TRACE IMPURITY ANALYZER[*]

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Summary

The desirability for long-term reliability of large scale helium refrigerator systems used on superconducting accelerator magnets has necessitated detection of impurities to levels of a few ppm. An analyzer that measures trace impurity levels of condensable contaminants in concentrations of less than a ppm in 15 atm of He is described. The instrument makes use of the desorption temperature at an indicated pressure of the various impurities to determine the type of contaminant. The pressure rise at that temperature yields a measure of the contaminant level of the impurity. A LN_2 cryogenic charcoal trap is also employed to measure air impurities (nitrogen and oxygen) to obtain the full range of contaminant possibilities. The results of this detector which will be in use on the research and development helium refrigerator of the ISABELLE First-Cell is described.

INTRODUCTION

The ISABELLE refrigeration system utilizes supercritical helium to supply refrigeration to nearly 1100 superconducting bending and focusing magnets. These magnets steer the proton orbits of the accelerator and are arranged into two interlocking rings, side by side in a common tunnel of 3.8 Km circumference. A single refrigerator capable of 18 kW at 3.8 K services the entire accelerator. This refrigerator is expected to operate longer and more reliably than present equipment in use and as such redundant components are added to insure continued operation. One drawback to long term reliability has been the buildup of impurities (air products, oil, etc.) which cause plugging and result in a reduction in capacity. Although several methods are available to identify impurities in a gas in a qualitative and quantitative manner, these methods in general require an expensive apparatus and instrumentation. In addition, these methods also tend to be time consuming and involve operation by skilled personnel. The trace impurity analyzer[1] described, circumvents these problems.

Method

A fixed volume of gas is drawn into an evacuated heated zone, maintained at an elevated temperature ($\approx$ 200°C) which reduces wall adsorption effects and prevents condensation or deposition of an impurity in the gas on the walls of the heated zone. After the sample has filled the heated zone, the zone is sealed and gas flow is stopped from going into or out of the zone. A small portion of the heated zone is cooled to liquid nitrogen temperatures (77 K) to condense the impurities in the gas sample. After the impurities have been deposited on the chilled sample container, the residual gas from which the impurities have been removed, including the chilled portion, is discarded by evacuation from the heated zone. The evacuated, heated zone is again sealed and the LN_2 coolant removed. The sample temperature is gradually warmed to vaporize the impurities deposited on the sample container in the order of their increasing boiling points. As the chilled portion is warmed, the temperature rise that vaporized the impurities causes the pressure to rise and serves to identify that impurity. The maximum pressure rise developed in the sealed heated zone quantifies that impurity.

When the pressure in the heated zone shows again an increase, with the rise in temperature of the small chilled container, the vaporization and desorption of another impurity is indicated. The temperature activating the pressure rise indicates qualitatively the other impurity while the new pressure increase provides a measure of the quantity of the new impurity. Figure 1 shows the pressure trace for three impurities.

FIG.1
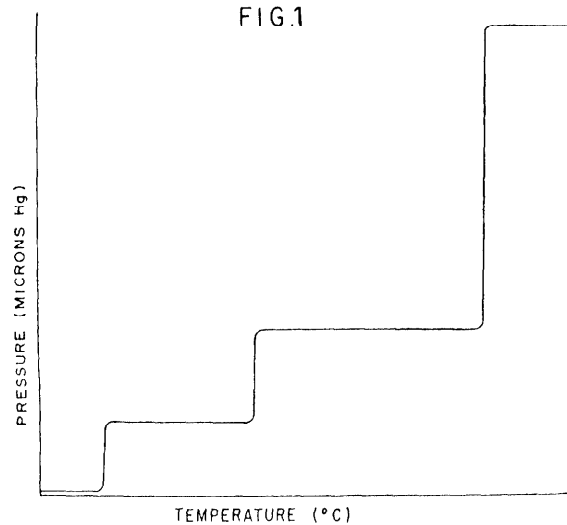
To detect trace amounts of noncondensible impurities such as air products, a molecular sieve adsorbent is placed in another segregable portion of the heated zone so that upon chilling the absorbent captures the impurities, thus separating them from the gas undergoing analysis. The chilling of the sample containing the absorbent to remove the impurities, the evacuation of the heated zone including the sample and the warming of the sample to desorb the impurities, is carried out in the same manner as described previously. Again, the temperature of the sample at the point where an impurity is desorbed from the adsorbent, identifies the impurity while the maximum pressure rise in the heated zone resulting from the desorption, indicates the quantity.

Results

To calibrate the detector, a known quantity of an impurity is added to a sample gas. The characteristic vaporization or desorption temperature is then established for that particular impurity. Two methods of desorption measurements have been used. The first consists of the method as described where dp/dt is determined. The second method continues evacuation of the sample and allows the peak of the desorption process to be observed by monitoring p(t) alone. Both methods permit calculation of the heat of vaporization of the desorption process from peak temperature (T_p) and peak width (Δw).

$$E/kT_p = \frac{\ln 2}{w} [1 + \varphi(w)]^{\begin{cases} dp/dt \text{ mode} \\ p(t) \text{ mode} \end{cases}}$$
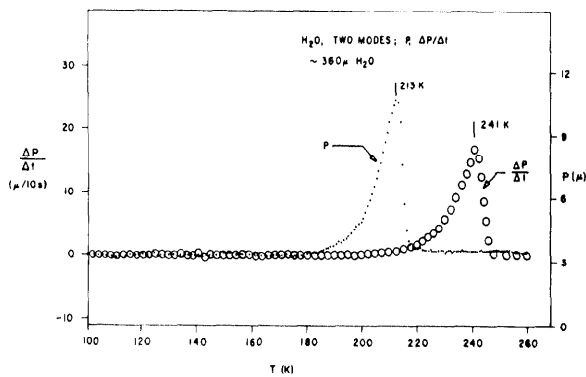
where $\quad w = \Delta w/Tp$.

Calculations of a number of known samples yields desorption energy values to within $\approx$ 1 K cal/mole of reference values. In Figure 2 is shown about 100 layers of water desorbed using the two methods.[2]

H₂O, TWO MODES; P, ΔP/Δt
~360μ H₂O

A detector has been put on line on the ISABELLE R&D Refrigeration system. The field results were confused due to virtual leaks in the system. A new detector is in the process of being constructed, designed to minimize the difficulties encountered with the first device.

Conclusions

A new detector capable of measuring impurities to less than a ppm is described. Experimental results have shown excellent correlation with published values of heats of vaporization. Employment of the detector under field conditions has been marred by virtual leaks. A new detector is undergoing fabrication.

Acknowledgments

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References

1. Patent applied for.
2. D. Edwards, Jr., J. Vac. Sci. Technol., (in press).