power from 1 KW was approximately 0.7 seconds. Therefore, if took approximately 1.71 seconds for the system to reach peak power from the initial subcritical state. This, however, is a lower limit because it is evident that the system's response was induced by a ramp rate of reactivity rather than by a step insertion.
CHAPTER 8.
CONCLUSIONS

The simulation of the Los Alamos accident was based upon the application of quasi-static thermodynamics and space-independent point kinetics. The accuracy of the simulation is hence limited to the application of both models. However, for a defined feedback coefficient of reactivity, this method successfully correlated the maximum reciprocal periods with the peak powers and pressures for both the KEWB and CRAC experiments. The application of the point kinetics and the quasi-static thermodynamics, therefore, has warranted further attention.

The uncertainty in the simulation lies in the uncertainty in the solution configuration during the accident. The paraboloid of revolution, assumed at the organic-aqueous interface, was a reasonable and convenient choice for the solution geometry. The minimal distortion of the interface, as evident in the selection of only four centimeters for the paraboloid height, is realistic when compared to the relatively short time scale of the accident (1.71 sec). However, the time scale also suggests an asymmetric geometry at the interface, because it is believed that this was not a sufficient amount of time for the solution to achieve a symmetric geometry. Only an experimental simulation of the tank system would reveal the true solution configuration.
Likewise, the maximum pressure created by the rapid deposition of radiative energy remains a mystery. It is apparent that a sufficient amount of pressure was required to displace the one ton tank. Until a more sophisticated hydro–dynamic model is developed, the quasi–static thermodynamic simulation can only generate a possible range of peak pressures for the accident.

Further attention should be made to the dissociation of kerosene by high–energy radiation. Although the principal product of radiolysis is hydrogen, a small fraction of hydrocarbons are produced. This small fraction may contribute to a supersaturated concentration of dissolved gas, which was absent in the simulation. If the supersaturated threshold were attained, fission track nucleation of radiolytic gas bubbles would provide additional negative reactivity. However, it is still uncertain whether the hydrocarbon fraction is significant.

The accuracy of the simulation hinges upon the reliability of the MCNP–derived volume coefficient of reactivity. The MCNP output and the least squares derivation of the feedback coefficient yielded reasonable results for the step insertion approximation. The power rose on a maximum reciprocal period of 15.37 sec\(^{-1}\) for a step insertion of 1.29 dollars above critical. It is intuitive, however, that the input reactivity was induced by a ramp rather than by a step insertion of reactivity. To seriously proceed with the accident simulation, an actual hydro–dynamic experiment must be performed to correlate the ramp reactivity rate with the distortion of the solvent layer. The author hopes that the results from this report will provide the astute observer the means to approximate possible ramp rates for future simulations.
APPENDIX A.
NOMENCLATURE

\[ A \equiv \text{Tank cross sectional area} \]
\[ a_0 \equiv \text{Quadratic regression coefficient} \]
\[ a_1 \equiv \text{Quadratic regression coefficient} \]
\[ a_2 \equiv \text{Quadratic regression coefficient} \]
\[ c(r, t) \equiv \text{Dissolved gas concentration outside a bubble} \]
\[ c_i \equiv \text{Precursor Density} \]
\[ c_{sat} \equiv \text{Saturated concentration of dissolved gas} \]
\[ c_T \equiv \text{Supersaturated concentration of dissolved gas} \]
\[ c_p \equiv \text{Specific heat at constant pressure} \]
\[ c_v \equiv \text{Specific heat at constant volume} \]
\[ D \equiv \text{Gas diffusivity} \]
\[ E \equiv \text{Nuclear energy in a pulse} \]
\[ f \equiv \text{Volume fraction of gas in bubbles} \]
\[ G \equiv \text{Radiolytic hydrogen formation constant} \]
\[ H_b \equiv \text{Maximum height of paraboloid of revolution} \]
\[ k \equiv \text{Effective multiplication factor} \]
\( \ell \equiv \text{Neutron generation time} \)

\( L \equiv \text{Thickness of solvent layer extension} \)

\( m \equiv \text{Mass of gas in a bubble} \)

\( M \equiv \text{Mass of solution} \)

\( n \equiv \text{Power} \)

\( n_0 \equiv \text{Initial power} \)

\( n_p \equiv \text{Neutron population} \)

\( N \equiv \text{Mass of gas in bubbles} \)

\( N_0 \equiv \text{Initial neutron population} \)

\( n_b \equiv \text{Total number of bubbles} \)

\( P \equiv \text{Liquid pressure} \)

\( P_i \equiv \text{Pressure in a bubble} \)

\( P_0 \equiv \text{Atmospheric pressure} \)

\( q_0 \equiv \text{Effective source strength} \)

\( r \equiv \text{Radial coordinate centered at bubble center} \)

\( r_b \equiv \text{Bubble radius} \)

\( r_0 \equiv \text{Initial bubble radius} \)

\( R_g \equiv \text{Hydrogen gas constant} \)

\( s \equiv \text{Laplace transform parameter} \)

\( t \equiv \text{Time} \)

\( t_N \equiv \text{Time of bubble nucleation} \)

\( T \equiv \text{Liquid temperature} \)
\[ u \equiv r(c-c_T) \]
\[ \bar{u} \equiv \text{Laplace transform of } u \]
\[ v \equiv \text{Specific volume of the solution} \]
\[ v_g \equiv \text{Specific volume of a gas} \]
\[ v_\ell \equiv \text{Specific volume of a liquid} \]
\[ v_z \equiv \text{Velocity of solution's center of mass in upward } z \text{ direction} \]
\[ V \equiv \text{Total solution/solvent volume} \]
\[ V_g \equiv \text{Total volume of gas in bubbles} \]
\[ V_0 \equiv \text{Initial solution/solvent volume} \]
\[ V_\ell \equiv \text{Total volume of liquid} \]
\[ x \equiv \text{Mass fraction of gas in bubbles} \]
\[ x_E \equiv \text{Mass fraction of dissolved gas} \]
\[ x_{sat} \equiv \text{Mass fraction of dissolved gas at saturation} \]
\[ x_T \equiv \text{Mass fraction of dissolved gas at supersaturated threshold} \]
\[ z \equiv \text{Vertical coordinate in upward direction} \]
\[ \beta \equiv \text{Isobaric compressibility of solution containing bubbles} \]
\[ \beta_d \equiv \text{Delayed neutron fraction} \]
\[ \beta_i \equiv \text{Individual precursor delayed neutron fraction} \]
\[ \beta_o \equiv \text{Isobaric compressibility of liquid} \]
\[ \kappa \equiv \text{Isothermal compressibility of solution containing bubbles} \]
\[ \kappa_o \equiv \text{Isothermal compressibility of liquid} \]
\[ \lambda_i \equiv \text{Decay constant for precursor density} \]
\( \Lambda \equiv \text{Momentum dissipation constant for the solution} \)

\( \rho \equiv \text{Total reactivity} \)

\( \rho_f \equiv \text{Feedback reactivity} \)

\( \rho_0 \equiv \text{Step input reactivity above critical} \)

\( \rho_b \equiv \text{Density of gas in a bubble} \)

\( \sigma \equiv \text{Liquid surface tension} \)

\( \phi_v \equiv \text{Density coefficient of reactivity} \)

\( \omega_m \equiv \text{Maximum reciprocal period} \)
REFERENCES


Thermodynamic Research Center, Thermodynamic Tables Hydrocarbons, Texas A&M University System, College Station, Texas, (6), p 1010 (1985).