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Correlation of Helium Solubility in Liquid Nitrogen

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Abstract

A correlation has been developed for the equilibrium mole fraction of soluble gaseous helium in liquid nitrogen as a function of temperature and pressure. Experimental solubility data was compiled and provided by National Institute of Standards and Technology (NIST). Data from six sources was used to develop a correlation within the range of 0.5 to 9.9 MPa and 72.0 to 119.6 K. The relative standard deviation of the correlation is 6.9 percent.

Introduction

Although liquid nitrogen (LN_2) is not a propellant, it is often used for ground-based testing of cryogenic propulsion systems. It is a safer and less costly alternative test fluid for liquid oxygen (LO_2) or liquid methane (LCH_4). Often LN_2 is substituted for other cryogenic propellants in the early stages of testing, and it is typically used in preparing test hardware for follow-on testing with the actual propellants of interest, such as LO_2, LCH_4, or liquid hydrogen (LH_2). Since LN_2 systems pressurized with helium are commonly tested, a convenient method of accurately estimating the amount of helium dissolved in the LN_2 would be useful. In a previous publication (Ref. 1), correlations for helium solubility in LO_2, LCH_4, and LH_2 were reported. In this report, we present a similar correlation for helium in LN_2.

Data Analysis and Correlation

As was the case for the previous work, the solubility data was provided by Lemmon at the National Institute of Standards and Technology (NIST), who compiled the data from various published sources and corrected the data to conform to the latest ITS-90 temperature scale (Ref. 2). The data was provided in tabular form consisting of total pressure (P, in MPa), temperature (T, in K), helium mole fraction in LN_2 (x), and the original data source.

Our correlation for helium-nitrogen is of the form

$$x = p* \cdot \exp\left(a_0 + a_1 T + b_0 p*\right) \tag{1}$$

where x is the mole fraction of helium in liquid nitrogen, $p*$ is a dimensionless pressure defined as $p* = (P - P_0)/P_{\text{ref}}$, where P is the total pressure (sum of liquid nitrogen vapor pressure plus the partial pressure of the helium pressurant gas), P_0 is the saturated vapor pressure of pure LN_2 at temperature T, P_{ref} is a reference pressure defined as 1 MPa, and a_0, a_1, and b_0 are empirically determined coefficients. The value of P_0 was determined using NIST's Standard Reference Database 23 software program REFPROP (Ref. 3). The coefficients a_0, a_1, and b_0 were obtained via multiple linear regression fitting of the data. The goodness of fit was evaluated by use of the relative standard deviation calculated as

$$\sigma_R = \sqrt{\frac{\sum (x_{\text{fit}} - x)^2}{N}} \tag{2}$$

where x_{fit} is the fitted mole fraction value, x is the reported value, and N is the number of reported values.

As done previously for LCH$_4$ and LO$_2$, we limited the pressure to 0 to 10 MPa and did not include data very close to nitrogen's critical temperature (126.2 K). Data below the normal boiling point (77.3 K) did not fit the correlation as well as higher temperature data, so data at temperatures significantly lower than the NBP were also excluded. Data from each individual source was first analyzed separately. Some of the data was deemed to be of lesser quality and was excluded. Reasons for exclusion were a large σ_R for data from an individual source or an insufficient number of reported significant digits in values of x. Data from six sources (Refs. 4 to 9) were used to develop the correlation. One of the data sets (DeVaney et al. (Ref. 6)) was substantially larger than the others due to a high number of repeated measurements (as many as six) at each individual pair of P and T. For this source, average values of x were computed for each (P,T) pair. Another source (Davis et al. (Ref. 5)) reported two measurements of x at each (P,T) pair; these were averaged as well. In some of the remaining data sets, there were occasional repeated tests with close values of pressure for the same temperature; these pairs were then averaged for P and x. Only two of the data sources (DeVaney et al. (Ref. 6) and Kharakhorin (Ref. 7)) contained data points for p^* greater than 7. However, these sources showed substantial scatter at low p^*, so only data from mid-to-high p^* was used. The low p^* data (with high scatter) from these two sources was omitted as other sources were available to provide data for the low p^* region.

The best-fit correlation for helium mole fraction in liquid nitrogen for the set of data described above is

$$x = p^* \cdot \exp\left(-9.727 + \frac{0.0472}{K} \cdot T - 0.025 \cdot p^*\right) \tag{3}$$

Data in the range 0.5 to 9.9 MPa and 72.0 to 119.6 K were used to obtain this correlation. The percent relative standard deviation of fitted values from the data considered is 6.9 percent ($N = 102$).

In Figures 1 to 3, the data is plotted as $\ln(x/p^*)$ versus p^*. In each figure, the solid lines representing fixed temperatures were generated from Equation (3). Each figure shows the temperature dependence of the solubility as well as the linear pressure dependence of $\ln(x/p^*)$. Figure 1 shows data that fits the correlation very well. Figure 2 shows data sets where the pressure dependence is slightly less than the correlation prediction and also shows the correlation over-predicts the solubility for two of the sources (DeVaney et al. (Ref. 6) and Skripka (Ref. 8)) at low temperatures. Opposite trends are shown in Figure 3, where the pressure dependence is greater than the correlation prediction and the correlation under-predicts the solubility for two different sources (Burch (Ref. 4) and Davis et al. (Ref. 5)) at low temperature.

The temperature dependence of helium solubility in LN$_2$ is shown in Figure 4. The correlation fits the data very well at higher temperatures, while at the low end of the temperature range, small differences are apparent, depending on the source of the data.

As an example calculation, consider LN$_2$ at its NBP (77.3 K) pressurized with helium to a total pressure of 1.72 MPa. Therefore, $p^* = 1.62$ and the equilibrium solubility mole fraction calculated from Equation (3) is $x = 0.36 \pm 0.025$ percent. Similar example calculations were given in Reference 1 for helium solubility in LO$_2$ and LCH$_4$ at the same pressure (1.72 MPa) and the respective NBP for each liquid. For both LO$_2$ and LCH$_4$ at these conditions, the helium mole fraction was $x = 0.13$ percent, thus indicating, on a mole fraction basis, that helium is almost three times as soluble in LN$_2$ as in LO$_2$ or LCH$_4$ for the example conditions.

Summary

Solubility data for the mole fraction of gaseous helium in liquid nitrogen from various sources has been correlated as a function of temperature and pressure. The correlation is valid over a broad range of temperature and pressure and should be valuable in the testing and analysis of cryogenic propulsion test systems when liquid nitrogen is utilized.

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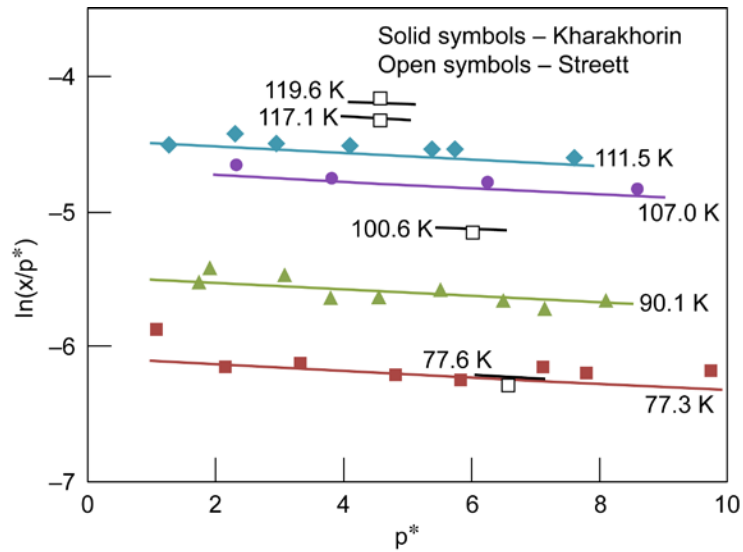
Figure 1.—Mole fraction solubility of helium in liquid nitrogen divided by dimensionless pressure as a function of dimensionless pressure. Data of Kharakhorin (Ref. 7) and Streett (Ref. 9).
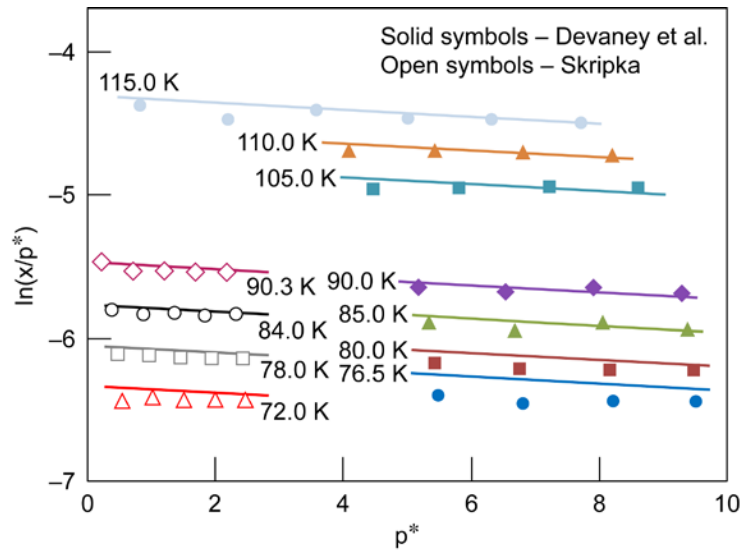

Figure 2.—Mole fraction solubility of helium in liquid nitrogen divided by dimensionless pressure as a function of dimensionless pressure. Data of Devaney et al. (Ref. 6) and Skripka (Ref. 8).
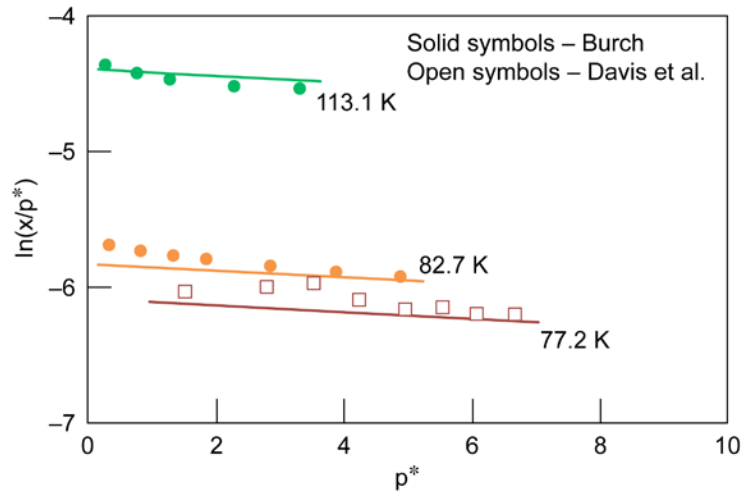
Figure 3.—Mole fraction solubility of helium in liquid nitrogen divided by dimensionless pressure as a function of dimensionless pressure. Data of Burch (Ref. 4) and Davis et al. (Ref. 5).
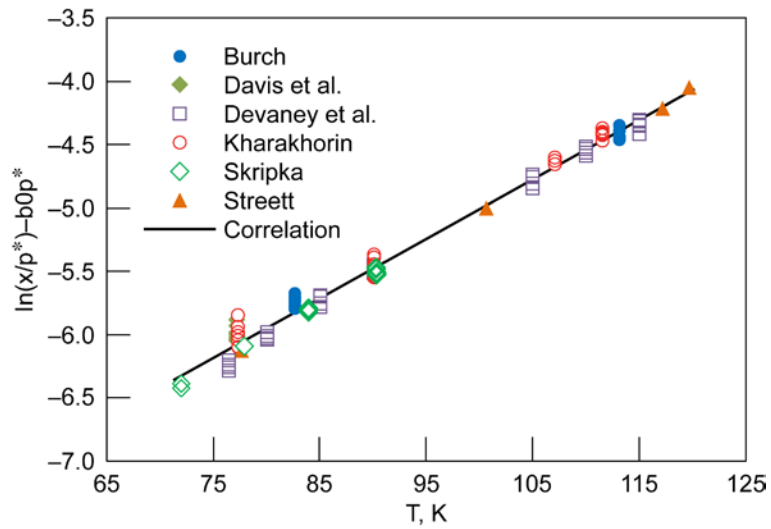

Figure 4.—Mole fraction solubility of helium in liquid nitrogen as a function of temperature.

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