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Physical Phenomena in Containerless Glass Processing

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Introduction

It has been suggested by scientists that certain new and unique glasses can be made in orbital laboratories because drops of liquid can be suspended and cooled without the need for a container (1-2). Glasses are amorphous substances formed by cooling a liquid rapidly enough to avoid the formation of a crystalline material. While common glasses can be made with relative ease, Topol et al. (1) found that certain rare earth oxides could not be cooled into a glass in a container because of container wall induced heterogeneous nucleation. However their experiments using a gas jet levitation scheme showed that the same substances could be cooled into an amorphous material when the container was avoided. Work by Day (3) is currently in progress on studying glass formation in a containerless mode in space.

Gas bubbles are commonly encountered in glass processing. They are released due to chemical reactions, and also are present because of gases trapped in the interstitial region among the grains of the raw materials. The bubbles, on earth, are removed by buoyant rise to the free surface as well as by dissolution. In orbit, the former mechanism is of negligible importance, and it is essential to explore mechanisms other than dissolution for the removal of bubbles from space-processed glasses.

Gas bubbles within liquid drops also are encountered in other applications on earth. For instance, in inertial confinement fusion, a hollow glass shell is filled with the fusion fuel and imploded in a laser or ion beam. The process appears to require shells with a uniform wall thickness. On earth, the shells are made in processes which are incompletely understood. Common to most of these is the appearance, at some point in the process, of a molten drop of liquid containing one or more gas bubbles. The drop typically settles in a gaseous medium in a furnace, and the bubbles within it execute motion and undergo possible size change. The drop solidifies into a glass shell at the bottom of the furnace. A surprisingly large yield of good shells (of relatively uniform wall thickness) apparently has been observed in this process. The reason for this self-centering of the gas bubbles within liquid drops is not clear. Oscillation has been suggested (4) and we have proposed drop rotation (5). Other candidates include expansion and contraction. Other applications in which gas or vapor bubbles might be encountered within drops of liquid include spray drying and encapsulation of volatile materials.

It is difficult to experiment on earth with a relatively large drop of liquid in a gaseous medium with gas bubbles present in it. Using orbital experiments on liquid drops containing gas bubbles it is possible to extract useful information concerning the behavior of bubbles in drops subjected to stimuli. Such information is of technological significance in the above applications.

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Objectives

The objectives of this investigation are

To develop an understanding of fluid motion and bubble and droplet motion and interaction when drops containing bubbles are subjected to stimuli such as surface tension gradients, rotation, oscillation, expansion and contraction.

Flight Experiments

It is envisioned that experiments will be conducted in orbit on drops of model liquids containing bubbles in a device which can hold and manipulate drops of liquid at room temperature using acoustic fields. Experiments also are planned on drops of glass melt, to occur at a later stage, in an appropriate apparatus. The model liquids chosen include Dow Corning silicone oils (dimethylsiloxane and similar polymers) and Union Carbide NIAX Polyols (Polypropylene Glycols). These fluids are sufficiently viscous to simulate the fluid mechanical phenomena expected in glass melts. In particular, the ranges of Reynolds and Prandtl numbers will be comparable to those expected in applications involving glass melts.

Initially, two types of experiments are planned, and are schematically illustrated in Figure 1. In one class, drops of liquid, within which one or more gas bubbles have been inserted, will be subjected to a beam of light (of suitable wavelength and intensity) which will be absorbed by the liquid near the surface. The resulting temperature gradients at the drop surface and at the drop-bubble interfaces will lead to surface tension gradients. The consequence will be motion within the drop and migration of the bubbles. The origin of such flows is termed thermocapillarity. It is expected that the bubbles will move toward the heated spot, and might subsequently be extracted from the drop. The shapes of the drop and the bubbles during the experiment also are of interest. The objective of the experiment is to compare the resulting motion of the bubbles with predictions from appropriate theoretical models. Ultimately, it is expected that such experiments might pave the way to the development of practical techniques for managing bubbles within liquid drops in orbit.

In the second category of experiment, similar drops containing bubbles will be subjected to rotation. The resulting pressure fields within the drop will drive the migration of the gas bubbles toward the center of the drop and also deform the shapes of the drop and the bubbles. In this case, the rotation speeds would be chosen to be sufficiently small to avoid shape bifurcations. The objective of this experiment is to compare the resulting trajectories of the bubbles as well as shapes with available theoretical models. This experiment can complement the previous one in providing techniques for bringing several small bubbles within a drop to its center to coalesce into a large bubble which might subsequently be extracted or otherwise manipulated within the drop.

The migration of the bubbles as well as shape information in the above experiments will be recorded via a video camera. Three orthogonal views will be recorded to extract full spatial information from the resulting images. The videotape will be brought back to earth for analysis.

The parameters in the experiments will be varied over a sufficient range either in a single flight or in multiple flights to gain good insight into the physical processes being studied. The experiments will have the best chance of success when they can be performed by a scientist in the space laboratory, and repeated.

The Drop Dynamics Module, designed and built at the Jet Propulsion Laboratory is considered ideal for the conduct of the above experiments. This device was flown on a Shuttle mission in 1985, and experiments on drop dynamics were performed by Dr. Taylor Wang of Jet Propulsion Laboratory who was an astronaut on this mission. His experiments were conducted in a shirt-sleeve environment, and repeated as desired.

The Drop Dynamics Module was not available to this investigation in its initial flight. Since it is well-suited for the conduct of the present experiments, it has been requested that the Clarkson experiments be accommodated on it during its next flight.

Supporting Ground-Based Studies

To date, five Ph.D. theses and five M.S. theses have been completed in support of our flight program. Currently, the program supports two graduate students. In addition, another student working on related problems is being supported as a NASA Graduate Student Researcher.

It is not possible to outline details of the past ground-based research program in this short document. The principal results have been published in the open literature beginning in 1979, and a complete bibliography has been provided separately along with the abstract. The bibliography lists 22 refereed papers published in journals such as J. Colloid and Interface Science, Chemical and Engineering Science, AIChE Journal, Physics of Fluids, J. American Ceramic Society, J. Fluid Mechanics etc., 21 Contributions to Conference Proceedings, and 54 talks presented at various meetings in the United States, Canada, and Western Europe. Copies of all the published papers have been provided routinely to the Microgravity Sciences Division offices at NASA Headquarters as well as to the Marshall Space Flight Center.

In the rest of this document, some interesting scientific problems currently being pursued on ground will be discussed.

Surface Tension Driven Motion

The literature on surface-tension driven motion has been reviewed by Scriven and Sternling (6), Levich and Krylov (7), and Schwabe (8). Some details also are discussed by Ostrach (9). Generally, the fact that gradients in interfacial tension at a fluid-fluid interface can drive fluid motion has been known since the last century. However, experimental measurements of surface tension driven flow velocities and the characteristics of such flows have been relatively rare, and substantially more attention has been devoted to stability problems. Initially in the present program, we undertook the task of making some measurements. The results from our experiments on capillary liquid bridges of silicone oils and glass melts in which flow was generated by surface tension gradients have already been reported in the literature.

In recent years, we have been investigating problems involving gas bubble and liquid drop migration in a temperature gradient. It is known that a gas bubble can be made to move by applying a temperature gradient. Briefly, the interfacial tension depends upon temperature and hence varies around the surface of the bubble. The resulting tangential stress acts upon the neighboring liquid causing it to move toward the cooler side of the bubble. By reaction, the bubble propels itself toward warmer regions.

The motion of gas bubbles due to the action of a temperature gradient was demonstrated by Young, Goldstein, and Block (10) in 1959. By applying a downward temperature gradient on bubbles contained in a capillary liquid bridge, Young et al. were able to arrest the normal buoyant rise of the bubbles, and make them move downward on occasion. These investigators also solved the governing field equations, and from a force balance on the bubble, extracted the steady migration velocity of the bubble. This was done in the limit of negligible convective transport of momentum and energy.

There were difficulties with the Young et al. experiment, not the least of which was the presence of a free liquid surface on the liquid bridge which experienced a temperature gradient and therefore a surface tension gradient. The temperature gradients in the liquid in the lateral direction must also have contributed to overall buoyant convection in it. Several bubbles were present at one time, and finally, the bubbles were observed through a curved liquid surface which must have introduced optical distortion.

Young et al. reported their data in the form of the temperature gradient necessary to arrest the motion of their bubbles plotted against bubble size. The data were considerably scattered, but demonstrated qualitative agreement with their theoretical model. Their experiment was later revived by Hardy (11) who built a refined apparatus at the National Bureau of Standards. Hardy eliminated most of the problems mentioned above.

Short duration (3-5 second) experiments on gas bubbles have been reported by Thompson et al. (12) who performed their study at the NASA Lewis Center's Drop Tower. The experiments, having been done in free fall, correspond to low gravity conditions. It is surprising that Thompson et al. report agreement of the migration velocity with the prediction of Young et al. for values of the Marangoni number as large as 1,200. The Marangoni number is a parameter which describes the relative importance of convective transport of energy to its molecular transport. The theory of Young et al. was developed for negligible values of the Marangoni number. We have extended the theory to accommodate moderate values of the Marangoni number, and our predictions indicate a substantial influence at these values of this parameter. Our predictions ignore inertial effects which were probably significant in the experiments of Thompson et al. The role of such effects would be to increase the deviation of the data from the predictions of Young et al. Further low gravity experiments are necessary to resolve the above discrepancy.

At Clarkson, an apparatus has been built which is modelled after the one used by Hardy. Air bubbles in the diameter range 50-200 micrometers are introduced into liquid contained in a rectangular cavity (20 x 20 mm cross-section, 5 mm high) and subjected to a vertical temperature gradient which can be as high as 10 K/mm. The refinements over Hardy's apparatus include active temperature control, the use of a nanoliter pump to inject bubbles reproducibly, and the facility to perform experiments on bubbles in the vicinity of a horizontal surface. Also, experiments are videotaped through a microscope and analyzed later on a frame-by-frame basis for bubble size and position as functions of time. Free convection velocities have been measured in the cell using a neutrally buoyant tracer to assure that such velocities are negligible compared to the bubble migration velocities.

Initially, experiments were performed on air bubbles in a Dow-Corning DC-200 series silicone oil of viscosity approximately three Poise. Data were taken on several isolated bubbles migrating in a downward temperature gradient. The bubbles, after injection, were found to grow throughout the experiment, roughly doubling in size over a two minute interval. In spite of this substantial size change, the experimental data on several bubbles were found to be consistent with predictions from the theory of Young et al. The Reynolds and Peclet numbers in these experiments were negligibly small. The growth of the bubbles is caused by the supersaturation of gas in the liquid, and will be an integral aspect of such experiments in space applications.

More recently, some preliminary experiments have been performed on air bubbles introduced in the vicinity of the bottom surface of the cell. A downward temperature gradient was used. The bubbles were found to grow as before as they migrated downward. Often they proceeded to become attached to the surface, grew further, and then became detached when sufficiently large. At this point they rose rapidly out of the field of view. The downward velocities have been observed to be substantially <u>larger</u> than those predicted by the theory of Young et al. The explanation lies in the nature of the influence of the surface on the velocity field components generated from the body force (gravity) and from the thermocapillary effect. A full quasi-steady theoretical analysis is planned and the results will be compared with the data. Additional experiments also are planned on interactions with a surface wherein the bubble moves upward.

Finally, it should be mentioned that we are continuing to develop new theoretical predictions on the subject of thermocapillary motion of drops and bubbles. An analysis of the thermocapillary motion of an isolated bubble/drop in the presence of an insoluble surfactant which forms a stagnant cap is in progress. Numerical work is under way for predicting the migration speed of a gas bubble in a vertical temperature gradient on earth under conditions of moderate Peclet number and negligible Reynolds number. The problem of predicting the migration speed of an isolated bubble in a temperature gradient under conditions of large Peclet number and no gravity is still unresolved. This is a difficult singular perturbation problem and the solution is not in sight yet. Finally, it is planned to begin posing the problem of predicting the direction and magnitude of the velocity of a bubble present in a drop subjected to an arbitrary non-uniform temperature field on its surface under conditions of negligible Reynolds and Peclet numbers. We solved the problem for axisymmetric fields in 1982, but are just beginning to consider the non-axisymmetric case.

Compound Drops

In another apparatus, drops of liquid are formed in a second immiscible liquid. While the drops are held at the tip of the injector, a gas bubble or an immiscible liquid droplet is introduced within the drop. Then the entire compound drop (drop containing droplet/bubble) is released. The resulting motion of the drop is recorded using an orthogonal pair of video cameras, and observation time is extended using a motorized platform which is moved upward at nearly the same velocity as that of the settling drop. An optical analysis has been developed to correct for the refraction effects through the drop's curved surface. Thus, from a frame by frame analysis of the videotape, suitably corrected, the bubble size and velocity (relative to the drop) can be obtained.

There are several reasons for initiating the compound drop experiments. Sadhal and Oguz (13) recently predicted that for certain ranges of parameters, the bubble velocity along a vertical drop axis can be zero. Some of these locations can be stable while others are unstable. This is an interesting prediction which can have important consequences on mass transfer to and from droplets/bubbles within a drop in various applications. Also, surface active agents (surfactants), if present, can influence the motion of the bubble within the drop. We have already observed, with certain fluid-fluid combinations, classical surfactant cap formation on the settling drops. Finally, the system also permits us to gain valuable experience on drop-bubble combinations of the types we expect to use in the flight experiments.

Bubble Migration in Rotating Fluids

A rotating fluid provides a gyrostatic pressure field which will cause less dense material such as bubbles to move inward toward the rotation axis. Experimental evidence of this fact was provided by Schrage and Perkins (14) who photographed the inward spiralling motion of gas bubbles in liquid contained within a cylinder. Our initial work on this topic was centered on confirming that this will happen in a fluid spun up from rest about a horizontal axis, and contained in a sphere. On earth, the trajectory of the bubble is influenced by gravity resulting in interesting oscillations of the radial position of the bubble as it approaches the axis of rotation. The final position of the bubble is determined by equilibrium among the forces acting on the bubble and may not be on the rotation axis, but some position off-set from it. Over the years, we have developed suitable theoretical descriptions of the trajectory of the bubble as well as its ultimate location. The predictions have generally been confirmed by the observations and the results have been reported in the open literature.

The shape of a rotating liquid drop has interested several prominent scientists since the last century. Initially studied by Plateau, the problem has been investigated analytically by Chandrasekhar (15) who studied the stability of equilibrium drop shapes. Brown and Scriven (16) have more recently constructed an exhaustive map of the stability diagram for a rotating drop using finite element methods.

Our plans are to work with relatively low rotation speeds so that in the experiments in orbit, there is sufficient time for observation of the bubble trajectories and drop and bubble shape changes. For such speeds, we do not expect complications from drop shape bifurcations. While a thorough stability analysis of the shapes of drops containing bubbles still has not been performed, the rotation speeds to be used will be an order of magnitude smaller than the speed required for liquid drops to exhibit shape bifurcation.

Serendipitously, it was observed that spin-up flows can cause a drop more dense than the surrounding fluid to migrate toward the rotation axis. After spending some time spinning at a location close to the rotation axis, the drop would migrate back toward the wall of the container. By using tracer particles to follow the shear front during spin-up, we have determined that the initial "wrong direction" motion of the more dense drops is due to the spin-up flows.

We also have performed bubble and drop migration experiments under low gravity conditions through the Marshall Space Flight Center KC-135 reduced gravity program. The bubble migration experiments provided data at low rotation rates on the shape and trajectories of large bubbles in a liquid-filled rotating sphere which were unobtainable in 1-g. Experiments on more dense liquid drops in a liquid-filled rotating sphere showed conclusively that gravity assists the process.

Because of the nature of the KC135 aircraft flight path, it was possible to conduct both sets of experiments repeatedly in alternating low-g and 1-g environments. The gas bubbles which adhered to the wall in 1-g were easily detached in low-g. The more dense drops which left the wall in 1-g adhered to the wall in low-g, and hence did not migrate at all.

Future experiments will focus on bubble and drop migration within contained rotating fluids wherein the size of the bubble/drop is not small compared to that of the container. Preliminary experiments on this subject have provided information on the shapes assumed by a bubble while it migrates toward the axis and reaches a stable equilibrium location. These shapes, determined by the complex flows within the system, are probably similar to those one might expect in the flight experiments. Another experiment currently being pursued involves a compound drop present in a more dense liquid contained in a sphere. As the sphere is spun up about a horizontal axis, the drop migrates toward the rotation axis, and upon reaching it, starts rotating. At this point, the bubble begins to spiral in toward the axis of the rotating drop. Work on this experiment has been at a preliminary stage for a long time due to wetting problems as well as problems with drop breakup. These problems are gradually being resolved.

SUMMARY

Flight experiments are planned on drops containing bubbles. The experiments involve stimulating the drop via non-uniform heating and rotation. The resulting trajectories of the bubbles as well as the shapes of the drops and bubbles will be videotaped and analyzed later frame-by-frame on ground.

Supporting ground based experiments are planned in the area of surface tension driven motion of bubbles, the behavior of compound drops settling in

an immiscible liquid and the shapes and trajectories of large bubbles and drops in a rotating liquid.

Theoretical efforts will be directed at thermocapillary migration of drops and bubbles, surfactant effects on such migration, and the behavior of compound drops.

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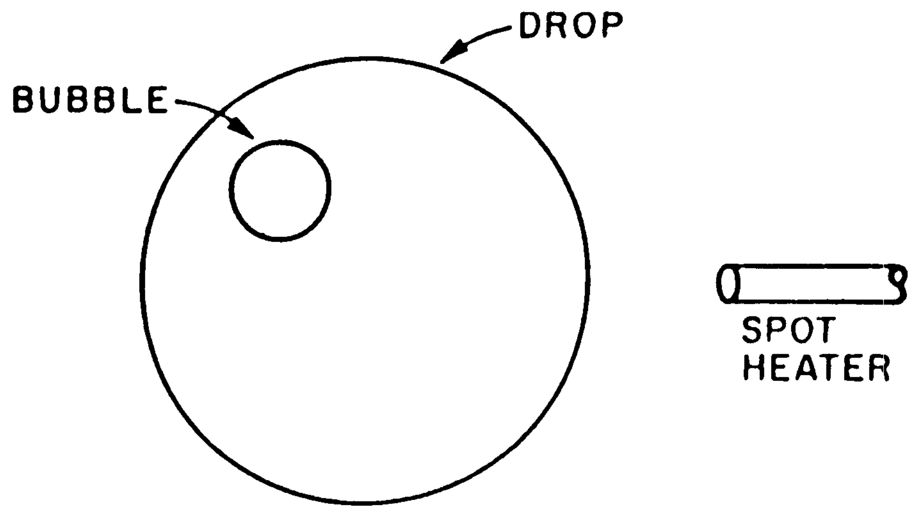
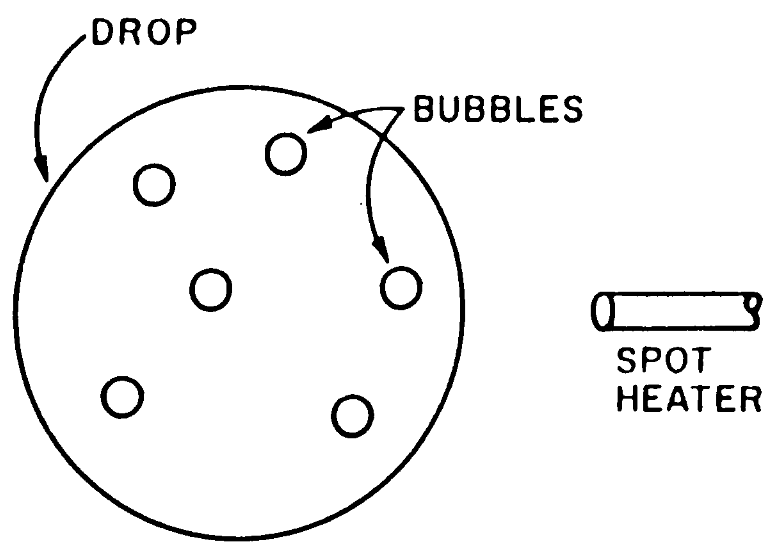


BUBBLE MIGRATION DUE TO
THERMOCAPILLARITY



CENTRIFUGAL FINING TECHNIQUE

Figure 1. Flight Experiments