FLUID DYNAMICS AND MASS TRANSFER
IN VERTICAL SUBMERGED GAS JETS

by

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ABSTRACT

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Submitted to the Department of Materials Science and Engineering on January 13, 1977 in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

The fluid mechanics of vertical gas jets injected into liquids have been studied using nitrogen, argon, and helium jets injected into water, liquid tin, liquid solder, and liquid iron. High speed films were taken of gas jets entering water. Two jet flow regimes were observed: one in which large oval shaped bubbles were produced at the nozzle which then shattered shortly after detachment, and one in which a steady cone of gas passed out of the nozzle and broke up continuously into small bubbles. The transition between the two flow regimes was gradual, with the jet fluctuating between the two over a wide transition range. The steady cone jet regime was generally predominant at mass flows through the orifice greater than 40 g/cm² sec.

A technique was developed to distinguish between the flow regimes by observing the sharp fluctuations of pressure in the nozzle chamber which occurred whenever a bubble was nucleated in the bubbling flow regime. This technique was tested by producing high speed films simultaneously with nozzle chamber pressure traces in the nitrogen-water system and recording the output of an electrical gas probe placed near the orifice in the gas-liquid solder system. The transition between the flow regimes in liquid metal systems is similar to that in water and takes place at approximately the same mass flow rate of gas.

The theoretical treatment used by Kumar and Kuloor to predict bubble sizes formed at nozzles at low flow rates was modified to account for the increased gas momentum forces and changes in bubble geometry in high speed jets. The modified equations were used to successfully predict the sizes of bubbles produced at the orifice in the bubbling
jet flow regime in gas-water and gas-liquid metal systems. A force balance approach assuming spherical geometry is proposed for determining which bubble size equations apply under various conditions.

The absorption and desorption of nitrogen in liquid iron was used to estimate the gas-liquid interfacial area produced by submerged gas jets injected into liquid iron. An interfacial area of 1.107 cm$^2$ per cm$^3$/sec of gas injected at standard temperature and pressure was observed. This figure is in good agreement with estimated interfacial areas in industrial systems.

Thesis Supervisor: T. B. King
Title: Professor of Metallurgy
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I. INTRODUCTION

In a number of important metallurgical processes, high speed reactive gas jets are injected beneath the surface of a metal bath for refining. Notable processes using this technique include the Q-BOP or OBM steelmaking process where oxygen is injected into the bath with natural gas shielding and the AOD process for stainless steel where oxygen or an argon-oxygen mixture is injected into the melt. The high speed jet breaks up into a large number of small bubbles giving a large interfacial area for mass transfer. In the course of the breakup there is considerable liquid phase turbulence which provides good mixing and causes accelerated refractory wear.

The mechanism by which the jet breaks up is not yet known, and it is not yet possible to predict from fluid dynamic considerations the interfacial area, the mass transfer rate, or the rate of wear of refractories in these systems. Instead, these must be estimated based on industrial experience. The theoretical understanding of submerged gas jet behavior as it applies to liquid metal processing systems must be improved substantially before it can be of use in the design and control of industrial scale apparatus. This study will attempt to develop new techniques for the study of submerged gas jets and to apply theoretical approaches
developed for bubble formation at lower flow rates to gas jet behavior. The rate of mass transfer between a submerged gas jet and liquid iron is used to estimate the interfacial area for mass transfer created in the system.
II. LITERATURE SURVEY

A. FLUID MECHANICS OF GAS STREAMS ENTERING LIQUID BATHS

1. Introduction

   When a stream of gas passes through a nozzle into a liquid bath, it breaks up into bubbles which then rise to the surface of the liquid. At low gas flow rates, bubbles form sequentially at the nozzle orifice and at high flow rates, the bubbles are formed by the breakup of a high speed gas jet. Although little information is available on the breakup of high speed gas jets, the fluid mechanics of sequential bubble formation have been the subject of considerable study.

   The size of the bubbles formed at the nozzle orifice can be calculated by balancing the forces acting on the growing bubble in the upward and downward directions. Forces acting upwards on a gas bubble forming at a horizontal orifice are produced by buoyancy and by the momentum of the gas passing through the nozzle. These forces are opposed by downward acting forces produced by the surface tension of the liquid, viscous drag in the liquid, and the inertia of the liquid which is displaced by the growing bubble.

   If the bubble is assumed to be spherical, the magnitude of each of the forces can be calculated from the flow
rate, orifice dimensions, and properties of the fluids so that a theoretical bubble size can be calculated. At a particular flow rate, some of the forces may be small enough that they may be neglected in the force balance. As the flow rate of the gas through a given orifice is increased, the relative magnitudes of the forces change. This causes the bubble size to pass through a sequence of flow regimes in which different properties affect the bubble size. The importance of the changing flow regimes on the bubble size was not apparent to early investigators, and as a result, a number of contradictory empirical equations have been proposed for calculating bubble size.

For gas flow into low viscosity liquids such as water or liquid metals, three flow regimes can be distinguished: one in which the force balance between buoyancy and surface tension forces controls the bubble size, one in which the force balance between buoyancy and liquid inertial forces controls the bubble size, and one in which the balance between gas momentum and liquid inertial forces controls the bubble size. Liquid viscous forces are never substantial in these systems. The first two of these flow regimes have been the subject of most of the work in the literature.

2. Buoyancy - Surface Tension Controlled Bubbles

The buoyancy force acting on a spherical bubble is
given by equation (1):

\[ F_b = \frac{4}{3} \pi R_b^3 (\rho_l - \rho_g) g = \frac{4}{3} \pi R_b^3 \rho_l g \]  \hspace{1cm} (1)

where:

- \( F_b \) = buoyancy force \hspace{2cm} \text{(dynes)}
- \( R_b \) = bubble radius \hspace{2cm} \text{(cm)}
- \( \rho_l \) = liquid density \hspace{2cm} \text{(g/cm}^3\text{)}
- \( \rho_g \) = gas density \hspace{2cm} \text{(g/cm}^3\text{)}
- \( g \) = acceleration due to gravity \hspace{2cm} \text{(cm/sec}^2\text{)}

\( \rho_l \) is typically larger than \( \rho_g \) by a factor of \( 10^3 \) or more.

The surface tension force acting on a spherical bubble is given by equation (2):

\[ F_s = 2\pi \sigma r_n \cos \theta \]  \hspace{1cm} (2)

where:

- \( F_s \) = surface tension force \hspace{2cm} \text{(dynes)}
- \( \sigma \) = surface tension \hspace{2cm} \text{(dynes/cm)}
- \( r_n \) = nozzle radius \hspace{2cm} \text{(cm)}
- \( \theta \) = contact angle between gas, liquid, and nozzle material.
Cosθ is generally taken as 1. Equating these two forces yields an equation for the theoretical bubble first given by Maier\textsuperscript{1}

\[ D_b = \left( \frac{6\delta d_n}{g \left( \rho_l - \rho_g \right)} \right)^{1/3} \]  

(3)

where:

- \( D_b \) = bubble diameter (cm)
- \( d_n \) = nozzle diameter (cm)

This equation has been confirmed by studies in a number of liquids including liquid metals\textsuperscript{73,74} and forms the basis of the bubble size method for measuring surface tension of liquids.

3. **Buoyancy - Liquid Inertia Controlled Bubbles**

As the flow rate of gas through the nozzle is increased, the bubbles become larger than the size given by equation (3), and a number of empirical equations have been proposed to correlate bubble sizes.\textsuperscript{2,3,5} This change in flow behavior is due to the effects of liquid inertia forces, which become larger than the surface tension forces at high flow rates.

A theoretical evaluation of the liquid inertial force
on a growing bubble was first given by Davidson and Schuler. Assuming that a bubble grows as a sphere, Davidson and Schuler made use of the virtual mass concept to calculate the rate of change of inertia of the rising bubble.

When a solid body moves within a body of inviscid fluid, a certain amount of fluid motion is produced by the motion of the body. For simple geometries, the effect of the fluid motion on the force required to accelerate the body can be conveniently treated as an added mass which is a function of the geometry of the system and the fluid density. The virtual mass of a body is the sum of the actual mass of the body and the added mass term.

For a bubble moving in a direction perpendicular to a nearby wall, the virtual mass is given by equation (4):

\[ M_V = \rho g V_b + \frac{4}{3} \rho_1 V_b + \rho V_b \frac{3R_D^3}{16b^3} \]  

(4)

where:

\[ M_V = \text{the virtual mass of the bubble} \quad (g) \]
\[ V_b = \text{the volume of the bubble} \quad (\text{cm}^3) \]
\[ b = \text{the distance from the wall to the center of the bubble} \quad (\text{cm}) \]
If the base of the bubble is fixed at the wall, \( R_b = b \) and equation (4) reduces to equation (5):

\[
M_v = \rho g V_b + \frac{11}{16} \rho_1 V_b \quad (5)
\]

A force balance can then be written between the buoyancy force on the bubble and the rate of change of momentum of the bubble.

\[
(\rho_1 - \rho) V_b g = \frac{d(M_v u_b)}{dt} \quad (6)
\]

where:

\[
u_b = \text{bubble velocity} \quad \text{(cm/sec)}
\]

Using the fact that \( V_b = Qt \) where \( Q \) is the volumetric flow rate of the gas (cm\(^3\)/sec), and \( t \) is the time elapsed since bubble nucleation (sec), Davidson and Schuler integrated equation (6) for the case of constant gas flow. With the assumption that the end of bubble growth occurs when the distance risen equals the bubble radius, they predicted a theoretical bubble volume given by equation (7):

\[
V_b = 1.378 \frac{Q^{6/5}}{g^{3/5}} \quad (7)
\]
The volume of the nozzle chamber may also affect bubble sizes in this flow range. A large nozzle chamber may act as a capacitor producing a fluctuating flow rate through the nozzle as the resistance to flow changes during the bubble growth cycle. Only bubble formation under constant flow conditions, which occur with high flow rates or small nozzle chambers, will be considered in this study.

Kumar, Kuloor, and co-workers\textsuperscript{11,12,13,14} have attempted to improve the accuracy and convenience of the Davidson and Schuler model by considering the bubble formation as a two stage process. During the first stage of growth, the downward acting forces are greater than the upward acting forces so that the bubble grows as a sphere with its base fixed at the orifice. The end of the first stage of growth is encountered when the upward and downward acting forces are exactly balanced. The second stage of growth consists of further spherical growth as the bubble rises, being connected to the nozzle by a thin neck. Detachment is assumed to occur when the bubble has risen a distance equal to the radius of the bubble produced in the first stage. The volume growth during this stage is then added to the volume of the force balance bubble to give the final bubble volume.

The volume of the bubble at the end of the first stage of growth, referred to as the force balance bubble,
is calculated by equating the buoyancy force with the rate of change of bubble momentum as in equation (6). The term on the right hand side of equation (6) becomes equation (8).

\[
\frac{d(M_v u_b)}{dt} = u_b \frac{dM_v}{dt} + M_v \frac{du_b}{dt}
\]  

(8)

The two terms on the right hand side of equation (8) can be evaluated. If the bubble grows as a sphere, the rate of change of bubble radius is related to the volumetric flow rate of gas by equation (9)

\[
Q = \frac{dV_b}{dt} = \frac{d(4/3\pi R_b^3)}{dt} = 4\pi R_b^2 \frac{dR_b}{dt}
\]  

(9)

If the bubble is much larger than the nozzle orifice then the bubble velocity during the growth stage is approximately equal to the rate of change of bubble radius.

\[
u_b \approx \frac{dR_b}{dt} = \frac{Q}{4\pi R_b^2}
\]  

(10)

\[
\frac{du_b}{dt} = \frac{d(4/3R_b^2)}{dt} = -\frac{2Q}{4\pi R_b^3} \frac{dR_b}{dt} = -\frac{Q^2}{8\pi^2 R_b^5}
\]  

(11)

The virtual mass of the bubble is given by:
\[ M_V = (\rho_g + 11/16\rho_1)(4/3)\pi R_b^3 \]  

(12)

Whereupon:

\[ \frac{dM_V}{dt} = (\rho_g + 11/16\rho_1)4\pi R_b^2 \frac{dR_b}{dt} = (\rho_g + 11/16\rho_1)Q \]  

(13)

Substitution of equations 10 through 13 into equation (6) gives equation (14):

\[ \frac{d(M_V u_b)}{dt} = \frac{Q^2}{12\pi R_b^2} (\rho_g + 11/16\rho_1) = (\rho_1 - \rho_g) V_V g \]  

(14)

This leads to a prediction for the radius of the force balance bubble given by equation (15):

\[ R_b = \frac{Q^{2/5}}{12g(\rho_1 - \rho_g)} \left(\rho_g + 11/16\rho_1\right)^{1/5} \approx \frac{33}{768\pi^2} \frac{1/5}{g^{1/5}} Q^{2/5} \]  

(15)

The further growth taking place during the second stage of growth brings the bubble to a final volume given by equation (16):

\[ V_b = 0.976 \frac{Q^{6/5}}{g^{3/5}} \]  

(16)

This equation is in better agreement with the experimental results than equation (7) proposed by Davidson and Schuler.\textsuperscript{8,9}
It is difficult to develop a theoretical equation which can predict bubble sizes in this flow regime with great precision in actual systems. Other forces besides the liquid inertia and buoyancy forces have small effects on the bubble size, and many studies have failed to account for the nozzle chamber volume effects. Reference 14 includes a large number of equations derived by Kumar and Kuloor and co-workers to account for these effects under a wide range of conditions.

In addition, the geometric assumptions involved in any theoretical model of bubble formation may not be exactly followed in an actual system.

A number of alternative approaches to the calculation of bubble volumes formed at an orifice have also been proposed. Wraith\textsuperscript{15} used a two-stage model in which the first stage of growth is assumed to take place in hemispherical geometry with a transition to spherical growth in the second stage. Kupferberg and Jameson\textsuperscript{16,17} and McCann and Prince\textsuperscript{18,19} used potential flow analysis of bubble growth coupled with simplified equations for nozzle flow and fluctuations in nozzle chamber pressure to analyze complex bubbling and liquid backflow behavior in the range where both liquid inertia and nozzle chamber volume effects are important. For the case of bubble growth under constant pressure conditions, Potter\textsuperscript{20} proposed a term to account for the
change in liquid height during bubble formation in a finite liquid container, and LaNauze and Harris\textsuperscript{21} proposed modifications to account for the effect of the radial acceleration of the liquid surrounding the bubble on the gas flow through the orifice.

The principal difference between the alternative theoretical approaches is that the approach of Kumar and Kuloor\textsuperscript{14} contains some assumptions about the stages of bubble growth, which are not entirely justifiable, in order to simplify the mathematics and permit the calculation of bubble volumes by algebraic equations. The approaches of the other investigators are more rigorous, but generally require the use of numerical integration techniques to predict bubble volumes and describe the dynamics of bubble formation.\textsuperscript{22}

These theoretical equations for the formation of gas bubbles at nozzles are relatively untested in liquid metal systems because of the difficulty of dealing with opaque, high temperature fluids. Only a few experimental studies have been carried out. Patel\textsuperscript{23} measured bubble volumes for argon injected vertically upwards in liquid iron at 1600°C through a 1.2 mm diameter nozzle by measuring the frequency of bubbles arriving at the surface. Fruehan\textsuperscript{24} used the same technique to measure the volume of argon bubbles injected horizontally into liquid iron at 1610°C.
through a 2 mm diameter nozzle. In both cases, the measured bubble volumes were much larger than predicted by equation (3).

Acoustical techniques, which make use of the sound produced by the detachment of the bubble, have been used to measure frequencies and volumes of bubbles injected vertically downward in cast iron at 1450°C by Berdnikov et al. Acoustical techniques have also been used by Andreini and Foster to investigate bubble formation in liquid lead and copper. Sano, Mori, et al. used the pressure pulse produced by the formation of the bubble to investigate vertically upward bubble formation in liquid mercury and silver. They found that when gas was injected into the metal through thick walled glass capillaries, the outer diameter of the capillary, rather than the inner diameter, should be used to calculate the bubble volume because the glass is not wetted by the liquid metals. They also investigated the effects of chamber volume and flow rate on bubble volumes.

4. **Bubble Formation at High Gas Flow Rates**

   No theoretical treatment has been developed to describe bubble formation at high gas flow rates where forces due to gas momentum must be considered. A number of experimental studies have been carried out in gas-water systems
which indicate the general features of high speed submerged jet behavior. The classical work in this area is that of Leibson et al.\textsuperscript{6}, which covered a wide range of flow rates in the air-water system. They produced high-speed motion picture films of bubble formation in the nozzle region and measured bubble volumes from photographs taken at a section approximately 3 feet (92 cm) above the orifice. They presented their results in terms of the Reynolds Number at the orifice ($N_{Re} = 4W/\pi d_n \mu_g$, where $W$ is the mass flow rate of the gas and $\mu_g$ is the gas viscosity) because they attributed the flow regime transitions that they observed to gas turbulence effects and to the effects of liquid circulation around the growing bubbles.

They observed that the bubbles formed at the orifice were elongated in the vertical direction at high Reynolds numbers. With the onset of turbulence ($N_{Re} > 2100$), large, irregular bubbles formed at the orifice, which then shattered apart into smaller bubbles at a height of 3 to 4 inches (7.6 to 10.2 cm) above the nozzle.

In fully developed turbulence ($N_{Re} > 10,000$), what appeared to the naked eye to be a continuous jet was observed by high speed motion pictures to consist of a series of closely spaced, irregular bubbles rising with a counterclockwise swirling motion. The large, irregular bubbles that formed at the orifice exploded at a point about
4 inches (10 cm) above the orifice and produced a large number of small irregularly sized bubbles. At the same time liquid was drawn into the jet by a swirling vortex action.

At Reynolds numbers above 30,000 with orifice sizes greater than 1/64 inch (0.04 cm), a ring of fine bubbles was observed suspended near the orifice. These bubbles rose slowly with some lateral pulsating motion. At Reynolds numbers above about 40,000 clouds of fine bubbles were produced which obscured the visibility of the jet.

No marked changes in flow behavior were observed when the nozzle pressure ratio was decreased below the critical pressure ratio for air. Straight walled nozzles were used so that no supersonic flow could have taken place, but the supercritical flow was not observably different from the subsonic flow. In systems where the surface tension was lowered by mixing 1.6% butanol with the water, clouds of very fine bubbles were produced at lower Reynolds numbers than in pure water. This may indicate that the bubble size in these systems is a function of the interfacial energy.

Leibson et al.⁶ correlated the results of their bubble size measurements in the laminar regime by equation (17)

\[ D_b = 0.18 \frac{d_n^2}{N Re}^{1/3} \]  

(17)
With the onset of turbulence, a randomization of bubble sizes was observed by Leibson et al.\textsuperscript{6}. However, they were able to correlate $D_{VS}$, the bubble diameter with a surface to volume ratio equivalent to that of the bubble size distribution, by equation (18) for Reynolds numbers greater than 10,000.

$$D_{VS} = 0.28 \ N_{Re}^{-0.05}$$

(18)

It should be noted that this bubble size distribution was measured at a height of 36 inches (92 cm) above the nozzle and therefore may not be representative of the bubble size distribution at other levels in the jet.

Oryall and Brimacombe\textsuperscript{76} have also filmed the formation of large irregular, bursting bubbles in horizontal air jets injected into water. They also used an integrating electrical gas probe to measure three dimensional gas fraction and bubble frequency profiles in horizontal air-mercury jets. The jet expansion and penetration that they observed in the air-mercury system could be best explained by bursting, pulsating behavior in the jet.

At still higher gas flow rates, the gas leaves the orifice in a continuous stream forming a cone which breaks up into small bubbles at some distance from the orifice. Turkdogan\textsuperscript{33} has proposed that in this flow regime, liquid is entrained in the gas stream from an unstable interface.
and the flow changes to gas bubbles in a liquid stream at a height where the gas has lost most of its kinetic energy. Turkedogan and Tien\textsuperscript{34} have proposed a theoretical analysis of the fluid mechanics of such a jet. The flow regime transition from irregular, bursting bubbles to a steady-walled cone of gas has not been studied and the circumstances under which each flow regime is stable are not clear.

The experimental work of Young\textsuperscript{35} in the steam-water system indicates the possible range of dynamic behavior in submerged gas jets. These studies revealed the existence of six different reproducible regimes of jet behavior which were identifiable by acoustic emissions and visual observations. Effects due to vapor condensation make this behavior more complicated than that expected in two component systems.

Young mapped the flow regimes observed in submerged steam jets as a function of the nozzle pressure ratio and the mass flow per unit area through the nozzle. The thrust produced by the jet action was measured and compared with the theoretical thrust given by equation (19).

\[
F_T = W(u_g - u_1) + (P_n - P_a) \pi r_n^2
\]  

(19)

where
\[ u_g = \text{gas velocity} \quad (\text{cm/sec}) \]
\[ u_l = \text{liquid velocity} \quad (\text{cm/sec}) \]
\[ P_n = \text{nozzle pressure} \quad (\text{dynes/cm}^2) \]
\[ P_a = \text{ambient pressure} \quad (\text{dynes/cm}^2) \]

The measured thrust agreed with the calculated thrust to within 5% for the quiet flow regimes at high and very low pressure ratios, but it was 30% less than the calculated thrust in the noisiest flow regime at intermediate pressure ratios. At the lowest pressure ratios, the steam jet envelope took on a steady bell shape which led Young to conclude that it acted as a supersonic diffuser for the exiting gas.

Bell, Boyce, and Collier\textsuperscript{36} also observed steady cone jets in horizontal nitrogen-water jets operated at 100 to 600 psig. A jet cone angle of approximately 24° was measured. These authors attempted to measure the rate of liquid entrainment in the jets by directing the jet through a hole in a plate which was positioned just far enough from the jet nozzle so that bubbles were not sheared off as the jet passed through the hole. By this technique, liquid entrainments up to 40% by volume were measured at jet lengths of about 20 nozzle diameters. In no case was any entrainment observed at jet lengths of less than 6 mm, after which the volumetric entrained fraction \( \frac{Q_w}{Q_g} \), was roughly
proportional to \((L-6)/d_n\)

Kennedy and Collier\textsuperscript{77} also investigated the behavior of nitrogen-jets injected into water at extremely high driving pressures in the range of 150 to 1900 psig. They photographed a jet at 900 psig in which the steady cone angle could be clearly observed. As the gas leaves the nozzle there is a short length in which the jet expands in a bell shape with a larger angle than the straight walled cone which is observed at higher lengths. The straight walled cone was found to have an angle of \(25^\circ \pm 5^\circ\) for most cases. Similar cone angles were measured when nitrogen jets were injected into ethanol. Kennedy and Collier proposed that the jet expands to ambient pressure in the bell shaped region and that the cone angle in the straight walled region is characteristic of the momentum transfer between the gas and liquid.

Bell, Boyce and Collier\textsuperscript{36} also investigated the structure of the jet cone region by traversing the jet with a photoelectric probe which indicated the frequency and approximate size of the entrained droplets. They found the mass mean drop size roughly equal to 250 microns, with most of the droplets found around the outside of the jet core region. A large number of droplets in the size range of
25 to 50 microns were also observed, which diffused into the core region.

The behavior described by Bell, Boyce, and Collier for the jet cone is similar to the behavior observed in the annular mist flow regime of two-phase, gas-liquid pipe flow. In this flow regime, which occurs at very high ratios of gas flow to liquid flow, nearly all of the liquid flows in a thin annulus between the wall of the pipe and the flow of gas along the center of the pipe. The gas carries some of the liquid within it in the form of entrained droplets. The droplets are believed to be entrained from the tips of Kelvin-Helmholtz instability waves traveling at the gas-liquid interface.

Although annular mist pipe flow behavior is very complex, it has been the subject of considerable study because of its engineering importance. Experience in this area provides the basis for a theoretical model by Chawla of the fluid mechanics of a steady cone submerged gas jet. A simplified analysis of the Kelvin-Helmholtz instability was used to derive a semiquantitative correlation which was combined with the data of Bell, Boyce, and Collier to predict liquid entrainment rates and entrained droplet sizes for submerged air jets into water under different conditions.

A time averaged approach to jet behavior can be
taken by considering the cone angle produced by the jet as it leaves the nozzle and using conservation of mass and momentum to calculate the fraction of entrained fluid and velocity as a function of distance from the nozzle. Donald and Singer\textsuperscript{28} measured jet cone angles photographically in gas to gas and liquid to liquid systems and concluded that the jet cone angle was a function only of the kinematic viscosity of the injected fluid. Themelis, Tarassoff, and Szekely\textsuperscript{29} used time lapse photography to measure jet cone angles in the air to water system and concluded that Donald and Singer's results were valid in gas to liquid systems as well.

Themelis, Tarassoff, and Szekely then deduced a formula to predict the trajectories of horizontal submerged jets based on the ratio of inertial and buoyancy forces on the jet. This analysis has recently been extended by Engh and Bertheussen\textsuperscript{30} to consider the presence of entrained particulates in the gas stream, as in processes where powdered fluxes are injected into the bath with the jet. However, recent experiments by Oryall and Brimacombe\textsuperscript{31,76} in which an electrical gas probe was used to plot jet trajectories in the air-mercury system have shown that gas properties alone do not determine jet cone angles. They observed jet cone angles of 150° to 155°, much larger than the 20° to 25° observed in air-water systems. They also
observed larger vertical components in the jets than would be predicted from the simple theory and back-penetration of the gas from the point of injection.

Igwe, Ramachandran, and Fulton\textsuperscript{32} carried out experiments on horizontal submerged nitrogen jets in several room temperature liquids. They found that for the nitrogen-water system, jet penetration was proportional to a power number defined by equation (20).

\[ N_p = P_u d_n \]  \hspace{1cm} (20)

where $P_u$ is the pressure upstream of the nozzle. Varying liquid density from 0.812 to 2.964 g/cm\textsuperscript{3} showed that increasing liquid density had no effect on jet penetration for densities greater than about 2.25 g/cm\textsuperscript{3}. They also observed that the amount of liquid splashed out of the container at a given gas flow rate could be reduced by increasing the number of submerged gas nozzles.

In these studies, however, time averaged, macroscopic properties of the jets have been used to predict macroscopic characteristics of the jets such as trajectories and penetration distances. To predict interfacial areas and mass transfer rates in submerged gas jets requires the knowledge of microscopic characteristics of the jets such as the mechanism by which liquid is entrained and the droplet and
bubble sizes produced in this process.

The principal problems remaining to be solved in the field of submerged gas injection are in the high flow rate regimes. These are also the areas of most commercial interest in process metallurgy. The behavior of high speed submerged gas jets is known largely on the basis of experiments in the air-water system, and the validity of the empirical relationships established in this system has not been tested in other systems. No theory has been proposed to explain the behavior of the irregular bursting bubbles reported by Leibson et al.\(^6\), and the theoretical understanding of the steady cone jet behavior is rudimentary. The transition between these two high speed jet flow regimes has not been studied and the conditions under which each regime is stable cannot be predicted.

B. NITROGEN ABSORPTION IN LIQUID IRON

The thermodynamics and kinetics of the absorption and desorption of nitrogen in liquid iron have been the subject of considerable study because of the effects of dissolved nitrogen on the properties of steel. Although the thermodynamics of the nitrogen solution reaction are well established, several aspects of the kinetics of the reaction remain unclear.
The rate of absorption of nitrogen into liquid iron is greatly reduced by the presence of surface active elements such as oxygen and sulfur in the iron. This effect has been investigated by Pehlke and Elliott using a Sievert's apparatus, Kozakevitch and Urbain using an apparatus where nitrogen was bubbled through a tube immersed 1 cm below the surface of a molten iron bath, and Schenck, Frohberg, and Heinemann using an apparatus where gas was blown onto the surface of an inductively heated melt. In all of these studies, the rate of absorption was observed to be first order with respect to the nitrogen concentration in the metal. Assuming that the surface active elements act as poisons to the nitrogen transfer by occupying potential adsorption sites on the metal surface, Swisher and Turkdogan proposed an interpretation based on the adsorption isotherm of oxygen on liquid iron at 1550°C which successfully explained the results of references 42, 43, and 44.

Later research, taking place principally in Japan, focused on the nitrogen desorption reaction. Ban-ya, Shinohara, Tozaki, and Fuwa found that the rate of nitrogen desorption was second order with respect to dissolved nitrogen concentration, as did Mori and Suzuki, and Narita, Koyama, Makino, and Okamura. All of these studies involved passing argon gas over inductively heated liquid iron which had previously been saturated with nitro-
gen. Shimmyo and Takami\textsuperscript{49}, using a levitation melting technique, also observed a second order process and analyzed the absorption reaction as second order as well.

Inouye and Choh\textsuperscript{50,51} disputed the use of the second order rate law to describe the nitrogen absorption reaction. Using an apparatus where nitrogen was passed over the surface of an inductively heated melt, the presented results showed that the first order equation gave a better fit to the nitrogen absorption data over a wide range of oxygen and sulfur concentrations.

The apparent asymmetry of absorption and desorption rates for the nitrogen solution reaction requires the proposal of a more complex reaction mechanism than either simple chemical reaction rate control or mass transfer control. A mixed control mechanism in which both a first order surface chemical reaction and liquid phase mass transfer affect the reaction rate has been proposed by Fruehan\textsuperscript{52} to explain nitrogen absorption rates in an experiment where nitrogen was bubbled slowly through a 7 cm deep melt. The asymmetry is better explained by the mixed control model proposed by Mori, Sano, and Suzuki\textsuperscript{53} in which the surface reaction is assumed to be second order with respect to the nitrogen concentration in the metal, and the diffusion of the dissolved nitrogen away from the reaction interface is described by a first order rate equation.
They used the model to explain a similar asymmetry in the absorption and desorption rates of oxygen in liquid silver and presented evidence that the same mechanism could be active in the nitrogen-iron system.

Mori, Sano, and Suzuki$^{53}$ proposed that at steady state the overall rate of nitrogen transfer is given by:

$$\frac{d\%N}{dt} = k_2 \frac{A}{V} (\%N_e^2 - \%N_i^2) = k_1 \frac{A}{V} (\%N_i - \%N) \quad (21)$$

where:

- $\%N$ is the bulk nitrogen concentration (wt\%)
- $\%N_e$ is the nitrogen concentration in equilibrium with the partial pressure of nitrogen in the gas phase (wt\%)
- $\%N_i$ is the nitrogen concentration at the gas-metal interface (wt\%)
- $t$ is time (seconds)
- $A$ is the area of the gas-metal interface (cm$^2$)
- $V$ is the volume of the bath (cm$^3$)
- $k_2$ is the second order rate constant (cm/sec\%)
- $k_1$ is the first order rate constant (cm/sec)

If the first order reaction is very fast, $\%N_i$ will be approximately equal to $\%N$ and the reaction will be described by the second order rate law. If the second order reaction
is much faster than the first order reaction, \( \%N_i \) will be approximately equal to \( \%N_e \) and the reaction will be described by a first order rate law. In intermediate cases, neither rate law should be sufficient.

If \( k'_1 \) is the apparent rate constant observed if the data is plotted according to a first order rate law and \( k'_2 \) is the apparent rate constant observed if the data is plotted according to a second order rate law, the apparent rate constants can be related to the actual rate constants of the two reactions by:\(^53\)

\[
k'_1 = k_2 \left( \frac{1}{\beta} + \frac{1}{\%N_e + \%N_i} \right)
\]

\[
k'_2 = k_2 \frac{\%N_e^2 - \%N_i^2}{\%N_e^2 - \%N^2}
\]

\[
\%N_i = \%N_e \frac{(\%N_e + \%N_i)}{\beta} + \%N
\]

where \( \beta \) is the ratio of the first order and second order rate constants. If \( \beta \ll \%N_e + \%N_i \), then \( k'_1 \approx k_1 \) and a first order rate law is observed. Because \( \beta \) is constant for a given system, experiments where nitrogen is absorbed from a pure gas would have a large \( \%N_e + \%N_i \) and appear to follow a first order rate equation. If \( \beta \gg \%N_e + \%N_i \), then \( \%N_i \approx \%N \) so that \( k'_1 \approx k_2 \) (\( \%N_e - \%N \))
and a second order rate law is observed. Experiments involving desorption of nitrogen to pure inert gases would reduce \( \%N_e + \%N_i \) so that the reaction would appear to follow a second order equation. If the driving force for the reaction is small as is generally the case for experiments in a Sieverts apparatus, \( \%N = \%N_e \approx \%N_i \). Under these circumstances \( k_1^{*} = k_2 / (1 + \frac{1}{2N_e}) \) and \( k_2^{*} = k_1 / (\beta + 2N_e) \) so that either a first or second order rate law could explain the data.

Although this theory gives a good qualitative explanation of the asymmetry effect, there are still some experimental results that have not been adequately explained. If it is assumed that the first order process is controlled by the diffusion of nitrogen in the liquid, then increasing the concentration of surface active elements in the bath should reduce the second order rate constant by reducing the number of surface reaction sites available to the nitrogen. The first order rate constant should be unaffected by the surface active elements since it would be dependent on the nitrogen diffusivity and mixing conditions in the bath. Changing the concentration of surface active elements should then change both the rate controlling step and the order of the reaction.

This interpretation is supported by the effect of increasing oxygen content on the activation energy of the nitrogen solution reaction. At low oxygen levels, the activation energy is approximately equal to the
activation energy for the diffusion of nitrogen in liquid iron of 17500$^{71}$ to 23000$^{72}$ calories per mole. At higher oxygen concentrations, however, it is greatly increased.$^{42,49,50}$ Table 1 shows a number of activation energies measured for the nitrogen solution reaction.

The change in reaction order with changing oxygen concentration that is predicted by this theory has been less firmly established. Inouye and Choh$^{50,51}$ observed simple first order behavior for nitrogen absorption at low oxygen concentrations, but at oxygen concentrations above 0.04% the rate of absorption became proportional to the nitrogen partial pressure rather than to the partial pressure of nitrogen to the one half power as Sievert's Law would imply. The data was still found to fit a first order rate law with respect to the nitrogen concentration in the metal, but the rate constant observed was found to be proportional to the square root of the nitrogen partial pressure. This effect could be due to the increasing importance of the second order reaction on the overall reaction rate, although it is not exactly as predicted in the theory.

Studies of the effect of surface active elements on nitrogen desorption rates have also given inconsistent results. Mori, Sano, and Suzuki$^{53}$ investigated the desorption of nitrogen from iron containing 0.036% to 0.038% O at 1600°C. They found that their results could be explained
by the mixed control model with $k_2 = 0.21 \text{ cm (sec)}^{-1}$ and $\beta = 0.11\%$ for nitrogen partial pressures of 0.02, 0.11, 0.21, and 0.41 atmospheres. Ban-ya, Shinohara, Tozaki, and Fuwa$^{46}$ investigated the nitrogen desorption reaction at the same temperature and oxygen concentrations and found that the simple, second order rate law effectively described the nitrogen desorption rate into atmospheres containing nitrogen partial pressures of 0.043, 0.085, and 0.125 atmospheres. These nitrogen partial pressures may have been too low for the deviation from second order behavior to be apparent. Shimmyo and Takami$^{49}$ reported second order behavior for both the absorption and desorption reactions over a wide range of oxygen concentrations and nitrogen partial pressures in their levitation apparatus.

The implications of the results of Sano, Mori, and Suzuki$^{53}$ on nitrogen desorption rates into pure argon are not consistent with the data for iron containing low oxygen concentrations. For example, the apparent second order reaction constant calculated from their data at $\%O = 0.036$ is $0.17 \pm 0.01 \text{ cm (sec)}^{-1}$ over the nitrogen concentration range from 0.02 to 0.005%. This could easily be mistaken for a pure second order reaction and is in agreement with the observed desorption rate. If the oxygen concentration of the melt is reduced to 0.006%, then the observed second order reaction rate is $0.668 \text{ cm (sec)}^{-1}$. If $k_1$ is unaffected
by the change in oxygen concentration as would be expected if it were controlled by the mass transfer in the liquid, then $k_2$ and $\beta$ must change to give the observed $k'_2$. $k_2 = 0.95 \text{ cm/}(\%\text{sec})^{-1}$ and $\beta = 2.34 \times 10^{-2} \%$ gives $k'_2 = 0.674 \text{ cm}(\%\text{sec})^{-1}$ at 0.005\%N. However, at higher nitrogen concentrations $k'_2$ should be much smaller as is shown in Figure 1. Similarly, $k'_1$ should be much larger at 0.02\%N than at 0.005\%N. The data of Mori and Suzuki\textsuperscript{47} indicates that $k'_2$ does not change significantly over this concentration range. If the mixed control model is valid, and the reaction steps are assumed to be a chemical reaction at the gas-liquid interface and mass transfer of dissolved nitrogen in the liquid metal then these results are inconsistent. The contradiction could be resolved by the selection of a different $k_2$ and $\beta$ at 0.036\%O.

The exact functional dependence of the rate constant on the oxygen concentration has also not been clearly established. The treatment of Turkdogan and Swisher\textsuperscript{45} predicts that the rate constant should be inversely proportional to the oxygen concentration to the first power. Experimentalists have reported that the inverse dependence is to be the first power\textsuperscript{47}, the second power\textsuperscript{46, 48, 49}, and to the two thirds power.\textsuperscript{50} The effect of sulfur on the rate constant is similar to the effect of oxygen except that higher concentrations are required to achieve the same
Figure 1. Calculated and observed effective second order reaction constants for nitrogen desorption at 0.0006% 0.
The reaction rate constant is also reduced by the presence of the surface active elements Te, Se, Sb, and Sn.\textsuperscript{49}

The presence of some non-surface active elements in the iron also affect the rate constant of the nitrogen solution reaction. The effect of carbon is of greatest interest in this study because of the tendency of the bath to accumulate carbon by erosion of the graphite components in the system. Ban-ya, Shinohara, Tozaki, and Fuwa\textsuperscript{46} proposed that the desorption rate was proportional to the square of the activity coefficient of nitrogen in liquid iron. Carbon substantially increases the desorption rate because of its positive interaction parameter with nitrogen, but this effect is reported to saturate at a carbon concentration of about 2%. Narita, Koyama, Makino, and Okamura\textsuperscript{48} found a similar but much smaller effect of carbon on the nitrogen desorption rate. Inouye and Choh\textsuperscript{50} found that carbon had no effect on the absorption rate of nitrogen in liquid iron.

All of the Japanese work was done in inductively stirred melts. Pehlke and Elliott\textsuperscript{42} reported a substantial effect of stirring on the rate constant measured in a Sievert's apparatus. Recent work by Bester and Lange\textsuperscript{54} also showed a very large effect of stirring in a Sievert's apparatus for iron containing less than 50 ppm O, the mass
transfer coefficient being reduced by a factor of 4 or 5 when the melt was completely shielded from induction stirring. For completely shielded melts, they found that the mass transfer in both directions was first order and controlled by nitrogen diffusion in the liquid metal. This may imply that the degree of mixing in induction heated melts is large enough that mass transfer coefficients measured in these systems may be as large as those in bubble stirred experiments.

The purity of the gases used in the study may also affect the observed reaction rates. Ban-ya, Shinohara, Tozaki, and Fuwa\textsuperscript{46} purified their gases in a complex gas train. Narita, Koyama, Makino, and Okamura\textsuperscript{48} used cylinder gases directly and observed lower desorption rates than the other studies. When they added 5\% $\text{H}_2$ to the argon stream in their desorption experiments they observed an increased rate which was in better agreement with the other studies. Since argon may contain as much as $10^{-5}$ atm of oxygen as it is supplied from the cylinder, this oxygen may adsorb on the liquid metal surface to an extent sufficient to reduce the nitrogen desorption rate even when the oxygen concentration in the metal is too low to produce an effect. The addition of 5\% hydrogen to the argon could lower the oxygen partial pressure enough to eliminate this effect.

In summary, although it has been the subject of
considerable study, our knowledge of the kinetics of the nitrogen solution reaction cannot be considered complete. The reaction may take place in two steps, one step probably being a second order chemical reaction on the metal surface and the other probably being the diffusion of dissolved nitrogen to and from the gas-metal interface. The reaction rate may be controlled by the slower of the two steps if one step is substantially slower than the other, or by the two steps in series if the rates are similar. The presence of surface active elements in the metal can slow the rate of the surface reaction step by occupying reaction sites and stirring of the melt can increase the rate of the mass transfer step, but at present no simple model explains the behavior of the system under all conditions. The apparent asymmetry of absorption and desorption rates observed in several experiments is contradicted by the principle of microscopic reversibility which demands that the same reaction step be rate controlling regardless of the direction of nitrogen transfer. The possibility of mass transfer effects in the gas phase affecting the apparent reaction rate cannot be ruled out in experiments where nitrogen is removed from metals by inert gas purging.

For the experiments carried out in this study, which were all either absorption from pure nitrogen or desorption to pure argon, the use of the mixed control model may not
be necessary. The reaction rates measured in previous studies may be used to estimate the gas-liquid interfacial area produced by the jet by comparing them with the reaction rates measured in this study. Since the liquid mixing conditions may be more turbulent in these experiments than in any others, it would be preferable to work under conditions where the reaction rate is controlled primarily by the surface reaction. This condition would be satisfied when the metal surface is saturated with oxygen, but under these conditions the reaction rate is very slow and it is difficult to measure the rate constants with certainty because of the scatter in the experimental points. Nitrogen desorption experiments under conditions of low oxygen concentration produce high rate constants, and since they fit, a second order rate law may be expected to be controlled by the surface reaction rather than by liquid phase diffusion. Several nitrogen absorption experiments were also carried out under low oxygen concentrations to determine how the mass transfer conditions in the submerged gas jet compared with those in other systems.
<table>
<thead>
<tr>
<th>Activation Energy (cal/mole)</th>
<th>%0</th>
<th>Experimental Apparatus</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>11500*</td>
<td>0.005</td>
<td>Sievert's Apparatus with shielded melt</td>
<td>54</td>
</tr>
<tr>
<td>31000</td>
<td>0.009</td>
<td>Gas blown onto melt</td>
<td>50</td>
</tr>
<tr>
<td>25000*</td>
<td>0.01</td>
<td>Sievert's Apparatus</td>
<td>42</td>
</tr>
<tr>
<td>36000*</td>
<td>0.03</td>
<td>Sievert's Apparatus</td>
<td>42</td>
</tr>
<tr>
<td>45000*</td>
<td>0.05</td>
<td>Sievert's Apparatus</td>
<td>42</td>
</tr>
<tr>
<td>63000*</td>
<td>0.07</td>
<td>Sievert's Apparatus</td>
<td>42</td>
</tr>
<tr>
<td>59000</td>
<td>0.08</td>
<td>Gas blown onto melt</td>
<td>50</td>
</tr>
<tr>
<td>40000</td>
<td>&quot;low&quot;</td>
<td>Bubbling</td>
<td>43</td>
</tr>
</tbody>
</table>

**B. Second Order Analysis**

| 60000 | 0.09 | Levitation melt | 49 |
| 96000 | 0.14 | Levitation melt | 49 |

**2. Nitrogen Desorption - Second Order Analysis**

| 31900 | 0.004-0.006 | Gas blown onto melt | 47 |
| 38600 | 0.006-0.011 | Gas blown onto melt | 48 |
| 68000 | 0.075       | Levitation melt     | 49 |
| 96000 | 0.138       | Levitation melt     | 49 |

*Activation energy applies to both absorption and desorption.*
### TABLE 2
REPORTED NITROGEN TRANSFER RATE CONSTANTS

1. **Nitrogen Absorption**

   **A. First Order Analysis**
   \[ \frac{d\%N}{dt} = k \frac{A}{V} (\%N_e - \%N) \]

<table>
<thead>
<tr>
<th>(k) (cm/sec)</th>
<th>(O) (wt%)</th>
<th>Experimental Technique</th>
<th>Temperature (°C)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>*32x10^{-3}</td>
<td>0.009</td>
<td>Sievert's Apparatus</td>
<td>1606</td>
<td>42</td>
</tr>
<tr>
<td>*20.2x10^{-3}</td>
<td>0.009</td>
<td>Sievert's Apparatus shielded melt</td>
<td>1606</td>
<td>42</td>
</tr>
<tr>
<td>*4.7x10^{-3}</td>
<td>0.08</td>
<td>Sievert's Apparatus</td>
<td>1606</td>
<td>42</td>
</tr>
<tr>
<td>*30.2x10^{-3}</td>
<td>0.005</td>
<td>Sievert's Apparatus</td>
<td>1600</td>
<td>54</td>
</tr>
<tr>
<td>*6.5x10^{-3}</td>
<td>0.005</td>
<td>Sievert's Apparatus shielded melt</td>
<td>1600</td>
<td>54</td>
</tr>
<tr>
<td>34.5x10^{-3}</td>
<td>0.005</td>
<td>Gas blown over melt</td>
<td>1600</td>
<td>50</td>
</tr>
<tr>
<td>28x10^{-3}</td>
<td>0.011</td>
<td>Gas blown over melt</td>
<td>1600</td>
<td>50</td>
</tr>
<tr>
<td>12x10^{-3}</td>
<td>0.0095</td>
<td>Bubbling gas</td>
<td>1610</td>
<td>52</td>
</tr>
<tr>
<td>2.2x10^{-3}</td>
<td>0.1495</td>
<td>Bubbling gas</td>
<td>1610</td>
<td>52</td>
</tr>
</tbody>
</table>

   **B. Second Order Analysis**
   \[ \frac{d\%N}{dt} = k \frac{A}{V} (\%N_e^2 - \%N^2) \]

<table>
<thead>
<tr>
<th>(k) (cm/sec)</th>
<th>(O) (wt%)</th>
<th>Experimental Technique</th>
<th>Temperature (°C)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>*0.32</td>
<td>0.005</td>
<td>Levitation</td>
<td>1600</td>
<td>49</td>
</tr>
<tr>
<td>*0.01</td>
<td>0.15</td>
<td>Levitation</td>
<td>1600</td>
<td>49</td>
</tr>
</tbody>
</table>

2. **Nitrogen Desorption - Second Order Analysis**

<table>
<thead>
<tr>
<th>(k)</th>
<th>(O) (wt%)</th>
<th>Experimental Technique</th>
<th>Temperature (°C)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.7</td>
<td>0.005</td>
<td>Gas blown over melt</td>
<td>1600</td>
<td>47</td>
</tr>
<tr>
<td>0.06</td>
<td>0.09</td>
<td>Gas blown over melt</td>
<td>1600</td>
<td>47</td>
</tr>
<tr>
<td>0.6</td>
<td>0.01</td>
<td>Gas blown over melt</td>
<td>1600</td>
<td>46</td>
</tr>
<tr>
<td>0.05</td>
<td>0.093</td>
<td>Gas blown over melt</td>
<td>1600</td>
<td>46</td>
</tr>
<tr>
<td>0.38</td>
<td>0.006</td>
<td>Gas blown over melt</td>
<td>1600</td>
<td>48</td>
</tr>
<tr>
<td>0.007</td>
<td>0.065</td>
<td>Gas blown over melt</td>
<td>1600</td>
<td>48</td>
</tr>
</tbody>
</table>

*Rate constant applied to both absorption and desorption.
III. OUTLINE AND PLAN OF WORK

The purpose of this research was to investigate the fluid dynamics of high speed submerged gas jets in liquid metals and to determine the parameters that affect the jet behavior in order to be able to predict the effects of operating variables on gas-liquid reaction rates and on refractory wear in steel making systems.

Since direct observation of gas jets in liquid metals is not possible, the first stage of the study was devoted to the observation of submerged gas jets in a water bath using the technique of high speed cinematography. Two distinct types of submerged gas jets were observed; one in which large oval shaped bubbles were formed at the nozzle orifice which then shattered into smaller bubbles shortly after detachment from the nozzle; and one in which an apparently steady cone of gas passed out of the nozzle and broke up continuously into small bubbles some distance from the nozzle.

Based on this observation, a number of experiments were carried out in which parameters such as the nozzle diameter, the ratio of the nozzle length to its diameter, the depth of nozzle submergence in the liquid bath, and the density of the injected gas were varied to determine their effects on the gas jet behavior. Several mass transfer
experiments were also carried out. These involved the absorption of SO₂ from a gas containing 1% SO₂ in an inert carrier gas injected into a water bath containing 0.3% H₂O₂. This reaction was selected for investigation because the rate had previously been shown to be controlled by diffusion of SO₂ in the gas phase under the conditions of the present experiments. The gas jet could be manipulated to give bubbling or steady cone behavior and the effects of the flow regime transition on the mass transfer rate could be observed.

A technique was developed for detecting bubble formation in the bubbling type jet by measuring the pressure in the nozzle chamber. The relationship between the pressure fluctuations in the nozzle and the behavior of the submerged gas jet was tested by simultaneously recording a sequence of pressure fluctuations in the nozzle on an oscilloscope and taking a high speed film of the jet in the nitrogen-water system. All gas-water experiments are described in Chapters IV-A and V-A.

With the technique established, the pressure pulse technique was then used to investigate jet behavior in gas-liquid tin and gas-liquid solder systems. An electrical gas probe was mounted on the nozzle near the orifice to check the relationship between nozzle chamber pressure fluctuations and actual jet behavior. These experiments
are described in Chapters IV-B and V-B.

After the technique had proven effective in the gas-
solder system, it was applied to the gas-liquid iron system. 
Mass transfer experiments were carried out on the absorption 
of nitrogen in liquid iron from submerged nitrogen jets 
under various flow conditions and the desorption of nitrogen 
from the metal to argon jets. The rate of the nitrogen 
dissolution reaction may be controlled by mass transfer in 
the liquid phase by a chemical reaction at the gas metal 
interface depending on the concentration of oxygen or other 
surface active solutes. Thus this reaction may be used to 
measure gas-liquid interfacial areas or to investigate mass 
transfer effects, depending upon the experimental conditions. 
Nitrogen absorption and desorption are also of considerable 
importance in several steelmaking systems. These experi-
ments are described in Chapters IV-C and V-C.

A theoretical model to explain the observed jet 
behavior in the bubbling jet regime was developed based on 
the concepts used to describe bubble growth in lower 
velocity, room temperature systems. Also in Chapter VI, an 
explanation for the transition between the two flow regimes 
is proposed and a qualitative description of the steady 
flow regime is given.

The kinetics of the nitrogen dissolution reaction in 
liquid iron and general considerations of mass transfer in 
submerged gas jet systems are discussed in Chapter VI as well.
IV. EXPERIMENTAL APPARATUS AND PROCEDURE

A. GAS-WATER EXPERIMENTS

1. Liquid Container and Gas Flow Apparatus

Water was contained in a rectangular, transparent tank, 30.5 cm wide by 30.5 cm deep by 71 cm tall. The walls of the tank were plexiglass, except for a glass front wall through which the films were taken. The walls were held together by a welded aluminum frame, and the seams between the walls were sealed with silicone rubber cement. The flow nozzle was mounted in the center of the bottom plate and a drain was placed in the bottom plate between the nozzle and a corner.

Brass flow nozzles were machined in the shape of hexagonal-headed bolts. The nozzle orifice was in the center of a 2.5 cm hexagon and the stem of the bolt was threaded to fit a 3/8 inch pipe fitting. When mounted in the tank, the nozzle orifice was 3.8 cm above the bottom of the tank. Orifice dimensions are shown in Table 3. All nozzles were straight-walled with the orifice diameter bored to the nearest British equivalent of the listed metric dimension.

Helium, nitrogen or argon was supplied to the nozzle from cylinders through copper or iron pipe of various sizes
ranging from 1/4 to 3/4 inch in different configurations depending on the particular experiment being carried out. High pressure flexible tubing was used where flexibility was required in the gas supply lines. Bourdon type pressure

<table>
<thead>
<tr>
<th>Diameter (mm)</th>
<th>Length (mm)</th>
<th>Length/Diameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>1</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>2</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>2</td>
<td>0.4</td>
<td>0.2</td>
</tr>
<tr>
<td>4</td>
<td>20</td>
<td>5</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>2.5</td>
</tr>
</tbody>
</table>

TABLE 3
NOZZLE ORIFICE DIMENSIONS

gauges were used to measure gas supply pressures, and rotameters were used to measure gas flow rates. The types of rotameters used are listed in Table 4 along with the usable flow range and method by which they were calibrated. Gas flow rates were controlled by varying the pressure in the gas supply line using the cylinder regulator.
In addition to the nozzles described in Table 3, several experiments were carried out using an annular nozzle in which the flow rates in the core and annulus of the jet could be varied independently. This nozzle was produced by sliding a collar into the 4 mm diameter, 20 mm long nozzle and centering a 30 mm length of stainless steel tubing of dimensions 2.7 mm inner diameter, 3.4 mm outer diameter in the orifice. The nozzle was then fitted into a 3/8 inch pipe fitting T joint, and the stainless steel tubing was soldered into a section of 1/4 inch hard copper tubing which passed into the T joint through a bored swagelock fitting. Separate gas cylinders and flow meters were then connected to the core and annulus sections of the nozzle.

**TABLE 4**

**ROTAMETERS USED FOR GAS FLOW**

<table>
<thead>
<tr>
<th>Rotameter Type (Fischer and Porter model number)</th>
<th>Flow Range (cm³/sec N₂ at STP)</th>
<th>Calibration</th>
</tr>
</thead>
<tbody>
<tr>
<td>5A-25-A</td>
<td>400 - 3000</td>
<td>Factory calibrated</td>
</tr>
<tr>
<td>3B-25</td>
<td>100 - 1000</td>
<td>Wet Test Meter * and Rotameter 5A-25-A</td>
</tr>
<tr>
<td>2B</td>
<td>10 - 200</td>
<td>Wet Test Meter and Rising Soap Film</td>
</tr>
<tr>
<td>B6A-25-A</td>
<td>5 - 100</td>
<td>Wet Test Meter and Rising Soap Film</td>
</tr>
<tr>
<td>10A3135</td>
<td>1 - 15</td>
<td>Rising Soap Film</td>
</tr>
</tbody>
</table>

*The wet test meter used in these calibrations was a Precision Scientific Co. Model 3114 with a 3 liter capacity.*
During the experiments in which nozzle chamber pressure was measured and used to detect changes in gas flow behavior, the nozzle chamber volume was one of the parameters varied. Therefore, it was necessary that the nozzle chamber be separated from the volume associated with the gas supply lines and flow meters. A needle valve was placed between the gas supply line and the nozzle chamber and a pressure drop of 7 to 15 psi was maintained across it. Since this pressure drop was more than an order of magnitude larger than the pressure fluctuations which occurred in the chamber, the flow rate of gas into the nozzle chamber was maintained approximately constant and the changes in nozzle chamber pressure could be directly related to changes in the resistance to gas flow into the liquid.

A small nozzle chamber with a volume of approximately 20 cm$^3$ was assembled from 1/4 inch copper pipe connecting the flow valve with the nozzle itself. A nozzle chamber of approximately 92 cm$^3$ volume was constructed using a section of 3/4 inch copper pipe between the flow valve and the nozzle. A large nozzle chamber, with a volume of approximately 1840 cm$^3$ was constructed by connecting an empty filter canister between the valve and the nozzle.

2. High Speed Film Equipment

In the first sequence of experiments, a series of
high speed films were taken of inert gas jets entering the water bath. A Hycam model K20S4E-115, rotating prism, 16mm, high-speed camera was used in these experiments. With this apparatus, films could be conveniently taken at framing rates of 100 to 6000 frames per second. The exact film speed was determined by use of a timing light which marked the edge of the film as it passed through the camera at 100 flashes per second.

For most experiments, Kodak 16 mm 4X reversal film Type 4xR430 with an ASA rating of 320 in tungsten light was used. This film had adequate speed and produced a positive image for viewing without requiring the additional step of printing from negative film.

Lighting was provided by four Mini-Pro, type 100-091, 650 watt quartz-halide lamps. Initially side lighting with a dark background was used, but later front lighting with a white background was found to give better delineation of the edges of the bubbles.

High speed films of submerged gas jets were produced by the following procedure:

1. The nozzle to be used was mounted in the tank, a cylinder containing the gas to be used was connected to the apparatus, and the water tank was filled with distilled water to the desired level.

2. The high speed camera was set up opposite the
glass face of the tank and the lighting and focusing adjusted.

3. With the gas flow set at a low rate, a meter stick was placed in the water tank alongside the nozzle. The camera was set to operate at 100 frames per second and a few feet of film were taken. The size of the meter stick in these frames then provided a distance scale for the high speed sequence.

4. The meter stick was removed from the tank, the camera was adjusted for high speed operation, the gas flow rate was increased to the desired level, and the film was taken. In most cases a film speed of 3000 frames per second was used, but in a few cases film speed of 5000 frames per second was used.

3. Piezoelectric Transducer

A Kistler, type 603A, piezoelectric transducer was used in these experiments. The output of the transducer was conducted to a Kistler, Model 504A, charge amplifier. When the setting of the amplifier is correctly matched with the characteristics of the transducer, the voltage output of the system is directly proportional to the pressure experienced by the transducer. This voltage was then measured on a Tektronix model 5031 dual beam storage oscilloscope and the oscilloscope trace recorded photographically.
The transducer was fixed into the system using a special adapter which was produced by plugging one side of a 1/4 inch copper pipe Tee joint, and then boring out the plug and threading it to fit the transducer. The adapter could be placed in the nozzle chamber at any location where a 1/4 inch pipe connection was available.

To record a nozzle pressure trace after the desired nozzle and flow conditions had been established, the oscilloscope was placed in the manually triggered, single sweep, storage mode. In this mode, a pressure trace could be recorded at the press of a button, then examined and recorded photographically before being erased in preparation for the next trace. A horizontal scale of 50 milliseconds per division (0.5 seconds full sweep) was found to give the best trace under most experimental conditions. This time scale gave sufficient separation of pressure pulses to make them easily distinguishable, yet sufficient resolution to determine their shapes. The vertical scale of the oscilloscope and the setting of the transducer amplifier were adjusted so as to give a good signal to noise ratio in the pressure trace. The exact settings used depended on the nozzle and gas being investigated, but ranges of 0.5 to 1 psi per division on the oscilloscope were typical.

Several experiments were carried out in which high speed films were taken and nozzle pressure traces recorded
simultaneously. The apparatus used is shown schematically in Figure 2. It includes a special electronic signal generating device, constructed by Mr. Penn Clower. This device used the gate output of the oscilloscope to trigger a flash bulb placed within the field of view of the high speed camera. The gate output of the oscilloscope is a signal of 5 volts emitted whenever the oscilloscope beam is on the screen and zero voltage whenever the beam is not on the screen. The signalling device used this signal to trigger a more powerful signal which fired the flash bulb.

For the simultaneous film and pressure trace experiments, the flow and photographic equipment were set up as in the high speed film experiments. The transducer and oscilloscope were adjusted and a fresh flash bulb was connected to the signalling device and placed where it would appear in the upper left hand corner of the movie frame. The camera was then started up, and, after allowing about 0.5 second to be sure that the film was up to speed, the oscilloscope sweep was started, triggering the flash bulb. The burning of the flash bulb takes place over an interval of about 25 milliseconds, and therefore covers about 75 frames of the film; however, it was possible to distinguish the frame in which the bulb first started to burn. This frame could then be matched up with the left hand side of the oscilloscope sweep. The timing marks on
Figure 2. Experimental apparatus for simultaneous recording of nozzle chamber pressure trace and jet behavior in gas-water systems.
the film provided a time scale to match the jet behavior against the peaks in the oscilloscope trace. The film ran for a period of 1.5 to 2 seconds, so that the entire 0.5 second pressure sweep could be observed. The flash bulb was not so bright as to completely overexpose the film, so the jet behavior near the nozzle could still be observed during the burning of the flash bulb.

4. **Sampling and Analysis of Liquid**

Several mass transfer experiments were carried out in a system in which 0.3% $\text{H}_2\text{O}_2$ was added to water and the inert gas contained 1% $\text{SO}_2$. In this range of $\text{SO}_2$ and $\text{H}_2\text{O}_2$ concentrations, the chemical reaction $\text{SO}_2 + \text{H}_2\text{O}_2 = \text{H}_2\text{SO}_4$ has been shown to be very fast so that the overall rate is controlled by the diffusion of $\text{SO}_2$ in the gas phase.$^{55}$

The effectiveness of mass transfer between the gas jet and the liquid bath was measured by drawing samples from the liquid at one minute intervals and analyzing them for sulfuric acid. Analysis of samples taken in various locations in the tank demonstrated that the liquid was well mixed and no substantial concentration gradients existed in the liquid phase.$^{56}$ Therefore, for convenience, all samples were taken from the drain hole located in the bottom of the tank.

Liquid volumes of 17.9 and 22.6 liters, corresponding
to nozzle submergence depths of 15.2 and 22.9 cm, were injected with gas jets of 1% SO$_2$ and 99% He or Ar carrier gas at a flow rate of 925 cm$^3$/sec through a 2 mm diameter orifice. This required a driving pressure of approximately 2.2 psig for the helium jet and 15 psig for the argon jet and produced a predominantly bubbling type jet in the helium case and a predominantly steady flow jet in the argon case. Sample volumes were 100 to 125 cm$^3$ and a total of 14 samples was taken in each run, so that the total volume of liquid removed during a run was small compared with the volume of the bath.

The samples were analyzed for sulfuric acid by titration with a standard sodium hydroxide solution using a methyl red indicator. The sulfuric acid concentration in the bath was plotted against time yielding a straight line. The slope of this line could be used to calculate the fraction of the injected SO$_2$ which was absorbed in the liquid using the following equation:

$$f_j = \frac{mV_bRT}{PQ}$$  \hspace{1cm} (25)

where $f_j$ is the fraction of the SO$_2$ reacted, $m$ is the slope of the concentration versus time plot, $V_b$ is the volume of the bath, $Q$ is the volumetric flow rate of SO$_2$ into the system, $R$ is the gas constant, and $P$ and $T$ are the pressure and temperature of the gas under metering conditions.
B. GAS-LIQUID TIN AND SOLDER EXPERIMENTS

1. Liquid Container and Gas Flow Apparatus

   Investigations of the behavior of inert gas jets into liquid tin and lead-tin solder baths were carried out in order to test the effectiveness of the nozzle pressure trace technique for detecting flow regime transitions in opaque systems and to determine the effect of large changes in liquid density and surface tension on the jet behavior. All the experiments were carried out in the apparatus shown schematically in Figure 3.

   The liquid metal container was a 38 cm length of steel pipe of dimensions 12.7 cm OD, 11.4 cm ID with a section of 0.95 cm thick steel plate welded to the bottom. Lengths of 1.27 cm diameter threaded steel rod were bolted into the container in holes bored 30.5 cm above the bottom of the tank. The container was supported by resting the rods in two notched wooden blocks. The rods could be clamped into the blocks to hold the system in a rigid upright position during the runs and loosened to allow the liquid metal to be poured out of the container at the conclusion of the run. A spout was welded into the top of the container to aid in pouring.

   A stainless steel nozzle of the same shape as those used in the water experiments was machined. The nozzle
Figure 3. Experimental apparatus for simultaneous recording of nozzle chamber pressure trace and electrical gas probe output in gas-solder systems.
diameter was 1 mm and the length was 10 mm. A 9/16 inch
diameter hole was bored in the center of the bottom of the
liquid container and tapped to accommodate the flow nozzle.
Since the stem of the flow nozzle was straight cut, it
could then be passed through the bottom of the liquid con-
tainer, making a tight seal, and connected to the gas supply
system in a 3/8 inch-pipe fitting. The nozzle orifice was
approximately 1.9 cm above the bottom of the liquid container
when installed. The volume of the nozzle chamber used in
these runs was 24 cm$^3$.

The system was heated by a Watlow 5641 BX band heater
of 900 watts capacity which was clamped around the outside
of the steel pipe near the bottom. Power to the band
heater was controlled by a Chromalox CH-152 120 volt
infinite control mechanism. This device is an off-on
controller which operates on a 30 second cycle using a
resistance heated bimetallic strip as a timer. The operator
controls the fraction of time in each cycle during which
current passes to the load. Due to the large thermal mass
of the system, temperature fluctuations during the 30
second cycle were negligible. The steel pipe and heater
were wrapped in several layers of 1/2 inch thick fiberglass
aircraft insulation to reduce heat losses.

In preparation for carrying out a run, the system
was preheated to 200°C, and the tin or solder to be used
was melted in stainless steel beakers over a gas flame. Pure tin and eutectic lead-tin solder (61.9% tin, 38.1% lead) were used in the experiments. The properties of these liquid metals are listed in Table 5. The quantity of metal melted in each experiment was varied to vary the head of liquid metal above the nozzle. The head of liquid metal used in these experiments was varied from 14.5 to 18.7 cm.

**TABLE 5**

**PROPERTIES OF LIQUID TIN AND LEAD-TIN ALLOY**

<table>
<thead>
<tr>
<th>Property</th>
<th>Pure Tin at 232°C</th>
<th>Lead-Tin Eutectic at 200°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density(^{57})</td>
<td>7.0 g/cm(^3)</td>
<td>8.05 g/cm(^3)</td>
</tr>
<tr>
<td>Viscosity(^{57})</td>
<td>0.0196 poise</td>
<td>0.0225 poise</td>
</tr>
<tr>
<td>Surface Tension(^{58})</td>
<td>565 dynes/cm</td>
<td>530 dynes/cm</td>
</tr>
</tbody>
</table>

The temperature of the molten metal was measured using a mercury thermometer with a range of 0 to 360°C. The temperature of the bath was held at approximately 235°C for the pure tin runs and at 200°C for the eutectic solder runs. Exact bath temperatures were measured at the end of each run and the metal was then poured back into the stainless steel beakers and cast into approximately 0.5 kg.
ingots for storage.

2. Piezoelectric Transducer and Electric Gas Probe

The piezoelectric transducer used in these experiments was the same as that used in the gas-water experiments described in section IV-A-3. The method of mounting the transducer in the nozzle chamber and recording the pressure traces was also the same. Since the transducer cannot function properly at temperatures above 300°F, cooling water was passed through 1/8" copper tubing wrapped around the transducer adapter before each run.

The electrical gas probe used in these experiments was of the type first described by Solomon\textsuperscript{59}, who used it to detect flow regime transitions in two phase gas-liquid pipe flow. It consists of a fine metal wire, in this case stainless steel, which is insulated from the liquid being investigated except at the tip. The probe wire is connected to a voltage source, in this case a dry cell, and the other terminal of the dry cell is grounded through a resistor. The voltage across the resistor is measured and recorded on an oscilloscope. Since the liquid metal container is grounded, whenever the tip of the probe is in contact with the liquid metal, current is able to flow through the electrical system and a voltage is measured on the oscilloscope. When the tip of the probe is in contact only with
gas, no current flows and no voltage is measured. Effectively, the bubbling action acts as a switch, closing the circuit whenever liquid is in contact with the probe and opening it whenever gas is in contact with the probe.

In order to obtain information on the flow regime of the jet, it was necessary to place the probe at a location where it would experience a noticeably different pattern of bubble contact in one jet flow regime than in the other. Gas had been observed to spread across the face of the nozzle only during periods of bubbling jet behavior. During periods of steady jet behavior, the nozzle face was relatively clear of gas bubbles. Therefore it was decided to place the gas probe on the top face of the nozzle near the orifice.

As shown in Figure 4, the gas probe was passed through the nozzle chamber and into the metal bath through a hole bored in the top of the nozzle. The probe wire was then bent at a 90° angle so that it was parallel to the nozzle face and brought close to the nozzle orifice. The probe was insulated from the nozzle by porcelain tubing and sealed into place using Sauereisen Type 1 high temperature cement. The stainless steel wire was 0.020" in diameter, and the distances from the probe to the nozzle were measured using an automotive spark plug gauge. For the jet cone to be in constant contact with the probe would require
Figure 4. Electrical gas probe used to investigate jet behavior in gas-solder systems.
a jet cone angle of at least 70°.

The stainless steel probe wire was soldered to a fine insulated copper wire which was sealed into the nozzle chamber by passing it through a short length of 1/8" diameter copper tubing. The copper tubing was connected to the nozzle chamber by a swagelock fitting and sealed at the opposite end with epoxy.

The probe used a 2200 ohm, 1 watt resistor and a 6 volt dry cell battery. The oscilloscope indicated a voltage of approximately 4.7 volts when the probe was immersed in liquid and zero voltage when the probe was in gas. The output of the probe was connected to trace 2 of a dual beam oscilloscope and the output of the pressure transducer was connected to trace 1. Since both oscilloscope beams were triggered by the same signal, simultaneous pressure and gas probe traces could be produced.

C. GAS-LIQUID IRON EXPERIMENTS

1. Liquid Container and Gas Flow Apparatus

A series of experiments were carried out in liquid iron to investigate the behavior of a submerged gas jet in a system of commercial interest and to measure the mass transfer rates between a submerged gas jet and a liquid metal. The apparatus used in these experiments is shown
schematically in Figure 5.

The liquid metal was contained in a Norton Type 70552 12 lb magnesium oxide crucible with a rounded bottom. It had an inside diameter of 8.9 cm, a wall thickness of 0.3 cm, and a height of 17.1 cm.

A 0.63 cm diameter hole was bored in the bottom of the crucible to accommodate the gas flow nozzle. In later experiments, a flat bottomed Norton type 70153A 12 lb alumina crucible was used. This crucible had an inside diameter of 8.9 cm, a height of 17.8 cm and a wall thickness of about 1.0 cm.

Special gas flow nozzles were machined from dense alumina to the design shown in Figure 6. Each of the nozzles was then sealed into a 6 inch length of 1/4 inch outer diameter alumina tubing with Norton type RA 1139 high temperature alumina cement. The assembled nozzle piece was then sintered into the bottom of the crucible using the same cement. The change from magnesium oxide to alumina crucibles was prompted by the failure of the alumina cement to adhere tightly to the magnesium oxide crucibles. Orifice diameters of 1.0, 0.8, and 0.56 mm were used in these experiments. In one case a copper nozzle was prepared by swaging the end of a section of copper tubing with a 1/8 inch outer diameter 0.018 inch wall thickness to an inner diameter of 0.80 mm.
Figure 5. Experimental apparatus used in gas-liquid iron experiments.
Figure 6. Alumina flow nozzle used in gas-liquid iron experiments.
A nozzle chamber was assembled using 1/4 inch pipe fittings to accommodate a Bourdon type pressure gauge, the piezoelectric transducer used in the previous experiments, and a control valve. Since the transducer has a maximum operating temperature of 300°F, it was necessary that the nozzle chamber be separated from the high temperature portion of the system. The chamber was connected through a swagelock fitting to a length of 1/8 inch outer diameter, 0.018 inch wall copper tubing. The copper tubing was then sealed into the bottom end of the alumina tubing attached to the nozzle using Sauereisen type 1 air hardening high temperature cement.

The system was heated by induction heating using a Tocco 50HO01A high frequency power supply capable of delivering 50 KVA at 10 kilohertz. When the metal was heated by direct induction, it was found that droplets of metal splashed from the bath and froze on the walls of the crucible, thereby reducing the effective volume of the bath. Therefore, a graphite susceptor was machined into a thin cylindrical shell to fit tightly around the outside of the crucible.

The lid of the system was fashioned from Babcock and Wilcox type K-30 firebrick and contained a large opening for gas exhaust and two smaller openings for metal sampling and temperature measurement. A 45 cm length of 1 1/2 inch
inner diameter alumina tubing was cemented over the gas exhaust to act as a stack and reduce the amount of metal ejected from the system during a run. The sampling hole was bored at a small angle to the vertical so that samples could be taken near the center of the bath. A graphite disc was cemented to the bottom of the lid to prevent erosion of the firebrick and plugging of the sampling hole by splashed metal.

Bath temperatures were measured using a Leeds and Northrup optical pyrometer reading through the sampling hole. The pyrometer was calibrated against a platinum-platinum 10% rhodium thermocouple before each experiment. Temperature during runs was controlled by adjusting the power input to the system in response to the temperature measurements which were made periodically throughout each run. Temperature fluctuations during the runs were approximately ± 20°C.

2. Materials

Pure Armco iron was used in these experiments. Pre-purified nitrogen and argon having the compositions listed in Table 6 were injected into the molten iron.
TABLE 6
GAS COMPOSITIONS

A. Nitrogen, Prepurified

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B. Argon

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3. **Sampling and Analysis of Liquid**

Mass transfer experiments were carried out to investigate the absorption of nitrogen in liquid iron from submerged nitrogen jets and the desorption of nitrogen from liquid iron to submerged argon jets. The apparatus was designed so that the composition of the injected gas could be changed quickly from one gas to the other. When the gas composition was changed a timer was started and the gas flow rate and pressure recorded. Metal samples and temperature readings were taken at intervals of about 2 minutes
for a 12 to 14 minute period after which a nozzle chamber
pressure trace was recorded. The gas composition could then
be changed again and the procedure repeated for the next
run.

Pin samples were taken using 6 mm vycor tubing
inserted into the melt through the sampling hole and air
cooled. The quality of the samples obtained was variable
with many samples being highly porous possibly as a result
of being taken from the bubbly part of the melt.

The chemical analysis of the metal samples was per-
formed by the analytical laboratory of the Allegheny Ludlum
Steel Corporation. All samples were analyzed for nitrogen
content. One sample from each run was analyzed for oxygen
content because dissolved oxygen has been shown to have a
strong effect on nitrogen transfer rates. One sample from
each run was also analyzed for carbon content because it
was found that there could be considerable carbon pickup in
the melt as a result of erosion from the graphite lid.
Carbon can affect the nitrogen transfer rates by altering
the activity coefficient of the nitrogen in solution.
V. EXPERIMENTAL RESULTS

A. GAS-WATER EXPERIMENTS

1. Characterization of Jet Flow Regimes

The characteristics of submerged gas jet flow regimes were determined by observation of high speed films of gas-water jets under various conditions. A total of 44 rolls of high speed film (each 100 ft.) were taken. The experimental variables in these films were the gas pressure and flow rate, gas composition, nozzle orifice diameter, nozzle length to diameter ratio, and nozzle submergence depth. The conditions for each film are listed in Table 7. All flow rates are given at standard temperature and pressure.

In spite of the large number of experimental variables, only two types of submerged gas jet behavior could be distinguished in the films: one in which separate bubbles were formed at the nozzle and shattered apart after detachment, and one in which a steady cone of gas passed out of the nozzle and broke up continuously into small bubbles. A third flow regime in which the walls of the gas cone pulsated and fluctuated was judged to be a transitional regime between the first two. Most submerged gas jets fluctuated between the three types of behavior and spent some time in each flow regime, although there were con-
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**Experimental Conditions for High Speed Films**

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ditions under which one flow regime was found to predominate.

The formation of the separate bubbles in the bubbling jet flow regime was found to be a two stage process taking place by the mechanism sketched in Figure 7. In the first stage of growth, the bubble is approximately spherical and grows with its base fixed at the nozzle orifice. In the second stage of growth, the horizontal dimension of the bubble remains approximately constant while the bubble grows by extending itself in the vertical direction. At the end of the second stage of growth, the bubble detaches from the orifice and, shortly afterwards, begins to shatter apart.

Figure 8 shows a sequence of bubbling type jet behavior in a nitrogen-water jet taken from film #38. As the sequence begins, a bubble has just begun the second stage of growth. It grows vertically until detaching from the nozzle after approximately 19 milliseconds. The nucleus of a second bubble is observed at 20.6 milliseconds. This bubble grows in approximately spherical geometry until the end of the first stage of growth occurs at about 22.9 milliseconds. It then grows in the vertical direction until its detachment after 42.9 milliseconds.

The details of the two stage growth pattern are more apparent in Figure 9, which shows the same phenomena in the helium-water system where the bubble size is much larger. A pulsation at the beginning of the sequence
Figure 7. Two stage bubble growth in bubbling jet flow regime.
recedes and a second bubble nucleates after 6.1 milliseconds. Spherical bubble growth then takes place until approximately 9.7 milliseconds. This is followed by second stage growth in the vertical direction until the bubble detaches after 42.6 milliseconds.

Figure 10 illustrates a case of pulsating type jet behavior. In this flow regime, the gas column emerging from the nozzle appears to be continuous, but its walls pulsate in approximately the same pattern as the bubbling type jets. At the beginning of the sequence a pulsation is growing at the nozzle. It reaches its maximum lateral expansion after 4.2 milliseconds and then the jet walls begin to recede. The pulsation nearly pinches off and forms a bubble after 12.7 milliseconds, but then begins to expand again. The growth is approximately spherical until 13.9 milliseconds when the pulsation has reached its maximum lateral expansion. It then grows vertically until 16.7 milliseconds and then begins to recede. The jet walls recede until 24.9 milliseconds and at 25.8 milliseconds another pulsation begins. This pulsation lasts until about 39.7 milliseconds and is followed by a smaller pulsation from 42.2 to 53.0 milliseconds.

Steady cone type jet behavior is illustrated in Figure 11. In this sequence, the jet exhibits steady cone type behavior until about 34 milliseconds into the film.
Figure 8. Bubbling type jet behavior in the nitrogen-water system. 2 mm diameter nozzle, 307 cm/sec, 2.1 psig.
Figure 8. (continued)
39.0
Time in milliseconds

42.9

Figure 8. (continued)
Figure 9. Bubbling jet behavior in the helium-water system 2 mm diameter nozzle, 3240 cm³/sec, 16 psig.
Figure 9. (continued)
Figure 9. (continued)
Figure 10. Pulsating jet behavior in the nitrogen-water system. 4 mm diameter nozzle, 2900 cm³/sec, 8 psig.
Figure 10. (continued)
Figure 10. (continued)
Figure 10. (continued)
Figure 11. Steady cone jet behavior in the nitrogen-water system. 1 mm diameter nozzle, 826 cm/sec, 80 psig.
Figure 11. (continued)
Figure 11. (continued)
It is significant that there is no contact between the gas and the flat surface of the nozzle during this period. At 34 milliseconds, a pulsation develops in the jet which lasts until approximately 43.6 milliseconds. During this period gas is ejected to the side of the jet across the nozzle surface. During the subsequent period of steady cone behavior, this gas coalesces into bubbles and rises from the nozzle surface. Cone angles observed during steady cone behavior fluctuated between 15° and 25° and were not substantially affected by any of the flow parameters investigated.

When a piezoelectric transducer is placed within the nozzle chamber during the operation of a submerged gas jet, sharp peaks are observed in the chamber pressure which are not observed when the jet is discharging into air. Several typical traces of gas jets discharging into air and water are shown in Figures 12 and 13, respectively. All of the traces shown are on channel 1 of the oscilloscope. The traces can be started at any location on the oscilloscope since only the deviation of the pressure from the average value is significant. The pressure fluctuations in the nozzle chamber are produced by the interaction between the gas jet and the liquid.

The nature of the relationship between nozzle chamber pressure fluctuations and the jet behavior was determined
Trace 1  
Trace 2  
Trace 3  
Trace 4
30 psig  20 psig  10 psig  4 psig
1645 cm³/sec  1291 cm³/sec  916 cm³/sec  590 cm³/sec

Figure 12. Nitrogen discharging to air through a 2 mm
diameter nozzle. Nozzle chamber pressure traces. 100 psi/volt
on channel 1.
Figure 12. Nitrogen discharging to air through a 2 mm diameter nozzle. Nozzle chamber pressure traces. 100 psi/volt on channel 1.
Diameter nozzle, 20 cm chamber, 100 psi/volt on channel A.

Inlet to water injection, 4 cm.

Figure 12.
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<th>Trace</th>
<th>Flow Rate (cm³/sec)</th>
<th>Pressure (psi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1760</td>
<td>4</td>
</tr>
<tr>
<td>2</td>
<td>2860</td>
<td>8</td>
</tr>
<tr>
<td>3</td>
<td>3670</td>
<td>12</td>
</tr>
<tr>
<td>4</td>
<td>4300</td>
<td>16</td>
</tr>
</tbody>
</table>

**Diagram Note:**
Diameter nozzle, 20 cm chamber, 100 psi/volt on channel 1.
Nitrogen discharge to water through a 4 mm nozzle chamber pressure traces.

*Figure 13*
by producing pressure traces which were synchronized with high speed films of the jets being studied. These are illustrated in Figures 14 and 15 for the bubbling type jet flow regime and Figures 16 and 17 for the predominantly steady cone jet flow regime. In Figure 15, detachment and nucleation of bubbles takes place at 8, 80, 107, 140, 182, 193, and 220 milliseconds from the ignition of the flash bulb. These times correspond with the 7 tall peaks in the left hand side of the nozzle chamber pressure trace in Figure 14. In Figure 17, the steady jet cone breaks down and produces bubbles or pulsations at 73, 78, 125, 180, 186, 190, 238, and 249 milliseconds after the ignition of the flash bulb. These times correspond to the 8 sharp peaks on the left hand side of the nozzle chamber pressure trace in Figure 16.

The effect of changing flow parameters on the nozzle chamber pressure traces was also investigated. The pressure traces in Figures 13, 14, and 16 were produced using N₂ gas injected through a 4 mm diameter nozzle with a 20 cm³ nozzle chamber. Figure 13 shows the effect of increasing gas flow rate on the pressure traces for a given system. As the flow rate is increased, the peaks become steeper and more closely spaced as the bubble frequency increases. At very high flow rates, the jet begins to spend substantial periods of time in the steady cone flow regime,
1.25 psig \quad 725 cm^3/sec

Figure 14. Nozzle chamber pressure trace. Nitrogen discharging to water through a 4 mm diameter nozzle. 20 cm^2 chamber, 100 psi/volt on channel 1. Synchronized with photographs of Figure 15.
Figure 14. Nozzle chamber pressure trace. Nitrogen discharging to water through a 4 mm diameter nozzle. 20 cm³ chamber, 100 psi/volt on channel 1. Synchronized with photographs of Figure 15.
Figure 15. Bubbling jet behavior in the nitrogen-water system. 4 mm diameter nozzle, 725 cm³/sec, 1.25 psig, synchronized with nozzle pressure trace of Figure 14.
Figure 15. (continued)
Figure 15. (continued)
Figure 15. (continued)
Figure 15. (continued)
Figure 15. (continued)
15 psig   4144 cm$^3$/sec

Figure 16. Nozzle chamber pressure trace. Nitrogen discharging to water through a 4 mm diameter nozzle. 20 cm$^2$ chamber, 100 psi/volt on channel 1. Synchronized with photographs of Figure 17.
Figure 16. Nozzle chamber pressure trace. Nitrogen discharging to water through a 0.4 mm diameter nozzle, 50 cm chamber, 100 psi/volt on channel 1. Synchronized with photographs of Figure 17.
Figure 17. Steady cone and pulsating type jet behavior in the nitrogen-water system. 4 mm diameter nozzle, 4144 cm³/sec, 15 psig, synchronized with nozzle chamber pressure trace of Figure 16.
Figure 17. (continued)
Figure 17. (continued)
Figure 17. (continued)
185.8  
Time in milliseconds

186.5  

189.3  

192.7

194.8  

195.8  

196.9  

199.6

Figure 17. (continued)
Figure 17. (continued)
Figure 17. (continued)
and the pressure peaks are separated by periods of undisturbed flow. Figure 18 shows the effect of changing the gas density with the same flow system. Increasing the density by injecting argon, as in Figure 18a, produces smaller, more closely spaced peaks, while reducing the density by using helium produces much larger peaks.

Reducing the nozzle diameter for a given nozzle chamber volume reduces the amplitude of the peaks in all cases as shown in Figure 19. Increasing the nozzle chamber volume to 92 cm$^3$ with the 2 mm diameter nozzle reduced the amplitude of the peaks as shown in Figure 20 so that only the helium bubbles could be detected. When the 1840 cm$^3$ nozzle chamber was used, the transducer detected periodic bursts of noise, as shown in Figure 21. These bursts of noise seem to be produced by the jet, since they could be detected more easily when the transducer was mounted near the nozzle. The patterns that they produce are similar to those produced by the pressure peaks in the small nozzle chamber volume runs, but high speed films taken simultaneously with such pressure traces did not show the bursts of noise to be synchronous with any observable jet behavior.

The nozzle chamber system can be considered analogous to an electrical RC circuit where the nozzle chamber is represented by a capacitor being charged with a constant current analogous to the constant gas flow into
Trace 1 16 psig 3600 cm$^3$/sec
Trace 2 12 psig 3200 cm$^3$/sec
Trace 3 9 psig 2450 cm$^3$/sec
Trace 4 4 psig 1470 cm$^3$/sec

Figure 18. Nozzle chamber pressure traces. a) Argon discharging to water. b) Helium discharging to water. 4 mm diameter nozzle, 20 cm$^3$ chamber, 100 psi/volt on channel 1.
<table>
<thead>
<tr>
<th>Trace</th>
<th>Pressure (psig)</th>
<th>Flow Rate (cm³/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>16</td>
<td>3600</td>
</tr>
<tr>
<td>2</td>
<td>12</td>
<td>3200</td>
</tr>
<tr>
<td>3</td>
<td>9</td>
<td>2450</td>
</tr>
<tr>
<td>4</td>
<td>4</td>
<td>1470</td>
</tr>
</tbody>
</table>

Figure 18. Nozzle chamber pressure traces. a) Argon discharging to water. b) Helium discharging to water. 4 mm diameter nozzle, 20 cm³ chamber, 100 psi/volt on channel 1.
Trace 1  1.25 psig  1956 cm$^3$/sec
Trace 2  0.9  psig  767 cm$^3$/sec

Figure 18. (continued)
Figure 19. Nozzle chamber pressure traces. Gas discharged into water through a 2 mm diameter nozzle, 20 cm chamber, 50 psi/volt on channel 1.

<table>
<thead>
<tr>
<th>Trace</th>
<th>350 cm/sec</th>
<th>3 psi</th>
<th>Trace</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Trace</td>
<td>600 cm/sec</td>
<td>6 psi</td>
<td>Trace</td>
<td>5</td>
</tr>
<tr>
<td>Trace</td>
<td>842 cm/sec</td>
<td>11 psi</td>
<td>Trace</td>
<td>2</td>
</tr>
<tr>
<td>Trace</td>
<td>1055 cm/sec</td>
<td>17 psi</td>
<td>Trace</td>
<td>1</td>
</tr>
</tbody>
</table>

Trace 3

NITROGEN

Trace 3

NITROGEN
Figure 19. (continued)

Trace 3  2.1 psi  425 cm³/sec
Trace 2  2.2 psi  252 cm³/sec
Trace 1  4.7 psi  430 cm³/sec

Helium

Argon
A 2 mm diameter nozzle. 70 cm distance, 20 ft/s valve on chamber I.

Plenum 20. Nozzle chamber pressure trace. Gas discharged into water through

<table>
<thead>
<tr>
<th>Trace</th>
<th>1 ft/sec</th>
<th>2 ft/sec</th>
<th>3 ft/sec</th>
</tr>
</thead>
<tbody>
<tr>
<td>Trace 5</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
</tr>
<tr>
<td>Trace 4</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
</tr>
<tr>
<td>Trace 3</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
</tr>
<tr>
<td>Trace 2</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
</tr>
<tr>
<td>Trace 1</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
<td>30 cm/sec</td>
</tr>
</tbody>
</table>

He:Jum

Nitrogen
Figure 20. Nozzle chamber pressure traces. Gas discharge to water through a 2 mm diameter nozzle. 92 cm chamber, 50 psi/volt on channel 1.

<table>
<thead>
<tr>
<th>Trace</th>
<th>Pressure (psi)</th>
<th>Flow Rate (cm³/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>1</td>
<td>412</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>1180</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>2082</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>2854</td>
</tr>
<tr>
<td></td>
<td>6.5</td>
<td>428</td>
</tr>
<tr>
<td></td>
<td>7</td>
<td>742</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>1008</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>1314</td>
</tr>
</tbody>
</table>

Helium

Nitrogen
A 4 mm diameter nozzle, 1840 cm chamber, 100 psig/volt on channel I.

Figure 2. Nozzle chamber pressure traces. Gas discharged through:

| Trace | 2.0 psig | 1535 cm/sec | Trace 1 2.5 psig | 2255 cm/sec | Trace 2 2.6 psig | 1735 cm/sec | Trace 3 1.5 psig | 936 cm/sec |

*Argon*

*Nitrogen*
chamber, and discharging through a resistance analogous to the resistance to flow through the orifice to a fluctuating voltage analogous to the fluctuating pressures produced by the growth of bubbles at the orifice. Since the duration of the pressure fluctuations produced by the growth of the bubbles is very short, the time constant of the nozzle chamber (defined by the volume divided by the volumetric flow rate) must also be short for the signal to be detected. This can be accomplished by reducing the nozzle chamber volume for a given system, but can be better accomplished by increasing the orifice diameter which both decreases the time constant of the chamber by increasing the volumetric flow rate at a given driving pressure and increases the duration of the signals produced since the bubble size is increased.

2. Liquid Velocities Within the Bubble Column

Velocities and mixing conditions within the bubble column region of the jet were investigated by injecting a sharp pulse of powdered graphite into the nozzle chamber just upstream of the orifice. The graphite was quickly transferred into the liquid phase and then carried up the jet, dispersing as it moves. The passage of the pulse along the jet was filmed. When the film was projected, a light meter which reads in a 1° solid angle circle was
Figure 22. Graphite injection in the gas-water system. Light intensity versus time at different heights in the jet.
directed at several points along the jet to measure the graphite concentration at these points. The resulting darkness vs. time curves are analogous to tracer concentration vs. time curves used in the analysis of chemical reactor systems. Several examples of these traces are shown in Figure 19. The time for the graphite pulse to reach various heights above the nozzle was measured and the velocities calculated are shown in Table 8. Unfortunately, because of variations in illumination along the length of the jet and interference due to the passage of bubbles across the measuring points, it was not possible to determine the dispersion coefficient in the jet from these experiments.

In film 17, the graphite entered the jet during a period of steady cone behavior and appeared simultaneously at heights of 0 to about 8 cm above the orifice. This may indicate the approximate height of the continuous gas region in this case.

3. **Bubble Size in Bubbling Jet Flow**

   The volume of the bubbles produced during the periods of bubbling type jet flow was measured by measuring the time interval from the initiation to the detachment of the bubble and multiplying this by the volumetric flow rate of the jet. The results of these measurements are shown
TABLE 8  
LIQUID VELOCITIES IN THE BUBBLE COLUMN

<table>
<thead>
<tr>
<th>Film #</th>
<th>Gas</th>
<th>Nozzle Diameter (cm)</th>
<th>Flow Rate (cm³/sec)</th>
<th>Height of Measurement (cm)</th>
<th>Velocity (cm/sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>16</td>
<td>Air</td>
<td>0.2</td>
<td>1127</td>
<td>3.38 to 8.86</td>
<td>914.0</td>
</tr>
<tr>
<td>16</td>
<td>Air</td>
<td>0.2</td>
<td>1127</td>
<td>8.86 to 14.07</td>
<td>186.0</td>
</tr>
<tr>
<td>16</td>
<td>Air</td>
<td>0.2</td>
<td>1127</td>
<td>3.38 to 14.07</td>
<td>314.4</td>
</tr>
<tr>
<td>17</td>
<td>Air</td>
<td>0.2</td>
<td>1127</td>
<td>7.82 to 15.62</td>
<td>173.3</td>
</tr>
<tr>
<td>22</td>
<td>N₂</td>
<td>0.2</td>
<td>2004</td>
<td>4.32 to 9.40</td>
<td>847.0</td>
</tr>
<tr>
<td>22</td>
<td>N₂</td>
<td>0.2</td>
<td>2004</td>
<td>9.40 to 14.73</td>
<td>666.0</td>
</tr>
<tr>
<td>22</td>
<td>N₂</td>
<td>0.2</td>
<td>2004</td>
<td>4.32 to 14.73</td>
<td>694.0</td>
</tr>
<tr>
<td>23</td>
<td>He</td>
<td>0.2</td>
<td>3240</td>
<td>2.41 to 7.37</td>
<td>238.0</td>
</tr>
<tr>
<td>23</td>
<td>He</td>
<td>0.2</td>
<td>3240</td>
<td>7.37 to 13.20</td>
<td>216.0</td>
</tr>
</tbody>
</table>

in Table 9. The bubble volumes were found to increase strongly with increasing nozzle diameter and decreasing gas density, but to increase only slightly with increasing flow rate for a given nozzle diameter and gas density.

Average volumes of 10 to 20 bubbles are given in Table 9 because there is a great deal of scatter in the volumes of bubbles produced in the bubbling jet systems. Much of the scatter seemed to result from variations in the amount of vertical growth taking place during stage 2
<table>
<thead>
<tr>
<th>Gas</th>
<th>Nozzle Diameter (mm)</th>
<th>Pressure (psig)</th>
<th>Flow Rate (cm³/sec)</th>
<th>Bubble Volume (cm³)</th>
<th>Standard Deviation (cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar</td>
<td>1</td>
<td>12.5</td>
<td>179</td>
<td>1.26</td>
<td>0.16</td>
</tr>
<tr>
<td>Ar</td>
<td>1</td>
<td>40</td>
<td>380</td>
<td>2.02</td>
<td>0.24</td>
</tr>
<tr>
<td>N₂</td>
<td>1</td>
<td>16</td>
<td>251</td>
<td>1.76</td>
<td>0.26</td>
</tr>
<tr>
<td>N₂</td>
<td>1</td>
<td>30</td>
<td>362</td>
<td>1.76</td>
<td>0.37</td>
</tr>
<tr>
<td>N₂</td>
<td>1</td>
<td>80</td>
<td>826</td>
<td>2.62</td>
<td>0.76</td>
</tr>
<tr>
<td>He</td>
<td>1</td>
<td>60</td>
<td>1825</td>
<td>23.2</td>
<td>6.18</td>
</tr>
<tr>
<td>N₂</td>
<td>2</td>
<td>2</td>
<td>453</td>
<td>11.2</td>
<td>3.37</td>
</tr>
<tr>
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<td>2</td>
<td>453</td>
<td>10.41</td>
<td>4.75</td>
</tr>
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<td>11.14</td>
<td>3.37</td>
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<td>777</td>
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<td>3.00</td>
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</tr>
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<td>2004</td>
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<tr>
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</tr>
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<td>Pressure (psig)</td>
<td>Flow Rate (cm$^3$/sec)</td>
<td>Bubble Volume (cm$^3$)</td>
<td>Standard Deviation (cm$^3$)</td>
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<td>Nozzle Diameter (mm)</td>
<td>Pressure (psig)</td>
<td>Flow Rate $^3$/sec)</td>
<td>Bubble Diameter (cm)</td>
<td>Standard Deviation (cm)</td>
</tr>
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<td>---------------------</td>
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<td>---------------------</td>
<td>----------------------</td>
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<td>0.143</td>
</tr>
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<td>0.115</td>
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<td>2.98</td>
<td>0.256</td>
</tr>
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<td>4586</td>
<td>3.72</td>
<td>0.309</td>
</tr>
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<td>1.25</td>
<td>724</td>
<td>2.76</td>
<td>0.148</td>
</tr>
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<td>Gas</td>
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<td>Pressure (psig)</td>
<td>Flow Rate (cm³/sec)</td>
<td>Bubble Diameter (cm)</td>
<td>Standard Deviation (cm)</td>
</tr>
<tr>
<td>-----</td>
<td>---------------------</td>
<td>----------------</td>
<td>---------------------</td>
<td>----------------------</td>
<td>------------------------</td>
</tr>
<tr>
<td>N₂</td>
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<td>2</td>
<td>1196</td>
<td>2.50</td>
<td>0.159</td>
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<td>8</td>
<td>2900</td>
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Double Nozzle Runs

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<th>Flow Rate (cm³/sec)</th>
<th>Bubble Diameter (cm)</th>
<th>Standard Deviation (cm)</th>
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<td>Flow Rate (cm$^3$/sec)</td>
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</table>
of the growth of the bubble. The diameter of the spherical bubble at the end of stage 1 was found to be more nearly constant for a given system. The bubble diameter at the end of the first stage of growth was also approximately equal to the horizontal dimension of the bubble during the second stage of growth and to the maximum lateral expansion of the jet during pulsating jet flow. This dimension was measured on a number of films and the average diameters are given in Table 10.

The ratio of the final bubble volume to the volume of the bubble at the end of the first stage of growth is equal to the ratio of the time that the bubble spends in the second stage of growth to the time spent in the first stage. It is also proportional to the axial ratio of the final bubble. These ratios are listed in Table 11. Although there is considerable scatter from one run to another there does not seem to be any systematic change in the ratio with changes in any of the flow parameters. The average ratio of the final bubble volume to the first stage bubble volume is 5.445.

4. Transition Between Bubbling and Steady Jet Flow

As the flow rate of a given gas into water through a given nozzle is increased, there is a gradual transition in flow behavior from the bubbling type jet to the steady
flow type jet. At relatively low flow rates, well formed bubbles are produced sequentially at the orifice and then break up after detachment. At higher flow rates, the detachment of a large bubble may be followed by a short period of steady flow behavior before the next bubble forms. At higher flow rates, bubbles sometimes coalesce or fail to detach completely, giving pulsating jet type flow. At even higher flow rates, when extended periods of steady flow type behavior are observed, there are still substantial periods of bubbling and pulsating flow behavior interspersed with the steady flow behavior. At the highest flow rates investigated in this study, the steady jets are occasionally interrupted by the formation of bubbles or pulsations at such high rates that they are nearly explosive.

This sort of flow regime transition is similar to the transition between flow regimes in two-phase gas liquid pipe flow. The flow regimes change gradually over a wide range of transitional fluctuating behavior.

The conditions for the flow regime transition can only be determined qualitatively. Different observers might select different points within the transition range as the flow regime boundary. Here the flow regime transition is considered to take place at the point where discrete bubble formation, characterized by breaks in the
continuity of the jet near the nozzle, become quite infrequent, with less than about 5% of the movie frames showing this behavior. At the same point, the steady flow cone becomes more prevalent, with more than about 30% of the movie frames showing this behavior. The nozzle chamber pressure traces produced by such jets contain occasional sharp peaks separated by periods of steady nozzle chamber pressure with the nozzle chamber pressure being steady over more than about 75% of the trace.

Observation of the 43 high speed films of gas-water jets showed that predominantly steady flow type behavior took place in every case where the momentum flux or mass flow per unit area through the orifice was greater than 40 g/cm² sec. This momentum flux was produced by a driving pressure of about 16 psig for nitrogen, 12.5 psig for argon, and 60 psig for helium. There seemed to be no noticeable effect of changing nozzle diameter or liquid submergence depth on the jet flow regime. The flow regime transition takes place at approximately the same orifice Reynold's number for a given nozzle diameter regardless of the gas injected, but will take place at a substantially lower Reynold's number if the orifice diameter is increased. The flow regime transition takes place at approximately the same modified Froude number for a given gas, regardless of the nozzle orifice diameter, but will take place at a
substantially lower modified Froude number if the density of the gas injected is increased. Neither of these dimensionless numbers can be used to predict which jet flow regime will be stable under all conditions.

5. Mass Transfer in Bubbling and Steady Jet Flow

Sulfuric acid concentration versus time curves resulting from mass transfer experiments where carrier gases containing 1% SO₂ were injected into a 0.3% H₂O₂ solution are shown in figures 20A through D. In each case, the total volumetric flow rate was held constant; the helium carrier gas produced a predominantly bubbling type jet and the argon carrier gas produced a predominantly steady flow type jet. The fraction of injected SO₂ reacted in each case is shown in Table 12.

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<tr>
<th>Carrier Gas</th>
<th>Submersion Depth</th>
<th>Fraction of SO₂ Reacted</th>
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<tr>
<td>He</td>
<td>15.2 cm</td>
<td>0.880 ± 0.05</td>
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<tr>
<td>He</td>
<td>22.6 cm</td>
<td>0.924 ± 0.05</td>
</tr>
<tr>
<td>Ar</td>
<td>15.2 cm</td>
<td>1.024 ± 0.05</td>
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<tr>
<td>Ar</td>
<td>22.6 cm</td>
<td>1.012 ± 0.05</td>
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</table>
Figure 23. $\text{H}_2\text{SO}_4$ concentration versus time for gas-aqueous systems.  a) Argon carrier gas, 15.2 cm submersion.  b) Argon carrier gas, 22.6 cm submersion.  c) Helium carrier gas, 15.2 cm submersion.  d) Helium carrier gas, 22.6 cm submersion.
B. GAS-LIQUID TIN AND GAS-LIQUID SOLDER EXPERIMENTS

1. Characterization of Jet Flow Regimes and Flow Regime Transition

Since no photographs could be taken in the liquid metal systems, characterization of the jet flow regimes had to be accomplished on the basis of nozzle chamber pressure traces and electrical gas probe results. Figure 24 shows the effect of increasing the nitrogen flow rate on the pressure traces produced when the gas is injected into pure liquid tin. There is a strong qualitative similarity between these pressure traces and those observed in the gas-water system at similar driving pressures. The magnitude of the pressure fluctuations is much greater, however, since the fluctuations produced by a 1 mm nozzle in the water system were too small to detect. Reducing the gas density by injecting helium instead of nitrogen produces much larger pressure fluctuations at the same driving pressure, as shown in Figure 25.

When nitrogen was injected into lead-tin solder at 200°C instead of pure tin there were no dramatic changes in the pressure trace, as indicated by Figure 26. As in the case of pure tin, injecting helium instead of nitrogen produces much larger pressure fluctuations, as shown in
Figure 27. No significant effect of changing the head of liquid metal above the nozzle was detected.

The results of the experiments where the electrical probe was used along with the pressure transducer were consistent with those where the pressure transducer was used alone. Figure 28 shows several such traces in the nitrogen-solder system. The upper sweep shows the nozzle pressure trace on channel 1 at 100 psi/volt and the lower sweep shows the output of the electrical gas probe on channel 2. The higher voltage indicates that the probe tip is in contact with liquid and the lower voltage that it is in contact only with gas. In the case of bubbling jet flow, as in Figure 28a, the probe tip is contacted by liquid only for very short periods at the beginning of each bubble. In the case of the higher flow rate jets, there are longer periods in which the probe is in contact with liquid. These correspond with the periods of smooth pressure in the jet nozzle chamber and are presumed to indicate steady flow jet behavior. Notice that a completely smooth pressure trace is not observed until the driving pressure is increased to 70 psig, giving a momentum flux of 108 g/cm$^2$ sec. Similar behavior is shown in Figure 29 for the argon-solder system.

2. **Bubble Size in Bubbling Jet Flow**

   The sizes of bubbles produced at the nozzle in the
through a 1 mm diameter nozzle. 2" cm chamber. 100 psi/volt on channel. I.

Figure 24. Nozzle chamber pressure traces. Nitrogen discharged to liquid tin.
Through a 1 mm diameter nozzle, 24 cm chamber, 100 psig. Note the channel 1.

Figure 24: Pressure chamber pressure traces. Hyperion characteristics to light.
through a 1 mm diameter nozzle, 24 cm chamber, 100 psi/volt on channel 1.

Figure 25: Nozzle chamber pressure traces. Helium discharge to 1400 psi.

Trace 4: 15 psi, 6.49 cm³/sec
Trace 4: 12 psi, 5.88 cm³/sec
Trace 2: 8 psi, 3.64 cm³/sec
Trace 1: 6 psi, 2.81 cm³/sec
Figure 26. Nozzle chamber pressure traces. Nitrogen discharged to junk.

Trace 2 6 cm/sec
Trace 3 8 cm/sec
Trace 4 15 psig
Trace 5 18 psig
Trace 6 22 psig
Trace 7 254 cm/sec
Trace 8 286 cm/sec

147
Figure 26. Nozzle chamber pressure traces. Hydrogen discharged to rig, 1.2 g/cm² sec.
Channel 1.

Solvent thrown in 1 mm diameter nozzle. 24 cm chamber, 100 psi on.

Figure 27. Nozzle chamber pressure traces. Helium discharged to 14.9 psig.

Trace 1 & Trace 2.

Trace 1: 16.7 cm Head
Trace 2: 14.5 cm Head

570 cm/sec, 525 cm/sec, 700 cm/sec, 875 cm/sec
Channel 1

Solder through a 1 mm diameter nozzle. 24 cm chamber, 100 psi/10 sec.

Pressure 27. Nozzle chamber pressure traces. Helium discharge to vacuum.

Trace 1: 50 cm/sec
Trace 2: 75 cm/sec
Trace 3: 125 cm/sec
Trace 4: 150 cm/sec

16.5 cm Head
14.5 cm Head
Figure 28. Nozzle chamber pressure traces and gas probe output. Nitrogen discharging to liquid nitrogen through a 1 mm diameter nozzle. 24 cm channel 1, gas probe output channel 2.
Transmission output at 1.0 sec./volt on channel I.  Current cu.-mm. in channel 2.
Figure 28. (continued)
This probe output on channel 2.

24 cm chamber. Transducer output on channel 1 at 100 V/ft.

Figure 29. Nozzle chamber pressure traces and gas probe output.

b) 32 psig  310 cm³/sec

a) 6.5 psig  88 cm³/sec
Figure 29. Nozzle chamber pressure traces and gas probe output.

2. Gas probe output on channel 2.
4 cm chamber. Transducer output on channel 1 at 100 psi/volt.
Argon discharge to liquid sulfur through 1 mm diameter nozzle.

(a) 3.5 psi 32 cm³/sec
(b) 8.2 psi 310 cm³/sec
### TABLE 13

**BUBBLE SIZES IN GAS-LIQUID TIN EXPERIMENTS**

<table>
<thead>
<tr>
<th>Gas</th>
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<th>Volume at Bath Temperature (cm³)</th>
<th>Diameter at Bath Temperature (cm)</th>
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TABLE 14

BUBBLE SIZES IN GAS-LIQUID SOLDER EXPERIMENTS

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<th>Diameter at Bath Temperature (cm)</th>
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Table 14 (cont'd.)

N\textsubscript{2}\textsuperscript{*} & 121.3 & 8.13 & 2.49 \\

*18.7 cm head
bubbling jet flow regime can be estimated by measuring the frequency of the pressure fluctuations in the nozzle chamber pressure traces and dividing this into the volumetric flow rate of the jet. If it is assumed that the bubbles are approximately spherical and that the gas quickly comes to the bath temperature after being injected into the melt, then bubble diameters can be calculated. These bubble diameters are given in Table 13 for the gas-liquid tin system, and in Table 14 for the gas-liquid solder system. Jets with momentum flux greater than 40 g/cm²/sec cannot be analyzed in this manner, since they are partially in the steady cone flow regime and their bubble frequencies may not be entirely dependent on the bubble size. Bubble sizes in jets with momentum flux greater than 30 g/cm²/sec may be slightly overestimated by this technique.

C. GAS-LIQUID IRON EXPERIMENTS

1. Characterization of Jet Flow Regimes and Flow Regime Transition

In liquid iron, neither photography nor electrical gas probe techniques could be applied to investigate the behavior of the submerged gas jets. Therefore, characterization of the jet behavior had to be accomplished on the basis of nozzle chamber pressure traces only. Figure 30
shows the effect of increasing the argon flow rate on the pressure traces when the gas is injected into iron through a 0.8 mm diameter nozzle. The pressure peaks are very small and indistinct because of the small size of the nozzle orifice and the effect of the length of copper tubing used to insulate the piezoelectric transducer from the high temperature region of the system. Figure 31 shows similar traces produced using a 1.0 mm diameter nozzle. Figure 32 shows the pressure traces produced when nitrogen was injected through a 0.8 mm diameter nozzle.

The effect of changing orifice diameter on the nozzle pressure traces is shown by Figures 30 and 31. The pressure fluctuations produced by the 0.56 mm diameter nozzle were nearly too small to detect in most cases. Because of the small amplitude of the pressure peaks, it is difficult to make firm statements about the changes in the jet behavior with increasing jet flow rates. Regularly spaced peaks continue to be present at a flow rate of 100 cm$^3$/sec for argon flow through the 0.8 mm diameter orifice and seem to be present at 152 cm$^3$/sec. These flow rates correspond to 31.7 and 48.6 g/cm$^2$ respectively and may indicate that the transition from bubbling to steady cone flow takes place at a higher flow rate in liquid iron than in the other systems investigated.
Figure 30. Nozzle chamber pressure traces. 3 Argon discharge to 149 psig

Channel 1.

Iron through 0.8 mm diameter nozzle. 10 cm chamber. 100 psig/volt on

18 psig  40 cm³/sec

93 cm³/sec  6 psig

(a)
Channel 1. Iron through 0.8 mm chamber nozzle, 10 cm chamber. 100 psi/volt on
Figure 30. Nozzle chamber pressure traces. Argon discharging to liquid.

93 cm³/sec
18 psi

40 cm³/sec
6 psi

158 INTENTIONAL DUPLICATE EXPOSURE
Figure 30. (continued)

30 psig

(c) 152 cm³/sec

40 psig

(d) 202 cm³/sec
Figure 30. (continued)

30 psig

c) 152 cm$^3$/sec

40 psig

d) 202 cm$^3$/sec
Figure 31. Nozzle chamber pressure traces. Argon discharge to 749.1 cm, 1000 psi, 10 cm. Chamber, 100 psi/volt on channel 1. Iron through 1 mm diameter nozzle, 10 cm chamber. 25 psi, 100 cm$^3$/sec. 5 psi, 15.8 cm$^3$/sec.
Channel 1. Ion through 1 mm diameter nozzle. 100 psf/volt on 10 cm chamber. 100 psf/volt on 1 mm chamber. 25 psig 1.58 cm/sec 5 psig 100 cm/sec

Figure 31. Nozzle chamber pressure traces. Aragon discharging to liquid.
Figure 32. Nozzle chamber pressure traces. Nitrogen discharging to liquid iron through 0.8 mm diameter nozzle. 12 psi/volt on channel 1. 76 cm³/sec.

9 psi/volt on channel 2. 65 cm³/sec.
Figure 32.
Nozzle chamber pressure traces. Nitrogen discharging to liquid iron through 0.8 mm diameter nozzle. 10 cm chamber.

12 psi
9 psi
76 cm³/sec
68 cm³/sec

a) 100
b) 100
2. **Bubble Sizes in Bubbling Jet Flow**

The frequencies of the pressure fluctuations in the nozzle chamber pressure traces could be used to estimate the size of the bubbles produced at the orifice. The same limitations on the validity of the results apply as in the gas-liquid tin and solder experiments. Table 15 shows the bubble frequencies observed in the gas-liquid iron systems along with the fluid flow conditions for each run. Table 16 shows bubble volumes calculated for the runs where a 0.8 mm diameter nozzle was used. The flow rates shown are at standard temperature and pressure and the volumes are calculated with the assumption that the gas reaches the metal bath temperature very quickly. The bubble diameters listed are calculated assuming spherical bubbles. The pressure pulsations produced by bubble formation at the 0.56 mm diameter nozzle were too small to be used to determine accurate bubble frequencies.

3. **Nitrogen Absorption and Desorption Rates**

   (a) **Nitrogen Absorption Rates at High Oxygen Concentration**

In two experiments, Runs 7 and 9, pure nitrogen was injected into iron that contained a high concentration of oxygen and the nitrogen absorption rate measured. The
<table>
<thead>
<tr>
<th>Gas Diameter (mm)</th>
<th>Pressure (psig)</th>
<th>Flow Rate (cm³/sec)</th>
<th>Frequency (sec⁻¹)</th>
<th>Iron Head Temperature (°C)</th>
<th>Liquid Iron Temperature (°C)</th>
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<tbody>
<tr>
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</tbody>
</table>

Bubble Frequencies in Liquid Iron

Table 15
### TABLE 16

**BUBBLE SIZES PRODUCED BY 0.8 MM NOZZLES IN LIQUID IRON**

<table>
<thead>
<tr>
<th>Gas</th>
<th>Temperature (°C)</th>
<th>Flow Rate (cm³/sec)</th>
<th>Volume (cm³)</th>
<th>Diameter (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar</td>
<td>1670</td>
<td>15.8</td>
<td>5.59</td>
<td>2.20</td>
</tr>
<tr>
<td>Ar</td>
<td>1550</td>
<td>38.8</td>
<td>10.0</td>
<td>2.67</td>
</tr>
<tr>
<td>Ar</td>
<td>1670</td>
<td>69.7</td>
<td>18.53</td>
<td>3.28</td>
</tr>
<tr>
<td>Ar</td>
<td>1570</td>
<td>100.5</td>
<td>28.64</td>
<td>3.79</td>
</tr>
<tr>
<td>Ar</td>
<td>1550 (Cu Nozzle)</td>
<td>45.4</td>
<td>13.92</td>
<td>2.98</td>
</tr>
<tr>
<td>Ar</td>
<td>1550</td>
<td>22.7</td>
<td>6.33</td>
<td>2.29</td>
</tr>
<tr>
<td>Ar</td>
<td>1550</td>
<td>29.7</td>
<td>12.92</td>
<td>2.91</td>
</tr>
<tr>
<td>N₂</td>
<td>1600</td>
<td>76.0</td>
<td>25.8</td>
<td>3.67</td>
</tr>
<tr>
<td>Ar</td>
<td>1575</td>
<td>93.1</td>
<td>35.7</td>
<td>4.08</td>
</tr>
<tr>
<td>N₂</td>
<td>1550</td>
<td>68.2</td>
<td>26.1</td>
<td>3.68</td>
</tr>
<tr>
<td>Ar</td>
<td>1550</td>
<td>152.7</td>
<td>52.02</td>
<td>4.63</td>
</tr>
<tr>
<td>Ar</td>
<td>1550</td>
<td>40.6</td>
<td>11.30</td>
<td>2.78</td>
</tr>
</tbody>
</table>
experimental conditions for these experiments are shown in Table 17 and the results are plotted in Figures 33 and 34.

Two equations have been proposed to predict the nitrogen absorption rate constants into iron at 1600°C containing more than 0.04% oxygen. That of Inouye and Choh: 50

\[
\frac{k = 1.325 \times 10^{-3} P_{N_2}^{1/2}}{(\%O_2)^{2/3}} \text{ cm/sec} \quad (26)
\]

and that of Swisher and Turkdogan: 45

\[
k = \frac{4.5 \times 10^{-4}}{(\%O)} \text{ cm/sec} \quad (27)
\]

The activation energy for nitrogen absorption at 0.14% O has been measured by Shimmyo and Takami 49 as 96000 cal/mole. Then for Run 7 where %O = 0.141 and T = 1555°C, according to equation (26)

\[
k_{1600} = 4.89 \times 10^{-3} \text{ cm/sec}
\]
\[
k_{1555} = 2.59 \times 10^{-3} \text{ cm/sec}
\]

For the same conditions equation (27) yields
### TABLE 17

**NITROGEN ABSORPTION AT HIGH OXYGEN CONCENTRATIONS**

<table>
<thead>
<tr>
<th>Run 7</th>
<th>Nozzle Area</th>
<th>5.19 x 10^{-3} \text{ cm}^2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Iron Head</td>
<td>6.73 \text{ cm} = 0.69 \text{ psi}</td>
</tr>
<tr>
<td></td>
<td>Nozzle Pressure</td>
<td>11 \text{ psig}</td>
</tr>
<tr>
<td></td>
<td>Flow Rate</td>
<td>38 \text{ cm}^3/\text{sec STP}</td>
</tr>
<tr>
<td></td>
<td>Bath Temperature</td>
<td>1555\text{°C}</td>
</tr>
<tr>
<td></td>
<td>Bath Composition</td>
<td>0.141%O, 0.011%C</td>
</tr>
</tbody>
</table>

**Measured Reaction Rate**

\[
\frac{kA}{V} = 4.45 \times 10^{-3} \text{ min}^{-1}
\]

**Predicted Areas:**

- by Inouye and Choh: \( A = 11.98 \text{ cm}^2 \)
- by Swisher and Turkdogan: \( A = 18.37 \text{ cm}^2 \)

<table>
<thead>
<tr>
<th>Run 9</th>
<th>Nozzle Area</th>
<th>5.19 x 10^{-3} \text{ cm}^2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Iron Head</td>
<td>6.73 \text{ cm} = 0.69 \text{ psi}</td>
</tr>
<tr>
<td></td>
<td>Nozzle Pressure</td>
<td>14 \text{ psig}</td>
</tr>
<tr>
<td></td>
<td>Flow Rate</td>
<td>52 \text{ cm}^3/\text{sec STP}</td>
</tr>
<tr>
<td></td>
<td>Bath Temperature</td>
<td>1575\text{°C}</td>
</tr>
<tr>
<td></td>
<td>Bath Composition</td>
<td>0.179%O, 0.014%C</td>
</tr>
</tbody>
</table>

**Measured Reaction Rate**

\[
\frac{kA}{V} = 9.25 \times 10^{-3} \text{ min}^{-1}
\]

**Predicted Areas:**

- by Inouye and Choh: \( A = 21.77 \text{ cm}^2 \)
- by Swisher and Turkdogan: \( A = 36.40 \text{ cm}^2 \)
\[ k_{1600} = 3.19 \times 10^{-3} \text{ cm/sec} \]
\[ k_{1555} = 1.69 \times 10^{-3} \text{ cm/sec} \]

For Run 9 where \( \%O = 0.179 \) and \( T = 1575^\circ C \) by equation (26)

\[ k_{1600} = 4.172 \times 10^{-3} \]
\[ k_{1575} = 2.94 \times 10^{-3} \]

and by equation (27)

\[ k_{1600} = 2.51 \times 10^{-3} \]
\[ k_{1575} = 1.76 \times 10^{-3} \]

The areas calculated using both correlations are unreasonably low considering the flow rates of nitrogen involved. At the end of the experiment in which these rates were measured, it was discovered that the alumina tube carrying gas to the flow nozzle had cracked. It was not clear whether the tube had cracked during the experiment and allowed most of the metered nitrogen to flow outside the crucible with only a small portion of it actually passing through the melt. It should also be noted that most of the experimental data on nitrogen in high oxygen melts has been taken in the range of 0.04\% to 0.10\% \( _O \) so
Figure 33. Nitrogen concentration versus time for nitrogen injection of iron baths containing high oxygen concentrations.
Figure 34. Reaction rate for nitrogen jets injected into iron baths containing high oxygen concentrations.
that the correlations may not be valid at oxygen concentrations as high as those in these experiments.

(b) **Nitrogen Absorption at Low Oxygen Concentration**

The results of nitrogen absorption runs 10 and 12, which were carried out at low oxygen concentration are shown in Figures 35 and 36. The experimental conditions are given in Table 18. The absorption curves in these experiments are distorted by the fact that the carbon content of the bath was continuously increasing during each run due to the erosion of the graphite lid. The carbon reduces the solubility of nitrogen in the metal so that the nitrogen concentration increases at the beginning of each run, then reaches a maximum and begins to decrease as the solubility drops. The nitrogen absorption rates are calculated from the samples taken during the first 5 to 6 minutes of each run where the driving force for mass transfer is large compared with the effect of the carbon increase on the solubility. The carbon concentration is taken as that at the beginning of each run.

The areas calculated from Inouye and Choh's\textsuperscript{50} data are not inconsistent with our perception of the fluid dynamic situation. They are slightly lower than the areas calculated from the nitrogen desorption experiments. The procedure by which the rates were calculated may not com-
### TABLE 18

**NITROGEN ABSORPTION AT LOW OXYGEN CONCENTRATION**

<table>
<thead>
<tr>
<th>Run 10</th>
<th>Nozzle Area</th>
<th>$5.19 \times 10^{-3} , \text{cm}^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Iron Head</td>
<td>$6.79 , \text{cm} \quad = \quad 0.69 , \text{psi}$</td>
</tr>
<tr>
<td></td>
<td>Nozzle Pressure</td>
<td>$12 , \text{psig}$</td>
</tr>
<tr>
<td></td>
<td>Flow Rate</td>
<td>$76 , \text{cm}^3/\text{sec STP}$</td>
</tr>
<tr>
<td></td>
<td>Bath Temperature</td>
<td>$1600 , \degree \text{C}$</td>
</tr>
<tr>
<td></td>
<td>Bath Composition</td>
<td>$0.0076 , % \text{O}, \text{ initially } 0.007% \text{C}, \quad 1.07% \text{C} \text{ at completion of run}$</td>
</tr>
</tbody>
</table>

**Measured Reaction Rate**

$$\frac{kA}{V} = 3.84 \times 10^{-1} \, \text{min}^{-1}$$

**Predicted Area:** by Inouye and Choh's data $k=27 \times 10^{-3} \, \text{cm}/\text{sec}$

$$A = 100 \, \text{cm}^2$$

<table>
<thead>
<tr>
<th>Run 12</th>
<th>Nozzle Area</th>
<th>$5.19 \times 10^{-3} , \text{cm}^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Iron Head</td>
<td>$6.79 , \text{cm} \quad = \quad 0.69 , \text{psi}$</td>
</tr>
<tr>
<td></td>
<td>Nozzle Pressure</td>
<td>$9 , \text{psig}$</td>
</tr>
<tr>
<td></td>
<td>Flow Rate</td>
<td>$68 , \text{cm}^3/\text{sec STP}$</td>
</tr>
<tr>
<td></td>
<td>Bath Temperature</td>
<td>$1560 , \degree \text{C}$</td>
</tr>
<tr>
<td></td>
<td>Bath Composition</td>
<td>$0.0083% \text{O}, 2.9 , % \text{C} \text{ initially, } 3.4 , % \text{C} \text{ at end of run}$</td>
</tr>
</tbody>
</table>

**Measured Reaction Rate**

$$\frac{kA}{V} = 0.208 \, \text{min}^{-1} \text{ using } e^C_N = 0.13563$$

**Predicted Area using Inouye and Choh's data $k_{1600} = 27 \times 10^{-3} \, \text{cm}/\text{sec}$ and $Q = 31000 \, \text{cal/mole}$**

$$A = 64.9 \, \text{cm}^2$$
Figure 35. Nitrogen concentration versus time for nitrogen injection of iron baths containing low oxygen concentrations.
contaminating low oxygen concentrations.

Figure 36. Reaction rate for nitrogen jets injected into iron baths.

\[
\left( \frac{N_{\%} - S_{\%}}{N_{\%} - S_{\%}} \right)_{\text{in}}
\]
pletely take account of the effects of the changing carbon concentration which would cause an underestimation of the reaction area.

(c) **Nitrogen Desorption Rates at Low Oxygen Concentration**

The experimental conditions for the nitrogen desorption experiments, Runs 1 and 11, which were carried out at low oxygen concentrations, are shown in Table 19. The results are plotted in Figures 37, 38, and 39. In both cases, there is little or no curvature when the results are plotted according to a second order rate law. This implies that even if a mixed control model is applicable to this reaction, the second order surface reaction is the principal resistance to mass transfer in this case.

The results of Narita, Koyama, Makino, and Okamura are used to calculate the effective area of the jet, because in their experiments, cylinder gases were used directly without additional purification. Cylinder gases were also used without additional purification in this study. It is suspected that the trace quantities of oxygen in these gases may produce lower reaction rates than those measured in experiments using purified gases. The results of these calculations are consistent with our expectations concerning the fluid mechanics of the system.
TABLE 19
NITROGEN DESORPTION AT LOW OXYGEN CONCENTRATION

<table>
<thead>
<tr>
<th>Run</th>
<th>Nozzle Area</th>
<th>5.19 x 10^{-3} \text{ cm}^2 (swaged copper)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Iron Head</td>
<td>7.42 cm = 0.75 psi</td>
</tr>
<tr>
<td></td>
<td>Nozzle Pressure</td>
<td>15 psig</td>
</tr>
<tr>
<td></td>
<td>Argon Flow Rate</td>
<td>45 cm$^3$/sec STP</td>
</tr>
<tr>
<td></td>
<td>Bath Temperature</td>
<td>1540°C</td>
</tr>
<tr>
<td></td>
<td>Bath Composition</td>
<td>0.0076 %O</td>
</tr>
</tbody>
</table>

Measured Reaction Rate \[ \frac{kA}{V} = 5.505 \text{ (min\%)}^{-1} \]

Predicted Area using data of Narita, Koyama, Makino and Okamura\textsuperscript{48}

\[ k = 1.19 \times 10^4 \exp\left(-\frac{38600}{RT}\right) = 0.264 \text{ cm/sec \%N} \]
\[ A = 160 \text{ cm}^2 \]

<table>
<thead>
<tr>
<th>Run 11</th>
<th>Nozzle Area</th>
<th>5.19 x 10^{-3} \text{ cm}^2 (alumina)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Iron Head</td>
<td>6.79 cm = 0.69 psi</td>
</tr>
<tr>
<td></td>
<td>Nozzle Pressure</td>
<td>18 psig</td>
</tr>
<tr>
<td></td>
<td>Argon Flow Rate</td>
<td>93 cm$^3$/sec STP</td>
</tr>
<tr>
<td></td>
<td>Bath Temperature</td>
<td>1570°C</td>
</tr>
<tr>
<td></td>
<td>Bath Composition</td>
<td>0.0073%O, %C, initially</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2.9 %C at end of run</td>
</tr>
</tbody>
</table>

Measured Reaction Rate \[ \frac{kA}{V} = 7.465 \text{ (min \%)}^{-1} \]

Predicted Area using results of Narita, Koyama, Makino and Okamura\textsuperscript{48}

\[ k = 1.19 \times 10^4 \exp\left(-\frac{38600}{RT}\right) = 0.496 \text{ cm/sec \%N} \]
\[ A = 105 \text{ cm}^2 \]
Figure 37. Nitrogen concentration versus time for argon injection of nitrogen saturated iron baths containing low oxygen concentrations.
Figure 38. First order reaction rates for nitrogen desorption from liquid iron to argon jets.
Figure 39. Second order reaction rates for nitrogen desorption from liquid iron to argon jets.
(d) Nitrogen Solution in Systems With 0.56 mm Diameter Nozzle

Nitrogen absorption and desorption experiments were also carried out using a 0.56 mm diameter nozzle under the experimental conditions listed in Table 20. The results of the absorption experiments are plotted in Figures 40 and 42 and the results of the desorption experiments are plotted in Figures 41 and 43.

The effect of carbon pickup on the desorption rates has been accounted for by using the results of Narita, Koyama, Makino, and Okamura. The results of the absorption experiments are distorted by the effect of carbon pickup by erosion from the lid of the system. The nitrogen desorption results do not perfectly fit either a first order or a second order rate law. This may be due to leakage of atmospheric nitrogen into the system during the argon injection or nitrogen pickup during sampling resulting in an equilibrium nitrogen concentration greater than zero in the metal.
TABLE 20
NITROGEN TRANSFER WITH 0.56 mm DIAMETER NOZZLE

<table>
<thead>
<tr>
<th>Run #</th>
<th>Pressure</th>
<th>Flow Rate</th>
<th>Temp.</th>
<th>%C</th>
<th>Reaction Rate</th>
</tr>
</thead>
<tbody>
<tr>
<td>14</td>
<td>15</td>
<td>N₂@49.7</td>
<td>1620</td>
<td>0.010</td>
<td>$kA = 0.1785 \frac{\text{min}^{-1}}{V}$</td>
</tr>
<tr>
<td>15</td>
<td>16</td>
<td>Ar@42.5</td>
<td>1630</td>
<td>--</td>
<td>$kA = 2.83(% \text{N} \text{ min}^{-1})$</td>
</tr>
<tr>
<td>16</td>
<td>20</td>
<td>N₂@68.8</td>
<td>1615</td>
<td>0.055</td>
<td>$kA = 0.3765 \frac{\text{min}^{-1}}{V}$</td>
</tr>
<tr>
<td>17</td>
<td>20</td>
<td>Ar@52.5</td>
<td>1620</td>
<td>0.51</td>
<td>$kA = 3.44(% \text{N} \text{ min}^{-1})$</td>
</tr>
<tr>
<td>19</td>
<td>30</td>
<td>Ar@117.8</td>
<td>1585</td>
<td>2.41</td>
<td>$kA = 15.44(% \text{N} \text{ min}^{-1})$</td>
</tr>
</tbody>
</table>

Calculated interfacial areas using data of Inouye and Choh\textsuperscript{50} for nitrogen absorption and Narita, Koyama, Makino, and Okamura\textsuperscript{48} for nitrogen desorption.

Run 14 44.72 cm\(^2\)
Run 15 47.7 cm\(^2\)
Run 16 96.24 cm\(^2\)
Run 17 61.66 cm\(^2\)
Run 19 133.13 cm\(^2\)
Iron baths through a 0.56 mm diameter nozzle.

Figure 4. Nitrogen concentration versus time for nitrogen injection of

Run 16
Run 14
Nitrogen saturated iron batch through a 0.56 mm diameter nozzle.

Figure 4.1. Nitrogen concentration versus time for argon injection of

Time (min)

Nitrogen Concentration (N)

Run 19
Run 17
Run 15
Figure 42. First order reaction rates for nitrogen injection of iron baths through a 0.56 mm diameter nozzle.
Figure 43. Second order reaction rates for argon injection of nitrogen saturated iron baths through a 0.56 mm diameter nozzle.
VI. DISCUSSION

A. INTRODUCTION

The most important flow regimes in submerged gas injection, in terms of commercial importance, are the high speed jet flow regimes where a large interfacial area for mass transfer is created from the breakup of the jet stream in the liquid metal. However, although some experimental work has been carried out at high gas flow rates, the bulk of the research and nearly all of the theoretical work on the fluid mechanics of gas streams entering liquid baths have been carried out at flow rates where single bubbles are formed at the nozzle orifice and rise, intact, to the surface of the bath. There are several reasons for this. From the experimental standpoint, the high speed of the phenomena and the loss of visibility due to turbulence make it difficult to examine the mechanics of jet behavior in detail.

From a theoretical standpoint, the problems are even greater. The bubbles formed at the nozzle in high speed jets are elongated in the vertical direction. This departure from spherical geometry would greatly complicate the application of the virtual mass models used to estimate the resistance to bubble motion due to liquid inertia. At
high flow rates, the effect of gas momentum on bubble behavior can no longer be neglected and gas compressibility effects must also be considered. Cooling of the gas as it expands through the orifice would be expected to take place and the behavior of the gas when it is injected at super-critical pressure ratios must be predicted. Most of the nozzles used in submerged gas injection are straight pipes, so that supersonic flow would not be expected. Instead, the gas must enter the bath at sonic velocity and elevated pressure and then expand to the bath pressure. The mechanism by which this would take place is not yet known.

The bubbles produced at the orifice in jet flow are not stable, but shatter apart into smaller bubbles shortly after detachment. A theoretical treatment with the goal of predicting gas-liquid interfacial areas must be able to describe the breakup as well as the formation of these bubbles. At very high flow rates, discrete bubbles are no longer formed at the nozzle orifice and instead, a steady cone of gas passes out of the nozzle and breaks up continuously. There are presently no theoretical criteria for the stability of these two flow regimes.

B. EXPERIMENTAL OBSERVATIONS OF SUBMERGED GAS JET BEHAVIOR IN THIS STUDY

Two important observations of submerged gas jet
behavior made in this study may be used to extend the theoretical understanding of this behavior. The mechanism of bubble formation at the orifice has been observed, and the conditions under which the jet flow regime changes from sequential bubbling to steady cone flow have been experimentally characterized.

The bubble formation at the orifice is observed to take place in two stages as is shown in Figures 7, 8, and 9. The first stage of growth consists of approximately spherical growth with the base of the bubble fixed at the nozzle orifice. In the second stage of growth, the base of the bubble remains fixed at the orifice and the horizontal dimension of the bubble remains constant, and the bubble grows by extending itself in the vertical direction. This mechanism of bubble formation was observed in all of the gas-water systems examined in this study at all of the flow rates used.

As shown in Figures 14 and 15, a sharp increase in nozzle chamber pressure takes place during the first stage of bubble growth. During the second stage of bubble growth, the nozzle chamber pressure returns to the background level. The magnitude of the pressure fluctuations (0.5 to 1 psi) is too great to result from surface tension effects (for this system $2\sigma/r_n = 0.01$ psi). Therefore, these pressure fluctuations must be produced by the radial
acceleration of the liquid surrounding the bubble as it grows from a nucleus with a diameter approximately equal to the orifice diameter to a larger sphere. The mode of bubble growth during the second stage does not produce liquid accelerations of sufficient magnitude to affect the nozzle chamber pressure. The single pressure peak observed during the formation of each bubble shows that the elongated bubbles are formed individually at the orifice rather than by coalescence of successive spherical bubbles. Figures 16 and 17 show that the mechanism of bubble formation is not affected by increasing the flow rate into the range where the bubbling type jet is only occasionally stable. The experiments of Oryall and Brimacombe\textsuperscript{76} have shown that the mechanism of bubble formation in horizontal submerged gas jets is similar.

Knowledge of the mechanism of formation of the elongated bubbles makes it possible to devise a theoretical model of the bubble growth. Since spherical geometry is observed in the first stage of growth, the theoretical equations developed for lower velocity systems can be applied to the first stage of growth with two major modifications. The momentum of the gas exiting from the nozzle must be considered in any force balance equation, and the compressibility of the gas cannot be ignored.

The two stage growth mechanism observed for these
bubbles is very similar to the two stage growth mechanism proposed by Kumar, Kuloor, et al.\textsuperscript{14} in lower velocity systems. The bubble diameter at the end of the first stage of growth in those systems could be calculated from a static force balance on the growing bubble. The same procedure has been applied in this system with considerable success (see discussion, sections C and D).

In the model of Kumar, Kuloor, et al.\textsuperscript{14}, the second stage of growth was assumed to consist of further spherical growth as the bubble rose with a thin neck connecting it with the orifice. The bubble position during the second stage of growth could be calculated as a function of time from the equation of motion. When the bubble reached a height above the orifice equal to the diameter of the first stage bubble, detachment was assumed to occur. The amount of growth which took place as the bubble rose through this distance could be calculated from the gas flow rate and the time required for the bubble to rise this distance. In the high speed system, this procedure would be much more difficult. Because the bubble grows by changing its shape, an iterative procedure would have to be used to account for the changes in geometry as the bubble grew. Since the base of the bubble remains fixed at the orifice until detachment, there is no apparent criterion which can be used to determine when the second stage of growth should
end.

The ratio of the final bubble volume to the first stage bubble volume was measured in this study for gas-water systems. Although there was considerable scatter, an average ratio of 5.445 was observed. In the absence of any theoretical model for the second stage of bubble growth, this ratio has been used as a multiplying factor to estimate final bubble volumes from calculated first stage bubble volumes.

It should be noted that there is a great deal of variation in the sizes of bubbles produced at the nozzle in high speed systems. Bubble size variations of more than ±50% would be expected even if an exact equation for the second stage of growth could be determined. Irregular interactions with previously formed bubbles and liquid phase turbulence have a randomizing effect on the bubble sizes in these systems. The bubble diameter at the end of the first stage of growth was much more predictable than the final bubble size.

A criterion for the transition between the jet flow regime where elongated, bursting bubbles were formed at the orifice and the jet flow regime in which a steady walled cone of gas passed out of the orifice was determined in this study. Although there was a very wide region in which the jet behavior fluctuated between the two flow regimes,
the steady cone type flow was observed to predominate when the gas mass flow per unit area through the nozzle was greater than $40 \text{ g/cm}^2\text{sec}$ in all of the gas-water and gas-liquid metal systems considered in this study. This observation has interesting theoretical implications.

There is presently no theory to predict the flow regime of the gas jet. The fact that the flow regime transition is not affected by nozzle size or liquid properties is a surprising result and it is difficult to imagine a theoretical treatment which would predict this result.

In the steady flow jet, a continuous gas region with roughly constant boundaries exists above the nozzle orifice. Gas passes through this region, entraining liquid droplets from the walls and exiting through the top of the region as bubbles passing into the liquid phase. In the bubbling jet flow regime, the gas is not able to pass out of the top end of the region, and instead, the entire gas packet detaches from the nozzle and breaks apart. The mechanism by which the gas forms bubbles at the top surface of the continuous gas region may be critical for determining which flow regime is stable. If the gas cannot leave the continuous region at the flow rate at which it is entering then the cone shape cannot be maintained and bubbling behavior must take place.

The formation of Taylor instabilities at the top of
a jet dome has been considered as a possible mechanism but does not seem promising (see Appendix A). Another possibility is the growth of Helmholtz type instabilities on the vertical walls of the continuous gas cone which may eventually bridge the region and produce bubbles at the top surface. In the highest velocity gas-water systems investigated where the steady cone behavior was generally stable, it was observed that whenever the steady cone behavior was interrupted a bubble or pulsation occurred (see Figure 11). The steady cone behavior then reestablished itself in the wake of the bubble. This behavior is consistent with a model which requires that vertical cone walls bearing instability waves be present for the steady cone flow regime to be stable. Our knowledge of the behavior of these instabilities is not yet sufficiently developed to predict what parameters would have the greatest effects on them.

In all of the cases examined in this study, inert gases were injected at a temperature near the bath temperature. In commercial systems with large diameter orifices, the gas would not be expected to be heated to the same extent in the tuyere. Most of the heating of the gas would take place after it has entered the bath, producing an increase in volume and in liquid inertial resistance to bubble motion. In addition, chemical reactions such as
the cracking of cooling agents would increase the gas volume. These factors could have an effect on the flow regimes and bubble sizes in commercial systems.

C. GENERAL MODEL OF BUBBLE FORMATION AT A SUBMERGED GAS ORIFICE

The two stage growth model of bubble formation proposed by Kumar, Kuloor, et al. assumes the existence of a first stage (force balance) bubble whose volume can be calculated algebraically by balancing the upward and downward acting forces acting upon it. This model has been criticized because the first stage bubble may not exist as a discrete entity in some systems and the dynamic force arising from the acceleration of the bubble during its growth is incorrectly treated as if it were a static force. Nevertheless, the calculation of the force balance bubble volume provides an easy means for estimating final bubble sizes and evaluating the relative importance of different fluid and system properties on the bubble size. In metallurgical processing, where the systems of interest cover an extremely wide range of fluid properties, orifice dimensions, and gas flow rates, an approximate bubble size equation based on this model would be of more value than a highly precise bubble size equation which is valid only for
a narrow range of properties and flow conditions.

Kumar, Kuloor, et al.\textsuperscript{14} calculated the volume of the force balance bubble by balancing three downward acting forces, a surface tension force, a liquid phase viscous drag force, and a liquid inertial force, against an upward acting force produced by buoyancy. To make this model valid at high gas flow rates, the upward acting force due to gas momentum must also be considered. The force balance equation then becomes equation (28).

\[ F_{\text{momentum}} + F_{\text{buoyancy}} = F_{\text{inertia}} + F_{\text{surface}} + F_{\text{viscosity}} \] \hspace{1cm} (28)

After making the assumptions that the bubble growth is spherical and that compressibility effects in the gas are not important, each of the five forces can be evaluated.

The surface tension force is given by equation (2) which has been given previously.

\[ F_{\text{surface}} = 2\pi \sigma r_n \cos \theta \] \hspace{1cm} (2)

The liquid inertia force is calculated from equation (29) which is the result of combining equations (10) through (13) given in the literature survey section.

\[ F_{\text{inertia}} = \frac{11 \, Q^2}{192 \pi R_b^2} \] \hspace{1cm} (29)
The viscous drag force on the growing bubble can be approximated by using Stokes law with \( u_b \) from equation (10).

\[
F_{\text{viscosity}} = \frac{3\mu L Q}{2R_b}
\]  
(30)

where:

\( \mu_L = \text{liquid viscosity} \) (poise)

The buoyancy force on the bubble can be calculated from equation (1) given previously.

\[
F_{\text{buoyancy}} = \frac{4}{3} \pi R_b^3 \rho L g
\]  
(1)

If compressibility effects are neglected, the force produced by the momentum of the gas leaving the nozzle can be calculated from equation (31).

\[
F_{\text{momentum}} = \frac{\rho g Q^2}{\pi r_n^2}
\]  
(31)

The force balance equation for the spherical bubble size is then given by equation (32).

\[
\frac{\rho g Q^2}{\pi r_n^2} + \frac{4}{3} \pi R_b^3 \rho L g = \frac{11}{192} \frac{\rho L Q^2}{\pi R_b^2} + 2\pi \sigma r_n \cos \theta + \frac{3}{2} \frac{\mu L Q}{R_b}
\]  
(32)
This equation can then be arranged to give a fifth order equation for $R_b$, the bubble radius, which can be solved by an iterative technique.

$$
R_b^3 \frac{33Q^2}{768\pi^2 R_b^2 g} - \frac{9\mu_1 Q}{8\pi \rho_1 R_b g} + \frac{3\rho g Q^2}{4\pi^2 r_n^2 \rho_1 g} - \frac{3sr_n \cos\theta}{4\rho_1 g} = 0
$$

(33)

It is not necessary to evaluate equation (33) in every case, however, since in many cases one or more of the five forces may be small enough to be neglected and simpler equations may be used in particular flow regimes. By examining the relative magnitudes of the terms in equation (32) one can estimate which forces are significant under different conditions.

Considering a given gas-liquid system where only the gas flow rate ($Q$) is allowed to vary, three gas flow regimes can be detected. At very low flow rates all of the terms containing $Q$ are small and the principal forces are those due to buoyancy and surface tension. For this case equation (33) reduces to equation (34).

$$
R_b = \left( \frac{3sr_n \cos\theta}{2\rho_1 g} \right)^{1/3}
$$

(34)

This equation is equivalent to equation (3) given previously.

At these low flow rates there is no second stage of growth so that the force balance bubble volume is also equal
to the final bubble volume. In this flow range, the bubble size is controlled by the liquid density, the surface tension, and the nozzle orifice diameter and is independent of the gas flow rate.

As the flow rate is increased, the other terms in equation (32) become significant. Because the liquid density is larger than the gas density by a factor of $10^3$ or more in most systems, the liquid inertia term becomes important at lower flow rates than the gas momentum term. The viscous drag force is always negligible in bubble formation in low viscosity liquids such as water or liquid metals.

When the liquid inertia force is large in comparison with the surface tension force, the bubble size is controlled by the balance between the buoyancy and liquid inertia forces and equation (33) reduces to equation (35).

$$R_b = \left( \frac{33 Q^2}{768 \pi^2 g} \right)^{1/5} \quad \text{or} \quad V_b = 0.1605 \frac{Q^{6/5}}{g^{3/5}} \quad (35)$$

This equation is equivalent to equation (15).

In this flow regime the bubble size is controlled by the gas flow rate only. Because further bubble growth takes place during the second stage of growth in this flow regime, the force balance equation does not give the final bubble volume. The final bubble volume can be calculated
by integrating the equations for bubble size and position until some detachment criterion is satisfied. Since the principal forces acting on the bubble are the same, the equations for the final bubble volume will be of the same form as equation (35), differing only by a constant.

Kumar and Kuloor\textsuperscript{14} predict that the final bubble volume is given by equation (16).

\begin{equation}
V_b = 0.976 \frac{Q^{6/5}}{g^{3/5}}
\end{equation}

(16)

Using a single stage bubble growth model, Davidson and Schuler\textsuperscript{8,9} predicted that the final bubble volume would be given by equation (7).

\begin{equation}
V_b = 1.378 \frac{Q^{6/5}}{g^{3/5}}
\end{equation}

(7)

Wraith\textsuperscript{15} proposed a two stage growth model where the first stage of growth was assumed to be hemispherical rather than spherical. He predicted that the final bubble volume would be given by equation (36).

\begin{equation}
V_b = 1.09 \frac{Q^{6/5}}{g^{3/5}}
\end{equation}

(36)

All of these equations are of the same form as equation (35) and are in fairly good agreement with one another.
It is also interesting to note that the empirical equation of Van Krevelen and Hoftijzer\(^3\) for bubble sizes in this flow regime is also of this form.

\[
V_b = 1.722 \frac{Q^{6/5}}{g^{3/5}}
\]  
(37)

The empirical equation of Leibson et al.\(^6\), given by equation (17)

\[
D_b = 0.18 d_n^{1/2} N_{Re}^{1/3} = 0.18 \left( \frac{4 \rho g}{\pi \mu g} \right)^{1/3} Q^{1/3} d_n^{1/6}
\]  
(17)

and of Davidson and Amick,\(^5\) given by equation (38) also predict that the bubble size is a function of the gas volumetric flow rate raised to an exponent very close to that predicted theoretically.

\[
D_b = 0.54 Q^{0.289} d_n^{0.145}
\]  
(38)

It should be noted that the gas density and viscosity were constant in all of Leibson's experiments and that the dependence of bubble size on nozzle diameter predicted by these equations is very slight.

Using the bubble formation model observed in this study for high speed bubble formation, the final bubble volume is larger than the first stage bubble volume by a
factor of 5.445. Multiplying the bubble volume predicted by equation (35) by this factor gives equation (39).

\[ V_b = 0.874 \frac{Q^{6/5}}{g^{3/5}} \]  

(39)

This equation is not substantially different from the bubble size equations derived with the assumption of spherical geometry for this flow regime.

The bubble sizes predicted by these equations will be formed if the bubbles grow in exact accordance with the geometric assumptions underlying each model. In actual systems, liquid phase turbulence will cause deviations from ideal geometry and there will be considerable scatter in the sizes of the bubbles produced. For a user interested in predicting bubble sizes in an actual system, the use of an approximate bubble size equation such as equation (40) could be a more practical solution than selecting one of the theoretical equations. Actual bubble sizes could deviate from predicted bubble sizes by as much as \( \pm 75\% \).

\[ V_b = \frac{Q^{6/5}}{g^{3/5}} \]  

(40)

If the flow rate through the nozzle is further increased, at some point the force produced by the momentum of the gas leaving the orifice will become greater than
the buoyancy force acting on the bubble. When this occurs, only the gas momentum and liquid inertial forces need be considered for calculating the force balance bubble size. Equation (33) then reduces to equation (41).

$$R_b = \left( \frac{11 \rho_l}{192 \rho_g} \right)^{1/2} r_n$$  \hspace{1cm} (41)

When the gas stream enters this flow regime, the bubble size again becomes independent of the gas flow rate and is controlled only by the orifice diameter and the ratio of the gas and liquid densities.

No previous theoretical studies have considered this flow regime, and its existence has only been reported in a qualitative way. Using the empirical correction factor of 5.445 observed in this study, the volume of these bubbles at detachment is predicted to be given by equation (42).

$$V_b = 0.313 \left( \frac{\rho_l}{\rho_g} \right)^{3/2} r_n^3$$  \hspace{1cm} (42)

To summarize, the overall shape of the bubble volume vs. gas flow rate curve predicted by equation (33) for a given system is as shown in Figure 44. The horizontal section of the curve at low flow rates is defined by equation (34) and the horizontal section at high flow rates is defined by equation (42). In the region between these
The general case.

Figure 44. Bubble diameter as a function of gas volumetric flow rate for
two horizontal sections, the bubble volume is dependent on the gas volumetric flow rate to the $6/5$ power and is given approximately by equation (40).

The flow rates at which the inflection points in the bubble size versus flow rate curve occur can be located by considering the ratios of the forces affecting the bubble size in each case. These ratios can be conveniently expressed as dimensionless numbers. For example, the first inflection point represents the change from surface tension to liquid inertia as the principal downward acting force on the growing bubble. The ratio of the liquid inertial force to the surface tension force is given by equation (43).

$$N_1 = \frac{11 \rho_1 Q^2}{384 \pi^2 \sigma R^2 r_n \cos \theta}$$  \hspace{1cm} (43)

Taking $\cos \theta$ as one and substituting equation (34) for $R_b$, since this ratio is to be evaluated near the surface tension controlled range gives equation (44).

$$N_1 = 0.022 \, g^{2/3} \left( \frac{Q}{\pi} \right)^2 \left( \frac{\rho_1}{\sigma r_n} \right)^{5/3}$$  \hspace{1cm} (44)

Bubble sizes given by equation (34) would be expected for $N_1$ substantially less than unity, and when $N_1$ is substantially greater than unity, first stage bubble sizes given by equation (35) growing to final bubble sizes given
approximately by equation (40) would be expected. Equation (33) would have to be solved for intermediate flow rates.

For most gas-liquid systems, this changeover would be expected to occur at very low flow rates. For a gas-water system with a nozzle diameter of 2 mm, \( N_1 \approx 1 \) when \( Q \approx 12 \text{ cm}^3/\text{sec} \). For a gas-liquid iron system with a nozzle diameter of 1.2 mm, \( N_1 \approx 1 \) when \( Q \approx 18 \text{ cm}^3/\text{sec} \) at the melt temperature. The latter prediction is in agreement with the recent experimental results of Sano and Mori\(^74\) in the liquid iron system.

The second inflection point can be located in a similar manner since it occurs when the upward acting gas momentum force becomes larger than the buoyancy force. The ratio of these two forces is given by equation (45).

\[
N_2 = \frac{3 \rho g Q^2}{4 \pi^2 \rho_1 g r_n^2 R_b^3}
\]  

(45)

Substituting equation (41) for \( R_b \) since this ratio is to be evaluated near the gas momentum controlled range gives equation (46).

\[
N_2 = \frac{54.7}{g r_n^5} \left( \frac{Q}{\pi} \right)^2 \left( \frac{\rho g}{\rho_1} \right)^{5/2}
\]  

(46)

Systems with \( N_2 \) substantially less than unity would be
expected to produce first stage bubble sizes given by equation (35) growing to final bubble sizes given approximately by equation (40), and systems with \( \text{N}_2 \) substantially larger than unity would be expected to produce first stage bubble sizes given by equation (41) growing to final bubble sizes given by equation (42). For intermediate flow rates, equation (33) would have to be solved.

Three important changes in flow behavior take place as the gas flow rate is increased whose occurrence is not indicated in Figure 44. Figure 44 shows only the sizes of the bubbles produced at the orifice, but at some point within the flow range where bubble sizes are given by equation (40), the bubbles produced at the orifice become unstable and break apart after detachment. This produces the column of rising froth commonly associated with submerged gas jets. Unstable bubbles were produced at the lowest flow rates examined in this study. When unstable bubbles are produced, the bubble size produced at the orifice can no longer be used to predict the gas-liquid interfacial area in the system.

Also at some point within the flow range where bubble sizes are given by equation (40), the shape of the bubbles formed at the orifice changes from spherical to the elongated shape observed in this study. Non-spherical bubbles were formed at the lowest flow rates examined in
the gas-water systems in this study, but the formation of spherical bubbles with volumes given approximately by equation (40) has been reported in the literature. This change in bubble shape does not seem to have a substantial effect on bubble size.

The departure of the bubbles from spherical geometry and the instability of these bubbles may be related. The departure from spherical geometry could be caused by the dynamic pressure of the gas leaving the nozzle and discharging against the upper surface of the growing bubble. According to Levich\textsuperscript{75}, the breakup of unstable bubbles is caused by the dynamic pressure of turbulent gas eddies within the bubbles. Turbulence capable of causing bubble breakup could be produced by the discharge of the jet against the growing bubble walls. While the bubble is growing, the positive pressure inside it keeps the bubble walls stable, but when bubble growth stops after detachment, the turbulence within the bubble causes it to break apart.

At some gas flow rate controlled by the mass flow per unit area through the nozzle, the jet will begin to spend time in the steady cone jet flow regime. In this flow regime, individual bubbles are no longer formed at the orifice, and equation (32) is no longer applicable. The beginning of steady cone flow may take place at \( N_2 \) less than unity or \( N_2 \) greater than unity depending on the liquid
density and nozzle diameter in the system. There is a wide transition range in which the jet fluctuates between bubbling and steady cone behavior. In this flow range, when bubbles are formed, their sizes are given by equation (33). Bubble frequencies, measured for example by the pressure pulse technique, cannot be used to measure bubble sizes in this flow range because the intervening periods of steady flow behavior will cause the size to be over-estimated.

As the gas flow rate is further increased, the fraction of time that the jet spends in the steady cone flow regime increases until the bubbling behavior is reduced to occasional bursts or pulsations in a generally steady jet. The behavior of the jet in the steady cone flow regime must be described by entirely different fluid dynamic equations than those that apply when sequential bubbles are formed. The approach of Chawla,37,38,39,40 makes use of Kelvin-Helmholtz instability theory to predict liquid entrainment rates and gas-liquid momentum transfer in the continuous gas region just above the nozzle. The mechanism by which the continuous gas region breaks up into small bubbles has not been determined or mathematically described.

Based on this theoretical treatment, none of the dimensionless numbers usually used to correlate fluid flow behavior (Reynolds number, Weber number, or Froude number
based on the nozzle orifice diameter) can be used to
describe completely the bubble formation in these systems.
Specifically, the Reynolds number used by Leibson et al.\textsuperscript{6} to correlate behavior in the air-water system cannot be used
to predict bubble sizes in fluids of varying density. The
orifice Reynolds number expresses the ratio of momentum
forces to viscous forces in the gas flowing through the
nozzle. The viscous forces in the gas, however, are
negligably small and do not play a role in the bubble
formation. Leibson et al.\textsuperscript{5} believed that the changes in
bubble size with increasing flow rate were the result of
the laminar to turbulent flow transition in the gas stream,
but this is not the case.

D. COMPARISON OF MODEL PREDICTIONS WITH EXPERIMENTAL
RESULTS OF THIS STUDY

Nearly all of the gas-water experiments carried out
in this study involved jets operating in the regime where
equation (42) predicts the bubble size. $N_2 \approx 1$ for
$Q \approx 200 \text{ cm}^3/\text{sec}$ for a nitrogen to water jet injected through
a 2 mm diameter orifice. The measured bubble diameters at
the end of the first stage of growth are in good agreement
with equation (41). Figure 45 shows the effect of changing
orifice diameter on first stage bubble diameter for nitro-
Figure 45. Bubble diameter versus nozzle diameter for the nitrogen-water system at 16 psig driving pressure.
gen jets operated at 16 psig. The results for 1, 2, and 4 mm diameter nozzles are in good agreement with the predictions of equation (41). Figure 46 shows the effect of changing gas density on first stage bubble diameter for gas injection through a 1 mm diameter nozzle. The results for nitrogen, argon, and helium are also in good agreement with equation (41).

In Figure 47, the first stage bubble diameters measured in the nitrogen-water system for 1, 2, and 4 mm nozzle diameters are plotted as a function of gas volumetric flow rate. The theoretical curve from equation (35) as well as the theoretical bubble diameters from equation (41) for 1, 2, and 4 mm diameter nozzles are also shown. The point at which the curve of equation (35) crosses the curve of equation (41) corresponds approximately to the flow rate at which \( N_2 \approx 1 \) for each nozzle diameter.

The experimental points are generally consistent with the predicted behavior considering the scatter in the data. The bubbles do appear to reach a slightly larger size than that predicted by equation (41) before leveling off. The bubble size also begins to increase again with increasing flow rate after following the prediction of equation (41) for a period. This is due to the failure of the simplified gas momentum model to account for compressibility phenomena. As the flow rate is increased beyond
Figure 46. Bubble diameter versus liquid to gas density ratio for systems using 1 mm diameter nozzles.
Figure 47. Bubble diameters in the nitrogen-water system as a function of gas volumetric flow rate.

Flow Rate (cm³/sec)

Bubble Diameter (cm)

1 mm nozzle model

2 mm nozzle model

4 mm nozzle model

Buoyancy

Data points for different nozzle sizes are plotted on the graph, with a trend line illustrating the relationship between flow rate and bubble diameter.
the sonic velocity, the jets become supercritical, and the gas issues from the nozzle at sonic velocity and a higher pressure than the liquid around it. The kinetic energy of the gas is thus lower than assumed in the model. After leaving the nozzle the gas expands to bath pressure. This expansion contributes to the downward acting force on the bubble by causing the motion of the liquid around it. It does not contribute to the upward acting force, so that the force balance is shifted in favor of larger bubbles.

In the liquid metal systems, first stage bubble diameters could not be measured directly. Instead final bubble volumes were calculated by dividing the gas volumetric flow rate by the bubble frequency indicated by the pressure pulse or electrical gas probe technique. The gas was assumed to be at the temperature of the bath in these calculations and these bubble volumes could not be measured in systems where steady flow behavior took place. The first stage bubble volumes predicted by equation (33) were multiplied by the correction factor of 5.445 to give final bubble volumes that would be comparable with the measured values. Although the bubbles produced at the orifice in the bubbling jet flow regime would not be expected to be spherical, equivalent spherical diameters are plotted for comparison purposes.

Figure 48 shows bubble diameters measured using
Figure 48. Bubble diameters produced at a 1 mm diameter nozzle in gas-liquid \textit{tin systems}. 

![Diagram showing flow rate vs. bubble diameter for different gases (He, Ar, N\textsubscript{2}) with associated data points.](image-url)
nitrogen, helium, and argon in the gas-liquid tin system as a function of volumetric flow rate along with the theoretical diameters calculated from equation (33) for nitrogen and helium. Equation (33) predicts that the argon bubbles should be approximately the same size as the nitrogen bubbles. The gases are assumed to leave the nozzle at the bath temperature in the force balance calculations. For all of the gases $N_2$ is less than unity over the entire flow range investigated so that the bubble sizes are increasing with flow rate. $N_2 \approx 1$ for $Q \approx 350$ cm$^3$/sec for nitrogen and $N_2 \approx 1$ for $Q \approx 1600$ cm$^3$/sec for helium. It is not possible to extend the measurements to higher flow rates than those plotted since extended periods of steady flow behavior are observed at higher flow rates and bubble frequencies are no longer proportional to the bubble sizes. Although the predicted values are slightly low in comparison with the measured values, the agreement is still quite good and the shape of the curve is consistent with the data. The most likely source of error is in the relationship between the first stage bubble diameter and the final bubble diameter. Figure 49 shows similar results taken in the gas-solder system. Changing the liquid head in the system does not affect the measured bubble sizes. This would be expected from the theoretical equations.

Figure 50 shows theoretical and measured bubble
Figure 4. Bubble diameters produced at a 1 mm diameter nozzle in gas-liquid solder systems.

Flow Rate (cm/Sec)

Nitrogen Model
HeIIium Model

Metal Head 14.6 cm, 18.7 cm

Bubble Size in Liquid Solder
Bubble Size in Liquid Iron

Figure 50. Bubble diameters produced at a 0.8 mm diameter nozzle in gas-liquid iron systems.
diameters as a function of the gas flow rate at standard
temperature and pressure in the gas-liquid iron system. For
the theoretical curve, the nozzle diameter is assumed to be
0.8 mm and the gas is assumed to leave the nozzle at 1550°C.
Again, the theoretical curve is in close agreement with the
data, although it does appear slightly low. Some of the
data was taken at temperatures higher than 1550°C and this
would partially account for this effect. Because of the
arrangement of the nozzle chamber required to protect the
transducer from high temperatures, it was difficult to tell
when periods of steady flow behavior were taking place
from the nozzle chamber pressure traces. This may have
causd the bubble volumes at high flow rates to be over-
estimated.

E. DYNAMICS OF THE FROTHER COLUMN REGION OF SUBMERGED GAS JETS

In both the elongated bubble and steady cone jet flow
regimes the region of the jet more than a dozen or so
nozzle diameters above the nozzle is occupied by a column of
rising froth. The froth column consists of a mixture of
gas bubbles and liquid rising together in turbulent motion.
Coalescence of the gas bubbles takes place as the mixture
rises. Many very fine bubbles (less than 1 mm in diameter)
are produced by the breakup of the steady cone or elongated
bubbles near the orifice, but very few bubbles this small can be found at a height of more than 15 cm above the nozzle.

If the jet is tall in comparison with the height of the bubbles produced at the orifice, then the majority of the gas-liquid interfacial area in the system is located in the froth column region. Therefore, most of the gas-liquid chemical reactions would also be expected to take place in this region. The behavior of the froth column is not substantially affected by the nature of the behavior taking place at the orifice. The breakup of the elongated bubbles and the breakup of the steady cone jet produce similar behavior in the froth column.

Jet behavior in the froth column was investigated by injecting a pulse of graphite into the jet and timing its passage between two points in the region. Velocities on the order of 200 cm/sec were measured for sonic air jets injected into water. This is drastically lower than the gas velocity at the orifice, but substantially greater than the terminal rise velocity of bubbles under buoyancy alone. The velocity drops to this level within 8 to 10 cm of the 2 mm diameter orifice and afterwards is approximately constant along the length of the jet. Effective diffusivities, which would have indicated the intensity of the turbulence in the froth column, could not be measured because of interference to the light intensity technique used to
measure the graphite concentration.

The average bubble size in the froth column may be controlled by the intensity of the turbulence in that region. As the bubbles are buffeted by the turbulent eddies, they break apart and coalesce so that after the bubbles have traveled a short distance in this region the bubble size distribution is likely to be independent of the original size distribution and controlled instead by the conditions in the froth column. This would be consistent with the observation of Leibson et al.\textsuperscript{6} that the average bubble size 92 cm above the nozzle in the air-water system was a function only of the orifice Reynolds number to the minus 0.05 power.

When the gas is injected at a higher mass flow per unit area, the velocity within the froth column is increased. Increasing the volumetric flow rate of the gas without increasing the mass flow rate by injecting helium did not increase the velocities in the froth column. These results indicate that the transfer of momentum from the gas to the liquid is important for determining the velocities and spreading angle which will be observed in the froth column.

The absorption of $\text{SO}_2$ by a solution of 0.3\% $\text{H}_2\text{O}_2$ was investigated for jets tall enough that most of the mass transfer in the system would be expected to take place in
the froth column. In spite of the higher column velocities, the jets with higher mass flows were found to have higher reaction rates for equal volumetric gas flow rates. This implies that the smaller bubble sizes and increased turbulence produced by the inertial forces in the higher mass flow rate jets more than compensates for the reduced residence time of the gas in these systems. This effect could not be investigated in the gas-liquid iron system because the bath depth was too shallow to allow the froth column region to fully develop.

F. NITROGEN ABSORPTION AND DESORPTION IN LIQUID IRON

The experiments carried out in this study have not been able to resolve the contradictions of the asymmetry of absorption and desorption rates because of the scatter in the data resulting from the difficulty of sampling from a turbulent, bubbly bath. Within the accuracy of the data, a first order rate law seems to apply to the nitrogen absorption reaction, and a second order rate law seems to apply to the nitrogen desorption reaction when the oxygen concentration in the iron is low. When the oxygen concentration in the metal is high, the results cannot distinguish between the two.

The principal difference between the nitrogen de-
sorption experiments of this study and those carried out in Seivert's apparatus is that the nitrogen is desorbed into an argon atmosphere in this case, while a partial vacuum is used in the Seivert's apparatus. Thus the diffusion of nitrogen molecules in the gas phase is a possible rate controlling step in this experiment which could account for the second order dependence of the rate on the nitrogen concentration in the iron.

In the Japanese experiments where second order dependence of the nitrogen desorption rate was also observed, argon was blown onto the surface of nitrogen saturated iron melts. To eliminate the effect of gas flow rate in these experiments, the argon flow rate was increased until there was no further effect of increasing it on the nitrogen desorption rate. This procedure may not completely eliminate the possibility of diffusion in the gas phase as a rate controlling step.

In the experiments of this study, the nitrogen desorption data still has some downward curvature when plotted as a second order rate law, possibly indicating that the order of the reaction is greater than two. A more likely explanation is that there was some leakage of nitrogen from the atmosphere into the system or nitrogen pickup during the sampling which resulted in the nitrogen concentration approaching a level higher than zero in the
argon purging experiments.

In the nitrogen absorption experiments, the results are distorted by the pickup of carbon by erosion from the graphite lid. Carbon reduces the solubility of nitrogen in liquid iron. In cases where carbon pickup was a problem, the reaction rate was calculated only from the first 5 to 6 minutes of the run where the increases in the nitrogen concentration were large in comparison with the decrease in nitrogen solubility due to carbon pickup.

The measured nitrogen absorption and desorption rates in the submerged jet system were compared with published nitrogen absorption and desorption rates for gas passing over inductively heated melts to calculate the effective area for mass transfer in the jet systems. The nitrogen absorption rates of Inouye and Choh \(^{50}\) and the nitrogen desorption rates of Narita, Koyama, Makino, and Okamura \(^{49}\) were used. Narita, Koyama, Makino, and Okamura \(^{48}\) measured lower reaction rates than did others using similar apparatus. It is felt that this effect may be due to the trace quantities of oxygen present in the cylinder gases which were used directly in their experiments. Since these experiments also used cylinder gases directly, their values were felt to be the most comparable. Narita, Koyama, Makino, and Okamura's \(^{48}\) results on the effect of carbon on the nitrogen desorption rate were also used.
The interfacial areas calculated from these runs are plotted in Figure 51 as a function of gas volumetric flow rate. The interfacial areas were found to have a linear relationship to the volumetric flow rates irregardless of the nozzle orifice diameter or gas driving pressure. There also appears to be no effect of whether the absorption or desorption rate is used for the measurement. An interfacial area of $1.107 \text{ cm}^2/(\text{cm}^3/\text{sec})$ of flow was observed in these systems. The results of Run Number 1 are not included in this correlation since the carbon content was not measured in that experiment.

The extremely large size of the bubbles produced at the nozzle in the liquid iron system provides a partial explanation for the low mass transfer areas measured in the nitrogen absorption and desorption experiments. The large bubbles produced at the nozzle in bubbling jet flow are unstable and the bulk of the gas-liquid interfacial area in these systems is produced when they break up shortly after detaching from the orifice. However, the bubbles produced in the liquid iron system using the 0.8 mm diameter nozzle are so large that if they are assumed to be approximately cylindrical with a diameter equal to the calculated first stage bubble diameter, then their height must be approximately 7 cm. This is very close to the bath depth used in these experiments. The bubbles produced
Figure 51. Gas-liquid interfacial area for submerged gas jets injected into liquid iron versus gas volumetric flow rate.
at the 0.56 mm nozzle are slightly smaller but still nearly as tall as the bath depth at the flow rates investigates. As a result the bubbles leave the bath shortly after detaching from the orifice and do not have an opportunity to break up and create a large surface area.

The good agreement between the results calculated from nitrogen absorption and desorption rates is surprising since the nitrogen desorption rate is believed to be controlled by a chemical reaction step at the gas-metal interface, while the nitrogen absorption rate is believed to be controlled by the diffusion of dissolved nitrogen away from the interface and into the metal bath.

The published absorption rates used in this study were measured in melts which were heated and stirred by induction heating. The recent studies by Bester and Lange of nitrogen absorption into melts which were shielded from the induction stirring effect indicates that the stirring greatly increases the reaction rate over the level observed in a stagnant bath. The fluid mixing near the gas-liquid interface in induction stirred melts may be comparable to the mixing around a rising bubble.

The agreement between the results measured with the 0.8 mm and 0.56 mm diameter nozzles indicates that the flow behavior in the two systems is approximately the same. The bath depth was approximately constant in all of the runs,
so in both cases large bubbles were produced at the nozzle which did not have time to break up completely before leaving the bath. The higher interfacial areas observed at the higher flow rates may be due to the increased agitation and mixing between gas and liquid as the bubbles pass through the bath at a higher rate.

G. APPLICATION OF JET BEHAVIOR MODELS TO PROCESS CONDITIONS

If typical industrial dimensions are inserted into the theoretical equations for the size of the bubbles produced at the orifice, then jet behavior in the large scale systems can be predicted. For a nozzle diameter of 1 cm, a bath depth of 1 meter, and oxygen injection at $4.17 \times 10^5$ cm$^3$/sec at room temperature, $N_2 = 18.3$, so that equation (41) would be expected to control the first stage bubble size. This bubble would be approximately 13.7 cm in diameter. Using equation (42), the volume of such a bubble would be about 7.34 liters. Assuming that this bubble has the same shape as those observed in the water system, this bubble would be approximately 50 cm tall, a substantial fraction of the total bath depth. It should be noted that this is a minimum size since the effects of heating the gas would tend to increase the bubble size.

The mass flow per unit area for such a jet would be
about 700 g/cm² sec so that the jet would be expected to be in the steady cone flow regime most of the time. Occasional fluctuations into the bubbling flow regime could take place. These fluctuations could be the source of the oscillatory accelerations of the converter vessel which Knauer et al.⁶⁴ reported could be analyzed to determine the temperature of the vessel.

When the correlation between the gas liquid interfacial area and the flow rate is extrapolated to process conditions such as the 7 x 10⁵ cm³/sec reported by Fruehan⁷⁰ for a 30 ton experimental Q-BOP, the predicted interfacial area is 7.75 x 10⁵ cm². This is in surprisingly good agreement with the interfacial area of 1.1 x 10⁶ cm² estimated by Fruehan from the nitrogen absorption rate at high oxygen concentrations.

The close agreement between the correlation obtained in the 8 lb, 7 cm deep system and the results obtained in the 30 ton, 75 cm deep system must be considered somewhat fortuitous because of the length of the extrapolation and our lack of knowledge of the mechanics of these systems. It is not reasonable to expect that such a simple correlation should be valid over such a wide range of variables. The good agreement may imply, however, that the systems are fluid mechanically similar in some important respects. Although the nozzle sizes and driving pressures were not published
for the 30 ton system, the jets in this system may be in the flow regime in which large bubbles are produced at the nozzles with heights on the same order of magnitude as the bath depth. The breakup of these bubbles as they leave the bath then produces most of the interfacial area in these systems. The use of smaller nozzles might be expected to produce a larger interfacial area per unit of gas flow by reducing the size of the bubbles produced at the orifice. This would increase the distance that the bubbles travel after detachment and give them more time to break up before reaching the surface of the bath. The bulk liquid mixing produced by the formation of the large bubbles may be more important to the process than is apparent, however, and the practical problems of subdividing the gas stream into many small gas streams may make such a furnace impractical.

The behavior of the graphite powder injected into the water bath may be similar to the behavior of powdered fluxes injected into liquid metal baths with gas streams for refining purposes. When the graphite was injected while the gas was in the steady cone flow regime, the powder was deposited quickly onto the surfaces of the cone and then carried up into the bubble column region. When the graphite was injected while the jet was in the bubbling jet regime, the graphite deposited in the liquid on the upper surfaces of the growing bubbles. When the
unstable bubbles shattered apart, the graphite was dispersed into the bubble column.

Dispersion coefficients in the bubble column could not be measured in these experiments because of changes in the illumination of the jet with height above the orifice and interference due to the passage of bubbles across the measuring points. The bubbling type jet seemed to produce a more rapid dispersion of the powder in the bubble column than did the steady cone type jet as well as a longer residence time because of the lower column velocity. This is consistent with the results of Kawakami\textsuperscript{62} which showed that the highest reaction rates between injected alkali powder and sulfur dissolved in the metal were achieved when the carrier gas flow rates were low.
VII. SUMMARY AND CONCLUSIONS

An experimental study of the fluid dynamics of submerged gas jets has been carried out on gas-liquid systems using helium, nitrogen and argon injected into water, liquid tin, liquid solder, and liquid iron.

High speed cinematography has been used to observe the flow regimes in gas-water jets. Two flow regimes were observed; one in which large bubbles were formed at the nozzle and shattered into many small bubbles after detachment, and one in which an apparently steady cone of gas passes out of the nozzle and breaks up continuously into small bubbles. At heights of more than a few centimeters above the nozzle both jets consist of a column of rising froth. Typical jets fluctuate between the two regimes, but with increasing mass flow rate of gas per unit area through the nozzle the steady walled jet becomes more prevalent. Jets with flows less than 40 g/cm² sec are predominantly bubbling while jets with greater flows are predominantly steady. This criterion adequately describes the flow regime transitions in all of the systems examined in this study.

A technique has been developed to investigate jet behavior in liquid metals. The technique makes use of the sharp increase in nozzle chamber pressure which occurs
whenever a bubble is nucleated in the bubbling jet flow regime. This technique can be used to distinguish between periods of each type of behavior and to measure bubble frequencies in the bubbling jet regime. The technique was tested by taking high speed film and simultaneously recording nozzle chamber pressure traces in the gas-water system and recording the output of an electrical gas probe simultaneously with the nozzle chamber pressure trace in the gas-liquid solder system.

A theoretical treatment of bubble formation at submerged orifices has been developed, based on concepts used to describe bubble formation in lower flow rate systems, which can be used to predict the sizes of bubbles produced at submerged orifices at all flow rates where discrete bubbles are formed. The predictions of this model are in good agreement with measured sizes of bubbles produced at the nozzle in the bubbling jet flow regime in all of the systems examined in this study.

The absorption of SO₂ in solutions of H₂O₂ has been used to investigate the effects of changing flow regimes on mass transfer rates in jets in aqueous systems. The steady cone type jet was found to be more effective for mass transfer than the bubbling type jet.

The absorption and desorption rates of nitrogen in liquid iron were studied in order to estimate the inter-
facial area created by submerged gas jets entering liquid iron. The apparent asymmetry of nitrogen absorption and desorption rates was observed in this study. A first order rate law was observed for nitrogen absorption and a second order rate law was observed for nitrogen desorption. Mass transfer of dissolved nitrogen in the liquid metal is believed to be the rate controlling step for nitrogen absorption while either a chemical reaction at the gas-metal interface or mass transfer in the gas may control the nitrogen desorption rate into argon jets. The reasons for the apparent asymmetry of absorption and desorption rates are not known and further study of this system is required. A correlation for gas-liquid interfacial area as a function of gas volumetric flow rate was developed predicting $1.107 \text{ cm}^2/(\text{cm}^3/\text{sec})$. This correlation is in agreement with estimated interfacial areas in industrial scale equipment.
VIII. SUGGESTIONS FOR FURTHER WORK

The force balance model for bubble sizes used in this study is only a simplified, approximate model for the bubble formation. A more accurate, but highly complex model could be produced by integrating the forces on the growing bubble with respect to time from the initiation to detachment. Such a model must also take the bubble shape changes and compressibility effects into account. In spite of its greater complexity, it is not clear that such a model would give a better prediction of bubble sizes in the jets because of the large scatter in the bubble sizes produced at the nozzle in a given system and the fact that it is the breakup of these bubbles that produces the bulk of the interfacial area in reactive systems. Some information helpful in the understanding of the flow regime transition might result from such a model. Production of such a model might best be carried out by fluid mechanics specialists, since the fluid dynamics of these systems become very complicated and no knowledge of metallurgy is required.

Further water model experiments could be undertaken to investigate the transition from spherical to elongated bubble formation at the orifice. In the lowest flow rates investigated in this study, elongated bubbles were already
being produced. No systematic changes in bubble shape were observed with changing flow parameters in this study, but the great scatter in these results makes this conclusion somewhat unsatisfactory. Further study of this problem could be undertaken.

Further study of the nitrogen absorption and desorption reactions is required to give a better explanation of the asymmetry of absorption and desorption rates. Experiments where the gas-liquid interfacial area and liquid flow conditions are tightly controlled would be required to achieve this.

Investigation of the dynamic behavior of the jet using the nozzle chamber pressure trace technique would also benefit from the use of larger experimental apparatus. Since there is a minimum practical size for the nozzle chamber volume, nozzle chamber pressure traces with greater amplitude and clarity could best be achieved by using larger diameter orifices. The application of this technique to a tuyere in an industrial system would also be of interest. Mass transfer rates between submerged gas jets and liquid iron could be measured more conveniently in a larger experimental system where metal splash problems are not so serious.
APPENDIX A
TAYLOR INSTABILITIES AT A JET DOME

One possible physical model of the steady cone type submerged gas jet assumes that the top of the continuous gas region is a dome and that gas passes out of the continuous gas region by forming bubbles in the dome.\textsuperscript{34} One possible mechanism for the formation of such bubbles is the growth of Taylor instabilities on the dome interface.

Taylor instabilities are instabilities which develop on the interface between two fluids of different densities placed one above the other. If the upper liquid has a higher density than the lower liquid and the viscous and surface tension forces are negligible, it can be shown that a sinusoidal disturbance in the interface between the two fluids will increase in amplitude with time.\textsuperscript{65} For the case of gases and liquid metals, the surface tension is significant but the viscosity is small. For this case, instabilities larger than a critical wavelength will increase in amplitude and instabilities smaller than this will decrease in amplitude with time.\textsuperscript{66} The bubbles produced as the result of the growth of such instabilities would be expected to have dimensions of the same order of magnitude as the wavelength of the instability. The critical wavelength for inviscid fluids with surface
tension is given by equation (1)

\[ \lambda_c = 2\pi \left( \frac{\sigma}{(\rho_1 - \rho_g)g} \right)^{1/3} \]  

(50)

Critical wavelengths for liquids used in this study are given in Table 21. The critical wavelength for water is clearly much larger than the diameter of the bubbles observed during steady cone flow in the gas-water systems. Therefore, growth of Taylor instabilities at a jet dome is not likely to be the mechanism by which these bubbles are formed.

### TABLE 21

**CRITICAL WAVELENGTHS FOR TAYLOR INSTABILITIES**

<table>
<thead>
<tr>
<th>Liquid</th>
<th>Critical Wavelength (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>1.70</td>
</tr>
<tr>
<td>Tin</td>
<td>1.78</td>
</tr>
<tr>
<td>Iron ((\sigma = 1600))</td>
<td>3.02</td>
</tr>
<tr>
<td>Iron ((\sigma = 800))</td>
<td>2.13</td>
</tr>
</tbody>
</table>
APPENDIX B
GAS FLOW TEMPERATURE IN LIQUID IRON SYSTEM

Heating effects in the gas after it has been injected into the liquid metal may be responsible for the difference in effects observed in liquid iron from those observed in lower temperature experiments. For this reason, it would be useful to be able to estimate the temperature of the gas as it leaves the nozzle. Since the sonic flow of a gas through a duct with heat transfer to the gas is controlled by the conditions at the exit from the duct, it should be possible to estimate the flowing temperature of the gas from the ratio of the flow rates produced through the nozzle by the same pressure difference at room temperature and at some higher temperature.

The adiabatic mass flow rate through a choked nozzle is given by:

\[ W = C_N P_1 A \left( \frac{\gamma MW}{RT} \right) \left( \frac{2}{\gamma+1} \right)^{\frac{\gamma+1}{\gamma-1}} \]  

(51)

Where:  
\( P_1 \) = the orifice pressure  
\( A \) = the nozzle orifice area for flow  
\( \gamma \) = the adiabatic exponent \( \frac{C_p}{C_v} \)  
\( MW \) = the molecular weight of the gas  
\( R \) = the gas constant in moalr units
\[ T = \text{the gas temperature} \]
\[ C_N = \text{the orifice discharge coefficient.} \]

For a monatomic gas where \( \gamma \) is not a strong function of temperature

\[
W_1 = C_{N1} P_1 A \sqrt{\frac{\gamma MW}{RT_1}} \left( \frac{2}{\gamma + 1} \right)^{\gamma + 1 \over \gamma - 1}
\]

\[
W_2 = C_{N2} P_1 A \sqrt{\frac{\gamma MW}{RT_2}} \left( \frac{2}{\gamma + 1} \right)^{\gamma + 1 \over \gamma - 1}
\]

and:

\[
T_2 = T_1 \left( \frac{W_1}{W_2} \right)^2 \left( \frac{C_{N1}}{C_{N2}} \right)^2
\]

(52)

Leibson et al.\(^6\) have shown that \( C_N \) is not affected by the presence of liquid on the discharge side of the nozzle, so it can be taken as a function of the gas flow parameters only. Table 22 shows orifice discharge coefficients from 0.8 and 0.56 mm diameter nozzles measured at room temperature. The discharge coefficient for a given nozzle appears to be an increasing function of orifice Reynolds number and a decreasing function of gas density. This is consistent with Leibson's results. Since increasing the gas temperature lowers both the gas density and the orifice Reynolds number, it may be expected that the discharge
<table>
<thead>
<tr>
<th>Pressure (psia)</th>
<th>Flow (g/sec)</th>
<th>Ideal Flow (g/sec)</th>
<th>Discharge Coefficient</th>
<th>Reynolds's Number</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.8 mm Diameter Nozzle, Argon Flow</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>29.7</td>
<td>0.2705</td>
<td>0.312</td>
<td>0.866</td>
<td>1.91 x 10^4</td>
</tr>
<tr>
<td>30.7</td>
<td>0.264</td>
<td>0.323</td>
<td>0.817</td>
<td>1.86 x 10^4</td>
</tr>
<tr>
<td>32.7</td>
<td>0.302</td>
<td>0.344</td>
<td>0.878</td>
<td>2.13 x 10^4</td>
</tr>
<tr>
<td>34.7</td>
<td>0.319</td>
<td>0.365</td>
<td>0.874</td>
<td>2.25 x 10^4</td>
</tr>
<tr>
<td>30.7</td>
<td>0.276</td>
<td>0.323</td>
<td>0.854</td>
<td>1.95 x 10^4</td>
</tr>
<tr>
<td>32.7</td>
<td>0.295</td>
<td>0.344</td>
<td>0.857</td>
<td>2.08 x 10^4</td>
</tr>
<tr>
<td>34.7</td>
<td>0.322</td>
<td>0.365</td>
<td>0.882</td>
<td>2.27 x 10^4</td>
</tr>
<tr>
<td>36.7</td>
<td>0.345</td>
<td>0.386</td>
<td>0.894</td>
<td>2.44 x 10^4</td>
</tr>
<tr>
<td>38.7</td>
<td>0.369</td>
<td>0.407</td>
<td>0.907</td>
<td>2.61 x 10^4</td>
</tr>
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<td>44.7</td>
<td>0.426</td>
<td>0.470</td>
<td>0.906</td>
<td>3.01 x 10^4</td>
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<tr>
<td>29.7</td>
<td>0.271</td>
<td>0.312</td>
<td>0.867</td>
<td>1.91 x 10^4</td>
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<td>30.7</td>
<td>0.280</td>
<td>0.323</td>
<td>0.867</td>
<td>1.98 x 10^4</td>
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<tr>
<td>32.7</td>
<td>0.304</td>
<td>0.344</td>
<td>0.884</td>
<td>2.15 x 10^4</td>
</tr>
<tr>
<td>34.7</td>
<td>0.323</td>
<td>0.365</td>
<td>0.885</td>
<td>2.28 x 10^4</td>
</tr>
<tr>
<td>0.56 mm Diameter Nozzle, Argon Flow</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>30.7</td>
<td>0.164</td>
<td>0.153</td>
<td>1.072</td>
<td>1.68 x 10^4</td>
</tr>
<tr>
<td>32.7</td>
<td>0.181</td>
<td>0.163</td>
<td>1.110</td>
<td>1.86 x 10^4</td>
</tr>
<tr>
<td>34.7</td>
<td>0.191</td>
<td>0.173</td>
<td>1.104</td>
<td>1.96 x 10^4</td>
</tr>
<tr>
<td>44.7</td>
<td>0.256</td>
<td>0.223</td>
<td>1.148</td>
<td>2.62 x 10^4</td>
</tr>
</tbody>
</table>
### 0.56 mm Diameter Nozzle, Argon Flow (continued)

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Mass Flow (g/min)</th>
<th>Velocity (m/s)</th>
<th>Mach Number</th>
<th>Pressure (Pa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>54.7</td>
<td>0.323</td>
<td>0.273</td>
<td>1.183</td>
<td>3.31 x 10⁴</td>
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<tr>
<td>64.7</td>
<td>0.394</td>
<td>0.322</td>
<td>1.224</td>
<td>4.04 x 10⁴</td>
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</tbody>
</table>

### 0.56 mm Diameter Nozzle, Helium Flow

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Mass Flow (g/min)</th>
<th>Velocity (m/s)</th>
<th>Mach Number</th>
<th>Pressure (Pa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30.7</td>
<td>0.0523</td>
<td>0.0485</td>
<td>1.080</td>
<td>6.13 x 10³</td>
</tr>
<tr>
<td>32.7</td>
<td>0.0585</td>
<td>0.0516</td>
<td>1.134</td>
<td>6.85 x 10³</td>
</tr>
<tr>
<td>34.7</td>
<td>0.0626</td>
<td>0.0549</td>
<td>1.143</td>
<td>7.33 x 10³</td>
</tr>
</tbody>
</table>
coefficients do not change substantially with increasing temperature.

Table 23 shows a number of flowing gas temperature estimates where the gas was discharged into air and the nozzle temperature was measured by optical pyrometer. Since the rotameter flow meters can only be read to an accuracy of about ±5\% in this flow range, these calculations can only estimate the gas flowing temperature to an accuracy of a few hundred degrees. However, these results do indicate that in cases where the orifice diameter is small, the gas leaves the nozzle at a temperature quite near the temperature of the nozzle. Therefore one would not expect the jet behavior to be significantly affected by heating of the gas within the metal bath. Table 24 shows temperatures estimated for argon jets discharging into liquid iron.
TABLE 23
FLOWING GAS TEMPERATURES

<table>
<thead>
<tr>
<th>Pressure (psia)</th>
<th>Flow Rate (cm(^3)/sec)</th>
<th>Gas Temperature (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A. 0.8 mm Diameter Nozzle, Argon, 1300°C Nozzle Temperature</td>
<td></td>
<td></td>
</tr>
<tr>
<td>29.7</td>
<td>81.5</td>
<td>1150</td>
</tr>
<tr>
<td>30.7</td>
<td>84.9</td>
<td>1130</td>
</tr>
<tr>
<td>B. 0.56 mm Diameter Nozzle, Argon, 1400°C Nozzle Temperature</td>
<td></td>
<td></td>
</tr>
<tr>
<td>30.7</td>
<td>40.0</td>
<td>1525</td>
</tr>
<tr>
<td>32.7</td>
<td>43.8</td>
<td>1550</td>
</tr>
<tr>
<td>34.7</td>
<td>47.5</td>
<td>1450</td>
</tr>
</tbody>
</table>

TABLE 24
FLOWING GAS TEMPERATURES TO LIQUID IRON

<table>
<thead>
<tr>
<th>A. 0.8 mm Diameter Nozzle, Argon</th>
</tr>
</thead>
<tbody>
<tr>
<td>32.7</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>B. 0.56 mm Diameter Nozzle, Argon</th>
</tr>
</thead>
<tbody>
<tr>
<td>30.7</td>
</tr>
<tr>
<td>34.7</td>
</tr>
</tbody>
</table>
APPENDIX C

RETARDED COALESCEANCE IN SUBMERGED GAS JETS

In one film experiment (film #34) nitrogen was injected into a concentrated sodium chloride brine with a specific gravity of 1.15. The original intention of this experiment was to investigate the effect of changes in liquid density on gas jet behavior, since NaCl has very little effect on the surface tension and viscosity of water.

When the jet was introduced into the brine, however, a cloud of very fine bubbles was produced which quickly filled the entire bath and obscured the jet itself from view. The bubbles produced were so fine that their rise velocity was too slow to allow them to leave the bath when the jet reached the liquid surface. This behavior is similar to that observed by Leibson et al.\(^6\) when surface active chemicals were added to the water. They attributed the effect to the lowering of the interfacial energy.

Sodium Chloride, however, has a slight negative surface activity and increases the interfacial energy by a small amount. The presence of inorganic salts in water has been reported to retard the coalescence of bubbles.\(^68,69\) This is believed to result from either an electrical double layer effect or a change in the surface viscosity of the water. Surface active chemicals also retard the coalescence
of bubbles in water.\textsuperscript{68}

If the only effect of inorganic salts is to retard bubble coalescence, then the production of a cloud of very fine bubbles when a jet is injected into a salt solution implies that bubbles of this size must be produced by jets into pure liquids as well. When the jets are injected into pure liquids the fine bubbles coalesce with larger bubbles in the jet column. Similar effects of retarded coalescence may occur in liquid metals containing surface active elements.
Several experiments were carried out investigating bubble formation and jet behavior in nozzles consisting of concentric orifices where the flow rate to either part could be varied independently. The mechanism of bubble formation in the bubbling jet flow regime was found to be the same in these jets as in the single orifice jets. The force balance used to calculate a predicted bubble size is more complex, however, because the momentum contributions of the two gas streams must be considered separately. For the momentum controlled case the predicted bubble size is given by:

$$\frac{\rho_1 Q_1^2}{A_1} + \frac{\rho_2 Q_2^2}{A_2} = \frac{11(Q_1 + Q_2)^2}{192 \rho_1 R_b^2}$$

(53)

where subscript 1 refers to the core gas and subscript 2 refers to the annular gas stream. The theoretical bubble diameters calculated by this equation for runs 35, 36 and 37 are 2.25, 2.67, and 1.73 cm, respectively. These are in reasonably good agreement with the measured bubble diameters listed in Table 10.

The flow regime transition in the double nozzle is similarly more complicated than in the single orifice
nozzles. The mass flow per unit area through each section of the nozzle must be considered and the effects must be weighed in proportion to the volumetric flow rate of the gas through the section of the nozzle considered. For example in the case of Film 36, where argon flows in the core and helium flows in the annulus the effective mass flow per unit area is given by equation (2).

\[
\frac{2210}{3880} \frac{3.64 \text{ g/sec}}{5.72 \times 10^{-2} \text{ cm}^2} + \frac{1670}{3880} \frac{0.275 \text{ g/sec}}{3.48 \times 10^{-2} \text{ cm}^2} = 39.6 \text{ g/cm}^2 \text{sec} \quad (54)
\]

The jets behavior in the film was observed to be similar to a single nozzle jet operating at about 40 g/cm² sec. In film 37, however, where the helium flow rate is very small in comparison with the argon flow rate, the jet behaves like an argon jet operating at about 63 g/cm² sec and the helium seems to be merely carried along. This type of behavior would be expected in most practical systems where the flow rate of the coolant gas is small in comparison to the flow rate of the reactive gas.

The fact that the weighting term needed to predict jet behavior in double nozzle systems is a function of the volumetric flow rates of the gases implies that the flow regime transition is controlled by the ratio of liquid inertia to gas inertia forces on the jet.
REFERENCES


51. Inouye, M., Choh, T., "Some Considerations on the Nitrogen Transfer Across Gas-Liquid Iron Interface,"


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