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Free Expansion Study of Liquid Nitrogen

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The phenomenas involved in the free expansion of liquid/supercritical hydrogen are of great importance in many space applications. In this study, liquid nitrogen was substituted for hydrogen due to practical reasons. An experimental setup was designed to study the free expansion process when liquid/supercritical nitrogen is injected into an evacuated chamber. The effects of nozzle size, upstream pressure, and downstream pressure were studied. During the experiment, three different nozzle sizes were tested. When the pressure inside the expansion chamber was below the triple point, the nitrogen injected into the chamber solidified in two different forms. For the smallest nozzle, the solid formation occurred inside the nozzle, whereas, for the larger nozzles, the fluid solidified after being sprayed into the expansion chamber. No solidification was observed at chamber pressures higher than the triple point pressure. It was also found that supercritical nitrogen acts like a liquid in the free expansion process.

Nomenclature

C_d = coefficient of discharge
d = nozzle orifice diameter
g = gravitational acceleration
L = length
$\dot{m}$ = mass flow rate
m_{N2} = mass of liquid nitrogen
P = pressure
P_c = critical pressure
R = gas constant of nitrogen
T = temperature
t = time
U = internal energy
V = volume of the expansion chamber
ρ = density

Introduction

IN some proposed space-based weapon concepts, high-power devices will be used to provide energy for the advanced weapon systems. The thermal management of these high-power devices is an important issue. Since hydrogen in liquid or supercritical form (high pressure and low temperature) is stored as an energy source for space applications, it can be used for cooling the high-power devices prior to being injected into the rocket engine. In a recent study on the thermal management of space-based high-power microwave tubes, one key conclusion was that direct cooling with the onboard liquid hydrogen was the best choice.[1] Similarly, onboard liquid hydrogen can be used for thermal management of space-based, high-power radio frequency (rf) sources.

A large plumbing system will have to be built to transport the liquid/supercritical hydrogen from its storage to the high-power devices. In case of space-based weapon systems, this manifold feeder pipe system will initially be under room temperature and vacuum. When the weapon system needs to be deployed, it has to be ready in as short a time period as possible. Therefore, the liquid/supercritical hydrogen has to be transported to the high-power devices as quickly as possible. Thus, the liquid/supercritical hydrogen undergoes the free expansion process in the plumbing system at the startup. The thermodynamic process of the free expansion, as well as the hydrogen mass flow rate in such a situation, has to be determined. Also, the possibility of solid hydrogen formation in the plumbing system is of great concern since the solid may block the supply of hydrogen to the devices.

Recognizing the need for an understanding of the thermal and fluid transients during the initial blowdown (startup) of liquid/supercritical hydrogen, this study tries to provide answers to the above concerns. Since the thermophysical phenomenas occurring during the free expansion process are varied and complex, it is necessary to study such phenomena experimentally. For safety reasons, liquid nitrogen is used instead of liquid hydrogen throughout this study. Although liquid hydrogen is used as the fuel in rockets and missiles, the use of liquid hydrogen could pose explosion and fire risks in a laboratory situation. It is highly flammable and explosive, and therefore, is extremely dangerous if not handled properly. Thus, to deal with hydrogen, without question, requires ultrasafe facilities. The facilities must be leakproof under all operational conditions. Elaborate ventilation systems, emergency exits, etc., have to be provided. The above precautionary measures are prohibitively expensive. Hence, it is preferable to use a suitable substitute fluid that is safer to deal with. Liquid nitrogen is a cryogen whose saturation temperature (77.3 K) is close to the saturation temperature of liquid hydrogen (22 K). Furthermore, the properties of nitrogen, such as viscosity ($\nu_{H2} = 1.9 \times 10^{-7}$ m^2/s, $\nu_{N2} = 1.1 \times 10^{-7}$ m^2/s at critical point) and surface tension ($\sigma_{H2} = 1.8 \times 10^{-3}$ N/m, $\sigma_{N2} = 6.1 \times 10^{-3}$ N/m, saturated state at 10% of its critical pressure), are closest to hydrogen among all cryogens. The reason for choosing the surface tension data at pressure equal to 10% of the critical pressure is due to availability of the surface tension data. Also, liquid nitrogen is much safer to work with. Therefore, in this study, liquid nitrogen is used instead of liquid hydrogen as the working fluid.

Experimental Design and Setup

The experiment was designed to study the free expansion of liquid/supercritical nitrogen injected into a pipe initially at

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vacuum. The phenomenas of interest are those occurring when the pressure inside the pipe is very low. The research focused on flow visualization and flow characterization by temperature and pressure measurements.

Since the final pressure of the free expansion process is below the triple point pressure (12.5 kPa), solid nitrogen formation is expected during the free expansion. Also, when nitrogen is near the triple-point region, a multiphase flow (a liquid–gas, a solid–gas, or a liquid–solid–gas mixture) will be seen in the expansion chamber. The critical pressure of nitrogen is 3.39 MPa. A large pressure drop from around critical point to below triple point takes place during the free expansion process. Thus, the solid nitrogen will break down and form very small droplets. The experimental setup was designed to allow the observation of these phenomena.

The free expansion process including the final state has to be thermodynamically determined. The most important parameters to be measured are the pressure and the temperature. One can assume that the liquid nitrogen injector and the expansion chamber form a closed system. The pressure in the expansion chamber will be near vacuum. Hence, conduction and convection heat transfer to the fluid injected into the chamber can be ignored. Radiation heat gain is estimated to be very insignificant when considering the large latent heat and moderate mass flow rate of nitrogen. Hence, the internal energy is constant during the free expansion process. Therefore, if the upstream temperature and pressure are measured, the internal energy of the initial state can be determined. Then, on the temperature-internal energy diagram, the final thermodynamic state inside the chamber can be determined, provided that the downstream temperature or pressure is measured. With these requirements in mind, a suitable experimental setup was designed.

Figure 1 is a schematic view of the experimental setup. A borosilicate glass cylinder made up the expansion chamber. This allowed clear visualization of the free expansion process. Borosilicate glass was chosen for its high strength and small thermal expansion coefficient. The expansion chamber was connected to a vacuum pump at one end and to the nitrogen injector at the other. The diameter of the chamber was chosen to be 203.2 mm (8 in.), this results in an expansion ratio of at least 500 (based on the diameter ratio of the chamber and the largest nozzle, 0.36 mm). A stainless steel flange system connected the injector–nozzle assembly to the expansion chamber. The injector was surrounded by a cooling chamber filled with liquid nitrogen in order to reduce the heat leakage into the fluid being injected. The size of the innermost cylinder (which carries the high pressure fluid) was 25.4 mm in diameter and 152.4 mm in length. One end of this cylinder leads to the nozzle while the other is connected to a small vertical cylinder, which in turn is connected to the high-pressure gas supply. The fluid was injected into the chamber by first filling the injector assembly with liquid nitrogen, then, allowing the high-pressure gas to pressurize the fluid, and finally, opening the valve leading to the nozzle.

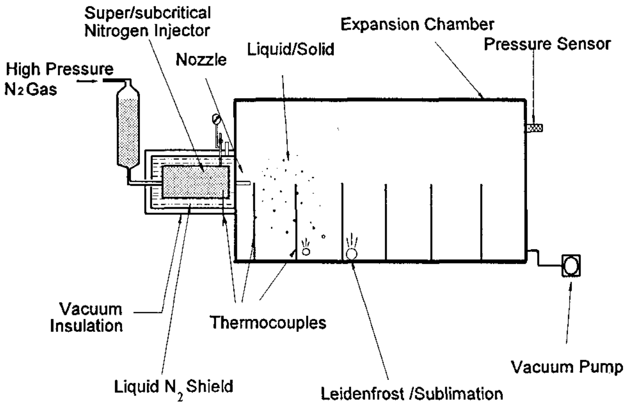


Fig. 1 Schematic of experiment arrangement.

All parts, except the glass chamber, were made of stainless steel. Indium gaskets and polytetrafluoroethylene (PTFE) jacketed spring-energized O rings provided seals between various parts of the injector assembly. The vacuum pump was connected to the opposite side of the expansion chamber via a quick-connect flange arrangement. This flange could be removed to allow the changing of the nozzle without disassembling the injector from the chamber. The pressure of the expansion chamber was read by a convectron gauge system with a range of 0.133 Pa to 133.3 kPa.

The purpose of this study was to investigate the phenomena at the startup for a long plumbing system initially at vacuum. Thus, the main region of interest was the period when the pressure inside the plumbing system is much lower than the triple point pressure of the fluid. Some experiments were also conducted with chamber closed-off after the initial evacuation. This allowed the chamber pressure to rise as the fluid being injected into the chamber evaporated.

Thermocouples were used to measure all the temperatures. The temperature of nitrogen inside the injector was expected to be between 78–130 K. Inside the glass chamber, the temperatures were expected to range from 50 to 300 K. For this range, E-type thermocouples were selected due to their high Seebeck coefficients. Since the experiment dealt with a transient process, the response time of the thermocouples had to be as short as possible. In other words, their thermal inertia had to be minimized. However, the strength of wire was also of concern since a very thin wire would be liable to break in the high-speed flows expected in this study. The diameter of the thermocouple wire was chosen as 0.0762 mm (0.003 in.); this provided a good compromise between strength and low mass. The thermocouples were formed by inert-gas arc-welding of the component wires. The resulting thermocouple bead was filed such that the thermocouple junction finally looked like a butt-weld.

Inside the chamber, six thermocouples were arranged axially in a row. They were positioned such that the thermocouple junctions were in the trail of the fluid exiting the nozzle. The first thermocouple was only 1 mm from the nozzle. The second one was 3 mm from the first thermocouple. From thermocouple no. 3 to no. 6, each was placed 5 mm further from the previous one. A pair of thermocouples was located inside the liquid nitrogen injector to measure the upstream temperatures. One was just prior to the nozzle inlet, and the other further upstream.

A special cryogenic probe (silicon diode, manufactured, and precalibrated by LakeShore Inc.) was used to calibrate all the thermocouples prior to their installation. The silicon–diode probe had excellent repeatability: ±10 mK at 4.2 K over repeated thermal cycling; and high accuracy: ±0.5 K in the 2.0–100 K range.

During the experiments, all measurements were taken by a data acquisition system and stored on a computer. Three different sizes of plain-orifice nozzles were tested. The orifice diameters for the nozzles were 0.18, 0.25, and 0.36 mm. Experiments were performed with upstream pressures of 0.7, 1.4, 2.1, 2.8, and 3.5 MPa (gauge pressure). The nitrogen was in supercritical state for the last case, and in liquid state for all the other pressures. During the start of each experiment, the pressure in the expansion chamber was about 133.3 Pa (1 mmHg). When the vacuum inside the chamber was continuously maintained, this pressure never rose above the 9 kPa (the triple point pressure of nitrogen is 12.5 kPa).

The mass flow rate could not be measured directly since the flow was multiphase. From the experiments with the closed chamber, it was observed that the pressure inside the chamber was below the triple point pressure for at least the first 5 s. During the experiments, the upstream pressure was always kept constant. Compared to the pressure difference across the nozzle (780 kPa at least), the variation in the downstream vacuum was very small. Therefore, the pressure difference across the nozzle can be considered constant for this first 5 s. Therefore, steady flow can be assumed. Thus, if the mass inside the chamber at the end of this time can be determined, the average mass flow rate can be computed.

To measure the mass flow rate, the expansion chamber was evacuated and the valve connecting the chamber to the vacuum pump closed. Any leakage into the expansion chamber was recorded. The initial pressure and temperatures inside the expansion chamber were recorded. The nozzle valve was then opened for t s (usually 5 s) to inject the liquid/supercritical nitrogen into the chamber. When steady state was reached inside the chamber, the pressure and temperatures were again recorded. Assuming the gaseous nitrogen occupying the chamber to be ideal, the equation of state can be applied to determine the average mass flow rate:

$$\dot{m} = m_{N2}/t = PV/tRT \qquad (1)$$

Results and Discussion

The free expansion phenomena inside the chamber were observed during the experiments. When the pressure inside the glass chamber was above the triple point (i.e., the pressure was higher than 12.5 kPa), the fluid sprayed out of the nozzle appeared to be a liquid–gas mixture (mist). No solid nitrogen could be observed anywhere in the chamber. The mist flow vaporized very quickly inside the chamber. Typically, the length of spray cone was less than 30 mm, i.e., the liquid drops vaporized completely within 30 mm of nozzle exit. The temperature recorded by the first thermocouple in the spray trail (the one closest to nozzle exit) was usually about 5 K higher than the saturation temperature (at the chamber pressure). This was probably due to the inability of the mist flow to cool the thermocouple junction down to the mist temperature during the short duration of the experiment.

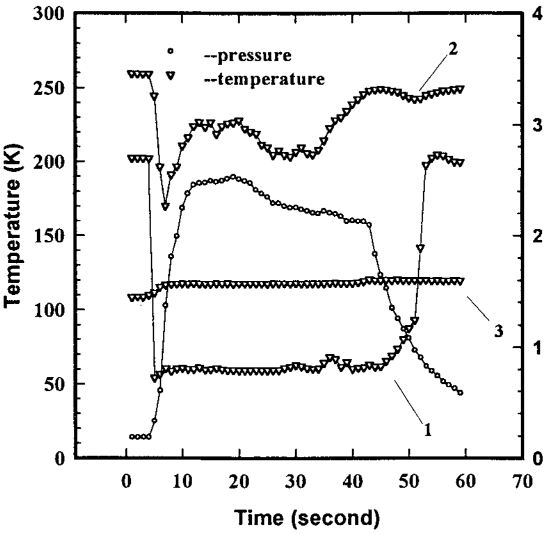
At chamber pressures lower than the triple point, the phenomena depended on the nozzle size. The following paragraphs describe the phenomena when the chamber pressure was maintained below triple point by continuous evacuation of the chamber.

The smallest nozzle (orifice diameter 0.18 mm) was tested first. When the injector valve was opened, solid nitrogen appeared immediately at the nozzle orifice. The solid appeared somewhat porous. It was very clear that the solid nitrogen was forming inside the nozzle since the solid nitrogen came out of the nozzle very slowly. The phenomenon was similar to toothpaste being squeezed out of a toothpaste tube. The solid nitrogen accumulated around the nozzle opening, increasing in size as the experiment progressed, and enveloping the first thermocouple. When the upstream pressure was low, i.e., 0.7 or 1.4 MPa, the solid block could grow up to 25 mm in length and 20 mm in diameter.

The temperature of the solid was below the triple point temperature (63 K), the pressure inside the chamber rose to a certain level and remained stable because the solid block could not melt or sublimate quickly. When the solid block grew so big that it could not be supported by the nozzle and the first thermocouple, it fell down to the bottom of the chamber. The temperature of the chamber wall was close to room temperature. Hence, the block of solid nitrogen floated on the wall due to the formation of a vapor layer between the solid nitrogen and the wall. A new solid block began to form at the nozzle orifice after the previous block had dropped off.

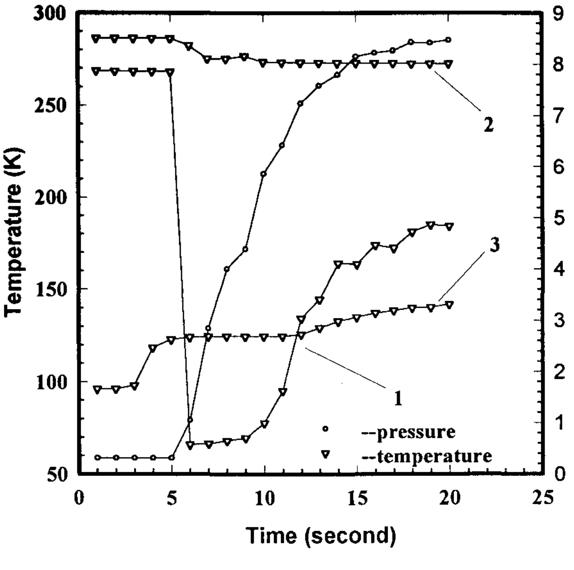
When the upstream pressure was increased, the solid forming process seemed to accelerate; a larger amount of solid nitrogen was pushed out of the nozzle. But since the pushing force was larger, the solid block could not grow to the same size before it was pushed away. The size of the blocks became smaller while the frequency of generation of the solid blocks became higher. This caused the pressure in the chamber to increase faster. The upstream pressure did not have a strong impact upon the solid formation process, it only affected the rate of solid ejection that was proportional to the upstream pressure.

The sublimation/melting process was very slow. The solid block could stay at the bottom of the chamber for a long time, usually until the experiment was ended. If the block did not fall to the bottom and touch the superheated wall, it would remain in solid form for a few minutes. Figures 2 and 3 show the temperature and pressure transients in the expansion chamber for upstream pressures of 2.1 and 3.5 MPa, respectively. In both figures, the opening of the injection valve is marked by very dramatic temperature drops accompanied by



nozzle: d = 0.18 mm
upstream pressure: 2.1MPa
1-first thermocouple in expansion chamber
2-second thermocouple in expansion chamber
3-temperature in injector

Fig. 2 Temperatures and pressures inside the expansion chamber.



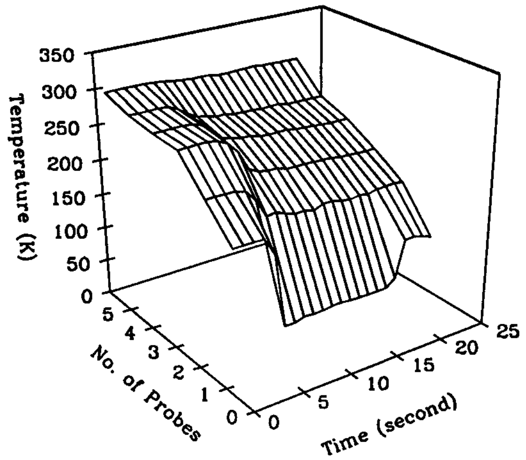
nozzle: d = 0.18 mm
upstram pressure: 3.4MPa
1--first thermocouple in expansion chamber
2--second thermocouple in expansion chamber
3--temperature in injector

Fig. 3 Temperatures and pressures inside the expansion chamber.

a pressure increase. In Fig. 2, at 45 s, the sudden pressure drop follows the closing of the nozzle valve (since the vacuum pump was always on). The trend of the temperature recovering in the expansion chamber can also be seen. Figures 4 and 5 show the transient temperature field for the same two cases. For the thermocouples further away from the nozzle, the temperature drops are not as dramatic as those closer to the nozzle. Also, there is a delay on the temperature drop for those further away.

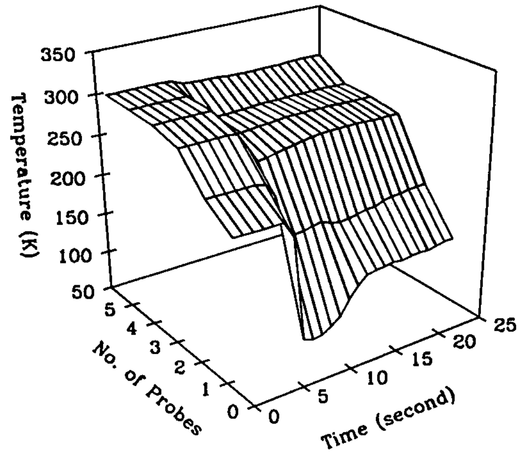
Experiments with the 0.25-mm-diam nozzle were performed next. The solid formation phenomenon was very different in this case. The solid nitrogen would form around the first thermocouple. No solid formation could be seen near the nozzle orifice. The spray from the nozzle could be seen very clearly. Once the nitrogen droplets touched the first thermocouple, solid accreted around the thermocouple. While the spray process went on, the solid block became bigger, but it could never reach the size of that formed around the small nozzle. When the pressure inside the injector was 2.1 MPa, the solid block was regularly blown away by the spray. Since the block was small, it melted into liquid droplets once it touched the bottom of the chamber. The droplets floated on the wall due to the Leidenfrost phenomenon.

Finally, the 0.36-mm-diam nozzle was tested in the system. In this case, the spray was much stronger. The solid nitrogen hardly accreted on the first thermocouple before it was blown away. The solid block could grow to about 1 mm at the maximum. The thermodynamic parameters such as temperatures and pressures in the chamber were still similar to those of the other two nozzles.



Nozzle: d=0.18 mm
Pressure in Injector: 2.1MPa

Fig. 4 Temperature field in the expansion chamber.



Nozzle: d=0.18 mm
Pressure in Injector: 3.4MPa

Fig. 5 Temperature field in the expansion chamber.

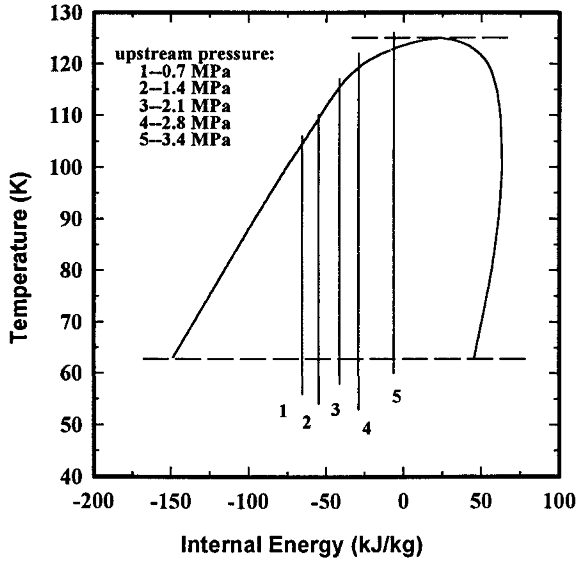


Fig. 6 Temperature vs internal energy results.

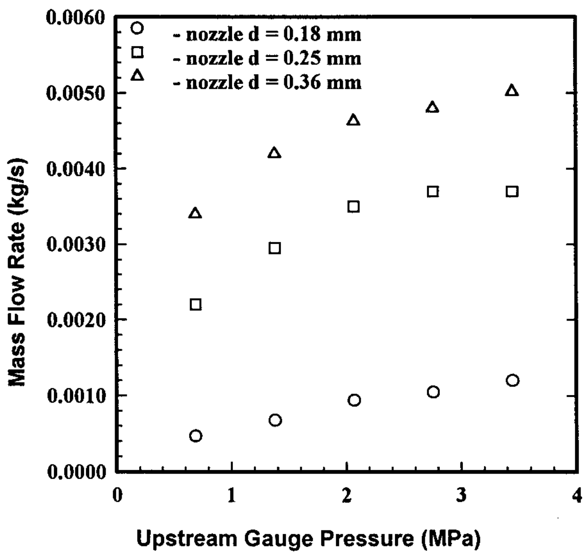


Fig. 7 Mass flow rate results.

As mentioned earlier, final state of the free expansion process can be determined on a temperature-internal energy diagram. As the temperature and pressure in the injector were obtained, the initial internal energy could be determined. Since the blowdown process was a constant internal energy process, as long as the temperature of the free expansion final state was measured, the final thermodynamic state could be located on the temperature vs internal energy diagram. With increasing upstream pressure, the internal energy of the liquid nitrogen increases as well, therefore, the constant internal energy line shifts to the right as the pressure goes up. The experimental results on the temperature-internal energy diagram are shown in Fig. 6.

The result of mass flow rate is presented in Fig. 7. For the purpose of comparing the nozzles, the coefficient of discharge can be obtained based on Eq. (2) (Ref. 2):

$$\dot{m} = C_d(\pi d^2/4)\sqrt{2\rho(\Delta P)} \qquad (2)$$

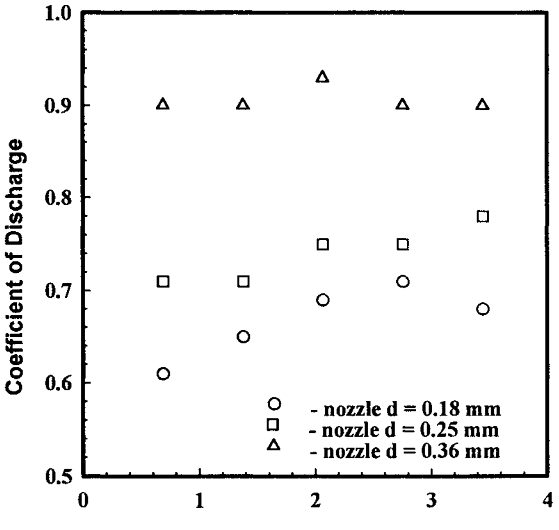


Fig. 8 Coefficients of discharge of the nozzle.

where ρ is the density of nitrogen and ΔP is the pressure difference across the nozzle. The C_d values for the three nozzles at different upstream pressures are shown in Fig. 8. The coefficient for the smallest nozzle ($d = 0.18$ mm) fluctuated since there was solid nitrogen coming through the nozzle.

From the experiments, it can be concluded that solid nitrogen formation never occurs if the pressure in the expansion chamber is higher than the triple point pressure of nitrogen, irrespective of the nozzle geometry and the upstream pressure. When the pressure inside the free expansion chamber was below 12.5 kPa, liquid nitrogen had the potential of forming solid nitrogen. The freezing process is affected by several parameters such as ambient temperature, droplet diameters, and nozzle geometry. The most important factor that affected the phenomenon of solid nitrogen formation in the experiment was the nozzle geometry. When the length of the nozzle is more than seven times the diameter, the nozzle can be termed a tube nozzle.[3] For the smallest nozzle in this study, the orifice diameter was 0.18 mm, whereas the orifice length was 1.4 mm. Thus, this nozzle was a tube nozzle. In case of a tube nozzle, the flow boundary layer fully develops inside the nozzle, and thus, the triple point pressure would occur somewhere inside the nozzle. Therefore, solid nitrogen would form inside the nozzle and be pushed out of the nozzle by the upstream pressure. The solid nitrogen formed in this case is called rime solid nitrogen.[3] Rime solid nitrogen, as seen from experiments, is a milky white, porous solid.

The nozzles with diameters of 0.25 and 0.36 mm do not meet the criteria for a tube nozzle. Therefore, the flow boundary layer could not be fully developed inside the nozzle and the resulting fluid pressure could not drop to the triple point before it exited the nozzle. When liquid was sprayed into the glass chamber, it went through a process of sudden exposure to the pressure below the triple point. The liquid nitrogen droplets would form tiny solid nitrogen droplets very quickly. But the liquid and solid nitrogen droplets could not be distinguished visually since they were too small. However, the spray was in fact made of solid nitrogen droplets. This could be seen from the solid nitrogen accretion on the first thermocouple.

The problem of freezing of liquid nitrogen upon a sudden exposure to vacuum has been studied in the past.[4] The icing time, i.e., the time it takes for a liquid drop to solidify, may vary slightly depending upon the downstream vacuum pressure. Another study[4] showed that there existed a maximum stable drop diameter for liquid nitrogen (2.7 μm when nitrogen at 77.4 K was subjected to sudden expansion). A temperature gradient within the droplet is established by rapid freezing of the surface. The solid/liquid interface moves toward the center of the drop until the sphere becomes completely solid. The mathematical model dealing with the above situation is described in that study.[4] It shows that for liquid nitrogen freezing at the triple point, the time required is less than 10^{-5} s. Accordingly, at a spray velocity of 100 m/s, the drops would freeze within 1 mm from the nozzle orifice (based on the energy equation, the maximum spray velocity in this study is expected to be less than 100 m/s). Thus, we can conclude that for the larger nozzles, tiny solid droplets were sprayed onto the thermocouple and accreted there.

Error Analysis

A vacuum gauge controller (VGC) with a convectron gauge was used to measure the pressure in the expansion chamber. The gauge range is 0.133–133.3 kPa. The accuracy is 3% of the reading. A Bourdon type gauge was used to measure the upstream pressure. The range of this gauge was 0–13.79 MPa, and the accuracy ±34.4 kPa.

The temperature measurements were carried out using E-type (nickel–chromium vs copper–nickel) thermocouples. The accuracy of this type of thermocouple is the larger of 1.0% or 1.7 K in the range of 30–300 K. The silicon diode probe used to calibrate the thermocouple has an accuracy of ±0.5 K in the range of 2–100 K. After calibration, the accuracy of the thermocouple is approximately ±1 K.

The uncertainty of internal energy calculation is mainly due to the errors of temperature and pressure measurement. The maximum uncertainty is less than 6%. The uncertainty in mass flow rate results is less than 6%. Finally, the uncertainty in the calculated coefficient of discharge is about 8%.

Conclusions

An experiment was designed to perform free expansion study with liquid/supercritical nitrogen. The experimental setup allowed for flow visualization and measurement of the temperature and pressure inside the expansion chamber.

It was found that the nitrogen solidified during free expansion only when the downstream pressure was lower than the triple point pressure. Depending on the nozzle size, the solid formation process fell into two categories. For the smallest nozzle, the solid formation occurred before the nozzle exit, whereas, for the larger nozzles the fluid solidified after being sprayed into the expansion chamber. It is clear that the nozzle geometry is an important factor in solid formation process. The smaller nozzle gets blocked easily, which could be detrimental to the system. The initial state of the fluid (liquid or supercritical) did not seem to have any impact on the solid formation process. Thus, we can say that the free expansion behavior of supercritical nitrogen is no different than that of liquid nitrogen. The final thermodynamic state of the free expansion process was determined based on the temperature vs internal energy diagram.

It is recommended that further studies be conducted to study the effect of nozzle size on free expansion process. Further increase or decrease of the nozzle diameter from this experiment is suggested. The maximum upstream pressure in this study was 3.5 MPa, i.e., the nitrogen was just within the supercritical region. In space applications, the hydrogen storage pressure could be as high as two times the critical pressure. Therefore, higher pressure experiments are recommended. Finally, although nitrogen is the best substitute for hydrogen, their properties are obviously not similar. Thus, it is recommended that experiments with hydrogen should be conducted.

Acknowledgments

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