Gas Pycnometry for Density Determination of Plutonium Parts

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A document prepared for 21ST AGING, COMPATIBILITY AND STOCKPILE STEWARDSHIP CONFERENCE at Albuquerque, NM, USA from 9/30/97 - 10/2/97.

DOE Contract No. DE-AC09-96SR18500

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Introduction

The traditional method for plutonium density determination is by measuring the weight loss of the component when it is immersed in a liquid of known density, Archimedes' Principle. The most commonly used heavy liquids that are compatible for plutonium measurement are freon and monobromobenzene, but these pose serious environmental and health hazards. The contaminated liquid is also a radiological waste concern with difficult disposition. A gaseous medium would eliminate these environmental and health concerns. A collaborative research effort between the Savannah River Technology Center and Los Alamos National Laboratory was undertaken to determine the feasibility of a gaseous density measurement process for plutonium hemishells.

Background

In the plutonium fabrication process the density of the plutonium serves as a quality assurance measurement on the finished product. In the surveillance program, the density of the aging components can be used as a quality measure on the long-term stability of the component. The traditional density measurement at Los Alamos National Laboratory (LANL) is performed by using a liquid immersion technique and Archimedes' Principle. The hemishell is weighed in air and in a liquid medium of known density and the density is computed from the weight change. The accuracy of this method has been estimated at 0.05%.

Since the hemishell is immersed in the liquid, the liquid becomes a hazardous waste stream and therefore generates a hazardous waste disposal problem. Monobromobenzene and freon, the liquids that are currently being used, are either a suspect carcinogen or are harmful to the environment. A survey of alternative liquids was performed to find a suitable replacement for these hazardous materials, but because of the requirements unique to plutonium, no other suitable liquid medium was found. Two techniques using a gaseous medium were identified as being possible alternatives for the liquid immersion technique, gas pycnometry and gas Archimedes. Of these two techniques, gas pycnometry was deemed the most feasible. It was determined that a gas medium would be used to perform a volume measurement with the weight in air to compute weight per unit volume or density.

Gas pycnometry is a gas displacement measurement technique that is used commonly throughout industry. The measurement technique makes use of the fundamental gas law. Equation 1 is the fundamental gas law at constant temperature and solving for the new volume yields equation 2.

\[ P_1V_1 = P_2V_2 \]
The standard industrial pycnometers were not applicable to high precision plutonium hemishell measurements because of the size and the self-heating properties of the plutonium. Therefore it was decided that a system designed for plutonium applications would be required. The Savannah River Technology Center entered into a collaborative research project with LANL to determine the feasibility of gas pycnometry for plutonium density measurements by designing, fabricating, and testing a gas pycnometer to be used to measure densities of surrogate parts. The following requirements were placed on the prototype system:

- accuracy within ±0.05%,
- no contamination of the part,
- measurement cycle of no more than 2 hours,
- operable in a closed hood environment.

Prototype Gas Pycnometer

The pycnometer chamber consists of two mating copper pieces with an inner machined chamber that closely matches the shape of the parts to be tested. Copper was chosen to take advantage of its high thermal conductivity. The outer surfaces are insulated to maintain isothermal conditions. The free volume in the chamber is twice that of the actual volume of the parts so an adequate pressure differential would be obtained for small volume changes. A schematic of the prototype system in provided in Figure 1. An inner O-ring seal is used to provide the main seal of the chamber. A bellows is used to provide the opening and closing of the chamber with uniform force. A feedback control loop is used to control the pressure of the bellows so it will maintain a constant force on the seal. Even small variations of the bellows pressure can translate into measurable and reproducible differences in the inner chamber volume.

An air operated piston is used to alter the measurement chamber volume. Temperature control of the piston allows precisely controlled and reproducible volume change.

![Diagram of prototype gas pycnometer](image_url)