STUDIECENTRUM VOOR KERNENERGIE

Beperkte verspreiding - Diffusion restreinte

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CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

WORKING GROUP "HOT LABORATORIES AND REMOTE HANDLING"
PLENARY MEETING 1986

A NEW TECHNIQUE FOR DENSITY MEASUREMENTS OF IRRADIATED NUCLEAR FUEL

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TEC/39.X8800/18/LS/hw/fq
April 4th, 1986

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ABSTRACT

A vacuum pycnometer with plunger displacement is described. It enables accurate determination of the open porosity density of solid samples. The operation of the home-made apparatus has been automated up to a high degree such that density measurements of radioactive fuel samples in the remote handling conditions of a hot cell environment are easily performed. The accuracy is better than 0.2 %.

INTRODUCTION

The basis for the pycnometry technique is the measurement of the volume of fluid displaced by the fuel sample the density of which is to be measured. To obtain the bulk density of porous materials, a high surface tension liquid which does not penetrate into the cracks and open pores has to be used. Due to its high surface tension, mercury is generally chosen as the sample-enclosing liquid. It can be used on all samples that do not amalgamate or react with nor wet the sample material. When these conditions are fulfilled, the volume actually measured is estimated as that volume bound by the exposed surface and the area over cracks and pores smaller than 14 µm wide (when working under atmospheric pressure). Since a classical glass pycnometer is not well suited for use in a master-slave manipulator equipped hot cell, a vacuum pycnometer with plunger displacement has been developed. A plunger of exactly known and uniform cross section is mechanically driven into a mercury chamber to a given pressure (± 1 atm.). The specimen volume is determined by the product of the plunger cross-sectional area and the difference in plunger displacement when filling the empty specimen chamber or when containing a sample.

APPARATUS (Fig. 1)

The main parts of the apparatus are :
- a measuring chamber block
- a high precision sealing valve system
- a high precision plunger system

a) The measuring chamber block (1) is a massive stainless steel block into which the specimen chamber cavity (about 2 cm^3) and the mercury plunger channel have been machined. Both are connected through a capillary bore; a second capillary bore connects the plunger channel with the pressure transducer (P.T.). A platinum resistance thermometer (T.C.) installed near the specimen chamber allows temperature control during the measurement.

b) A double-sealed high precision valve system (2) allows the specimen chamber to be always closed in a fixed reference position. Translational movement of the inner teflon valve is achieved through rotation of the drive screw at a 2 mm displacement rate per rotation. As the azimuthal position can be controlled to within ± 0.18°, by means of a vernier, the teflon plunger displacement, sealing the specimen chamber, is to within ± 1 µm. This corresponds to an uncertainty of the volume sealed of to ± 0.18 mm^3 (surface of plunger : 180 mm^2).

c) Translation movement of the motor driven high precision plunger (3) displaces mercury in the teflon sealed plunger channel. Before entering this channel, the plunger first passes a small vacuum chamber to avoid air inlet into the mercury chamber.
The motor drive mechanism allows a plunger displacement of 1.33 µm for each motor rotation.
Plunger displacement is measured by means of an electronic device - magnetic recording principle - allowing a position accuracy of ± 1 µm. Since the plunger has a diameter of (5.0517 ± 0.0010) mm, the minimum travel corresponds to a minimum volume change of 0.020 mm^3.

WORKING PROCEDURES

Filling procedure

At first the apparatus has been tested on its vacuum tightness. A vacuum better than 0.1 Pa is readily obtained and the leak rate determined by means of a helium leak detector is $< 10^{-6}$ Pa.l.s^{-1}.
The pycnometer is filled with about 11 cm^3 pure mercury via a filling device.
This device is a cylindrical block of plexy glass which fits on top of the measuring chamber block creating a vacuum seal. In the centre there is a mercury reservoir which can be unlocked so that the mercury can fill in the specimen chamber cavity when a vacuum is created in the pycnometer (pressure $\leq$ 1 Pa). The quantity of mercury present in the apparatus can be estimated within $\pm$ 0.2 cm^3 from the mercury level in the measuring chamber.

Measuring procedure

Each density measurement consists of two consecutive operations : a blank plunger displacement measurement and a subsequent sample plunger displacement measurement.
For the blank plunger displacement, no sample is loaded into the pycnometer. The apparatus is sealed with its measuring chamber empty and vacuum is pumped to evacuate any trapped air or moisture. When a vacuum of $<$ 1 Pa has been reached, the teflon valve is closed and the exact position of the activating drive screw is noticed. The plunger drive system is activated and the pressure transducer signal is recorded as a function of time. This operation is continued until a constant reading of a $\sim 10^5$ Pa pressure is obtained during 5 minutes at a given plunger position. Then the plunger is withdrawn again, the teflon seat is lifted and the pycnometer is vented. Before proceeding to the subsequent sample plunger displacement determination, it is carefully checked that no mercury is splashed out of the measuring chamber and that no mercury is adhered to the teflon seat. In either case, a new blank determination is required.

The subsequent plunger displacement with the sample in the measuring chamber is performed in the same way as the blank determination, except that now a sample (standard or fuel sample) is loaded into the measuring chamber of the pycnometer. Care is taken that exactly the same position of the teflon valve is each time installed. It is also taken care of that exactly the same final pressure is adjusted at each measurement.
The difference of travel of the plunger in the blank and sample determination is a measure of the mercury volume displaced by the sample. The density may then be determined by dividing the sample weight by the measured sample volume :

$$D_T = \frac{W}{(L_o - L) \times A - V_{Hg} \cdot \beta \cdot (T - T_o)} \quad (g.cm^{-3})$$

where D_T equals the density in $g.cm^{-3}$ at temperature T

W equals the sample weight in g

L_o equals the distance of travel of the plunger in the blank determination in cm

L equals the distance of travel of the plunger in the sample determination in cm

A equals the cross-sectional area of the plunger (0.02043 ± 0.00006 cm^2)

V_{Hg} equals the volume of mercury in cm^3 present in the apparatus

β equals the thermal expansion coefficient of mercury ($\beta = 0.0001822$ $°C^{-1}$ between 20 °C and 30 °C)

T equals the temperature at the time of the sample determination

T_o equals the temperature at the time of the blank determination

At the beginning of each series of measurements and at regular intervals afterwards, the performance of the apparatus is checked by measuring the density of stainless steel standards.
When measuring irradiated nuclear fuel density, fuel samples are selected in a random manner by removing the fuel portion from the fuel rod cross section. If the sample is fragmented, it is sieved to remove particles with a cross-section less than 2 mm. The remaining fuel is rinsed in acetone, allowed to dry and weighed to the nearest 0.1 mg.

ACCURACY

Several factors affect the accuracy of the density measurement. Among these, the most important are :

- The sample chamber seal. As described above a seal reproducible to within ± 0.18 mm^3 can be established.
- The precision by which the plunger displacement can be regulated and measured. This was pointed out above to be ± 1 µm what leads to an uncertainty in the volume measured of ± 0.020 mm^3.
- The change in volume of mercury due to a temperature change. Since the mass of the measuring chamber block of the pycnometer is large and a blank reading is taken every time a sample is measured, the temperature of the mercury will not change enough to interfere.
  However, a very highly radioactive sample may require some correction as pointed out in the formula above. The greatest error that may arise from this point of view is coming from the estimate of the mercury quantity present in the block (± 0.2 cm^3) what leads to an uncertainty in volume increase due to a temperature change of 1 °C of ± 0.036 mm^3.

Taking into account all these factors, an overall theoretical accuracy of ± 0.24 mm^3 can be expected.

Some cold runs have been performed to control the accuracy (Table 1).

- To control the overall reproducibility, several successive blank determinations (with intermediate venting and opening of the pycnometer) have been performed. The results indicated a measurement precision of ± 0.26 mm^3 (2σ).
- Two stainless steel standards (both with a density of 7.7221 ± 0.0015 $g.cm^{-3}$) were measured several times :

Table 1. Cold standard measurements

STANDARD I (2.2715 g)			STANDARD II (7.7191 g)		
MEASURED DENSITY ($g.cm^{-3}$)	ACCURACY ($g.cm^{-3}$)	(%)	MEASURED DENSITY ($g.cm^{-3}$)	ACCURACY ($g.cm^{-3}$)	(%)
7.7199	- 0.0022	- 0.029	7.7369	+ 0.0148	+ 0.190
7.7220	- 0.0001	- 0.002	7.7224	+ 0.0003	+ 0.003
7.7215	- 0.0006	- 0.008	7.7196	- 0.0025	- 0.033
7.7204	- 0.0017	- 0.022	7.7196	- 0.0025	- 0.033
7.7209	- 0.0011	- 0.015	7.7204	- 0.0017	- 0.023
7.7225	+ 0.0004	+ 0.005	7.7196	- 0.0025	- 0.033
7.7257	+ 0.0036	+ 0.046	7.7345	+ 0.0124	+ 0.160
7.7214	- 0.0006	- 0.008			
Mean :	Precision ($g.cm^{-3}$)	(%)	Mean :	Precision ($g.cm^{-3}$)	(%)
7.7218	± 0.0018	0.023	7.7247	± 0.0076	0.098

Foregoing results clearly demonstrate that the measured reproducibility is corresponding very well with the theoretical expected one. So it can be concluded that the home-made apparatus functions very well. No error sources other than expected on base of the conceptual design of the apparatus are introduced during normal operation. The accuracy of the density determination is better than 0.2 %, while the precision is better than 0.1 %.

The last experimental step to check the performance of the pycnometer is to investigate if the remote handling conditions of a hot cell influences the accuracy or not. Therefore the same standards were measured again with the pycnometer installed in a mock-up cell equipped with manipulators (Table 2) :

Table 2. Measurements performed with the remote set-up in a mock-up hot cell.

STANDARD I			STANDARD II		
MEASURED DENSITY ($g.cm^{-3}$)	ACCURACY ($g.cm^{-3}$)	(%)	MEASURED DENSITY ($g.cm^{-3}$)	ACCURACY ($g.cm^{-3}$)	(%)
7.7220	- 0.0001	- 0.001	7.7284	+ 0.0063	+ 0.082
7.7226	+ 0.0005	+ 0.006	7.7172	- 0.0049	- 0.063
7.7301	+ 0.0080	+ 0.104	7.7189	- 0.0032	- 0.041
7.7366	+ 0.0145	+ 0.188	7.7226	+ 0.0005	+ 0.006
7.7086	- 0.0135	- 0.175	7.7233	+ 0.0012	+ 0.016
7.7333	+ 0.0112	+ 0.145			
7.7212	- 0.0009	- 0.012	Mean :	Precision	
7.7307	+ 0.0086	+ 0.0111		($g.cm^{-3}$)	(%)
7.7250	+ 0.0029	+ 0.038			
			7.7221	± 0.0044	0.056
Mean :	Precision				
	($g.cm^{-3}$)	(%)			
7.7256	± 0.0083	0.11			

These results show that the remote handling conditions do not affect the performance of the highly automated mechanical pycnometer, as was intended from its original design.

CONCLUSION

A fully remotely working mechanical mercury pycnometer has been designed. It allows accurate determination of the density of highly radioactive samples in a relatively simple manner without any loss of accuracy. The accuracy is experimentally verified to be < 0.2 % while the precision equals 0.1 %.

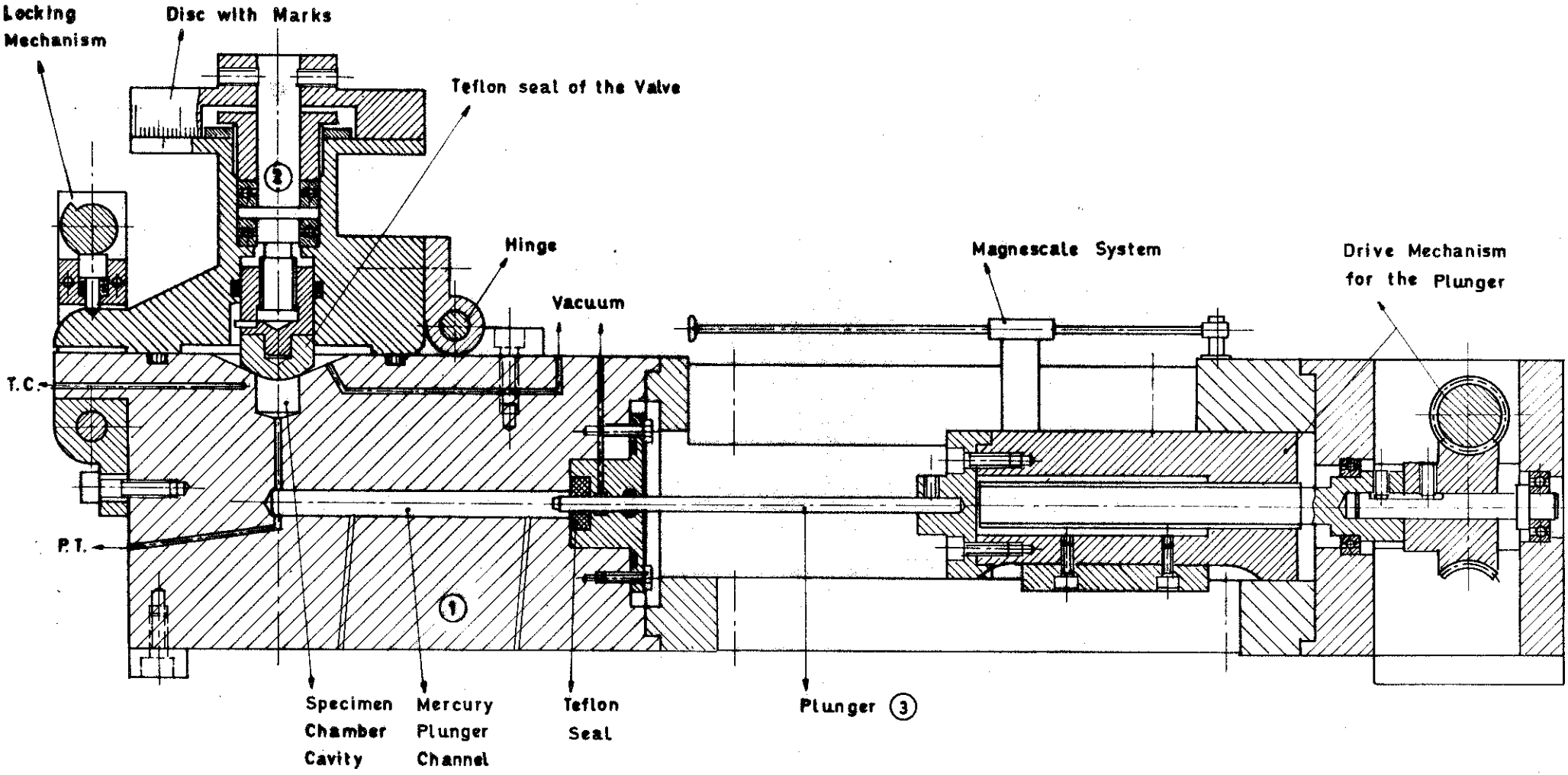

Fig. 1. Cross-section of the Pycnometer : (1) Measuring Chamber Block
(2) High Precision Valve
(3) Plunger Drive System

T.C. Pt 100 Resistance Thermometer

P.T. Pressure Transducer