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INEXPENSIVE, OFF-THE-SHELF HYBRID MICROWAVE SYSTEM

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A hybrid-heating microwave oven provides the energy to heat small 10-gram samples of spent metal tritide storage bed material to release tenaciously held decay product ^3He. Complete mass balance procedures require direct measurement of added or produced gases on a tritide bed, and over 1100°C is necessary to release deep trapped ^3He. The decomposition of non-radioactive $CaCO_3$ and the quantitative measurement of CO_2 within 3% of stoichiometry demonstrate the capabilities of the apparatus to capture generated (released) gases.

I. BACKGROUND

Savannah River National Laboratory (SRNL) is developing a modern gas assay method for spent tritium storage beds for Sandia National Laboratory/Ca. Currently, helium from tritium decay remains trapped after delivery of the stored gas, so a complete characterization of the storage bed is not obtained from the delivered gases. A technique is under development that releases the helium content from the spent bed material in a way that allows quantitative measurement with the same accuracy as that measured for other gas species. SRNL has successfully identified the right materials of construction and process configuration to heat small samples of spent palladium storage bed material in an inexpensive commercial "hybrid" microwave oven in a controlled manner to a high enough temperature to cause the desired effect of releasing the helium.[1] After a few successful tests using zirconia as a surrogate storage bed material, the first test using representative (cold) palladium storage bed material was performed. A temperature of 1200°C obtained in a few minutes after starting the microwave oven, and controlled for over half an hour.

It was then desirable to produce a dry gas in the unit and measure the moles of gas produced to demonstrate the ability to quantify a non-condensable gas produced in the system. The decomposition of $CaCO_3$ to CO_2 was used to test the system without using tritium contaminated samples. This proved to be quite acceptable since the measured number of moles of CO_2 matched the calculated number of moles based on stoichiometry to within 3%.

II. SYSTEM CONFIGURATION

The hybrid microwave unit for gas measurement consists of the microwave oven, the susceptor unit and insulation, the plasma shield, the cooling fin(s), the charge tube, and the gas manifold. A thermocouple, vacuum pump, pressure gauge, gas supply, and calibrated bomb tank are connected to the gas manifold. Flexible hoses between the manifold and the vacuum pump, pressure gauge, and calibrated bomb tank allow freedom of movement of the part of the manifold that holds the charge tube. Fig.1 shows a picture of the prototype microwave oven with the charge tube inserted into the unit. A 7.4-inch tall charge tube reaches 2.25 inches into the susceptor encasement, which is 5 inches above the bottom of the microwave cavity.

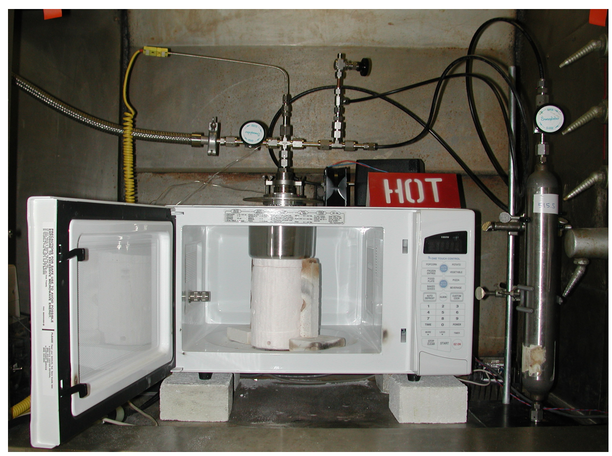

Fig.1. Prototype Microwave Oven

The charge tube is made of a 1” OD quartz tube and is sealed by an ultra-Torr® fitting at the top. A thermocouple extends down through the center of the tube that has an alumina filter attached to it to shield the coupling at the top from the heat shine below. The single fin with fan keeps the temperature at the fitting below 50°C. If the fan fails, the coupling temperature reaches almost 80°C at an operating temperature of 1200°C in the charge tube. The maximum temperature for the o-ring in the ultra-Torr Seal® fitting is 204°C. Fig. 2 shows the charge tube fully removed above the microwave oven.

The microwave susceptor is 200 grams of granular SiC (silicon carbide) encased in a 6” tall annular quartz encasement around a 1” center hole where the charge tube slides in and out. The 2” OD quartz encasement is set in a 2” ID x 4” OD x 8” tall alumina insulation tube that has a 1” thick bottom to it.

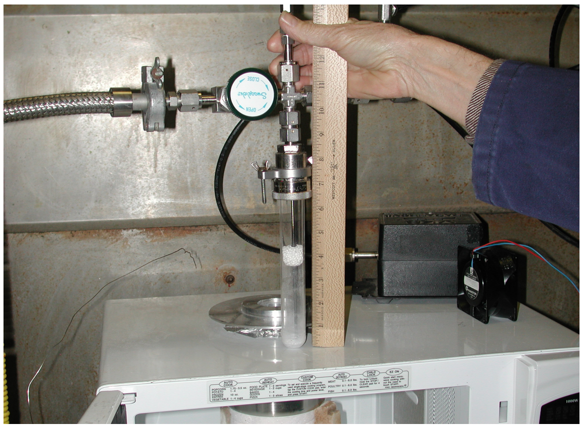

Fig.2 Charge tube with alumina foam filter

Once the quartz encasement is inserted into the insulation tube, a 1” thick alumina insulation ring is placed on top of the quartz encasement and some alumina wool is stuffed above that to ensure no heat shine reaches the metal above. The 3” tall stainless steel plasma shield goes around the outside of the insulation and is positioned against the top of the microwave oven cavity. The bottom of the shield is about 3/8” below the top of the SiC in the susceptor unit. These pieces are shown in Fig.3.

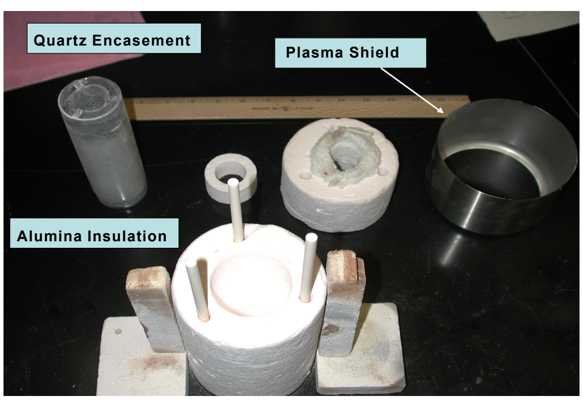

Fig.3 Plasma Shield and Quartz Encasement

The plasma shield extends down from the top of the microwave cavity to about 1/4” below the top of the susceptor. The purpose of this shield is to reduce the direct impingement of microwaves into the charge tube to prevent a plasma from forming inside the charge tube at low pressures. A plasma causes uncontrolled rapid heating of the tube and its contents. Without the plasma shield, a plasma can form at pressures from 3 Torr up to 80 Torr. With the plasma shield, a plasma can only form at pressures less than 9 Torr. As a result, the heating process can be started with about 15 Torr of an inert gas.

III. SYSTEM CONTROL

III.A. Microwave Heating Control

The magnetron produces the microwaves in the oven. Control of the magnetron was accomplished by inserting an ON/OFF relay in the line going to the step up transformer prior to the magnetron. Automatic computer control of the relay was based on either the pyrometer temperature reading or the charge tube thermocouple temperature reading versus an operator set point. Manual computer control of the relay was based on a percent output inputted by the operator. The cycle time is 32 seconds, so a 50% output would produce 16 seconds ON, then 16 seconds OFF and so on. To operate the oven, the operator would still have to enter a time on the oven front panel and press the start button. All original safety interlocks on the oven remained in use, which includes the door interlock and the cavity over-temperature interlock. Both interlocks shut off the oven.

III.B. Temperature Response

Using the magnetron control scheme described above, the temperature of the sample in the charge tube rapidly heats to process temperature and remains at temperature for the duration of the measurement. Fig. 4 shows the pyrometer (IR2) reading for a process run, and the pressure increase in the charge tube for an initial pressure of 400 Torr.

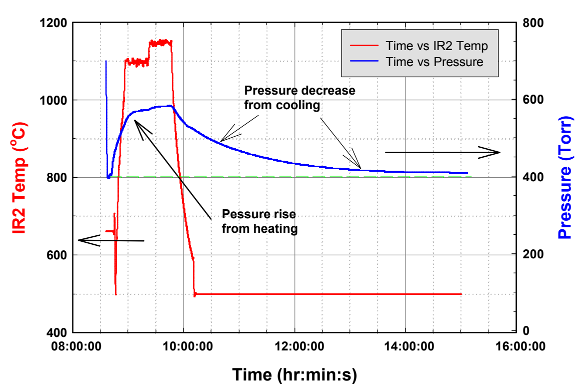

Fig. 4 Pressure rise due to gas heating

The sample temperature rose to 1100°C, remained for about 30 minutes, and was increased to 1150°C for another 30 minutes. Power was turned off and the system cooled for about 4 hours. Notice the pyrometer does not begin to read until 600°C, necessitating the use of an internal thermocouple to measure the full range of temperatures of the sample.

While the sample is at temperature, there is a good bit of radiant "shine" emanating in all directions. The alumina foam filter (Fig.2) attached to the internal thermocouple effectively blocks the shine and helps maintain the temperature of the ultra-Torr Seal® o-ring connection to the quartz tube at or near ambient. Fig. 5 shows the IR2 pyrometer reading during a process run again at 1100 -1150°C, along with the measured temperatures of the upper and lower parts of the o-ring connection. The heat-sink and instrument fan help remove the heat, maintaining 29 to 33°C at the o-ring.

The temperature rise is rapid for these process runs. The observed increase based on the pyrometer is about 80°C per minute from about 500 to 1100°C. The temperature control using the ON/OFF power control of the magnetron is within ±5°C at 1100°C.

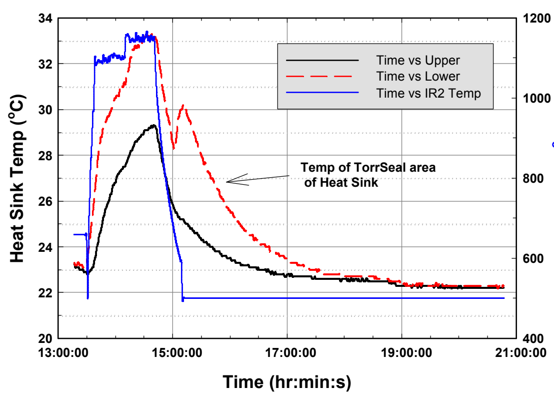

Fig.5 Temperature range of o-ring

IV. MEASUREMENT OF A GAS

The quantitative measurement of a gas requires the knowledge of the total system volume, the charge volume, the hot volume, the initial cold temperature, the initial pressure, the final pressure, the final cold temperature, and the hot temperature. The total system volume and the hot volume are calibrated once and are constants throughout successive runs.

IV.A. Hot Volume Determination

During operation, part of the system is heated to about 1200°C with a temperature gradient to the temperature of the rest of the system, which is near ambient temperature. For purposes of determining moles of gas, the system will be described mathematically as having two distinct temperature zones that are the hot volume *Vh*, and the cold volume, *Vc*, which is the measured manifold temperature.

Using the Ideal Gas Law,

$$\frac{P_1 V_1}{T_1} = \frac{P_2 V_2}{T_2} \qquad (1)$$

Eq. (1) can be divided into hot and cold volumes

$$m = \frac{P_1}{P_2} = \frac{\left(\frac{V_C}{T_O} + \frac{V_H}{T_O}\right)}{\left(\frac{V_C}{T_O} + \frac{V_H}{T_H}\right)} \qquad (2)$$

where V_C is the cold volume, V_H is the hot volume, T_O is the cold temperature, T_H is the hot temperature, P_1 is the pressure when the charge tube is at the hot temperature, and P_2 is the pressure when the charge tube is at the cold temperature. Cold temperature is usually the room, or starting temperature. Solving for the hot volume,

$$V_H = \frac{V_T T_H (m-1)}{m(T_H - T_O)} \qquad (3)$$

where ***m*** is the ratio of the system pressure at the hot temperature to the pressure at the cold temperature (Eq.2), and V_T is the total volume, $V_C + V_H$. To determine the hot volume, a known ZrO_2 charge and an initial pressure of N_2 at room temperature was heated to 1200°C and obtained steady state pressure. This was repeated for several starting pressures. The data are in Table I.

TABLE I. Hot Volume

P_1 (Torr)	*Vh* (mL)
269	53.9
338	54.1
378	54.7
451	54.2
489	52.0
629	52.6
733	51.7
Average	**53.3**

IV.B. Gas Production Determination

The decomposition of $CaCO_3$ to CO_2 was used to demonstrate that a gas can be produced in the system and quantitatively collected. A total of eight runs were performed for determining gas production. In each run, various amounts of ZrO_2 and $CaCO_3$ were charged to the tube. The system was evacuated and purged with N_2 three times. The charge volume was determined, and the system was charged with enough N_2 to raise the pressure to 15 Torr to prevent plasma from forming during initial heating. The system was heated to 1200°C and held there until the pressure came to steady state. Then the data were collected to determine the number of moles of CO_2 formed from the $CaCO_3$ both by chemical stoichiometry and by the ideal gas law, and are in Table II.

Table II. $CaCO_3$ Decomposition Data

#	$CaCO_3$ gms	P_1 Torr	CO_2 calc	CO_2 meas	%
1	1.575	495.2	0.0158	0.0157	-0.28
2	1.568	496.2	0.0157	0.0157	-0.22
3	1.575	498.8	0.0158	0.0158	0.27
4	1.580	504.1	0.0158	0.0160	0.95
5	1.574	510.2	0.0157	0.0161	2.52
6	0.926	314.2	0.009	0.001	5.57
7	0.926	314.0	0.009	0.001	5.32
8	2.235	705.0	0.022	0.023	0.99

The error results were analyzed against final system pressure and volume of inert charge. Neglecting the data of runs 6 and 7 (final pressure around 300 Torr seemed to have an issue), the measured moles of CO_2 were accurate at worst to within about 3%. The data were independent of the volume of the charge.

V. CONCLUSIONS

- The hybrid microwave system is capable of heating samples to greater than 1200°C.
- The quartz components can withstand 1200°C for at least several hours without any apparent change. (Quartz annealing temp: 1215°C; softening temp: 1885°C)
- The quartz components in the susceptor remain usable after 26 runs.
- The unit can be controlled to prevent plasma from occurring.
- The unit can control the temperature ± 25°C at 300°C and ± 5°C at 1200°C.
- The system can measure non-condensable gases to within less than 3% error when the final pressure is 490 to at least 700 Torr. Note that the o-ring was tested at the high pressure test lab and held pressure up to 50 psig at room temperature. The expectation is to run the unit where the maximum expected pressure is below 1000 Torr.
- Because of the massive amount of insulation around the charge tube and susceptor, the oven cavity temperature becomes warm (about 50°C), but does not follow the temperature of the charge tube.

The system protocol described here is but one of many that can be programmed to use the hybrid-heating microwave oven. For example, intermediate temperature stops can be included to soak a sample at temperature before continuing to a higher temperature. Also, the sample gas released can be evaluated after the charge tube and sample return to ambient, removing the need to calculate a "hot volume". However, we have demonstrated that the division of the manifold into a hot section and a cold section does return reasonable results, to within about 3% once the calibration curve has been obtained.

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