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Improvement and Verification of
Fast Reactor Safety Analysis Techniques

Progress Report

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and Dean J. Bybee

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ABSTRACT

This report deals with the simulation of a HCDA in a LMFBR using chemical heating. The chemicals used were acetyl chloride and dimethyl sulfoxide in a benzene base. In our last report, it was shown that for high concentrations of the two chemicals, vapor velocities above 8 cm/sec and void fractions above 50% were obtained in a closed system. However, at lower concentrations, such as 4 M, no void fraction was obtained. During this period of work, a closed system with a cooling coil was used to provide a larger heat sink. It was shown that for the 4 M concentration void fractions above 50% was obtained and for the 9 M concentration the void fraction was higher and stayed dispersed much longer than without the cooling coil.

I. INTRODUCTION

During this period of contract work, research was continued on the closed system simulation of a HCDA in a LMFBR, this time using a cooling coil to provide a larger heat sink. Also included in this report is Paul Wheeler's dissertation. This report contains the following:

1) experimental apparatus and procedure; 2) the results, 3) the conclusion, and 4) the dissertation.

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II. APPARATUS AND PROCEDURE

The experimental apparatus used in this study is shown in Fig. 1. It consisted of a glass test tube 200 mm long and 32 mm in diameter, a rubber stopper, three stainless steel tubes used for injection and temperature-pressure measurements, a 4 mm glass tube wrapped around the test tube and used as a cooling coil and a differential amplifier was used to measure the difference between the inlet and outlet temperature. Thermistors were used to measure temperatures and a pressure transducer used to measure pressure. These signals were recorded on voltage recorders.

To proceed with the experiment, the test tube was placed on a piece of styrofoam to cushion it and held upright by a clamp. A small amount of bakelite was then added to give the fluid nucleation sites. Ten ml of acetyl chloride was placed in the test tube and the stopper secured in place by a clamp. Ten ml of dimethyl sulfoxide (DMSO) was next put in the syringe, the syringe connected to the injection line and then placed in a syringe pump. The pressure release valve was closed and the syringe pump started. The camera was started and also a timer. After the reaction was completed the apparatus was cleaned and another run made. This was done for both 4 and 9 molar concentrations and again with different flow rates of water flowing in the cooling coil.

III. RESULTS

The vapor velocities, shown in Fig. 2 were calculated using the following equation

$$U_V = V/A((fmgCg/m_V h_{fg} - 1/T)dT/dt = dP/Pdt)$$

(See Nomenclature)

developed in our last report², and then adding the amount of heat removal by the coiling coil. Curve A shows the 4 m run without cooling and Curve B shows the run with cooling. The temperature change in the coiling coil was .18°C with a water flow rate of 5.81 ml/su. This removal heat at the rate of 1.05 cal/sec. This 14% increase in an upward heat sink produced vapor velocities above 8 cm/sec. Curve C and D show the 9 m runs without and with cooling, respectfully. For the cooling the temperature change was .75°C with a flow rate of 5.81 ml/sec or a heat removal of 4.36 cal/sec. This was a 12% increase and produced higher vapor velocities for a longer period of time. From the films, it was seen that for the 4 m without cooling, no void fraction was observed and with cooling, void fractions above 50% were obtained for an extended period of time. The 9 m with cooling resulted in a higher void fraction and also for a longer period of time than the 9 m without cooling.

IV. CONCLUSIONS

In conclusion, this period of contract work has shown that in a closed system using an internally heated fluid void fractions above 50% can be obtained using a small upward heat sink where normally no void fraction was observed. For a system where a void fraction exists, a small upward heat sink can increase the void fraction and extend the duration of the boil up.

NOMENCLATURE

U_v	Vapor Velocity	cm/sec
V	Volume of Container	cm^3
A	Cross-Sectional Area	cm^2
f	Fraction of Test Tube	.25
M_g	Mass of Test Tube	gm
L_g	Heat Capacity of Test Tube	cal/gm°C
M_v	Mass of Vapor	gm
h_{fg}	Heat of Vaporization	cal/gm
T	Temperature	°C
dT	Change in Temperature	°C
P	Pressure	mm/Hg
dP	Change in Pressure	mm/Hg
dt	Change in Time	sec

Figure 1. Closed Reaction Vessel with Cooling

Figure 2. Vapor Velocities with Cooling

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APPENDIX

CHEMICAL SIMULATION MODELING
OF FUEL MOTION IN LMFBRs

A Dissertation
Presented to the
Department of Chemical Engineering
Brigham Young University

In Partial Fulfillment
of the Requirements for the Degree
Doctor of Philosophy

by
Paul A. Wheeler
December 1978

CHEMICAL SIMULATION MODELING
OF FUEL MOTION IN LMFBRs

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Department of Chemical Engineering

Ph.D. Degree, December 1978

ABSTRACT

Chemical reactions were used to simulate fuel motion in LMFBR core disruptive accidents. The superficial vapor velocity necessary for transition between bubbly flow and dispersed flow was calculated for the chemical system with the Kutateladze stability criterion. The actual vapor velocity at transition was measured experimentally in an open vessel. It was 50-100% higher than calculated.

Closed vessel experiments showed a decrease in vapor velocity for a given concentration due to pressurization. A small heat sink added sufficient condensation to allow full dispersion of fluids even in a pressurized vessel. These results were compared to accident sequences in an LMFBR core.

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This dissertation by Paul A. Wheeler, is accepted in its present form by the Department of Chemical Engineering of Brigham Young University as satisfying the dissertation requirement for the degree Doctor of Philosophy.

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NOMENCLATURE

<u>Symbol</u>	<u>Definitions</u>
A	Cross sectional area (cm^2)
A	Antoine equation constant
A	Amplitude in normal curve
A_1	Vessel inside surface area (cm^2)
A_2	Vessel outside surface area (cm^2)
B	Antoine equation constant
C	Antoine equation constant
c_g	Glass vessel specific heat ($\text{cal} / \text{gm}^\circ\text{C}$)
c_l	Liquid specific heat ($\text{cal} / \text{gm}^\circ\text{C}$)
f	Fraction of vessel condensing vapor
g	Gravitation constant (cm/sec)
h_1	Heat transfer coefficient, liquid to vessel ($\text{cal}/\text{cm sec}^\circ\text{C}$)
h_2	Heat transfer coefficient, vessel to air ($\text{cal}/\text{cm sec}^\circ\text{C}$)
h_{fg}	Latent heat of vaporization (cal/gm)
I	Electric current (Amps)
I_h	Electric current through heater (Amps)
K	Rate equation constant
k	Kutateladze number
m_g	Mass of glass vessel (gm)
m_l	Mass of liquid (gm)
m_v	Mass of vapor (gm)

$\dot{m}_v$	Mass vaporization rate (gm/sec)
P	pressure (psi or mm-Hg)
P_h	Power in heater (Watts)
$\dot{Q}$	Heat generation rate (cal/sec)
R	Electrical resistance (Ohms)
R_s	Electrical resistance, standard resistor (Ohms)
T	Temperature ($^{\circ}\text{C}$ or $^{\circ}\text{K}$)
T_1	Liquid temperature ($^{\circ}\text{C}$ or $^{\circ}\text{K}$)
T_2	Vessel temperature ($^{\circ}\text{C}$)
T_a	Air temperature ($^{\circ}\text{C}$)
t	Time (sec)
U_v	Superficial vapor velocity (cm/sec)
V_h	Voltage across the heater (Volts)
V_s	Voltage across the standard resistor (Volts)
V	Vessel volume (cm^3)
v_{fg}	Change in vapor specific volume (cm^3/gm)
v_v	Vapor specific volume (cm^3/gm)
w	Rate equation constant
x	Rate equation constant
α	Void fraction
δ	Position of NMR peak
μ	Mean in normal curve
ρ_c	Density of continuous phase (gm/cm^3)
ρ_h	Density of heavy phase (gm/cm^3)
ρ_l	Density of light phase (gm/cm^3)
σ	Surface tension (dyne/cm)

GLOSSARY

AC-	Acetyl Chloride
BWR-	Boiling Water Reactor
CRBR-	Clinch River Breeder Reactor
DBA-	Design Basis Accident
DMSO-	Dimethyl Sulfoxide
FCC-	Federal Communication Commission
FFTF-	Fast Flux Test Facility
FSAR-	Final Safety Analysis Report
HCDA-	Hypothetical Core Disruptive Accident
HTGR-	High Temperature Gas Reactor
LGR-	Light Graphite Reactor
LMFBR-	Liquid Metal Fast Breeder Reactor
LOF-	Loss of Flow Accident
LWR-	Light Water Reactor
MFCI-	Molten Fuel Coolant Interaction
NMR-	Nuclear Magnetic Resonance
NRC-	Nuclear Regulatory Commission
OSHA-	Occupational Safety & Health Administration
PAHR-	Post Accident Heat Removal
PLBR-	Prototype Large Breeder Reactor
PSAR-	Preliminary Safety Analysis Report
PWR-	Pressurized Water Reactor
TOP-	Transient Overpower Accident

CHAPTER 1

INTRODUCTION

All engineering structures, whether buildings, dams, vehicles or power plants, present some inherent risk to their operators or to the public. The responsibility for protection of the public lies with the local or national government. The Nuclear Regulatory Commission (NRC) maintains authority, from the Federal government, for the regulating and licensing of nuclear power plants. Because of the strict, conservative requirements imposed by the NRC, there has not been a death nor injury from a nuclear accident in the twenty years of existence of commercial power plants. The continuation of this excellent safety record in new and larger plant designs requires extended research.

The Liquid Metal Fast Breeder Reactor

Approximately 95% of the commercial nuclear power plants in operation or in the licensing process in the United States are light water reactors (LWRs). Light water (as opposed to heavy water containing the isotope deuterium) is used as a coolant and moderator. When neutrons are produced in fission their energies average about 1 MeV. The moderator slows down these fast neutrons to the thermal range of about 0.02 eV before they produce further fissions. The two types of LWRs in current use in the United States are the

pressurized water reactor (PWR) and the boiling water reactor (BWR).

There are a few high temperature gas reactors (HTGR) in the licensing process. They are helium cooled, graphite moderated reactors. One light graphite reactor (LGR), a light water cooled, graphite moderated reactor, is operating.

The liquid metal fast breeder reactor (LMFBR) has a design which is vastly different from other reactor designs. The first difference is that the LMFBR is designed to operate with fast neutrons rather than thermal neutrons. This precludes the need of a moderator. Since the fission cross section (the ability to use neutrons for fission) is less for fast neutrons than for thermal neutrons, the core density is greater in an LMFBR than is a thermal reactor.

The second difference between the LMFBR and other reactors is its breeding capability. Where other reactors only consume fuel, the LMFBR is designed with a blanket of ^{238}U which produces ^{239}Pu upon absorbing a neutron and subsequent decay. The ^{239}Pu is a fuel that can be used in other reactors. The process of producing more fuel than is consumed is called breeding.

The last major difference between the LMFBR and other reactors is that it uses liquid sodium as a coolant. These fundamental differences present unique safety considerations in analyzing LMFBR accidents.

The current LMFBR program in the United States consists of three stages. The first stage is the production of the Fast Flux

Test Facility (FFTF). This facility, currently under construction at Richland, Washington, is designed to test the response of materials to a flux of fast neutrons. It will not produce any electrical power.

The Clinch River Breeder Reactor (CRBR) is the second step in LMFBR production. The CRBR is the first large-scale (350-400 MW) demonstration breeder nuclear power plant. It is presently in the licensing stage with plans for building near Oak Ridge, Tennessee.

The Prototype Large Breeder Reactor (PLBR) is the last step toward full commercialization of LMFBRs. Now only in the design phase, the PLBR will be a full scale commercial plant.

Licensing of Fast Reactors

With more than sixty commercial light water reactors presently operating in the United States, the main emphasis in safety research for them is to streamline licensing procedures and verify the safety guidelines already established.

The situation is somewhat different with the licensing of commercial LMFBRs. Since there has been only limited experience in licensing fast reactors in this country, specific guidelines are still in the negotiating stage. The Clinch River Breeder Reactor will set the precedent for future licensing of LMFBRs. Extensive research is needed to answer the various safety questions raised during the licensing procedure.

In the licensing process a design basis accident (DBA) is identified. A DBA is a low-probability accident with major

consequences (such as a loss of coolant accident in LWRs), which becomes the explicit basis for design (1). The accident is studied in detail, and the power plant is designed to withstand the accident.

A statement of safety compliance with NRC regulations is required in two reports: the preliminary safety analysis report (PSAR) filed prior to receiving a construction permit, and the final safety analysis report (FSAR) filed prior to receiving an operating licence (2). These reports explain the DBA used in design and the facility's ability to withstand it.

Since the core melt accident is not included as a design basis accident for light water reactors, the NRC has decided not to include the hypothetical core disruptive accident (HCDA) as a design basis accident in LMFBRs. An HCDA is an accident in which the original geometry of the core is altered by melting or an energetic disruption. The philosophy of the NRC is that LMFBRs should achieve a level of safety comparable to current generation LWRs. It must be shown that the probability of an HCDA is low enough to not be considered a design basis accident (3).

Since the CRBR is the first commercial reactor of its type to apply for licensing, there has been no precedent set in terms of risk analysis. There is no "Wash 1400" for LMFBRs and little operating experience for this design. For this reason the NRC has set ultra conservative requirements before the CRBR can be licensed. Besides the preliminary and final safety reports, which state the ability of the CRBR to handle a DBA, the NRC has required the CRBR-3, which is a two volume topical report assessing the ability

of the CRBR to accommodate an HCDA. This report establishes both a thermal margin and a structural margin beyond the DBA. The thermal margin calls for total containment of core materials for 24 hours in a hypothetical accident leading to total core melt in 1000 seconds. The structural margin requires structural integrity of the reactor vessel for an accident leading to 1200 Megajoules of work energy (4).

There is presently a strong debate over the energy containment required for proper structural margin. The industry contests that a 661 MJ margin is sufficient for any credible accident sequence (5), whereas the NRC maintains the need for a 1200 MJ margin (6). In reality the scenario of these hypothetical accidents is uncertain. The numbers presented are the results of extended analytical models in the form of large computer codes. Many of the assumptions and input parameters used in these codes have not, as yet, been experimentally verified.

Statement of Problem

One of the greatest uncertainties in the work-energy produced by a nuclear accident is the location and motion of the fuel after the initial geometry of the core has been lost (7). Several analytical and theoretical models have been proposed to describe the motion of the fuel during an accident. There has been, in the past, insufficient experimental data to verify the models. In-pile experiments are prohibitively expensive; therefore, it was necessary to simulate the accident with non-nuclear materials to verify previous models.

A unique feature of nuclear energy is that the heat source is volumetric. Experiments used to model reactor accidents must also have volumetric heating. In the past, electrical and microwave heating were used in modeling. The first objective of this dissertation is to show the feasibility of using chemical reactions to model full core accidents in LMFBRs.

The main question to be answered is the nature of the two-phase flow for an internally heated fluid in both an open vessel and a sealed system. Another question is the minimum power needed to maintain a dispersed flow regime in both an open and a closed system. The second objective of this dissertation is to determine if internally heated fluids are by nature dispersive even at low power levels.

Summary of Chapters

A description of the hypothetical core disruptive accident is presented in chapter 2. Starting with the accident initiation phase, the accident may terminate early or undergo a type of core disruption. The last part of any accident evaluation is final heat removal and damage assessment.

Since this research was to determine the nature of fuel motion, chapter 3 discusses the current study of flow regimes. Fauske's model of dispersed flow is presented, along with problems encountered by his model. Other analytical models are also discussed. Experimental models relating to fuel motion are presented.

Chapter 4 evaluates the role of chemical reactions for modeling fuel motion in an HCDA. Chemical properties are

evaluated along with advantages and disadvantages of using chemicals as an internal heat source.

The experimental portion of the research is presented in the next three chapters. Chapter 5 covers the calorimeter studies performed. Chapter 6 presents the experiments and results carried out in an open vessel system. Chapter 7 reports closed vessel experiments.

The final chapter presents conclusions of the research and future research possibilities using chemical reaction modeling.

CHAPTER 2

THE HYPOTHETICAL CORE DISRUPTIVE ACCIDENT

To insure safety in an LMFBR, three levels of safety design are employed. The first level provides reliable plant operation during normal operating conditions. This is accomplished through quality assurance, redundancy, and in-service inspectability. The second level protects against anticipated and unlikely faults that could occur during the lifetime of the reactor plant. Loss of coolant flow and reactivity insertions are examples. Protection is provided on the second level through redundancy of components, protective features which prevent fault propagation, and control systems for reactivity and coolability. The third level of design deals with very unlikely events, such as severe earthquakes or floods, which are not expected to occur within the lifetime of the plant. A design basis accident is set up which assumes design sufficient to protect the public in event of a level three accident (8).

To guarantee defense-in-depth for the Clinch River Breeder Reactor, design features and safety margins beyond the third level were provided (5). This is the category of the hypothetical core disruptive accident (HCDA), which has very small probability but potentially large consequences. The goal of HCDA research is to insure a probability of less than 10^{-6} per year of a loss of coolable geometry (8). With this low probability established, the HCDA

would not need to be considered in future licensing beyond the Clinch River Breeder Reactor.

HCDA Overview

An overview of the hypothetical core disruptive accident was presented by Jackson, et al. (9). Figure 1 shows a schematic diagram of possible accident sequences.

The starting point of the accident is defined by certain initiating events, such as loss of power to coolant pump or a reactivity insertion. The conclusion of the accident occurs when the core materials are no longer moving so they can cool in place. From this point a source term for radiation can be determined to evaluate the consequences of the accident.

HCDA's are usually discussed in terms of three types of initiating events: the transient overpower accident, in which sudden increase in reactivity (or neutron density) increases the reactor power; the loss of flow accident due to pipe break or pump failure; and the loss of heat sink accident. Each of these accident types could occur with or without a scram (reactor shutdown).

Early termination of an accident is possible if scram occurs and the cooling mechanism remains intact. In this case the core retains, to a great extent, its original geometry. The accident analysis mainly deals with removal of heat after the accident.

If scram does not occur and the cooling mechanism does not remain intact, the initiating events lead to a full core disruption, in which the original core geometry is lost. The severity of the accident is discussed in terms of ramp rate. When sufficient

Figure 1. Schematic Diagram of HCDAs

reactivity is introduced for the reactor to become supercritical, the reactor power will increase exponentially with a period determined by the excess reactivity. Large reactivity insertions are expressed in terms of dollars and cents, where one dollar is prompt critical. At prompt critical the reactor power would increase with a period of about 50 msec. The ramp rate (expressed in units of dollars per second) is the rate the reactivity is increasing.

For large ramp rates (greater than \$20/sec) there is enough fission energy to result in mechanical disassembly. Fuel and steel vapor is produced so fast that the pressures created expand the core and support structure, energetically dispersing the core materials. After the mechanical disruption, the nuclear materials are spread out enough to stop any further nuclear reaction. The structural damage must be evaluated and decay heat removed.

For small ramp rates (less than \$20/sec) there is a gradual meltdown of the core rather than an energetic disruption. Since the LMFBFR core is not in its most reactive state (10), several accident paths are possible. The transition phase treats the accident during the period of melting, mixing, and fuel motion. Once the core has melted, about 60% of the fuel must be removed for the core to become permanently subcritical (11). With various paths possible, the transition phase can itself lead into a mechanical disassembly if the fuel compacts while in motion producing high ramp rates or the fuel can gradually boil out until a permanent subcritical state is reached.

Once sufficient fuel is removed from the core for the reactor to become subcritical, damage to the reactor vessel must be assessed. The vessel should be designed to remain intact (not allowing the radioactive fission products to escape into the containment building) following a mechanical disassembly. The appropriate structural margin needed to contain the accident, 661 MJ or 1200 MJ, is still debatable.

After the work-energy is dissipated, decay heat is still being produced in the core. The reactor vessel should be able to contain the heat produced without letting the core melt through the reactor bottom. This last phase of accident analysis is called post accident heat removal.

The analytical complexity of the HCDA requires several elaborate computer codes to analyze the accident sequence (12). This chapter will discuss the various phases of the HCDA in detail.

Accident Initiation

The initial phase of the HCDA describes the three initiating events: the transient overpower accident (TOP), the loss of flow accident (LOF), and the loss of heat sink accident. The loss of heat sink accident will not be discussed in detail, since it is not treated extensively in the literature and the consequences are enveloped by the LOF and TOP accidents (13).

The TOP and LOF accidents could occur with or without scram. In the event of scram the reactor will be shut down and cool in place. The initiating events of interest are the TOP without scram and the LOF without scram. The two events can be described as an

unprotected maximum-speed rod withdrawal (about 10¢/sec) and an unprotected pump coastdown, respectively.

There are three possible paths for the TOP to follow. There are only two for the LOF (6). Early termination could result from a TOP if small amounts of fuel are swept out of the core from local melting, leaving the rest of the core undisturbed. A mechanical disassembly results from large ramp rates, whereas lower ramp rates will lead to a core melt and the transition phase.

Large amounts of sodium must be voided rapidly from the core for the LOF to go directly into a mechanical disassembly. Although some accidents, in which sodium is removed from the core by boiling, result in high ramp rates, the major concern is with hydrodynamic removal of fuel. When the reactor experiences large power excursions near prompt critical with the core region unvoided, the sodium can be driven from the core by means of fuel-coolant interaction (a sudden vaporization of sodium when it comes in contact with molten fuel) or fission gas release. This voiding effect, due to massive fuel pin failure, is called the LOF driven TOP (14).

For a TOP accident to go into direct mechanical disassembly, small amounts of sodium must be voided while the fuel motion reactivity is zero or positive. One scenario leading to this state is caused by pin failure near the core midplane. Molten fuel moves inside the fuel pin to the failure site and into the channel, causing a fuel coolant interaction which suddenly voids the core. When the reactivity insertion is not large enough for mechanical disassembly, propagation of fuel pin failure leads to the transition phase (5).

The scenario which is usually analyzed, because of its possible consequences, is the LOF accident leading to a total core melt. A summary of the accident sequence following a coolant pump failure without scram was presented by Fauske (15),

The sequence of events leads to a slow but progressive melting of the core. The special sequence is as follows: The flow coasts down and the sodium starts to boil. The power has remained near full power in the absence of scram. After the sodium boils and dryout occurs, the cladding melts, and then the fuel begins to melt. The subassemblies successively progress through the stages of coolant voiding and melting, but the pressures generated are too low to cause a massive dispersal of molten fuel from the core region (absence of hydrodynamic disassembly conditions). The subassembly duct walls melt, and a growing region of molten fuel and steel begins to form in the hottest portion of the core. Enough local axial fuel dispersal has taken place by this time to lower the reactivity and to bring the reactor subcritical. Extended fuel dispersal is inhibited by blockages that form near the axial ends of the core because molten core materials refreeze in the colder regions near the axial ends. Continued decay heating causes further core melting and also causes the melted core materials to boil up and fill the available volume. If paths are available through the upper and lower structures, material will begin to be ejected from the core. If the passages are blocked, rapid melting attack on the upper and lower structures will begin, allowing passages to be open for ejection of molten core materials from the core. If this ejection occurs as a result of the boil-up mechanism, it should have little or no damage potential. Some of the materials may mix with the above core sodium pool and come to rest upon the upper vessel surfaces where they are coolable. The remaining core materials will eventually come to rest on lower surfaces where the material also is coolable.

The analysis of accident initiation is performed by the computer code SAS-3A (16). Preaccident conditions and fault conditions are input into the code. The SAS-3A code follows the accident sequence to the point where the original geometry is lost. The

resulting ramp rate, fuel temperature distribution, power and reactivity worth distributions are calculated.

Initiating events leading to early termination do not cause an accident sequence that disrupts the full core. The core geometry remains intact and can cool in place. For initiating events leading to mechanical disassembly, the output of the SAS-3A code is used as the input for the VENUS-II code (17). The VENUS-II code handles the accident hydrodynamically for large ramp rates (greater than \$20/sec). No codes have been developed that model the transition phase as well as SAS-3A and VENUS-II model the initial and the disassembly phases (12).

Early Termination

The most probable outcome of initiating events is an early termination because of the reliability of the scram systems. If a scram occurs, the neutronics will stop and the core will cool in place without full core involvement. Even in an LOF accident the core would likely cool by natural convection of the sodium (18).

The scenario of the no scram TOP was discussed earlier. The main difficulty in analyzing early termination is assessing the localized damage and final heat removal.

Mechanical Disassembly

The mechanical disassembly occurs when there is a large ramp rate (greater than \$20/sec). The initial conditions leading into the disassembly are given by the results of the SAS-3A code or by engineering hypothesis. The mechanical disassembly is modeled

by the VENUS-II code which calculates the fission energy deposited in the fuel up to the point where Doppler and core expansion feedback shut the core down.

The initiating events can lead directly into disassembly or pass through the transition phase of gradual melting. A recriticality event during the transition phase could produce a ramp rate large enough to disassemble the core (19).

Transition Phase

The transition contains more uncertainties than any other portion of the HCDA. Though SIMMER-I attempts to model the transition phase, there is not a code, as yet, that can handle all of the interacting variables involved during this phase. There is a three-phase, three dimensional mixture of fuel, steel, and fission products. Melting, freezing, condensation and vaporization are all taking place at once, each affecting the heat transfer and pressure production. The fuel motion is uncertain, making the reactivity difficult to follow.

The transition phase occurs when an initiating event, without scram, leads to a low ramp rate and gradual meltdown of the core. Because of the low boiling point of sodium, it is assumed that the core is void of coolant. Steel cladding and fuel melt and relocate. Figure 2 shows a diagram of the FFTF core and support structures which helps illustrate the problems involved.

The radial reflector and shield are made up of stainless steel, as is the lower axial shield. This massive structure tends to inhibit radial and downward motion. The fission gas plenum

Figure 2. FFTF Core and Support Structure

contains hollow stainless steel tubing, providing a large heat transfer area. Any fuel or molten steel motion would likely be in the upward direction.

As steel and fuel melt, they will freeze somewhere in the fission gas plenum, creating a blockage to future fuel movement. Since there are many uncertainties to the accident sequence, several scenarios have been proposed. There is a continual need for experimental research in this portion of the HCDA (12).

The most pessimistic scenario would lead to recriticality. In this scenario, molten fuel is ejected upward by pressures produced in the mid-core regions. The fuel cannot pass out of the core because of blockages that arise from the freezing of molten materials. There is also a possibility of pressurization from the molten fuel coolant interaction. Fuel and steel have also frozen in the lower axial region to cause a completely sealed system.

A sample recriticality calculation was performed by Broudreau and Jackson (19). In this example only the central most subassemblies were molten. The outer subassemblies remained intact. Half of the fuel from the molten subassemblies was injected upward into the fission gas plenum, (stopping due to blockages), while the rest of the fuel formed a pool in the lower core region (see Figure 3). The fuel slug was allowed to fall back into the core region. The fall could be from gravity alone or due to pressure gradients. As the slug of fuel falls back into the core, the reactivity increases as shown in Figure 4, which shows a plot of the reactivity as a function of penetration depth, or the distance the slug

Figure 3. FFTF Core Prior to Recriticality

Figure 4. Excess Reactivity vs Penetration Depth

has fallen toward the pool of fuel. The ramp rate of the recriticality is the product of the slug velocity in cm/sec and the slope of the curve in $\$/\text{sec}$ at the point of prompt criticality.

Many uncertainties exist in the calculation of a criticality model. It is very unlikely that a slug of fuel would be ejected upward and fall coherently. The incoherence of the system depends upon many parameters. Experiments have indicated that severe pressures due to molten fuel coolant interaction is unlikely. The credibility of slug flow is seriously questioned (7).

The recriticality event is the most pessimistic scenario of the HCDA. It has been studied for both the FFTF and the CRBR plants (21-23). Some have claimed that recriticality is a dead issue, but, if it is a dead issue, the body should be exhibited to the public (24).

The accident scenario presented by Fauske (15) suggests fuel dispersal which shuts the reactor down until enough blockage is melted to pass the fuel out of the core. This proposed scenario makes several assumptions that require experimental verification. The first assumption suggests that molten fuel is basically dispersive in nature, therefore, would not slump or compact enough to cause recriticality even at low decay heating levels. The next assumption is that the fuel is unable to mix with the coolant well enough to cause an energetic vapor explosion. Finally, the fuel is coolable on surfaces outside the core region. This means that the fuel would come to rest in a coolable state and not melt through the reactor vessel.

This scenario identifies three key areas where experimental research is needed for HCDA analysis. Fuel flow regimes is one of the most important areas, since it is the fuel motion which determines the reactivity of the system. Fuel flow is treated in detail in the next chapter.

The final event of the transition phase is the removal of fuel from the core to insure permanent shutdown. Some fuel may be removed in the initial phases of the HCDA, but once the whole core has melted, about 60% of the fuel must be removed to insure there is no possibility of future recriticality. As the sealed structure melts, openings will occur in the blockages. Fuel will be ejected through the passages and allowed to cool in the above-core sodium pool.

Damage Evaluation

Because of the many uncertainties involved in the previous phases of the HCDA, the damage evaluation is performed by parametric studies in an attempt to envelope possible events. For a mechanical disassembly, the VENUS-II code calculates the work energy of the event. A conservative estimate of work energy is made by calculating the core pressure then allowing the core volume to expand to atmospheric pressure.

The work energy results of an accident determine the mechanical strength for the reactor vessel to contain an accident. The question of the 661 MJ standard proposed by industry -vs- the 1200 MJ imposed by NRC may be resolved as the most pessimistic events are found experimentally to be not credible.

Post Accident Heat Removal

The HCDA leading to a mechanical disassembly occurs in the order of seconds. The transition accident may extend to minutes. After the fuel is removed from the core so permanent shutdown is achieved, the decay heat keeps the reactor in an unstable state by continued melting. The final result of any nuclear accident is the release of radioactive materials to the atmosphere. If the reactor vessel contains the work-energy released, the only way for the radioactive materials to escape is by melting through the reactor vessel bottom.

The core materials will continue to melt through the lower axial support structure, and possibly even the cement and ground. The post accident heat removal phase attempts to evaluate the heat transfer after the end of the accident until the whole reactor system has cooled. Many experiments have been performed in post accident heat removal (25-27).

The thermal margin set for the CRBR is total containment of core materials within the reactor vessel for 24 hours after an accident that produces a full core melt in 1000 seconds. Much theoretical and experimental research is ongoing in post accident heat removal as well as other phases of the HCDA.

CHAPTER 3

FUEL FLOW REGIMES

The problem of recriticality in an LMFBR accident has received considerable attention in HCDA analysis, mainly because of its potential consequences. Various accident scenarios have been proposed to show that a molten fuel pool will not lead to a recriticality event. The arguments of no-recriticality depend on either extensive fuel removal from the core or a dispersed flow regime due to internal heat generation. Approximately 60% of the fuel in the core would need to be removed to remain permanently subcritical. This dissertation treats the nature of fluid flow as it relates to dispersion of molten fuel in an accident condition.

Theoretical Flow Models

Because of the complexities in the analysis of boiling two-phase flow, it is difficult to define its flow regime. Four major characteristic flow regimes have been identified. They are, in order of increasing vapor content, bubbly flow, slug flow, annular flow, and dispersed flow.

Hosler (28) presented a brief summary of the flow regime studies. The boundary between flow regimes is not well defined because the experimental conditions, leading to empirical correlations, vary from one investigator to another. There is a need for

analytical models defining the transition from one flow regime to another. No clear model has been developed for the molten fuel-steel mixture.

Several experiments and analyses have been performed with low density, gas-liquid systems in order to evaluate the transition from one flow regime to another (29-35). Most of the work was done with air-water systems in vertical channels, both annular and rectangular. Little work has been done with metallic fluids (36). The result of research has defined the cause of flow transitions. Transition from bubbly flow to slug flow is due to collisions between small bubbles, with coalescence of large fractions of these bubbles. The Helmholtz hydrodynamic instability of the liquid film to the bubble interface produces the transition from slug to dispersed flow. Slug flow is a transition of bubbly and annular flow.

There have been many empirical correlations suggested for the initiation of flooding, where the vapor phase rises fast enough to not allow the liquid phase to drain back to lower regions (37-44). Fauske (38) used a nitrogen-water countercurrent flow in vertical tubes to predict upward clad motion. Majumdar (39) used a different interface friction factor to improve Fauske's model. Wallis (42) proposed a correlation based on dimensionless groups which showed that the flooding is dependant on the momentum flux and hydrostatic forces. Kutateladze (43) defined flow regimes in terms of a dimensionless parameter known as the Kutateladze number where

$$k = \frac{U_v \sqrt{\rho_c}}{\sqrt{4\sigma g (\rho_h - \rho_l)}}$$

U_v is the superficial vapor velocity. σ is the surface tension of the liquid. The acceleration of gravity is g , and the heavy, light, and continuous phase densities are ρ_h , ρ_l , and ρ_c .

Kutateladze proposed that the overall flow stability is determined by the stability of the individual elements in the flow system; that is droplets, bubbles, and films. The disturbing factor is the dynamic vapor head in contact with the droplets. The stabilizing factor is the surface tension. The stability criterion is the ratio between the two factors. Table 1 summarizes the value of k as it relates to the flow regime transition.

TABLE 1
VALUES OF PARAMETER k

Nature of Process	k
Breakdown of churn turbulent flow -	$\cong 0.14$ ($\rho_c = \rho_l$)
Drop fluidization of a heavier fluid by a lighter fluid	
Breakdown of bubbly flow	$\cong 0.3$ ($\rho_c = \rho_h$)
Flooding of liquid films	$\cong 3$ ($\rho_c = \rho_l$)
Entrainment of liquid films	$\cong 3.7$ ($\rho_c = \rho_l$)

Besides the empirical correlations obtained through experiments with low density gas-liquid systems, several analytical models have been presented to model internally heated boilup. Lipkis, Liu, and Zuber (45) investigated volume-heated boiling pools by predicting the transient development. The study was limited to early stages of the transient. Neglecting bubble rise effects, they extended the analysis of the growth of a single bubble to a boiling liquid of a known

bubble distribution by summing the contribution of the density change of each bubble. This method agreed with measurements they took in early stages of the density transient.

Condiff and Epstein (46) carried the theory beyond the transient to approach a steady-state limiting pool. In this case bubble travel into and out of a region could not be ignored. They found an analytical solution for the transient of a volume-heated boiling pool using the method of characteristics. They obtained the results of variation of vapor void fraction with time at a given height, the time required to reach steady-state, and the final steady-state height of the pool.

This model was extended to a boiling pool with foaming by Condiff, Epstein, and Grolmes (47). They used kinematic wave and discontinuity analysis for the one-dimensional model. Later they extended their one-dimensional "drift flux" model to a closed system (48). An understanding of the boiling process in a sealed enclosure is necessary in order to determine the pressure response to internal heat generation. The heat removal capability is coupled to flow regime characteristics. They determined that thermal radiation was the predominant heat removal mechanism.

An early computational model of fuel motion from a neutronic point of view was presented by Ostensen and Jackson (49). They developed a model with molten fuel and steel, and steel vapor bubbles. Molten fuel was dispersed uniformly in droplets throughout the fuel. Steel vapor bubbles were in thermodynamic equilibrium with molten steel. A detailed energy balance was performed for each mesh cell.

First order perturbation theory was used to determine the reactivity of the pool. The molten pools were dynamically unstable so that small reactivity perturbations induced rapidly growing oscillations leading to prompt critical excursions.

A second computational model was presented by Abramson (50). He investigated the importance of fuel-to-steel heat transfer on the response of a boiling pool to a uniform overpressure. With high heat transfer, the internally generated pressures overcame the overpressurization in the vessel, retaining dispersion. Pool collapse was predicted if the heat transfer was not high enough.

Experimental Fuel Flow

Several experiments have been performed to evaluate the correlations presented for two-phase flow. Theofanous (41) used both an alcohol/air system and a (Bi-Sn) eutetic/air system to compare models. The Wallis model (34) represented both the alcohol data and the liquid-metal data. The Kutateladze model represented very well the liquid-metal data.

The experimental models of most interest are those using simulant materials to produce internal heat. Three heat generation systems are available for internal heating. They are: electrical (or Joule) heating, microwave heating, and chemical heating.

Although experiments have been carried out for PAHR with Joule heating (26), the major area of interest for the transition phase is fuel flow modeling. Kazimi and Chen (52) used Joule heating to study the void distribution in internally heated boiling pools. The vertical profile of the void fraction and the average void fraction

in an aqueous solution of ZnSO_4 was measured. An analytical correlation was found for the data. Extending the results to a boiling pool of UO_2 they concluded that substantial voiding would occur for even a very small fraction of nominal power in a nuclear reactor. Lipkis, Liu, and Zuber (45) also used electrical heating to verify their model of early transients of boiling pools.

Farahat, Henry, and Santori (53) used microwaves to simulate boiling pool stability for open and "bottled up" systems. Water was heated in a microwave oven. Open pools were dispersive at vapor production rates equivalent to 2% of nominal reactor power.

As far as can be determined, Brigham Young University is the only one to be using chemical reactions to simulate dispersed flow. The results of chemical heating experiments support the findings of heating data collected by others.

Fauske's Fuel Flow Model

Because of the difficulties and expense of working with nuclear fuels, there is a lack of experimental data using actual molten fuel. It is also difficult to extrapolate the existing flow regime models for analyzing boiling fuel-steel mixture pools. However, the analysis of flow regime transition of a boiling LMFBR core with existing models has been attempted.

Fauske (54) used the Kutateladze model (43) to predict the bubble rise velocity necessary for fuel dispersal as about 200 cm/sec. Kutateladze obtained $k=0.14$ for the onset of drop fluidization in his water-mercury system. Wallis (55) also observed droplet dispersion at $k=0.14$ where air is blown through liquids. Catton (56), however,

predicted the bubble rise velocity of about 3220 cm/sec needed for fluid dispersal using the Collier model (42). The superficial vapor velocity at the top of the pool can be evaluated from energy and mass balances for a system saturated at one atmosphere, and is expressed by

$$U_v = \dot{Q} v_v / (A h_{fg})$$

where $\dot{Q}$ is the power going into vapor production, v_v is the vapor specific volume, A is the cross sectional area, and h_{fg} is the specific heat of vaporization.

Fauske and Catton each used this equation to calculate the minimum fractional full power required for fuel dispersal in the core of the FFTF. Fauske claimed that the core would disperse when only about 2% of full operating power remained, whereas Catton claimed that the full power was needed to disperse the fuel. The discrepancy stems from the different bubble rise velocities used.

Two questions exist in relation to the dispersed flow regime of a fuel-steel pool. Is a pool with internal heat generation dispersive by nature? A pool open to the environment would be expected to produce a high enough vapor generation to disperse, but a pool in a closed system might not.

At what minimum power levels will a core remain sufficiently dispersed to remain subcritical? Even after the neutronics have stopped, between 10 to 15% of full power is produced by fission products.

Fauske presented several phenomenological arguments to show that recriticality would be avoided, since the core would remain

dispersed even at low decay-heat levels (54-57). Because the boiling point of steel is about the same as the melting point of fuel, there could be a significant amount of steel vapor produced in the core. The steel vapor would keep the liquid fuel dispersed throughout the core. A flow regime of continuous vapor and liquid fuel droplets would be formed in the core region. Figure 5 represents Fauske's picture of the conditions in a boiled up core. An idealized closed system with no heat losses is assumed. Even at 1% of full reactor power, the void fraction would be well within the dispersed regime.

The reasoning leading to Fauske's conclusions was as follows: Fuel can only support a small amount of superheat, about 10°C for UO_2 . Nucleation occurs throughout the liquid if this superheat is exceeded. The nucleation causes fragmentation of the liquid into smaller masses until the drops are small enough to conduct the heat to the drop surface where it is carried away by convection and evaporation. The heat transfer from the droplets is conduction limited, because of the poor thermal conductivity of liquid fuel. A 10°C superheat would lead to a droplet about 1 mm in diameter. This mechanism rapidly disperses the pool and prevents any recompaction of fuel.

By extrapolating experimental data based on low density gas-liquid systems, Fauske (58) proposed flow regime maps for boiling sodium and fuel as shown in Figure 6. The units are in normalized power and length. In the case of molten fuel, the dispersed regime is entered directly from the bubbly regime with no intermediate slug flow regime. The slug flow would be eliminated in

Figure 5. Boiled-up Core Conditions

(a)

(b)

Figure 6. Flow Regime Maps for (a) Boiling Fuel and (b) Boiling Sodium.

internally heated fluids by fragmentation needed to avoid superheating since the liquid would be at the saturation temperature.

The transition between the bubbly flow regime and the dispersed flow regime was needed. The vapor velocity necessary to cause instability in the bubbly flow regime was determined by the Kutateladze stability correlation, which is valid for low viscosity fluids. A dimensionless group related to the ratio of the dynamic head of the vapor phase and the surface tension of the liquid phase is used to define the stability of the system. The vapor phase acts upon the liquid phase due to the vapor velocity, which tends to tear the droplets apart. The liquid phase counteracts this force by means of surface tension. The ratio of these forces is proportional to the square of the Kutateladze number, k^2 .

The Taylor Stability Criterion was next used by Fauske to determine the stable dimensions for the droplets in the hydrodynamic system. It is based upon balancing buoyant and surface tension forces in a liquid-vapor system. The maximum radius that the surface tension can contain within the liquid was determined. Experimental data were needed to determine the vapor velocity necessary to cause instability in the liquid phase. Since k is a dimensionless number it should be constant regardless of the physical parameters. Experiments serve to correlate all possible parameters for a given situation.

To determine k for an internally heated boiling pool, Fauske related dispersed flow for internally heated fluids to film boiling for externally heated fluids for which k had been determined. In both

cases the bubbly flow regime undergoes a transition due to increased vapor production. In the case of internally heated pools, the bubbly to churn flow transition would be followed by dispersed flow, rather than slug flow. The value of k used by Fauske was based on experimental data on the pool boiling crisis. The critical vapor velocity lead to a Kutateladze number of 0.14 .

The analogy of internally heated pool with pool boiling crisis leads to a critical vapor velocity of 200 cm/sec for fuel. If the vapor velocity exceeds this value the pool will stay dispersed. The model excluded stainless steel for conservatism. If it were included, a lower value of critical vapor velocity would be expected.

Problems With Fauske's Model

Several problems and unanswered questions have been presented (59-60) because of Fauske's model. It is not the purpose of this dissertation to analyze these problems in detail. It is sufficient to recognize that there are some unanswered questions, and to identify some of them.

First, what happens to the vapor? Enough vapor must be generated to disperse the fuel. If the vapor cannot be condensed fast enough, the vessel will pressurize. If the vapor does not condense enough to prevent overpressurization in a closed system, can the fuel recompact?

A related question is how the heat transfer and radiation of the pool affect dispersion. The effect of fuel crust formation at the edges of the core where the fuel comes in contact with the stainless steel is uncertain. The full mechanism of heat removal is unknown.

One question involves the boiling pool analogy: Can the data for bubbly film boiling actually be extrapolated to internally heated pools? Little data presently exists for internal heating.

Use of the Kutateladze number also requires the use of Taylor's Stability Criterion. There is an apparent conflict between the characteristic dimension derived from Taylor's Stability Criterion and that derived from thermodynamic arguments.

In summary, there are several explanations of fuel flow regimes in the HCDA. Fauske's approach to the problem is credible, but there are still many questions to be answered. Much of the research presently being carried out in LMFBR safety is aimed at answering these questions. The major difficulty has been insufficient experimental data in modeling the internally heated system. Questions are being answered in many research facilities, and will be answered as the experimental basis expands to fill the gaps.

CHAPTER 4

THE USE OF CHEMICALS FOR FUEL FLOW MODELING

One of the predominant thermal characteristics of a nuclear reactor is its internal heat source. In-pile safety experiments to model whole core melt are not practical because of the expense involved. Experimentation is needed, however, to complement analytical analysis of reactor accidents. Three types of internal heating can be used to simulate whole core accidents in the LMFBR. They are electrical (or Joule) heating, microwave heating and chemical heating. Each has inherent properties which create difficulties in simulant modeling.

Comparison of Internal Heat Sources

Joule heating is produced in a fluid by passing an electric current through an electrolyte. If two electric plates form parallel walls of a vessel containing an electrolyte, the fluid will heat up because of the resistance of the electrolyte. For a constant voltage across the plates, the power generated in the solution is equal to $I^2 R$, where I is the current passing through the fluid and R is the resistance of the electrolyte. The power is easy to measure and can be controlled continuously. Problems exist in using Joule heating, however. One main problem is created by void in the liquid when it boils up. The vapor does not conduct electricity, so the droplets suspended in vapor will not be producing heat. This will eliminate

droplet fragmentation which may occur in molten fuel. A second problem encountered with Joule heating is caused by geometry. For volumetric heating the plates should be parallel, which requires rectangular geometry. The LMFBR is designed in cylindrical geometry. Cylindrical geometry in modeling would therefore be preferable. A third difficulty with Joule heating is in its uneven (non-volumetric) heating. Actually the LMFBR core is not expected to produce true volumetric heating. Hot and cold spots are anticipated. The analytical models, however, use volumetric heating terms.

Internal heating from microwaves is produced by dielectric loss in vibrating molecules in response to electromagnetic stimulation. Experiments are usually carried out in a microwave oven. The problem of uneven heating from cavity resonance is removed by the use of a scatterer in the oven. Fairly constant heating rates can be measured anywhere in the oven. Three main problems exist with microwave heating. Because the electromagnetic radiation could interfere with communication systems, microwave emission is carefully controlled by the FCC. This has several consequences. First, the frequency is carefully controlled. The heating response is frequency dependent, which may limit the materials to be used for simulation. The power output of the microwave tube is set, so the power density must be adjusted by using different volumes of solution for each run. This means that the amount of simulant material is different for each power desired. Regulations are also set on the oven container. To prevent the microwaves from causing injury to the user of the oven, shielding is set up around the oven cavity,

making photographing and instrumentation more difficult. The second major problem with microwave simulation is the response of metals to microwaves. Metal instrumentation cannot be used in the oven. Continuous temperature measurements must be replaced by spot readings while the oven is off. Third, the dielectric loss is temperature dependant. There could be a positive or negative temperature coefficient, depending upon the material used and the temperature range. These problems are not insurmountable, however, in producing good experimental data with microwave heating.

Chemical heating is a third method of producing internal heat for fuel motion simulation. The heat is produced by exothermic chemical reactions. Several properties of chemical reactions make chemical heating unique. First, there are always multiple components. Joule heating and microwave heating can use a single solution to be heated. A complexity is added when a multi-component system is used, however, the core being modeled is itself a multi-component system.

Power generation can be controlled in two ways, by injection speed and by concentration. To be able to predict the power generated in a given experiment, information is needed about the reaction mechanism and kinetics, as well as the heats of reaction. This information can be measured. The volumetric uniformity of the power is uncertain. Volumetric heat generation would be expected with proper mixing, which is predicted in a boiling system. The reactants would produce heat even while the liquid is dispersed in the

vapor. Although the complexity is increased by using a multi-component system, the ease of experimental setup makes chemical heating attractive.

Selecting a Simulant Reaction

Since there are numerous chemical reactions from which to choose, a set of criteria must be established to choose a reaction that will properly model the molten core. Once the reaction is chosen, it can be analyzed in more detail to determine the specific properties of the reactants.

The first step in reaction identification was to present the reactor conditions to be modeled. Since the accident sequence is so complex, certain simplified conditions were used. The model was a homogeneous molten pool of fuel and stainless steel in a solid cylindrical container. Sodium had been removed from the core, as had the non-condensable fission products, so the only gases present would be from fuel and steel vapor. Heat is produced by decay of fission products distributed homogeneously throughout the pool. To maintain two-phase modeling, melting and freezing were not considered. The pool was dispersed within the core region due to decay heat. Two systems are possible, an open system vented to the atmosphere and a closed system sealed off on top. The fission product impurities allow volumetric nucleation.

This model specifies certain properties which the chemical reaction must have to simulate the reactor conditions. The most notable is that the reaction must be highly exothermic. Because it was assumed that there were no non-condensable fission products

present, the reaction must be a liquid-liquid reaction. If gases were given off in the reaction, an artificial boilup would occur. All vapor production should be from boiling rather than from chemical reaction. A precipitate was allowed, however, since it would add to volumetric boiling, though a totally liquid system was preferred. There are many exothermic reactions that produce sufficient heat to disperse the fluid, but react so quickly that data are difficult to collect. Decay heat would maintain a dispersed flow regime into steady-state. The reaction must produce enough heat for long enough periods of time to simulate a steady-state condition. A final requirement was safety. Though the power production is high, explosive reactions could not be used. Also extremely hazardous or toxic chemicals were avoided.

To select a suitable reaction, four major reaction types, which are known to produce heat, were investigated: dilution, neutralization, oxidation-reduction, and organic.

The heat of dilution is the heat produced by diluting some chemical with a solvent. The greatest heat of dilution was found when sulfuric acid was diluted with water. There may be chemicals which produce greater heat when diluted in organic solvents, but the sulfuric acid dilution was chosen because of the data available on the reactants. The kinetic rate is very fast, so continuous injection must be used to obtain a semi-steady-state. Vigorous boiling was obtained when water was injected into sulfuric acid. It stopped as soon as the injection stopped. Since the kinetic rate is very fast, a problem occurred in uniform mixing.

An acid and a base will neutralize, forming water and a salt along with heat. Various neutralization reactions were inspected before choosing the sulfuric acid - sodium hydroxide reaction. Sodium sulfate was the salt produced in the form of a precipitate. As in the sulfuric acid - water reaction, there was a high heat production and very fast kinetics.

Two main oxidants in oxidation-reduction reactions are peroxides and permanganates. The reactions encountered in this category tend to produce non-condensable gases, not suitable for simulant modeling.

There are many organic reactions which produce large amounts of heat. Two well-known reactions are the Friedel-Crafts and the Grignard reactions. A reaction involving a chlorine exchange similar to the Friedel-Crafts reaction, but not requiring a catalyst, is the reaction of dimethyl sulfoxide (DMSO) with an acid halide. The reaction of DMSO with various acid halides is known to be very vigorous (61). Preliminary studies reacting DMSO with acetyl chloride produced a high degree of dispersion for long periods of time. This reaction met the criteria established to model fuel flow and was therefore chosen to be used for the simulant experiments in the remainder of this research. Other reactions could be used for future verification of the results presented here.

Properties of the DMSO-Acetyl Chloride Reaction

When DMSO reacts with an acid chloride, chloromethyl methyl sulfide is produced with the corresponding carboxylic acid. The carboxylic acid which corresponds to acetyl chloride is acetic

acid. The DMSO-acetyl chloride reaction would then proceed as follows (62):

Chloromethyl methyl sulfide was observed as the product of this reaction, however, no acetic acid was observed until hours after the reaction cooled down. The acetic acid reacted with the direct products to form two liquid phases, a lighter clear phase consisting mainly of benzene, and a heavier yellow phase containing unknown products. The second direct product, produced with chloromethyl methyl sulfide, has not been identified.

Yields of chloromethyl methyl sulfide above 90% are obtained if DMSO is added to acetyl chloride in an inert diluent. If acetyl chloride is added to DMSO the reaction is more complicated, since the excess DMSO reacts with the product, producing a mixture of secondary products (63). Chloromethyl methyl sulfide also reacts with any water available.

Because of the complexity of the reaction obtained when acetyl chloride is added to DMSO, care was taken to add DMSO to acetyl chloride when modeling the HCDA. The physical properties of interest are given in Table 2 for the primary reactants, primary products, and the solvents used. The values, for the most part, were obtained from various handbooks (63-66). The remaining properties were calculated from correlations presented by Reid, Prausnitz, and Sherwood (67). The heat of vaporization of chloromethyl methyl sulfide was estimated by the Vetere correlation

TABLE 2
CHEMICAL PROPERTIES

Chemical	Structure	Mol. Wt.	Temp boil (°C)	Density ₃ (gm/cm ³)	Vap Heat (cal/gm)	Heat Cap (cal/g-mK)	Sur Tens ₂ (dyne/cm ²)
DMSO	CH ₃ SOSH ₃	78.13	189	1.1014	144.6 [@]	37.5 [@]	40 [@]
Acetyl Chloride	CH ₃ COCl	78.50	50.9	1.1051	78.84 [@]	30.06 [‡]	26.7
Benzene	C ₆ H ₆	78.12	80.1	0.87865	94.14 [#]	32.4	27.56
Toluene	C ₆ H ₅ CH ₃	92.15	110.6	0.8669	86.8 [#]	37.3	27.4
Chloromethyl methyl sulfide	CH ₃ SCH ₂ Cl	96.58	107.1 [*]	1.7773 [*]	104.8	32.7 [‡]	182.8 [‡]
Acetic Acid	CH ₃ CO ₂ H	60.05	117.9	1.0492	96.75 [#]	29.4	24.8

Main information from reference (64)

* from reference (65)

@ from reference (63)

from reference (66)

‡ calculated as explained in text

because critical properties were unknown. The heat capacities of acetyl chloride and chloromethyl methyl sulfide were estimated by the Chueh-Swanson group contribution method. The parachor formula was used to estimate the surface tension for chloromethyl methyl sulfide.

To reduce the complexity of parameters used in chemically simulating reactor accidents, several simplifying assumptions were made. Only pure components were considered in the first case. The effects of solvent properties were treated later. Products were assumed to form slowly enough to be neglected. Since the boiling temperature for acetyl chloride is much lower than for DMSO it was assumed that the vapor production was produced from vaporizing acetyl chloride while DMSO remained in a liquid state. These assumptions define the set of parameters which are valuable for each component. The significant properties for acetyl chloride are: boiling temperature, latent heat of vaporization, and density of gaseous phase. The properties which relate to DMSO are: liquid density, heat capacity, and surface tension.

Properties of Mixtures

During the actual data collection, the reaction was carried out in a solvent rather than as pure components. The effect of mixtures was therefore taken into account. Benzene was used as the main solvent. After new OSHA regulations restricted the use of benzene in experimental laboratories, toluene was substituted as the solvent. The results obtained using toluene were not consistent with those run earlier in benzene. This is because of the lower

volatility of toluene. After compliance with OSHA regulations, benzene was used in the remainder of the experiments. The properties of both benzene and toluene are presented, whereas the actual data collected were only for the benzene system.

The properties of an acetyl chloride solution or a DMSO solution vary as a function of concentration as they are diluted with either benzene or toluene. The acetyl chloride solution was assumed to boil off while the DMSO solution stayed as a liquid. The mixed properties for each were determined. Nuclear magnetic resonance (NMR) spectra were run of the vapor and the liquid phases after the reaction took place to obtain some idea of the constituents actually present in the two phases.

The boiling temperature of acetyl chloride solution was assumed to be a linear variation between benzene (80.1°C) or toluene (110.6°C) and pure acetyl chloride (50.1°C). The expected boiling temperature as a function of concentration is shown in Figure 7. The high boiling temperature of solutions in toluene presented experimental difficulties so benzene was used for the experiments. The latent heat of vaporization for the acetyl chloride solution as a function of concentration is shown in Figure 8. Because of the possible presence of acetic acid, a molar fraction estimation was used rather than the Trouton method to determine vaporization heats.

A linear variation was used for the liquid density of DMSO solution. Since benzene and toluene have close to the same densities, no distinction was made. The Maclead-Sugden correlation, as suggested by Reid, Prausnitz, and Sherwood (67), was used to

Figure 7. Boiling Temperature of Acetyl Chloride Solution

Figure 8. Heat of Vaporization for Acetyl Chloride Solution

determine the surface tension as a function of concentration. The results showed an almost linear variation in surface tension. NMR tests run later showed no DMSO remained in the liquid, so further adjustments were made.

It was beyond the scope of this research project to look at the DMSO - acetyl chloride reaction in detail. The purpose was to model the fuel flow in an HCDA. However, there have been several questions asked about the accuracy of the assumptions that only the acetyl chloride solution boils, and the effect of products on the results. To answer these questions several NMR spectra were run on the vapor produced and the liquid remaining.

The NMR spectrum is produced by nuclear magnetic resonance in the protons of hydrogen atoms when placed in a magnetic field. By varying the magnetic field strength, protons show up as peaks of the spectrum. The number of peaks are determined by the number of non-equivalent protons (or protons of the same kind). The position of the peaks represents the electronic environment of the protons. The intensity of the signals, or areas under the peaks, indicate how many protons there are of each kind. With extended analysis the components of a solution could be determined by their NMR spectrum (68).

The NMR spectra of the vapor and the liquid products produced by the DMSO - acetyl chloride reaction is shown in Figures 9 and 10. The reactants were in a 9M solution with benzene. The data collected by the NMR spectra are listed in Table 3.

Figure 9. NMR Spectrum of Vapor Products

Figure 10. NMR Spectrum of Liquid Products.

TABLE 3
NMR RESULTS

Peak		Area	H	Norm A	Mol. %	Comp.
Vapor						
1	10.5	2	2	1	4	HCOH
2	6.9	58	6	9.7	38	C ₆ H ₆
3	4.1	4	2	2	(8)	CH ₂ Cl
4	1.9	30	3	10	39	CH ₃ COCl
5	1.7	6	3	2	8	CH ₃ S
6	1.5	8	3	2.7	11	?
Liquid						
1	7.0	56	6	9.3	41	C ₆ H ₆
2	4.2	12	2	6	(26)	CH ₂ Cl
3	1.8	18	3	6	26	CH ₃ S
4	1.6	23	3	7.7	33	?

For the vapor (Figure 9), there were six main peaks, with several smaller peaks called spinning side bands. The position of the peak is presented in terms of δ , the magnitude of the magnetic shift. The area under the peak is proportional to the number of equivalent hydrogen atoms. From this the molar percent was determined for each component.

The benzene and acetyl chloride make up almost 80% of the vapor (peaks 2 and 4). The chloromethyl methyl sulfide forms two peaks (3 and 5), one for the methyl group and the other for the chloromethyl group. Peak 6 represents the unidentified product which appears to have a methyl group because of the position of the peak. Some of the chloromethyl methyl sulfide reacts with DMSO to form formaldehyde which shows up as peak 1. Less than 4% of the total vapor is formaldehyde.

The DMSO was totally reacted and therefore did not show up in the liquid product. Only three components were detected in the liquid: benzene, the major component (peak 1), chloromethyl methyl sulfide (peaks 2 and 3), and the unknown product (peak 4).

The heat of vaporization for the reaction will be about midway between that expected for pure acetyl chloride and pure benzene (about 86 cal/gm). The heat capacity is about the same for benzene and chloromethyl methyl sulfide. The fluid density is about 1 gm/cm³. The surface tension increases from that of DMSO to about 85 dynes/cm².

Without a detailed evaluation of the actual reaction used to carry out simulant experiments, the properties of the system can be

estimated. Because of the complexity of the reaction, some of the parameters are only order of magnitude approximations. This was sufficient for the scope of this research, however, refinements could be made where needed for researchers continuing beyond this project, by making extensive analysis of the reaction.

CHAPTER 5

CALORIMETRY

One of the most important parameters to be measured in internally heated two-phase flow is the heat generation term. The heat of chemical reactions is usually measured with a calorimeter. In a precision calorimeter the reactants are diluted so that the temperature change will be small, keeping heat leakage very small. There is a problem, for some reactions, in extrapolating data obtained in dilute solution to a more concentrated solution. The region of interest for HCDA modeling is the boiling range. Precision calorimeters are not designed to allow boiling, for this reason two additional calorimetric methods were used, one to measure the power in approach to boil and another to take into account the vapor production in the form of pressure increase.

Precision Calorimetry Measurements

The first piece of equipment used in these calorimetric studies was a precision titration calorimeter furnished by the Center of Thermochemical Studies at Brigham Young University. The block diagram in Figure 11 illustrates the main components of the calorimeter (69). The titrant, containing one of the reactants, is injected from the buret into the reaction vessel. The reaction vessel, shown in Figure 12, contains the other reactant. Both the

Figure 11. Schematic diagram of a titration calorimeter apparatus. A= reaction vessel with temperature sensor. B= buret (constant rate). C= temperature measuring circuit. D= amplifier. E= data output (strip chart recorder). G= environmental control circuit.

Figure 12. Titration Calorimeter Reaction Vessel

buret and the reaction vessel are placed in a constant temperature bath. The temperature change of the reaction is monitored with a thermister while the stirrer keeps the reactants well mixed. The reaction vessel is a Dewar flask which minimizes the amount of heat leakage. The water bath temperature is controlled by a control circuit. The reaction temperature is converted to a voltage by means of a Wheatstone bridge. The voltage is amplified and recorded on a strip chart recorder. A heater circuit is used to calibrate the reaction, see Figure 13. A known power is used to heat up the products to compare the temperature rise due to the heater to the temperature rise from the reaction.

The operation of the titration calorimeter is explained by Eatough, Christensen, and Izatt (70). The two reactants are diluted with a solvent. One reactant is placed in the buret, the other in the reaction vessel. Both buret and vessel are lowered into the constant temperature water bath. The reactants are allowed to come to thermal equilibrium with the bath. Any deviation from equilibrium will show up as an increasing or decreasing slope on the temperature plot. Heat leaking into the reaction vessel will show an increase of temperature whereas heat loss will show a decrease of temperature.

The titrant is injected into the other reactant at a known speed. The temperature rise is followed to the completion of the reaction. Since heat is added by the reaction, the vessel will no longer be in thermal equilibrium with the bath, therefore any heat leakage from the vessel is detected as a negative slope on the temperature plot. The average heat loss (or gain) is determined

Figure 13. Typical Calorimeter Heater Circuit.

from the average slope of the line before the reaction begins and after the reaction ends. This value is added to (or subtracted from) the total heat of reaction.

The readout on the temperature plot is in millivolts and must therefore be calibrated in terms of temperature or heat. The circuit in Figure 13 is used for this purpose. The procedure of calibration is the same as for reacting chemicals except that the heat input is electrical. The total power dissipated in the heater, which must be absorbed by the liquid, is $P_h = I_h V_h = V_s V_h / R_s$. The voltage across the heater V_h is measured with a voltmeter. The current through the heater I_h is the same as the current through the standard resistor R_s and is determined by measuring the voltage across the standard resistor V_s and knowing the value of the standard resistance. For a given calorimeter and liquid in the reaction vessel the results of the calibration gives the energy needed to raise the temperature plot one millivolt. The actual temperature is not needed since the energy is now known in joules per millivolt.

For the DMSO - acetyl chloride reaction, 5 ml of 0.1M DMSO solution was injected into 50 ml of 0.01M acetyl chloride solution. DMSO was injected into acetyl chloride to simplify the reaction kinetics. By making the DMSO solution ten times as concentrated as the acetyl chloride solution, only one tenth the volume of DMSO solution was needed to maintain equal molar concentration. Benzene was used as the solvent in both cases. The reaction continued for more than one hour, giving a total heat of reaction of -14.70 Kcal/mole (61.5 Kjoules/mole). The heat

generated as a function of time was not linear (Figure 14) so the power density during the first few minutes of reaction was much greater than during the last part as seen in Figure 15.

The purpose of the first phase of calorimeter studies was to determine the total heat of reaction of the chemicals used. The parameter needed is the power generated. Dilute reactants will not react as fast as concentrated reactants. The total heat per mole should be the same, but the kinetics may vary. To determine the mechanism and kinetics of the reaction, further calorimetric studies were performed with the precision calorimeter. It was not within the scope of this research to fully characterize the mechanism or the kinetics of the chemical reaction used, however, insight into the reaction helped to determine the power produced.

A series of runs were carried out to learn about the kinetics of the reaction. If the reaction rate had simple kinetics, the reaction rate could be written by the rate equation

$$\text{rate} = K (\text{DMSO})^w (\text{AC})^x.$$

A base case (case A) was performed by injecting 5 ml of 0.1M DMSO into 50 ml of 0.01M acetyl chloride at the rate of 24 ml/min. In case B the concentration of DMSO was quadrupled while leaving the acetyl chloride concentration unchanged. In case C the acetyl chloride concentration was quadrupled while the DMSO was the same as in case A. Case D showed the effect of quadrupling both the DMSO and the acetyl chloride concentrations. The results of this test series is shown in Figure 16. The abscissa is in units of temperature, calibrated such that one unit change requires one

Figure 14. Sudden Titration of DMSO in Acetyl Chloride

Figure 15. Change in Power Density as a Function of Time.

Figure 16. Concentration Effect (non-equimolar).

calorie of added energy deposited in the liquid. The tabulated results are shown in Table 4.

As can be seen in case A, the reaction started out fast and leveled off after the first half minute. By increasing the concentration of DMSO, the initial power doubled but decreased greatly after the first half minute. The increase of acetyl chloride concentration did not cause such a large initial power rise but maintained a steady rise over a period of minutes. By increasing the concentration of both the DMSO and acetyl chloride there was a strong initial power burst which gradually leveled off.

The tests demonstrated that the reaction was a stepwise reaction containing at least two steps so it did not follow simple kinetics. The first step, dominated by the DMSO concentration, produced the initial power burst. The second step was a slower reaction which continued to add heat for several minutes.

In order to completely characterize the reaction and thereby predict the heat produced as a function of time, a more detailed analysis of the reaction kinetics and mechanism would be needed.

The information actually required was the effect of increasing the concentrations of DMSO and acetyl chloride while maintaining equal molar concentrations. By injecting 5 ml of 0.4M DMSO into 50 ml of 0.04M acetyl chloride, an equal molar concentration of each (0.022 moles) was reacted while requiring the injection of only 5 ml of solution. In the second series of tests, the concentration of each reactant was increased in five steps as shown in Figure 17.

Table 5 tabulates the results.

TABLE 4
CONCENTRATION TESTS (non-equimolar)

Case	DMSO Con.	AC Con.	Power	P. Increase
A	0.1 M	0.01 M	0.35 W	base case
B	0.4 M	0.01 M	0.49 W	1.4
C	0.1 M	0.04 M	0.70 W	2.0
D	0.4 M	0.04 M	0.73 W	2.1

TABLE 5
CONCENTRATION TESTS (equimolar)

Case	DMSO C.	AC Con.	Power	P. Incr.	Cons. Incr.
L	0.4M	0.04M	0.95W	base case	base case
M	1 M	0.1 M	2.85W	3.1	2.5
N	2 M	0.2 M	5.23W	6.0	5
O	4 M	0.4 M	14.4 W	4.3	10
P	8 M	0.8 M	44.5 W	17.2	20
Q	14 M	1.4 M	-	-	-

TABLE 6
INJECTION SPEED TESTS

Case	Inj. Speed	Size(ml)	Power	P. Incr.	Speed Incr.
K	1.5	5	38 mW	base case	base case
H	3.75	5	94.7mW	2.5	2.5
G	7.5	5	203mW	5.3	5.0
F	15	5	401mW	10.6	10.0
I	24	10	150mW	4.0	16
J	60	10	707	18.6	40

Figure 17. Concentration Effect (equimolar)

An obvious problem can be noted by observing Figure 17. There are various methods of deriving the power for each curve. The power rise could be described as the energy produced in the first minute, the first two minutes, or as the average slope of the line over any interval. The length of the initial power burst varied with each curve. For case P it lasted for about 40 sec whereas a rise was observed for 2 min in case N. Because of this difficulty the tables represent the average power from the start of the reaction until the reaction begins to level off. The actual time period varies for each case. Case Q showed localized heating near the thermistor from not enough mixing. For this reason the power was not calculated.

In the tables the power increase and concentration increase is the power or concentration for the given case divided by the power or concentration for the base case. Table 5 shows that the initial power increase was approximately proportional to the concentration increase within this limited range of concentrations. The concentration range of interest in HCDA modeling was between 1M and 14M. A boiling calorimeter was needed to measure initial power for concentrations in this range.

The effect of injection speed upon initial reaction power was determined by injecting 5 ml of 0.1M DMSO into 50 ml of 0.01M acetyl chloride while varying the injection speed. A variable speed syringe pump was used to make the injection. The results are shown in Figure 18 and Table 6. The speed increase is the speed of the case of interest divided by the base case speed in ml/min.

Figure 18. Injection Speed Effect.

At slower speeds using a 5 ml syringe (cases F, G, H, and K) the initial power was exactly proportional to injection speed. The reaction rate was fast enough to occur as fast as the reactant was injected. After injection the secondary reaction took place. For case I, the injection lasted for only 0.2 min while the initial reaction lasted for about 0.7 min, before the secondary reaction predominated. In this case the reactant was injected faster than the initial reaction could take place. The proportionality of power and injection speed was limited to injections slower than the reaction rate of the initial reaction.

Case J was the upper limit of the injection. The DMSO was injected as fast as possible by hand. There were localized heating effects as well as viscous heating from the injection. A test of viscous heating was made by injecting benzene into benzene. With an injection rate of 24 ml/min, about half a calorie was produced by viscous heating. At higher concentrations the heat produced from the reaction was several orders of magnitude higher than that produced from viscous heating, showing the lack of importance of viscous heating error in the measurements.

Several conclusions were made as a result of the precision calorimeter studies. First, the reaction is a complex stepwise reaction. Much more information would be required to completely characterize the reaction. The path of the reaction depends upon which reactant is the most concentrated. Given equal molar concentration the initial power generation is approximately proportional to the concentration within a limited range of concentrations.

The initial power is also proportional to the rate of injection, as long as injection does not occur faster than the first reaction takes place.

Boiling Calorimetry Measurements

A precision calorimeter does not allow boiling. Because of the non-linear nature of the DMSO - acetyl chloride reaction, the power generation data obtained with the titration calorimeter at low concentrations could not be extrapolated to the concentrations of interest without undue analysis of the reaction kinetics. A new calorimeter was built to make measurements of the reaction power in the actual region of interest, boiling. The data from the new calorimeter were not as precise as the titration calorimeter but were within about 10%.

The basis of design for the calorimeter was to measure the liquid temperature rise in the range of boiling. Several assumptions were made in the design. A precision calorimeter can measure small temperature changes, therefore small heat sources and sinks must be taken into account, such as stirring heat, viscous heat from injection, and heat flow through the flask. In high power reactions the power produced is large compared to these errors, therefore, no constant temperature bath was needed.

The calorimeter built is shown in Figure 19. The reaction takes place in a one liter Dewar flask. The instrument head is supported by a steel stand. A motorized stirrer is used for even mixing. The titrant is injected by means of a syringe pump through the titrant tube. Thermister and heater leads feed into a control

Figure 19. Boiling Calorimeter

box which is connected to a plotter and a digital voltmeter, all of which are located on a counter away from the reaction vessel.

There is a vent at the top of the vessel to allow the vapor to escape. The instrument head is seated into the flask with a rubber stopper. A plastic guide was first used to support the wires and the stirrer, but later replaced with glass when the plastic melted from the heat.

The control box has several functions. It contains the on/off switch for the heater. It also contains the Wheatstone bridge which converts the resistance change of the thermister to an output voltage. A selection switch chooses the voltage to be indicated on the voltmeter: the bridge voltage, heater voltage, or power supply voltage.

The reaction power is measured as the average rate of temperature increase from room temperature to boiling temperature. This represents the initial power burst of the reaction.

This method cannot measure the actual heat of reaction, but it is the reaction power which is of interest. The measurements made were somewhat crude compared to precision calorimetry, but the 10% error is smaller than the power information during the transition phase of an LMFBR accident.

In order to obtain an idea about the accuracy of the boiling calorimeter, a 3M concentration comparison run was made. The power density of the boiling calorimeter had a -10.5% error when compared with that measured with the precision calorimeter. The power density was used because of the volume difference of the two reaction vessels. There was nothing to keep the reactants the

same temperature prior to reacting in the boiling calorimeter. This could account for the 10% difference measured. A 3M solution was used for comparison because it was a concentration close to boiling but still measurable in the precision calorimeter.

As the concentration was increased to approach boiling, the measurements taken also increased as expected. When boiling was reached, it was difficult to get an accurate measurement since only the temperature increase was measured. When the transition to boiling occurs, the void fraction increases rapidly providing a large cooling area. Heat not only goes into vapor production, but also into heating up the vessel and instrumentation inside the vessel. The range of greatest interest is at transition from no boil to dispersed flow. This occurs between 3M and 4M in an open vessel system. The power produced was measured for several runs in the 3.1M to 3.5M range. The mean power measured was 60.5 cal/sec with a 95% confidence interval between 52.1 and 68.9 cal/sec.

There is little heat loss from the Dewar vessel. Since there is a heat loss in the actual reaction vessel, the lost heat was measured by comparing the temperature rise in the reaction vessel to that of a Dewar flask. A 42% loss was measured. With this loss accounted for, there would be 35.1 cal/sec available for vapor production with a 95% confidence interval between 30.2 and 40.0 cal/sec. Actually some of the heat goes into heating up the instrumentation and vessel so that not all of this heat produces vapor.

Even the boiling calorimeter did not give information about how much heat actually was used in vapor production, only how

much was available for vapor production. To determine the power which generates vapor, closed vessel pressure measurements were used.

Closed Vessel Pressure Measurements

The reaction vessel used in the closed vessel pressure measurements is shown in Figure 20. It consists of a stainless steel reaction vessel 25 cm long, 5 cm in diameter, and with a wall thickness of 0.8 cm. Brass bolts capped the openings on the ends of the reaction vessel. Entrance holes for reactant injection and pressure/temperature measurements were silver soldered to 3/16 in stainless steel tubing. A release valve was used to depressurize the vessel. A pressure transducer was used to measure the pressure. A thermister was used for temperature measurements.

Figure 21 shows a block diagram of the equipment used. The voltage measured by the transducer was amplified then recorded on a two-channel recorder. The thermister forms one arm of a balanced bridge. The thermister was inserted into a temperature well in the reaction vessel. The bridge output was also plotted on the two-channel recorder. A thermocouple was attached to the outside of the reaction vessel to measure the temperature rise of the vessel itself. Reactants were injected into the vessel by means of a syringe pump.

The purpose of the closed vessel pressure tests was to predict the amount of heat actually producing vapor as compared to the sensible heat. The temperature and pressure rise are shown

Figure 20. Stainless Steel Pressure Vessel

Figure 21. Block Diagram of Pressure Vessel Setup.

in Figures 22 and 23 for three different concentrations. The temperature rise for the outside of the vessel is also shown in Figure 22.

The sensible heat of the liquid was measured by the temperature difference in the first ten seconds, knowing the heat capacity of the liquid. A power of about 30 cal/sec went into the liquid. The vessel was treated as a lumped body, with a temperature equal to the outside temperature. Since only the bottom portion of the vessel heated up, the vessel mass used in the calculation was only a third of the full mass of the vessel. The power going into heating up the vessel was about 12 cal/sec.

The heat going into vapor was calculated by using the pressure increase, knowing the heat of vaporization. A power of about 17 cal/sec was measured for heat producing vapor. This is about half as much as the sensible heat.

Several difficulties arose in using a pressure vessel to measure the power production. The main problem was that the closed vessel suppresses vapor production while allowing more heat to go into sensible heat. In fact, active boiling did not occur at 4M in a closed vessel where it did in the open vessel. The heat loss through the metal container was different than through the glass, so the measurements would be different. A third problem was due to other heat sinks. Only the heat for vapor production, liquid temperature rise, and vessel temperature rise were measured. The heat capacity of the other instruments would also affect the heat loss. In spite of these difficulties, the power measurements made

Figure 22. Temperature Change in Closed Vessel Tests.

Figure 23. Pressure Buildup in Closed Vessels.

in the pressure vessel were remarkably close to those made in the boiling calorimeter. The total power measured in the pressure vessel was 59 cal/sec as compared to 60 cal/sec in the boiling calorimeter.

CHAPTER 6

OPEN VESSEL SIMULATION EXPERIMENTS

The first condition to be simulated in the transition phase of an LMFBR accident is the possible fuel motion in an open top core, where there has not been sufficient freezing of materials to block the top of the core. Experiments were performed to measure the superficial vapor velocity at the transition between bubbly and dispersed flow. The vapor velocity was compared with that predicted by Kutateladze. A comparison was made with the LMFBR.

Theory

The superficial vapor velocity as evaluated from energy and mass balance is

$$U_v = \dot{Q} v_v / A h_{fg} \quad (1)$$

where $\dot{Q}$ is the heat generation rate, v_v is the vapor specific volume, A is the cross sectional area of the vessel, and h_{fg} is the latent heat of vaporization of the liquid which is vaporized. By increasing the concentration, the heat generation rate increases until there is a transition between bubbly and dispersed flow. The superficial vapor velocity at that point is called the critical superficial vapor velocity. This is the minimum velocity necessary for the vapor to disperse the liquid droplets.

Several empirical correlations have been proposed for the initiation of flooding. The Kutateladze stability criterion, which predicts the transition between flow regimes, correlated data for liquid metal flow systems very closely. The predicted critical superficial vapor velocity is given by

$$U_v = \frac{k \sqrt{4\sigma g(\rho_h - \rho_l)}}{\sqrt{\rho_c}} \quad (2)$$

where k is the dimensionless parameter known as the Kutateladze number as given in Table 2. The densities are ρ_h for the heavy phase, ρ_l for the light phase, and ρ_c for the continuous phase. The liquid surface tension is given by σ , and the acceleration of gravity by g .

Prior to the research reported here, very little experimental work had been done to verify the applicability of the Kutateladze stability criterion to internally heated fluids. The previous correlations presented were for external heating, that is, for boiling systems from a flat heated surface and for pool stability of gas-liquid systems where the gas was injected through the liquid. Farahat, Henry, and Santori (53), however, successfully used microwave heating of water to study the use of the Kutateladze number for internal heating.

Apparatus and Procedures

The basic setup for the open system simulation runs is shown in Figure 24. The reaction vessel was a 250 ml graduated cylinder held in place by a laboratory stand. A digital clock was located next to the cylinder to time the sequence of the reaction.

Figure 24. Experimental Setup - Open System

All of these materials, as well as a photographic background, were placed in a standard chemical hood to remove fumes. The DMSO was injected into the cylinder by means of a syringe pump located outside the hood. A teflon tube lead from the syringe into the cylinder. A super-8 movie camera was placed on a tripod in front of the hood. Standard photo floods were set at a 45 degree angle from the front of the cylinder to provide illumination during photographing the reaction.

The chemical reaction used in this experiment was the injection of DMSO into acetyl chloride. Concentrations ranging from 1M to 14M were obtained by diluting the reactants in benzene. Since the boiling temperature of acetyl chloride is very much lower than for DMSO, acetyl chloride and benzene were the working fluids. For this reason, together with the complexity of the reaction in excess DMSO, the DMSO was used as the titrant rather than acetyl chloride.

For each run, Bakelite was placed in the bottom of the cylinder to assure sufficient nucleation for smooth boilup. After placing 10 ml of acetyl chloride solution in the cylinder, 10 ml of equal concentration DMSO solution was injected into the acetyl chloride at a rate of 24 ml/min. Moving pictures were taken of the reaction at normal speed (18 frames/sec) with a super-8 camera. The void fraction was determined from the movies. The total height and the liquid height were measured. The void fraction is the height difference (total minus liquid) divided by the total height.

To determine the effect of reaction vessel diameter on the vapor velocity, the procedures were repeated in a larger vessel. Again the reaction was photographed and the void fraction determined.

Experimental Results

When DMSO reacts with acetyl chloride, a large amount of heat is liberated. The heat causes vigorous boiling. Droplets of liquid are entrained by the moving vapor dispersing the liquid in the cylinder. The void fraction is the volume percent of void in the total height of dispersed liquid. The vapor phase height is the total height minus the liquid height which is measured from the movies. The void fraction is then the vapor height divided by the total height. Figure 25 shows the liquid in the cylinder prior to boilup. Figure 26 shows a 4M concentration resulting in a void fraction of 60%. Figure 27 shows a 9M concentration producing a 93% void fraction.

The void fraction varies as a function of time, depending upon the heat produced and heat lost. Lower concentrations obtain a peak void fraction then quickly taper off, never really obtaining a steady-state value. Higher concentrations maintain a dispersed state for much longer periods of time. Figure 28 shows the void fraction buildup as a function of time for both 4M and 9M concentrations.

Since the higher concentrations maintained their peak void fractions into a condition approaching steady-state, the peak void fraction was also used for lower concentrations.

Figure 25. 4M Solution, $\alpha = 0$, Prior to Boil.

Figure 26. 4M Solution, $\alpha = 60\%$.

Figure 27. 9M Solution, $\alpha = 93\%$

Figure 28. Void Fraction Buildup.

It was difficult to observe the transition between flow regime. Only two flow regimes appeared in the small vessel, non-boil and dispersed flow. There was a point at which a few bubbles from boiling would appear on the surface, but there seemed to be a continuous increase in void fraction with no definite steps of transition. In the large vessel there seemed to be three flow regimes. There was a no-boil regime, bubbly flow regime with void fractions around 10-20% and a dispersed flow regime with void fractions above 50%. The transition between bubbly flow and flooding was chosen at the point of rapid increase in void fraction to exceed 50%.

The plots of peak void fractions as a function of concentration are shown in Figures 29 and 30. Figure 29 shows the void fraction for the smaller vessel. There is a sharp increase in void fraction between 3M and 4M. The void fraction for the larger vessel is shown in Figure 30. In this case the increase from bubbly flow (around 20%) to dispersed flow (above 50%) occurs at about a 5M concentration.

The critical superficial vapor velocity, or the vapor velocity at which transition takes place, was calculated by using equation 1 and the properties of fluids at the concentration of interest. The applicability of the Kutateladze stability criterion was determined by calculating the vapor velocity at transition using equation 2. This value should be close to the critical vapor velocity measured by experiment and calculated by equation 1.

Figure 29. Steady State Void Fraction (small vessel)

Figure 30. Steady State Void Fraction (large vessel)

Table 7 contains a list of parameters used in the open vessel calculations. It shows the mixture values at 3M and 5M concentrations as determined from Figures 7 and 8 and similar calculations. The 3M represents the small diameter vessel, whereas the 5M represents the larger vessel. The range represents possible values for pure components (benzene or acetyl chloride/product). For example, the range for heat of vaporization would be 78.8 cal/gm for pure acetyl chloride to 94.1 cal/gm for pure benzene, with the actual value somewhere in between depending upon the concentration. The surface tension is 27 dynes/cm for pure benzene and 182.8 dynes/cm for pure chloromethyl methyl sulfide. Table 7 also shows typical values of these parameters for an oxide fuel. The error introduced by not properly accounting for change in properties due to mixing is less than 10% for the experimental vapor velocity and less than 20% for the Kutateladze derivation of vapor velocity.

The heating rate of vapor production was determined from the calorimetric studies. The heat used to increase the liquid temperature (as measured in the boiling calorimeter, and accounting for heat loss to the glass reactor vessel) was 35 cal/sec. The closed vessel calorimetric runs showed that about 56% as much heat went into vapor production as into sensible heat, so about 20 cal/sec were used to vaporize the liquid in the small vessel. The boiling calorimetry tests also showed a 78% power increase between the 3M and 5M concentrations, which gives the vapor production power of 36 cal/sec for the large vessel. Since the

TABLE 7

PARAMETERS USED IN OPEN VESSEL CALCULATIONS

Parameters	3M Value	5M Value	Range	Fuel
$\dot{Q}$ (cal/sec)	20	36	15-55	
v_v (cm ³ /gm)	365	361	341-372	666.7
A (cm ²)	9.6	17.4		11310
h_{fg} (cal/gm)	93.4	92.3	78.8-94.1	301
σ (dyne/cm)	60.3	71.1	27-182.8	450
ρ_h (gm/cm ³)	0.93	0.96	0.88-1.1	8.4
ρ_l (gm/cm ³)	2.74×10^{-3}	2.78×10^{-3}	$2.7-2.9 \times 10^{-3}$	1.5×10^{-3}
$k=0.3$ $g=980$ (cm/sec ²)				
U_{vexp} (cm/sec)	8.14	8.09	5.7-13.7	
U_{vk} (cm/sec)	4.76	4.92	3.97-5.35	4.54

actual power needed to generate vapor was not measured in each case there was room for possible error. The lowest power expected is about 15 cal/sec for the 3M case and the largest about 55 cal/sec for the 5M case. Errors as high as 40% may occur because of the power measurements.

The critical superficial vapor velocity as calculated by using the Kutateladze number and equation 2 was 4.76 cm/sec for the small vessel and 4.92 cm/sec for the large vessel. The vapor velocity as determined from void fractions and equation 1 was 8.14 cm/sec for the small vessel and 8.09 cm/sec for the large. The experimental vapor velocities were about 70% higher than predicted by the Kutateladze number. Farahat, Henry, and Santori measured a vapor velocity in a microwave oven that was 50% lower than predicted.

The vapor velocity for fuel as predicted by the Kutateladze number (equation 2) is 4.54 cm/sec. The possible error of heat measurements using chemical reactions could lead to measurements in the range of 50% to 100% higher than predicted. If the actual vapor velocity of liquid fuel were 100% higher than predicted by the Kutateladze number then it would be 9.08 cm/sec. Using equation 1, the power necessary to give the fuel this vapor velocity was calculated as 46.4 Kcal/sec (0.2 MJ) which is less than 0.5% of decay heat levels. Even very low decay heat would be sufficient to keep the core dispersed in an open system.

Suggestions for Future Work

There are three main areas which could be developed or improved in chemical simulation of HCDAs. They are: better calorimetry to reduce the error in heat generation measurements, the use of other chemicals to simulate HCDA conditions, and the verification of correlations other than the Kutateladze number. Further refinement in determining the effects of mixture properties for the DMSO - acetyl chloride reaction is not needed for the open vessel tests. If chemicals were used with widely differing properties, there would be more error of mixture introduced.

Calorimetric measurements are the main source of error in these experiments. The main problem was that the calorimetric measurements were made in a different environment than the experiments, then adjustments were made to account for the difference. Future measurements could be made in a special vessel designed for constant pressure tests, constant volume tests, and calorimetric measurements. The vapor would be collected and condensed during calorimetry runs to determine the heat going into vapor production. The same vessel should be used for all experiments. Thermocouples on the outside of the vessel would measure the heat going into the vessel.

Only one chemical system was used in the tests reported here. Several other chemical reactions have been identified that could also be used to simulate reactor accidents. The reaction of DMSO with acetyl bromide as well as other acid halides is possible. Each reaction tested would give further verification of the type of

of flow expected for internally heated systems. If comparable results are obtained for chemicals with different properties, then there will be more confidence in the results.

The results of the open vessel tests showed that the measured critical superficial vapor velocity is near that predicted by the Kutateladze number. Other empirical correlations could also be used to compare with the results here.

CHAPTER 7

CLOSED VESSEL SIMULATION EXPERIMENTS

The second condition to be simulated in the transition phase of an LMFBR accident is the possible fuel motion in a sealed off core, where freezing of materials has completely blocked fuel exits, so the reactor is allowed to pressurize. Experiments were performed to determine the difference in void fraction and vapor velocity between an open and closed vessel for a given concentration of reactants. The temperature and pressures in the constant volume system was compared with a theoretical model. The effect of radial cooling on dispersion was also determined.

Theory

The vessel modeled was an upright glass cylinder shown in Figure 31. The fluid within the cylinder is heated internally with a power of $\dot{Q}$. The heat produced in the liquid goes into increasing the temperature of the liquid, vaporizing the liquid, and transfer to the vessel. The heat transferred to the vessel can increase the vessel temperature or leak out into the surrounding air. The energy equations are:

$$\dot{Q} = m_l c_l \frac{dT_l}{dt} + h_{fg} \dot{m}_v + h_l A_l (T_l - T_2) \quad (3)$$

$$h_l A_l (T_2 - T_1) + h_2 A_2 (T_2 - T_a) = -m_g c_g \frac{dT_2}{dt} \quad (4)$$

Figure 31. Glass Cylinder Model.

The temperatures T_1 , T_2 , and T_a are of the liquid, vessel, and ambient air. The inside and outside vessel areas are A_1 and A_2 . The masses and specific heats of the liquid and glass are m_l , m_g and c_l , c_g . The heat of vaporization and mass flux are h_{fg} and $\dot{m}_v$. Two heat transfer coefficients are used. The heat transfer from the liquid to the container (h_1) is from forced flow and condensation. The heat transfer from the vessel to the air (h_2) is from free convection. The inside heat transfer is of the order $1 \text{ cal/cm}^2 \text{ sec}^\circ\text{C}$ whereas the outside is about $10^{-4} \text{ cal/cm}^2 \text{ sec}^\circ\text{C}$. Because of the low heat transfer to the environment, h_2 was neglected.

If the vessel maintains the same temperature as the liquid equations 3 and 4 reduce to

$$\dot{Q} = (m_l c_l + m_g c_g) \frac{dT}{dt} + h_{fg} \dot{m}_v \quad (5)$$

If the vapor is assumed to be a perfect gas then

$$\dot{m}_v = m_v \left(\frac{1}{P} \frac{dP}{dt} - \frac{1}{T} \frac{dT}{dt} \right) \quad (6)$$

Substitution of the Clasius-Clapeyron relation yields

$$\dot{m}_v = \frac{m_v}{T} \left[\frac{h_{fg}}{P v_{fg}} - 1 \right] \frac{dT}{dt} \quad (7)$$

where v_{fg} is the specific volume increase at vaporization. The final equation for the instantaneous rate is obtained by substituting equation 7 into equation 5:

$$\dot{Q} = \left\{ (m_l c_l + m_g c_g) + \frac{m_v h_{fg}}{T} \left[\frac{h_{fg}}{P v_{fg}} - 1 \right] \right\} \frac{dT}{dt} \quad (8)$$

Of the energy deposited in the liquid, only a fraction is used to produce vapor. The energy resulting in sensible heat in the

upper fraction of the vessel (f) is supplied by condensing vapor.

The superficial vapor velocity in a closed system is due to that required for the upper axial and radial heat sinks and that required for pressurization.

$$U_v = \frac{v_v}{A h_{fg}} \left\{ f m_g c_g + \frac{m_v h_{fg}}{T} \left[\frac{h_{fg}}{P v_{fg}} - 1 \right] \right\} \frac{dT}{dt} \quad (9)$$

The pressure rise rate can be evaluated in thermal equilibrium is assumed. An Antoine equation (71) was used to determine the vapor pressure as a function of temperature.

$$\log P = A - B/(T+C) \quad (10)$$

where A, B, and C are constants determined from experimental data for pure components and P and T are the liquid temperature and the vapor pressure.

Measurements may be taken of either the pressure rise or the temperature rise to determine the vapor velocity. The vapor velocity resulting from temperature data will be the same as from pressure data if, in fact, there is thermal equilibrium.

In the use of the Clausius-Clapeyron relation thermal equilibrium is assumed. If the reaction is not in thermal equilibrium, both pressure and temperature data are needed to determine the vapor velocity. In this case equation 6 is substituted into equation 5 to yield

$$\dot{Q} = (m_l c_l + m_g c_g - \frac{h_{fg} m_v}{T}) \frac{dT}{dt} + \frac{h_{fg} m_v}{P} \frac{dP}{dt} \quad (11)$$

using the heat required for vaporization (pressure plus condensation) the resulting vapor velocity is

$$U_v = \frac{V}{m_v h_{fg} A} \left[f m_g c_g \frac{dT}{dt} + h_{fg} m_v \left(\frac{1}{P} \frac{dP}{dt} - \frac{1}{T} \frac{dT}{dt} \right) \right] \quad (12)$$

As can be seen from equations 9 and 12, an increase of pressure will decrease the vapor velocity, whereas an increase in heat sink (condensation) will increase the vapor velocity.

Apparatus and Procedures

Two different reaction vessels were used in the closed system simulation runs. The first vessel (Figure 32) was made from a 38 x 200 mm pyrex test tube. A #7 1/2 rubber stopper was used to seal off the top. A 1/16 inch stainless steel injection tube, a 3/16 inch pressure measurement tube, and a 3/16 inch temperature well were attached to the reaction vessel through the rubber stopper. The rubber stopper was clamped to the vessel to hold the pressure.

The injection tube was connected to a syringe pump that can deliver a constant reactant injection rate up to a back pressure of 50 psi. The pressure measurement tube connected to a pressure relief valve and a pressure transducer which was connected to a linear amplifier and a strip chart recorder. A thermister was placed into the temperature well to measure the liquid temperature during the reaction.

The second vessel used in the closed system tests was the same as the first vessel except it was slightly smaller (32 x 200 mm). A cooling coil was wrapped around the vessel to provide a heat sink (Figure 33). Thermisters were placed in the inlet and outlet of the cooling tube to measure the incoming and outgoing temperature of

Figure 32. Closed Reaction Vessel

Figure 33. Closed Reaction Vessel with Cooling

the cooling water. The two thermistors fed into a differential bridge so the temperature difference was recorded.

In both cases the reaction vessel was placed in a standard chemical hood. A digital clock and a yard stick were placed next to the reaction vessel to show the time sequence and height of dispersion. A super-8 camera and photo floods were used in the same fashion as in the open vessel experiments.

Bakalite was placed in the bottom of the vessel for smooth boilup. After placing 10 ml of acetyl chloride solution into the reaction vessel, the pressure relief valve was closed to seal off the system. Ten milliliters of DMSO solution was injected into the acetyl chloride at the rate of 24 ml/min. Moving pictures were taken of the reaction at normal film speed (18 frames/sec). The void fraction was determined from the films in the same manner as in the open vessel system.

Concentrations ranging from 3M to 9M (diluted in benzene) were used for the closed vessel experiments. Concentrations lower than 3M did not boil in the open system so were not used in the closed system. Pressurization was too great above 9M for the syringe pump to produce a constant injection rate. Temperature and pressure were measured as the reaction was photographed.

The reaction vessel with the cooling tube was used to study the effect of heat sink. Two runs were made for both 4M and 9M concentrations. First, a closed system test without cooling was run to compare with the large vessel. Next, the same condition was set up with water flowing through the cooling tube to act as a

Figure 34. Closed Vessel Temperature (no cooling)

heat sink. The heat removed was measured by the temperature difference of the coolant and the coolant flow rate.

Experimental Results

Three sets of data were collected in the closed vessel experiments; pressure, liquid temperature, and void fraction as determined from liquid heights. The closed system temperature and pressure plots are shown in Figures 34 and 35 for each of the concentrations used. This first set of runs were carried out in the uncooled vessel. The data are consistent with the general shape of the open system temperature. Both temperature and pressure increase as a function of concentration.

The first assumption tested was the validity of thermal equilibrium. If the reaction were in thermal equilibrium, equation 9 could be used to calculate the superficial vapor velocities. Either the temperature or the pressure could be measured, calculating the other with the proper Antoine equation.

The validity of the thermal equilibrium assumption was tested by measuring both the temperature and the pressure. If the reaction were in thermal equilibrium, the temperature calculated from pressure data using the Antoine equation would be the same as the temperature measured. The constants used in the Antoine equation was reported by Boublik, Fried, and Hala (72). For acetyl chloride they are $A=6.94887$, $B=1115.954$, and $C=223.554$. For pure benzene they are $A=6.89272$, $B=1203.531$, and $C=219.888$. The NMR results showed the vapor was a mixture of acetyl chloride and benzene in roughly equal proportions. Since there is no heat of

solution between acetyl chloride and benzene, the total pressure is the sum of the partial pressures of the two components. The constants $A=6.9208$, $B=1159.743$, and $C=221.721$ represent the vapor pressure for the mixture.

The liquid temperature of the 9M reaction is plotted in Figure 36 along with the temperature calculated from pressure data by the Antoine equation. During the first part of the reaction the calculated temperature leads the actual temperature, possibly because of uneven mixing. It lags during the later part of the reaction sequence. Thermal equilibrium is never really achieved. Axial temperature differences could be the cause of this. The effect of increasing benzene in the calculation is to raise the calculated temperature. An increase of acetyl chloride results in a lower calculated temperature. Similar results were obtained for each concentration measured.

Since the reaction was not in thermal equilibrium equation 12 was used to calculate the superficial vapor velocity. Both the pressure and the temperature measurements were used in the calculation. The smallest vapor velocity is produced in an adiabatic system so the vessel does not heat up by condensing the vapor. With no external heat sink, the greatest vapor velocity possible occurs when all of the vessel is exposed to vapor which condenses the vapor while heating up the vessel. The actual value is somewhere between the adiabatic and full condensing systems. Condensation was observed for about 2 inches above the liquid surface. This represents about 25% of the vessel heating up which gives a constant

Figure 35. Closed System Pressure (no cooling)

of $f=0.25$ in equation 12. The resulting vapor velocities are shown in Figure 37. Four of the concentrations (6M, 7M, 8M, and 9M) have vapor velocities which exceed 8 cm/sec, the vapor velocity required for dispersion in an open system. The void fractions for these concentrations are shown in Figure 38. Each of these concentrations had void fractions in excess of 50%. Concentrations lower than 6M did not boil in the closed system.

A full range of vapor velocities are possible between the adiabatic system and the system with full heat sink. Table 8 shows the results for closed system tests. The second column shows the maximum adiabatic vapor velocities obtained for the various concentrations. The next column shows the vapor velocities if one fourth of the vessel is allowed to heat up from vapor condensation. The heat removal needed above the adiabatic to disperse the fluid (8 cm/sec) is shown in column 4 with the actual heat removed in heating the vessel in column 5. Column 6 shows the peak void fractions observed. Sufficient heat was removed in all the cases that showed dispersion. The 5M was very close to dispersing but did not. An additional heat sink would be needed.

The vapor velocities and the void fractions remained high for only a short period of time in the closed vessel tests. The power produced in a chemical reaction is not constant with time; it increases to a peak value, then diminishes. As the power declines, the pressure inhibits dispersion, therefore, in the closed system tests, a dispersion above 50% was maintained for only a few seconds.

Figure 36. Test for Thermal Equilibrium.

Figure 38. Closed System Void Fractions (no cooling)

Figure 37. Closed System Vapor Velocities (no cooling).

An additional heat sink would be needed to reduce the pressure sufficiently to maintain dispersion for a longer period of time.

The question arose whether the data obtained from the calorimeter runs would predict the results of the closed vessel system. Two problems occur in this comparison. First, the power curve is not a convenient function of time. For computational purposes the 8M power curve obtained from calorimeter data was modeled in two segments. The first segment (0 to 14 sec) was fit with a normal distribution curve where $\mu=8$, $\sigma=2$, and $A=90$. The second segment (14 to 40 sec) was represented by a constant power (2.9 cal/sec) which was the average power for that segment. The actual power curve and the modeled curve are shown together in Figure 39.

The second problem was the assumption of thermal equilibrium. Thermal equilibrium between phases must be assumed to predict the temperature and pressure rises in a closed system from calorimeter data. In reality the reaction was not in equilibrium. Equations 3 and 4 were solved numerically for the conditions $h_2 = 10^{-4}$, $h_1 = 1 \text{ cal/cm}^2 \text{ sec}^\circ\text{C}$, and $\dot{Q}(t)$ is described as modeled in Figure 39. The predicted temperature compared to actual temperature is shown in Figure 40. The pressure comparison is shown in Figure 41.

The general shape of the predicted temperature curve is similar to that measured in the closed system tests. The disagreement can be explained by two factors. First, the normal curve only approximates the power generation measured in the calorimeter. The real power starts sooner, rises less sharply

Figure 39. Power Generation Curve.

Figure 40. Predicted and Actual Temperatures.

Figure 41. Predicted and Actual Pressures.

and falls off more sharply than the normal curve. Second, the heat transfer is much greater in an open vessel system, used to measure the power curve, than in the closed vessel system being modeled. Because of heat loss, the power generation term starts later, rises more sharply and drops off much more sharply than the closed vessel tests. These effects are clearly observed in Figure 40. The calculated curve starts out more slowly than the measured curve. Once the power starts rising it does so rapidly, but the extra heat loss slows down the sensible heat.

The effect of non-thermal equilibrium is seen in Figure 41. Curve A shows the pressure measured in the closed vessel. Curve B shows what the pressure would be as calculated from temperature data assuming thermal equilibrium. Curve C shows the pressure calculated by the computational model from calorimetric data. The pressure is clearly difficult to predict when the system is not in thermal equilibrium. The best procedure to follow is to make the experiments as close as possible to the actual conditions simulated.

To complete the closed vessel studies of dispersed fluids, a cooling jacket (Figure 33) was used as a heat sink to allow additional condensation which increases the vapor velocity. Water was run through the cooling tube at different flow rates. The resulting vapor velocities are shown in Figure 42 for a 4M and a 9M reaction.

Curve A shows a 4M reaction without cooling. The vapor velocities were greater in the new vessel than measured previously for two reasons. First, the additional glass used in the cooling coil increased the mass of the vessel and therefore the heat capacity was

Figure 42.. Vapor Velocities with Cooling

higher. Second, the smaller cross sectional area of the vessel increased the vapor velocities by a factor of 1.2 . Curve B shows a 4M reaction with cooling. The temperature change was 0.18°C with a water flow rate of 5.81 ml/sec which removed heat at a rate of 1.05 cal/sec. This small heat removal (only a 14% increase) produced vapor velocities above 8 cm/sec for about 25 sec. The void fraction was zero in the case of 4M without cooling but stayed above 50% for 4M with cooling.

Curve C shows a 9M reaction without cooling. Curve D shows the 9M reaction with cooling. The temperature change was 0.75°C with a flow rate of 5.81 ml/sec removing 4.36 cal/sec (a 12% increase). The vapor velocity remained above 8 cm/sec for much longer when cooled than when not cooled. The void fraction also stayed above 50% for much longer in the case of cooling.

The pressure rise rate for the reactor system at full power is approximately 0.45 MPa/sec. The pressure rise rate for the 4M reaction was about 0.007 MPa/sec for the non-cooled and 0.010 MPa/sec for the cooled system. This represents a pressurization rate of 1.6% and 2.2% of the reactor rate produced by the 4M chemical reaction. The 9M rates were 0.036 and 0.038 which is 17% and 19% of the reactor. To produce a 0.45 MPa/sec pressurization rate, the reactor must have a vapor velocity of about 53 cm/sec. Vapor velocities in the range of 16 to 19 cm/sec were obtained in the chemical systems. A chemical reaction producing only about 2% of nominal reactor power maintained a void fraction high enough to be dispersed in the closed system. Again, decay

heat levels would be sufficient to produce the heat necessary to disperse the fuel even in a closed system. Any axial or radial heat sink would greatly increase the amount of dispersion.

Suggestions for Future Work

The main difficulty encountered in the closed vessel experiments was the non-thermal equilibrium condition. Large errors were avoided by measuring both the temperature and the pressure. Other errors were present, however, because of assuming lumped capacities. There are thermal gradients across the liquid, vapor, and vessel, especially in the axial direction since the heat is produced only in the bottom of the vessel.

The experiments could be improved by collecting more data for each run. Thermocouples could be attached to the outside and inside of the glass vessel in several axial positions to determine axial temperature differences and the heat transfer to and from the vessel. Several temperature measurements could be made in the liquid and in the vapor to evaluate the degree of deviation from thermal equilibrium.

In addition to overcoming the non-equilibrium problems, the range of pressures could be extended by using a high pressure vessel and syringe pump. This would allow more energetic reactions to be used.

CHAPTER 8

CONCLUSIONS

The research presented in this dissertation established two facts; first, chemical reactions can be used for modeling full core accidents in LMFBRs, and second, internally heated fluids are by nature dispersive, even at low heating rates.

Value of Chemical Modeling in HCDA Analysis

Chemical reactions have properties that have certain advantages and some disadvantages over other simulant systems for modeling internally heated fluids. The main advantage is that the chemical simulation can be performed in a small laboratory without the use of expensive equipment. Also, there are a variety of reactions that can be used, each with its unique properties.

It is somewhat difficult to evaluate the power generation term in the region of interest. Multi-component systems are also difficult to handle, though the reactor itself is a multi-component system. Chemical reactions make it possible to have two fluids present, one fluid with a low boiling point (the working fluid) and the other fluid with a higher boiling point, which will disperse. The two component system is much closer to what really occurs in a reactor than a single component system.

Chemical reactions can be controlled by varying the concentration or the injection speed. Large ranges of heating rates can be obtained. The reactions can simulate transients, such as in the transition phase of the HCDA, or slow heating, such as in PAHR.

Chemical reaction simulation of LMFBR accidents can give new insight into the accident because of the unique properties of chemicals. It can also be used to verify conclusions obtained from microwave and Joule heating.

Conclusions of Reactor Simulation Tests

Reactor simulation tests were performed in both open and closed vessel systems. In the open vessel system the applicability of the Kutateladze stability criterion to internally heated fluids was evaluated. The Kutateladze stability criterion predicted a vapor velocity of 4.76 needed to initiate dispersion in the chemical system. The vapor velocity measured experimentally was 8.14 for start of dispersion which is 71% larger than predicted. If the actual vapor velocity needed for fuel to disperse were twice as high as predicted by the Kutateladze stability criterion only 0.20 MJ would be required to disperse the fuel in an open system. This is less than 1% of decay heat levels.

Pressurization in a closed system reduces the vapor velocity. Non-equilibrium conditions present in the closed vessel made it difficult to model. Vapor velocities above 8 cm/sec allowed dispersion in the closed vessel as well as the open vessel. A very small heat sink provided enough condensation to obtain vapor velocities high enough for dispersion.

Even though the vapor velocity is reduced by pressure in the sealed off reactor, small amounts of heat transfer allows condensation to occur so a dispersive flow can be maintained. Dispersion occurred in the chemical system with powers as low as 2% nominal reactor power.

Proposal for Future Research

The main emphasis of this research has been to model the two-phase flow in the transition phase of the HCDA. Many refinements were suggested at the end of chapters 6 and 7 to improve the experiments in open and closed vessel systems. Chemical heating could also be used in the related areas of heat transfer and fluxes during the transition phase and initial part of the PAHR phase.

Baker, et al. (26), used Joule heating and microwave heating to study the effect of internal heating on downward, sideward, and upward heat fluxes. Soluble and non-soluble waxes were used in these simulant material experiments. Others (73-75) have also used simulant materials to model melting in HCDAs, but no one, to our knowledge, has used chemical reactions.

To test the feasibility of using chemical heating for heat transfer analysis, a scoping experiment was performed. A cylindrical container of paraffin was formed to represent the stainless steel lower axial and radial portions of a reactor (see Figure 43). Ten milliliters of DMSO was allowed to react with 10 ml of acetyl chloride. The result was a vigorous boilup of materials. The wax melt pattern is also shown in Figure 43. A photograph of the wax

Figure 43. Wax Melting with Chemical Reaction.

after the reaction is complete is shown in Figure 44. As the reaction cooled, a layer of wax floated to the top of the remaining liquids and solidified.

Wax could be used to simulate core structure melt by exposing it to exothermic reactions. Vertical melting through a horizontal surface, two-dimensional melt: axial and radial, and melting patterns in a sealed system would be studied. Soluble as well as insoluble waxes could be used to model the fuel soluble concrete or insoluble steel.

At the present time a proposal for continued funding from the Department of Energy is pending to continue research in chemical simulation of reactor accidents. The proposed research would include a literature survey of prior experimental techniques used to model heat transfer and melting during HCDAs. Waxes would be identified with the required properties needed for experimental modeling. Vertical melt through a horizontal surface, two-dimensional melt, melt in a closed system with dispersed materials are three of the conditions to be modeled. Conclusions from these experiments would relate to melting patterns in HCDA scenarios.

Figure 44. Actual Sample of Melted Wax.

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