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FUSION TARGET PELLETS

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MASS SPECTROMETER DETERMINATION OF ARGON CONTENTS
IN LASER FUSION TARGET PELLETS*

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ABSTRACT

A system for measuring argon contents in individual laser fusion targets using a getter pumped closed volume quadrupole mass spectrometer has been developed. Accuracy of 10% can be obtained, limited by the calibration standard and electron multiplier drift. Measurements are simple, unambiguous and can be performed at up to one per hour with a sensitivity equivalent to 1×10^3 Pa (.01 atm) argon in a 140 μ m internal diameter sphere.

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1. INTRODUCTION

Argon is added to the fill of laser fusion targets as a compression diagnostic and to allow imaging of the imploded core⁽¹⁾ through measurements of the emission spectra. Production of specific argon fills and non-destructive measurement techniques require accurate measures of the fill of an individual selected sphere, since spheres produced in developmental argon filled droplet generator systems have content variations within any batch. Current gas analytical techniques utilizing crushing 10^5 spheres in the inlet to a standard mass spectrometer result in only averages and no information on the fill of an individual sphere.

Techniques for measuring Deuterium-Tritium (DT) contents in individual spheres rely on calibrating the flow properties and pumping of gas from a crushed sphere inlet into a pumped quadrupole mass spectrometer.^(2,3) Clark has used an alternate method for small volume gas measurement using a closed chamber to measure $^3\text{He}:^4\text{He}$ ratios in seawater.⁽⁴⁾ We have developed a similar technique using a closed volume mass spectrometer in which the argon partial pressure reaches a steady state for the measurement and active gases are pumped by a zirconium-aluminum getter. This permits accurate, repeatable readings and easy calibration.

2. MEASUREMENT SYSTEM

A UTI Model 100, open source quadrupole residual gas analyzer with an argon pressure sensitivity $> 10^{-15}$ Pa and an emission stability better

than 1% is used in the experimental system shown in Figure 1. The analyzer and an ion pump, a cold trapped diffusion pump, and a chemical getter pump are connected with bellows valves to the three liter all metal system. The ion pump is used during stand-by to keep the system evacuated and minimize hydrocarbon background. After closing off the ion pump, a cold trapped diffusion pump is used to evacuate the chamber before and after argon measurements. This reduces argon instabilities in the ion pump. During calibration and sample measurement, both pumps are closed off. Normally, in an unpumped vacuum system, the hydrogen, water, carbon dioxide, and residual hydrocarbon pressures rapidly rise above the tolerance of a mass spectrometer. In this application a chemical getter pump is used which selectively removes the active gases and does not pump noble gases, allowing argon partial pressures to stabilize.

A valved ion gauge, sampling fixture, and argon calibrated leak are connected to the chamber. This system is sufficiently sensitive that leaks in the 10^{-15} Pa - m³/sec range can be detected so it is important that all fittings and valves be metal sealed and checked. Pumping of the ion gauge induces a significant error requiring that it must be valved out, also baking and good vacuum practice are essential for stability.

A Hastings-Raydist 4.59×10^{-8} std cc/sec capillary leak is connected for calibration. This leak has been certified by the manufacturer to within 10% with a measured stability of 2%.

2.1 Chemical getter pump

Chemical getters have been shown to pump active gases by their combining with the getter elements while hydrogen goes into solution.⁽⁵⁾ A heated 100 L/sec Zirconium-Aluminum chemical getter pump manufactured by S.A.E.S. getters is used. Evaluations of these pumps by one of the authors has demonstrated that the Zirconium-Aluminum pump will readily remove most active gases and has virtually no pumping speed for argon.⁽⁶⁾ Prior to analysis, the getter is activated by heating to 800° C for 15 minutes which releases hydrogen in solution and reactivates the surface. During analysis, a temperature of about 250° C is maintained, being chosen as the lowest temperature which gives no discernible reduction in argon measurement over time frames of more than 15 minutes.

2.2 Sampling system

A crushing fixture built into a $\frac{1}{8}$ -inch metal sealed valve is used for sampling. The sphere is mounted using .1% Collodion on a stainless steel disc and crushed against a stainless plug inserted in the valve seat. The sorption roughing pump is used to evacuate the fixture from atmosphere until the diffusion pump can complete pump down.

3. TECHNIQUE

This technique is based on the principle that the system pressure change ΔP (Pa) caused by an argon increase is related to the quantity of

argon in the sphere Q ($\text{Pa} \cdot \text{m}^3$) and the system volume V (m^3) by,

$$\Delta P = \frac{Q}{V} \quad (1)$$

Quadrupole sensitivity is expressed as S (amperes/Pa) of the electron multiplier output current so the equivalent quadrupole current change ΔI (amperes) for a pressure rise is,

$$\Delta I = \Delta PS = \frac{S \cdot Q}{V} \quad (2)$$

Using a calibrated leak L ($\text{Pa} \cdot \text{m}^3/\text{sec}$) and assuming no argon pumping, the rate of change of pressure $dp/dt = L/V$ and the quadrupole current change is therefore,

$$\frac{dI}{dt} = \frac{SdP}{dt} = \frac{SL}{V} \quad (3)$$

Substituting sensitivity in (1,2), the quantity of argon in a sphere is thus

$$Q = \frac{\Delta I \cdot L}{\frac{dI}{dt}}$$

It is only necessary to know the argon calibrated leak rate L , the quadrupole current change rate during calibration dI/dt , and the current change ΔI resulting from the sphere breakage for a measurement. As long as the volume is held constant, there is no need to determine actual volume and there is also no need to refer to any transient flow phenomena. In practice, there is a small argon background which must also be subtracted.

4. TEST PROCEDURE

A mounted sphere is placed in the crushing valve attached to the test chamber and sorption pumped until the pressure is 10^{-5} Pa. One hour is then usually sufficient to diffusion pump the pressure to the low 10^{-10} Pa region. However, longer times can result in an order of magnitude improvement in background since this is a function of pumping time.

Both regular pumps and the ion gauge are valved off and the argon leak valve opened with the activated getter pump open. The resulting rate of quadrupole output current rise at the argon mass of 40 is used as the calibration. After valving off the argon leak, the system is again diffusion pumped and the argon background monitored. Once the argon partial pressure indicates less than 10^{-12} Pa, the diffusion pump is valved out and the argon pressure is noted as the background. With an acceptable background, the sphere is crushed, and the resulting output

current step is measured for at least a minute to assure stability. The argon pressure normally stabilizes after a few seconds and the rate of increase is strictly due to background. Any decrease would indicate argon pumping and is not usually observed.

The argon is again pumped out and another calibration is performed completing the analysis. Averages of the two calibrations are used to calculate the argon fill and compensate sensitivity drift.

It is important that the calibration before analysis be performed not only to check the instrument but to condition the system components and reduce pumping by clean chamber walls. Typically, backgrounds are less than 1% of the measurement being consistent with the stability of the entire system. Sensitivity is observed to continuously drift probably due to aging of the electron multiplier. Samples can be analyzed at up to one an hour with a sensitivity better than 10^3 Pa pressure in a 140 μ m ID sphere which is sufficient to measure the argon content of spheres produced in air.

5. RESULTS

Argon analyses of eight spheres from one batch performed over a period of three days is presented in Table 1 below. All readings are made to 1%.

Measurement repeatability should be within 1%; however, major system errors are caused by sensitivity drift and the argon calibrated leak. A 5% error from non-uniform sensitivity drift is indicated appearing to be due primarily to electron multiplier aging but also including some additional short-term phenomena which may be related to the drift in the spectrometer peak position. Pulse counting is necessary to overcome electron multiplier effects permitting evaluation of any other problems. Major systematic errors are due to the calibrated leak resulting in an absolute measurement of no better than 10%. If a more accurate leak calibration is obtained and pulse counting electronics are used, the precision of measurement should be increased to at least 5%.

Components of the continual argon background are unclear. Valves, fittings, and connections are all metal, either stainless steel or copper, although many of the welded sections are made with an argon purge gas. The chamber has had frequent bake outs which reduces the level to a degree. Some background is probably due to slight leaks and exchange of adsorbed argon with the system walls. In any event, the background is less than 1% of most readings and is subtracted during calculation.

6. OTHER GASES

This system works with other noble gases such as xenon, neon, and krypton. However, calibrated leaks of these gases are difficult to obtain. One advantage of the technique is that for unpumped, inactive

gases in the absence of a specific calibration, the argon sensitivity can be adjusted by published factors for the spectrometer and a rough measurement made.

With a very well baked system and long sample pump downs, it is possible by closing all pump valves to totally observe the components resulting from a ball break. Argon analysis can then be performed by opening the getter pump valve. Many factors relating to wall gas exchange effects and source pressure sensitivity need to be evaluated before this can be considered as an additional quantitative measurement technique.

7. SUMMARY

We have built a closed volume mass spectrometer with active gas pumping by a chemical getter to obtain a stable system for the measure of argon fills in laser fusion target spheres. Measurement accuracies of 10% are achieved, limited by the calibration leak itself. Analysis is simple and unambiguous and can be performed on other noble gases. This system is used to understand the argon fill mechanisms of spheres and provide a means to accurately calibrate non-destructive measurement techniques.

REFERENCES

- 1) V. W. Slivinsky, G. R. Leipelt and J. D. Eckels, "A Crystal Spectrograph for Imaging High-Density Targets", presented to the APS Plasma Physics Meeting, Atlanta, Georgia, November 5-11, 1977. Lawrence Livermore Laboratory report UCRL-79787.
- 2) M. A. Ebner and W. B. Rensel. Technique for Sampling Gases in Hollow Microspheres", Rev. Sci. Instrum. 46, No. 9, September 1975, Pg. 1290.
- 3) H. W. Deckman and G. M. Halpern, "Analysis of the Gaseous Content of Microballon Laser Fusion Targets", in the Technical Digest of the Topical Meeting on Inertial Confinement Fusion, February 7-9, 1978, San Diego, California. Digest No. 78 CH1310-2QEA, sponsored by the Optical Society of America.
- 4) W. B. Clarke, M. A. Beg, and Harman Craig, "Excess ^3He in the Sea: Evidence for Terrestrial Primordial Helium", Earth and Planetary Science Letters 6 (1969), (213-220), North Holland Publishing Company, Amsterdam.
- 5) Aldo Barosi and T. A. Giorgi, "A Non-Evaporable Getter for Low Temperatures". Presented at the Second Annual Conference on Ultra High Vacuum. Sponsored by the Vacuum Group Institute of Physics, University Collete, Sivansea, April 17-19, 1972.

- 6) L. E. Bergquist, "Chemical Pump Study", Martin Marietta Aerospace, Denver, Colorado, June, 1973. NASA CR-114594. Prepared under Contract NAS 2-7383 for Ames Research Center.

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FIGURE 1. Argon measurement system.

<u>Sample #</u>	<u>Sensitivity Before Analysis</u>	<u>Sensitivity After Analysis</u>	<u>Background</u>	<u>Current Increase After Break</u>	<u>Calculated Argon Fill</u>	<u>Volume Corrected Argon Fill In Std Atm</u>
500	1.08×10^{-7} amp/sec	9.88×10^{-8} amp/sec	1.30×10^{-9} amp	9.88×10^{-7} amps	4.18×10^{-7} cc	0.21 atm
501	8.60×10^{-8}	8.3×10^{-8}	9.0×10^{-10}	1.90×10^{-7}	1.04×10^{-7}	0.10
502	7.48×10^{-8}	6.76×10^{-8}	3.0×10^{-10}	1.89×10^{-7}	1.90×10^{-7}	0.17
503	6.47×10^{-8}	6.46×10^{-8}	3.52×10^{-10}	3.84×10^{-7}	2.79×10^{-7}	0.25
504	4.81×10^{-8}	4.46×10^{-8}	3.50×10^{-10}	0.99×10^{-7}	9.49×10^{-8}	0.08
505	3.99×10^{-8}	3.33×10^{-8}	2.3×10^{-10}	8.10×10^{-8}	1.10×10^{-7}	0.12
506	3.15×10^{-8}	3.01×10^{-8}	7.5×10^{-11}	1.60×10^{-7}	2.39×10^{-7}	0.21
507	3.06×10^{-8}	2.81×10^{-8}	6.0×10^{-11}	7.99×10^{-8}	1.25×10^{-7}	0.11

Standard leak 4.59×10^{-8} std cc/sec

TABLE I. Argon analysis of eight spheres.