MASTER

Improvement and Verification of
Fast Reactor Safety Analysis Techniques

Progress Report

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ABSTRACT

The critical superficial vapor velocity was measured for an open vessel system with chemically produced internal heat. The measured value was within experimental error of the value predicted by the Kutateladze stability criterion. The greatest error induced was in the measurement of the energy generation rate. The effects of mixtures were taken into account. Further work suggestions are also presented.

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SCOPE

During this period of contract work, research was completed on the open-system verification of Kutateladze stability for internally heated fluids. Further refinements and optional research in this area are suggested. The results of this phase of research is outlined as follows:

1. Introduction
2. Experimental Procedure
3. Void fraction results
4. Effects of mixtures
5. Calorimetry
6. Experimental results
7. Possible future work

INTRODUCTION

One of the unanswered questions in analyzing the transition phase of a hypothetical core disruption accident (HCDA) is fuel motion. Little work has been done in violent boilup of internally heated pools. The stability of the two-phase flow regime has been determined for flat-plate boiling and systems where gas is injected through a fluid. The ratio of the dynamic and surface tension forces is given by a parameter called the Kutateladze number (1)

$$K = \frac{U_D^* \sqrt{\rho_c}}{4\sqrt{g\sigma(\rho_H - \rho_L)}}$$

Fauske suggested the use of the Kutateladze stability criterion to analyze flow regime maps in HCDA's (2).

Using a microwave oven to generate internal heat in water, Farahat, Henry and Santori compared the critical superficial vapor velocity predicted by the Kutateladze number with that measured experimentally (3). The superficial vapor velocity as evaluated from energy and mass balances is

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

The critical superficial vapor velocity (vapor velocity at the breakdown of bubbly flow) was measured at approximately 0.05 m/sec. The calculated critical superficial vapor velocity for a water system was predicted to be 0.074 m/sec.

The purpose of the present research was to use chemical reactions as a source of internal heat to verify the applicability of the Kutateladze stability criterion to internally heated fluids. The advantage of using chemical reactions to model a reactor system is the mixed components. A reactor will have fuel and stainless steel present. The dynamics of a multicomponent system is different from a pure component system. The greatest disadvantage is the ability to predict the exact power generation at any given time.

EXPERIMENTAL PROCEDURE

The basic experimental setup is shown in Figure 1. The reaction vessel is a 250 ml graduated cylinder held in place by a laboratory stand. A digital clock is located next to the cylinder to time the sequence of the reaction. All of those materials as well as a photographic background is placed in a standard chemical hood to remove fumes. One of the reactants is injected into the cylinder by means of a syringe

Figure 1. Experimental Setup.

pump located outside the hood. A teflon tube leads from the syringe into the cylinder. A super-8 movie camera is placed on a tripod in front of the hood. Standard photo floods are set at a 45° angle from the front of the cylinder to provide illumination during photography.

The chemical reaction used in this experiment was the injection of dimethyl sulfoxide (DMSO) into acetyl chloride

Concentrations ranging from 1 molar to 14 molar were obtained by diluting the reactants in benzene. Since the boiling temperature of acetyl chloride is much lower than DMSO, acetyl chloride is the working fluid. For this reason DMSO was used as the titrant rather than acetyl chloride.

For each run, Bakalite was placed at the bottom of the cylinder to assure sufficient nucleation for smooth boilup. Ten ml of acetyl chloride solution was placed in the cylinder. Ten ml of equal concentration DMSO solution was injected into the acetyl chloride at the rate of 24 ml/min. Moving pictures were taken of the reaction at normal speed (18 frames/sec) with a super 8 movie camera. From the movies, the void fraction was determined.

VOID FRACTION 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 head of dispersed liquid. Figure 2 shows liquid in the cylinder prior to boilup. Figure

Figure 2. 4 M Solution, $\alpha = 0$, Prior to boil

Figure 2. 4 M Solution, $\alpha = 60\%$

3 shows a 4m concentration resulting in a void fraction of 60%. Figure 4 shows a 9 m concentration with a 93% void fraction.

The void fraction will vary as a function of time, depending on the heat produced and heat lost. Lower concentrations will obtain a peak void fraction then quickly taper off. Higher concentrations maintain a dispersed state for much longer periods of time. See Figure 5.

A plot of steady state void fraction as a function of concentration shows the concentration of which there is a transition from no flow and dispersed flow. Figure 6 shows a sudden increase of void fraction between a concentration of 3 m and 4 m.

EFFECTS OF MIXTURES

As mentioned in the introduction, the superficial vapor velocity can be calculated from the Kutateladze number .

$$U_u^* = \frac{K \sqrt{g\sigma(\rho_H - \rho_L)}}{\sqrt{\rho_c}}$$

or be determined experimentally from the energy balance.

$$U_u = \frac{\dot{Q}}{V_v/A h_{fg}}$$

In this reaction system there are three initial components: DMSO, acetyl chloride, and benzene. Table 1 shows each of the factors used in the equations, with the units used, and typical values for pure components.

Certain assumptions must be made in order to deal with many components. Since the boiling temperature of acetyl chloride is much lower than DMSO, it was assumed that only the acetyl chloride boiled,

Figure 4. 9 M Solution, $\alpha = 93\%$.

Figure 6. Steady State Void Fraction as a Function of Concentration

Figure 5. Void Fraction Buildup as a Function of Time.

Table 1

<u>Parameter</u>	<u>DMSO</u>	<u>AC</u>	<u>Benzene</u>	<u>3M Value</u>
σ Surface tension (dyne/cm)	40	26.7	26.7	31.5
ρ_H Density, heavy phase (g/cm ³)	1.1014	1.105	0.87865	0.96
ρ_L Density, light phase (g/cm ³) $\times 10^{-3}$	2.06	2.45	2.70	2.74
$\rho_C = \rho_H$ Density, continuous phase (g/cm ³)	1.1014	1.105	0.87865	0.96
V_v Vapor specific volume (cm ³ /g)	485.2	338.7	370.9	365
h_{fg} Latent heat of vaporization (cal/gm)	144.6	78.84	103.6	98.3
Tb Boiling temperature (°C)	189	51-52	80.1	74

creating vapor. The remaining liquid was DMSO which was entrained. Since the density for DMSO is about the same as acetyl chloride, there are only two parameters affected by this assumption. The surface tension and latent heat of vaporization. Since the surface tension is taken to the fourth root, only a 2.4% error is created by entraining acetyl chloride, reducing the calculated vapor velocity accordingly. An 83% increase of latent heat of vaporization caused by vaporizing DMSO instead of acetyl chloride could reduce the experimental vapor velocity. Where the boiling temperature of DMSO is so much higher than obtained in the experiment, the latent heat of vaporization will be mainly that of the acetyl chloride solution.

It was also assumed that not enough products are formed during the energetic phase of the reaction to alter the parameters. This is not necessarily a good assumption. Since the reaction is complex, producing a multitude of unknown products, at unknown concentrations, with unmeasured properties, the affect of products was neglected for simplicity. Future research into the mechanism and kinetics of the reaction may refine the results.

Since the reactants are diluted in benzene, an idea of mixing effects on component properties as a function of concentration is needed. The mixing of DMSO in benzene will decrease the density and surface tension. It was assumed that the liquid density would vary linearly between 0.87865 g/cm^3 to 1.1016 g/cm^3 . The surface tension was calculated by using the Macleod-Sugden correlation (4). This also was almost linear with concentration between 27 and 40 dyne/cm.

The vapor specific volume was calculated for the acetyl chloride solution by use of the ideal gas law. Trouton's rule was used to

calculate the latent heat of vaporization as a function of concentration. Though Trouton's rule is inaccurate for polar compounds and in the presence of acetic acid, it gave an indication of the variation.

The values at a 3 molar concentration are shown in Table 1. This is approximate transition from bubbly to dispersed flow.

CALORIMETRY

To determine the experimental vapor velocity, the energy generation rate producing vapor must be measured. Prior calorimeter studies at low concentration (below one molar, in the nonboiling range) with a precision calorimeter disclosed the nonlinearity of the reaction kinetics. Without studying the reaction dynamics and kinetics in detail, the reaction powers could not be extrapolated into the regions of interest. Since the precision calorimeter could not handle boiling without damage it was necessary to build a nonprecision calorimeter to make these measurements, Figure 7. A comparison run of 3 m concentration was about 10% lower in the boiling calorimeter than in the precision calorimeter, which is close enough for the rough measurements being made.

The calorimeter actually measures only sensible heat. An adiabatic system is desired, but with large heat flux there will be some leakage. To compute the heat loss expected in the non-adiabatic void fraction experiments comparison runs were made in the reaction vessel. There was a 42% decrease in sensible heat in the non-adiabatic reaction vessel as compared to the calorimeter.

The actual value wanted is the energy generation rate producing vapor at the transition point between no boil and dispersed flow.

Figure 7. Calorimeter.

The power was measured as the rate of sensible energy prior to boiling. After boiling begins the energy will go into vapor production rather than increase of temperature. The heat loss is increased during boilup because of the increase of surface contact area.

The energy generation rate at transition was about 56 cal/sec as measured in the calorimeter. Since 42% is lost in the non-adiabatic conditions about 33 cal/sec are available for vapor production. Actually some of this will be lost from the increased areas from boilup, 20 cal/sec may be more realistic.

EXPERIMENTAL RESULTS

Using the mixed parameters for a 3 M concentration and $A = 9.62 \text{ cm}^2$, $K = 0.3$, $g = 48.07 \text{ cm/sec}^2$; the calculated critical superficial vapor velocity is 4.02 cm/sec using 20 cal/sec as the energy generation rate, the experimental critical vapor velocity is 7.7 cm/sec. This value is definitely within the experimental error band. The values obtained at ANL for the microwave system was 5.0 cm/sec and 7.4 cm/sec in a water system. Future research could produce refinements in measuring.

POSSIBLE FUTURE WORK

The main area for needed refinement is power measurement. If the reaction were studied in detail so the actual power and products were clearly known, the results would have less error. Some refinement would be realized by running more calorimetry tests and closed vessel pressurization tests to better predict heat losses and heat producing boiling.

The properties of DMSO and acetyl chloride were convenient because of similar molecular weights and densities, but widely differing boiling points. The high energy and long lasting reaction were favorable. Other reactions, such as DMSO and acetyl bromide, could also be studied.

The affect of heat loss on dispersion height in an open vessel would also give more insight into the problem.

Chemical reaction heating can given insight in experiments modeling of HCDA's.

REFERENCES

1. S.S. Kutateladze, "Elements of the Hydrodynamics of Gas-Liquid Systems," Fluid Mechanics - Soviet Res., 1,4,29 (1972).
2. H.K. Fauske, "Boiling Flow Regime Maps in LMFBR HCDA Analysis," Trans. Ans. Vol. 22, 385 (1975).
3. M. Farahot, R.E. Henry, and J. Santori, "Fuel Dispersal Experiments with Simulant Fluids," CONF-761001, International Meeting on Fast Reactor Safety and Related Physics, Chicago Oct. 5-8, 1976.
4. R.C. Reid, J.M. Prausnitz, and T.K. Sherwood, The Properties of Gases and Liquids, McGraw-Hill Book Co. (1977).