Integrated refrigeration and storage of LNG for compositional stability

L Rose¹, A M Swanger¹, W U Notardonato¹, J E Fesmire¹, J Gleeson², and R Carro²

¹ NASA Kennedy Space Center, Cryogenics Test Laboratory, UB-R1, KSC, FL 32899
² Bionetics Corp. at NASA Kennedy Space Center, KSC, FL 32899 USA

E-mail: laura.p.rose@nasa.gov

Abstract. Growing interest in liquefied natural gas (LNG) as a rocket fuel necessitates a greater technical understanding of the compositional changes due to preferential boil-off (or weathering) that occurs during long duration storage. The purity of methane in LNG can range from 90 to 98%, and is subject to preferential boil-off due to its low boiling point compared to other constituents despite the use of high-performance thermal insulation systems. Active heat extraction (i.e. refrigeration) is required to completely eliminate weathering. For future operational safety and reliability, and to better understand the quality and efficiency of the LNG as a cryofuel, a 400-liter Cryostat vessel was designed and constructed to measure the composition and temperatures of the LNG at a number of different liquid levels over long durations. The vessel is the centerpiece of a custom-designed lab-scale integrated refrigeration and storage (IRaS) system employing a pulse tube cryocooler capable of roughly 50 W of lift at 100 K. Instrumentation includes ten temperature sensors mounted on a vertical rake and five liquid sample tubes corresponding to five liquid levels. Two modes of operation are studied. The first is without refrigeration in order to determine a baseline in the change in composition, and to study stratification of the LNG. The second is performed with the cryocooler active to determine the operational parameters of the IRaS system for eliminating the weathering as well as stratification effects in the bulk liquid. The apparatus design and test method, as well as preliminary test results are presented in this paper. As a bonus in cost-saving and operational efficiency, the capability of the IRaS system to provide zero-loss capabilities such as zero boil-off (ZBO) keeping of the LNG and zero-loss filling/transfer operations are also discussed.

1. Introduction

The plans for a number of future launch vehicle propulsion systems include the use of liquid oxygen (LOX) and liquefied natural gas (LNG) cryofuels [1-3]. The use of LNG rather than liquid methane (LCH₄) as a rocket fuel necessitates a greater technical understanding of the compositional changes due to preferential boil-off (or weathering) that occurs during long duration storage. The purity of methane in LNG can range from 90 to 98%, and is subject to preferential boil-off due to methane’s low boiling point compared to other constituents despite the use of high-performance thermal insulation systems. Active heat extraction (i.e. refrigeration) is required to completely eliminate weathering [4].

The technology of integrated refrigeration and storage (IRaS) can provide key benefits for both operational efficiency and flight performance in the area of LOX and LNG storage and transfer systems. Operational efficiency benefits could include long-term storage with consistent composition of the

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cryofuels through the zero boiloff mode of operation. Flight performance benefits include simplified vehicle loading (providing the enthalpy margin for heat gain) and increased energy density through the densification mode of operation [5]. The IRaS technology offers full control over the state of the cryofuel using the automatically managed addition and removal of heat (energy). Conventionally, the liquid evaporates to the point where the relief valve opens, or the tank is vented, which is inherently limited and provides minimal “control” of the system, but no control whatsoever of the state of the fluid.

Although LNG (or even LCH₄) has never been adopted as a rocket fuel before, it is now becoming a popular choice as it is highly efficient and widely available at a low cost. As a fuel, LNG has clean combustion characteristics to improve engine refurbishment and reusability. And unlike kerosene, LNG can be used to self-pressurize the tank’s ullage, thus eliminating the demand for the Earth’s limited and expensive helium supply. For future operational safety and reliability, and to better understand the quality, efficiency, and performance potential of LNG as a cryofuel, a laboratory research program has been initiated by the Cryogenics Test Laboratory at NASA Kennedy Space Center (KSC).

2. Experimental approach and apparatus
The experimental platform of this research is a cryostat apparatus with an integrated cryocooler. The system was designed and constructed to measure the composition and temperatures of the LNG at different locations within the bulk liquid. The experimental plan and purpose of this study are centered around three main objectives: 1) How is the mixture composition and temperature changing as a function of time and liquid level while in a steady state of boil-off, as compared to in a steady state of zero boil-off?; 2) What is the feasibility and cost-effectiveness of long-term storage using active refrigeration (zero boil-off)?; 3) What is the feasibility and benefit of densified LNG? The in-situ liquefaction of natural gas and/or methane can also be performed. The mitigation of adverse weathering effects is also a prime area of investigation.

The primary equipment consists of a 400-liter vertical vacuum-jacketed cylindrical vessel (cryostat) with an integrated pulse tube cryocooler providing approximately 50 W of heat rejection at 100 K (Cryomech PT30). The IRaS technology is implemented by the custom designed interface; a cryogenic refrigerator to a cryogenic storage tank via an internal heat exchanger. The Cryostat is shown in figure 1.

![Figure 1. Cryostat with 400-liter vessel and integrated cryocooler for LNG studies.](image-url)
The connecting flange of the cryostat vessel was designed for minimal heat leak. The primary design consideration was the cryocooler cold head extension into the bulk liquid. A new IRaS heat exchanger was designed to maintain steady state zero boiloff (ZBO) with this cold head.

Two modes of operation are addressed in this study. The first mode is without refrigeration in order to determine a baseline in the change in composition, and also to study stratification of the LNG by long-duration boiloff testing. The second mode is performed with the cryocooler active to determine the operational parameters of the IRaS system for eliminating the weathering as well as stratification effects in the bulk liquid (ZBO test).

The cryostat includes a vertical rake located along the centerline of the tank. Instrumentation includes ten RTD temperature sensors mounted on the vertical rake, two RTDs on the cold head, and five liquid 1.6-mm diameter sample tubes corresponding to five liquid levels. During testing, the tank’s ullage is monitored for pressure with both a transducer and analog gauge. The details of the temperature and pressure instrumentation are provided in table 1 and table 2, respectively.

| RTDs  | Location                        |
|-------|---------------------------------|
| T1    | 0% (bottom of the tank)         |
| T2    | 25%                             |
| T3    | 50%                             |
| T4    | 75%                             |
| T5    | 100%                            |
| T6    | Cold head                       |

| Instrumentation | Pressure range |
|-----------------|----------------|
| Pressure transducer | 0-150 psig    |
| Pressure gauge       | 0-200 psig     |

During the test, the composition is measured two different ways. One way is with the Precisive 5-283 Gas Analyzer from MKS. This infrared/optical analyzer is capable of measuring the gases in Table 3 [6]. The other method of sampling is a mass spectrometer configured in-house at NASA/KSC (see Table 4 for the gases this mass spectrometer will sample). Using the gas analyzer in conjunction with the mass spectrometer allows for the differentiation of the CO\textsubscript{2} under the propane signal that cannot be seen using the mass spectrometer (because a mass spectrometer cannot differentiate CO\textsubscript{2} and propane).

| Gases measured | Calibration Concentration | Accuracy |
|----------------|---------------------------|----------|
| Methane        | 50-100%                   | +/- 0.3% |
| Ethane         | 0-20%                     | +/- 0.2% |
| Propane        | 0-10%                     | +/- 0.2% |
| Iso-Butane     | 0-5%                      | +/- 0.1% |
| n-Butane       | 0-5%                      | +/- 0.2% |
| Pentanes       | 0-2%                      | +/- 0.1% |
| CO\textsubscript{2} | 0-20%                   | +/- 0.2% |

Table 3. Gas Analyzer gases measured with calibration and accuracy.
Table 4. Mass spectrometer gases measured, with calibration and accuracy for each.

| Gases measured | Calibration Concentration | Accuracy relative to the measurement |
|----------------|---------------------------|--------------------------------------|
| Methane        | 54-91%                    | 10%                                  |
| Ethane         | 5-25%                     | 15%                                  |
| Propane        | 5-12%                     | 15%                                  |
| n-Butane       | 1-7%                      | 15%                                  |
| n-Pentane      | 1-2%                      | 15%                                  |

Each of the five sampling stations and a portion of the vent flow is connected to a sampling manifold mounted outside of the Cryostat. Each of these six inlets to the sampling manifold has an isolation valve to allow controlled sampling of each station.

A back pressure regulator is set to approximately 70 kPa (10 psig) and used to maintain constant pressure in the cryostat and to provide a slight backpressure to aid in the sampling out of the 1.6-mm diameter sampling ports (because the gas analyzer does not include a vacuum pump). Full details of the test setup are given by the schematic in figure 2. A cutaway of the cryostat apparatus is given in figure 3.

![Figure 2. Test schematic of LNG cryostat apparatus.](image-url)
3. Experimental test plan and initial results

Due to the flammable nature of LNG and to protect for an over pressurization event, the system was designed for worst case heat flux due to loss of vacuum [7].

There are two ways for the cryostat to vent. Under normal operations (for the boiloff test), the vent flow will utilize the boiloff vent stack which is downstream of the vent valve and the 70 kPa back-pressure regulator. The other vent was designed for emergency flow which is downstream of a 793 kPa (115 psig) relief valve sized for a loss-of-vacuum event.

The cryostat is situated outdoors under a canopy to allow for venting to the atmosphere while protecting the cryostat, equipment, and sensors. There is a single liquid fill/drain line to the bottom of the inner vessel and a pressurization/vent port connected to the ullage region. This fill/drain port allows the cryostat to be serviced with LNG from a mobile trailer.

Prior to performing the LNG boiloff test and zero boiloff test, a liquid nitrogen (LN$_2$) boiloff test is performed. The cryostat is filled with LN$_2$ and allowed to boil off and vent to atmosphere. During this test the mass flow rate from the vent port is measured as well as the barometric pressure and liquid level for consistent results. This baseline test provides a good approximation for the heat leak of the cryostat’s thermal insulating performance [8]. The heat leak for the cryostat with LN$_2$ for the near-full condition is approximately 21.5 W as shown in figure 4.

![Figure 3. Cutaway view of LNG cryostat apparatus.](image)

![Figure 4. Heat leak of cryostat from the LN$_2$ boiloff test.](image)
The LNG test begins by filling the cryostat and line with LN$_2$ in order to pre-cool to 77 K. Following the cooldown, the LN$_2$ is vented/drain; before the cryostat temperature reaches 100 K, the cryostat is filled with LNG. During the boiloff test, the vent is left open and the test is allowed to continue until all the LNG has evaporated.

Following the completion of the boiloff test, the vent is closed and the cryostat is filled with LNG again (the Cryostat must again be below 100 K). By turning the heater controls on/off, the cryocooler is adjusted so that the heat extraction matches the heat leak, without allowing the pressure to drop below atmospheric pressure. The ZBO condition is maintained for two weeks after which the cryocooler is then shut off and the vent valve opened to allow the LNG to evaporate.

During the boiloff test, the output of the vent is continuously analyzed with the mass spectrometer (not including the ZBO test when the vent will be closed). Three times a week, for both the boiloff and the ZBO tests, each of the five liquid levels is sampled one at a time. During sampling, the vent is isolated from the sampling manifold.

The gas analyzer is used once a week to also sample the gas. One at a time, each of five liquid levels and the vent (only during the boiloff test) will be connected to the gas analyzer via the sampling manifold to record the composition of the gas at each of these stations.

4. Discussion and future experimental investigations

At this time, the cryostat apparatus is built and in place. All instrumentation and equipment is installed and the gas analyzer, RTDs, and pressure transducer are being connected to the DAQ for data recording. Upon completion of this final setup, the LNG will be delivered and the first test (boiloff) will begin.

One of the challenges to this experimental apparatus was finding a correctly sized cryocooler. The original cryocooler was capable of roughly 300 W of lift at 100 K. With the LN$_2$ baseline test performed and the heat leak found to be <22 W, this cryocooler was determined to be too large, resulting in the search for the new cryocooler (50 W of lift at 100 K).

Other challenges were the sampling method to be used. Originally, methane, ethane, propane, nitrogen, CO$_2$, and argon were the desired compounds to be measured. However, the gas analyzer does not measure nitrogen or argon and the mass spectrometer cannot differentiate CO$_2$ from propane. It is suspected that nitrogen and argon may evaporate first, so they were not chosen to be sampled despite the fact that, per the military specification for grade-A methane propellant, the fluid can have a maximum of 5000 ppmV of nitrogen and 5000 ppmV of other gaseous impurities (argon is among these gaseous impurities)[9].

A future challenge that may arise, is the accuracy of the sampling as the methane preferentially evaporates and the percentage of other compounds in the cryostat become larger than the calibrated range of the mass spectrometer or gas analyzer.

5. Conclusion

For future operational safety and reliability, and to better understand the quality and efficiency of the LNG as a cryofuel, a 400-liter cryostat vessel with integrated cryocooler was designed and constructed to measure the composition and temperatures of the LNG at a number of different liquid levels over long durations. LNG is a cryofuel rocket fuel of the future as evidenced by the move to developing LOX/LNG based rocket engines. Learning and characterizing the composition changes, weathering, and stratification are therefore of basic and fundamental importance. Also important is providing an experimental basis for the future performance enabling capability of rocket vehicles and other enhancements afforded by cooling LNG below its normal boiling point to produce densified LNG or slush LNG.

6. References

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