Apparatus to Measure Low Level Helium for Neutron Dosimetry

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An apparatus to measure low level helium in a solid sample for neutron dosimetry in the practical use such as area monitoring in the long-term and reactor surveillance was reported. In our previous work, the helium atoms measurement system (HAMS) was developed. A sample was evaporated in the furnace and the released gas from the sample was analyzed with the mass spectrometer of the system to determine the amount of helium contained in it. The system has been improved to advance the lower helium measurement limit in a solid sample for its application to an area monitoring system. The mass of a solid is up to 100mg. Two important points should be considered to advance the lower limit. One was to produce a high quality vacuum in the system chamber for suppressing background gases during the sample measurement. The other important point was to detect very small output from the mass spectrometer. A pulse counting system was used to get high sensitivity in the mass 4 analyzing.

1. Introduction

The helium atoms measurement system (HAMS) was developed to measure He production cross sections in our previous work[1]. We obtained He production cross sections for several elements by measuring He atoms down to 10^{10} He atoms in a solid sample[2, 3] by using the system. The purpose of this work is to advance the lower helium measurement limit of the HAMS to less than 10^{7} He atoms in a solid sample. The improvement is for applying this system to a new type of area monitoring system to which a He gas measurement method is employed. In the method, the amount of He produced in a dosimeter by neutron irradiation is measured with the absolutely calibrated mass spectrometer. The amount of He is in proportion to neutron fluence at the dosimeter-setting position and is independent of variation in neutron flux. The only He production cross section is, moreover, required to obtain absolute neutron fluence value.

Two important points should be considered to achieve the lower limit described above. One is to produce a high quality vacuum in the system chambers to decrease the background gas during the sample measurement. The other is to detect very small output from the mass spectrometer. A pulse counting system is applied to get high sensitivity in the mass 4 analyzing. This paper presents the detail description of architecture of the improved HAMS and preliminary results of performance tests.

2. Apparatus

A block diagram of the HAMS is shown in Fig.1. The HAMS is composed of three blocks, which are a gas releaser, a mass spectrometer, and a standard He supply. The measurement procedure is described briefly as follows. A solid sample containing He is set in the furnace of the gas releaser and is evaporated. The released gases are purified with a trap of the gas releaser and then are introduced into the mass spectrometer to measure the amount of He in the sample. The mass spectrometer has a quadrupole mass spectrometer (QMS), a digital electrometer, a multichannel analyzer and a personal computer which controls the QMS and stores data from the digital electrometer or the multichannel analyzer, and additional information. The sensitivity of HAMS to He is calibrated by measuring standard He. The standard He is produced by using the standard He supply.

2.1. Vacuum evacuation

Vacuum vessels of the HAMS are redesigned to decrease background gases at the He measurement. They are made by welding their parts at the inside of the parts and their inside walls are cleaned by emery polishing and electric polishing. The total volume of the HAMS vessels is decreased and the pumping speed from the vessels is increased. The three blocks (the gas releaser, the QMS, and the standard He supply) have individual turbo-molecular pumps to keep them at the high quality vacuum. The QMS block, especially, has tandem jointed turbo-molecular pumps and is evacuated to lower than 1.2×10^{-8} Pa. The tandem jointed pumps also pump the gas releaser. Two Ti-getter pumps cooled by liquid nitrogen are attached to the gas releaser and the QMS to trap the background gases and purify the released gas. Valves employed in this system are metal seal valves to prevent He permeation.

2.2. Gas releaser

The gas releaser is composed of a furnace, a sample loader, and a trap. The furnace is a device for releasing He from the solid sample by vaporizing it. The furnace has a pair of electrodes and a shield. An evaporating boat which is heated electrically is located between the electrodes which are made of stainless steel 304. The material of the evaporating boat is selected from tungsten, molybdenum, tantalum and graphite and is chosen according to the melting point and the element of the sample.

2.3. Mass spectrometer

The mass spectrometer is composed of a quadrupole-type mass spectrometer (QMS), a personal computer, a digital electrometer, a multichannel analyzer, and a trap. The QMS is adjusted to be able to measure the mass from 1 to 6 in atomic mass units with the mass analyzing power of 0.025amu(10% valley).

The released gas is introduced into the QMS from the furnace, is purified again with the trap in the QMS, and is then analyzed. The output current is amplified by a pulse counting type of secondary electron multiplier (gain: $<1 \times 10^{8}$) with which the QMS is equipped, and led to the digital electrometer or the multichannel analyzer. The improved HAMS operates in the current measurement mode or the pulse counting mode. The pulse counting by a multichannel analyzer is applied to measure the very small amount of He of less than 10^{10}. The computer code for these procedures can control and compute the timing for measurements and the processing for preparations of the standard He gas when we input the operating conditions.

2.4. Traps

The traps are Ti-getter pumps cooled with liquid nitrogen, which have a nominal total pumping speed of 11.5 l/s, and have two kinds of roles to purify the sample gas and to keep the devices at an ultra-high vacuum during He measurement. When we operate HAMS, the QMS and the furnace are isolated from the turbo-molecular pumps in order to increase the sensitivity to gases.

The Ti-getter pump works in either a dynamic mode or a static mode. In the dynamic mode, the Ti-filament is emitting Ti vapor and is producing new Ti-film on the inside wall of its vessel all through the He measurement. In the static mode, a fresh Ti-film is produced once just before isolating the pump and is used throughout the operation. The dynamic mode is preferable because the pumping speed is expected to be steady. The flashing of the Ti-filament, however, generates the background of the mass-4 equivalent to about $1x10^8$ He atoms. Therefore, The static mode is used in He measurement of less than $1x10^{10}$ He atoms. In this mode, we must pay attention to the amount of gases released from a sample because excess sample gases sometimes can not be pumped out and then saturate the QMS.

2.5. Standard He supply

Figure 2 shows a block diagram of the standard He supply producing the standard He which is used to calibrate HAMS absolutely. Standard He in the range of 1×10^9 to 5×10^{15} He atoms can be produced in a routine manner described below.

The standard He supply comprises four vessels: the glass standard volume vessel (V1), the sub-standard vessel (V2), the inlet vessel (V3), and the dilution vessel (V4) and three absolute pressure gauges. The furnace is used as the fifth vessel in producing the standard He, so it is called V5. These vessels are made of stainless steel and connected with all metal-sealed valves and flanges except the glass standard vessel and its cock valve, which are made of Pyrex glass. Farrar IV et al.[4] used aluminosilicate glass for their vessels which are long-term storage vessels for standard He instead of Pyrex glass to prevent He contained in the atmosphere from permeating through the vessel wall. The permeating He increases the amount of the standard He and causes undesirable uncertainty. In our apparatus, Pyrex glass is employed only for the glass standard volume vessel, which is not used in preparing the standard He but just in determining the volume of the other vessels.

To prepare the standard He, we must know the volumes of the five vessels, V1 ~ V5, whose volumes are also represented as V1 ~V5, respectively. The volume of V1 is 20.254 cm^3 determined by measuring the net weight when it is filled with distilled water. The volumes of V2 ~ V5 are determined by comparing the pressure of filled He and that after expanding the He into the relating volume and referring to the volume of V1 with Boyle's law. It can be assumed that the experimental conditions about the temperature are the same. The absolute pressure gauges are two diaphragm-type gauges and a spinning-rotor gauge. One of the diaphragm-type gauges has a measurable maximum range of ~ 1330 Pa with the uncertainty of 0.3% and the other has a range of ~ 13.3 Pa with 0.1%. They are suitably used to prepare the standard He so as to decrease the uncertainty for the resultant standard He. The spinning-rotor gauge is used to calibrate other gauges because of its good long-term stability.

3. Performance

Many standard He gases in the range of 1×10^{10} ~1×10^{13} He atoms were produced with the standard He supply and were measured with the HAMS in the current measurement mode. These results with a regression curve are shown in Fig.3. QMS output is converted into the real number of He atoms by this curve. The curve indicates a good linearity and fluctuation of the He measurement efficiency of the HAMS is improved.

Figure 4 shows a result of background measurement in the pulse counting mode at mass number of 4.000amu. The result contains both pulses caused by electric noise and ones by background gases. The total number of pulses corresponds less than $1x10^{9}$ He atoms. It is important to eliminate the electric noise and to optimize some parameters such as SEM gain and time constants of pulse shaping, for achieving reliable measurement.

References

[1] Takao Y., Kanda Y.: Rev. Sci. Instrum., **67** 198(1996).

[2] Takao Y., Kanda Y., Uenohara Y., Takahashi T., Itadani S., Iida T., Takahashi A.: J. Nucl. Sci. Technol., **34**[1], 1(1997).

[3] Takao Y., Kanda Y., Hashimoto H., Yamasaki K., Yamaguchi K., Yonemoto T., Miwa M., Etoh H., Nagae K.: J. Nucl. Sci. Technol., **34**[2], 109 (1997).

[4] Farrar IV H. and Oliver B. M.: J. Vac. Sci. Technol. A **4**, 1740(1986).

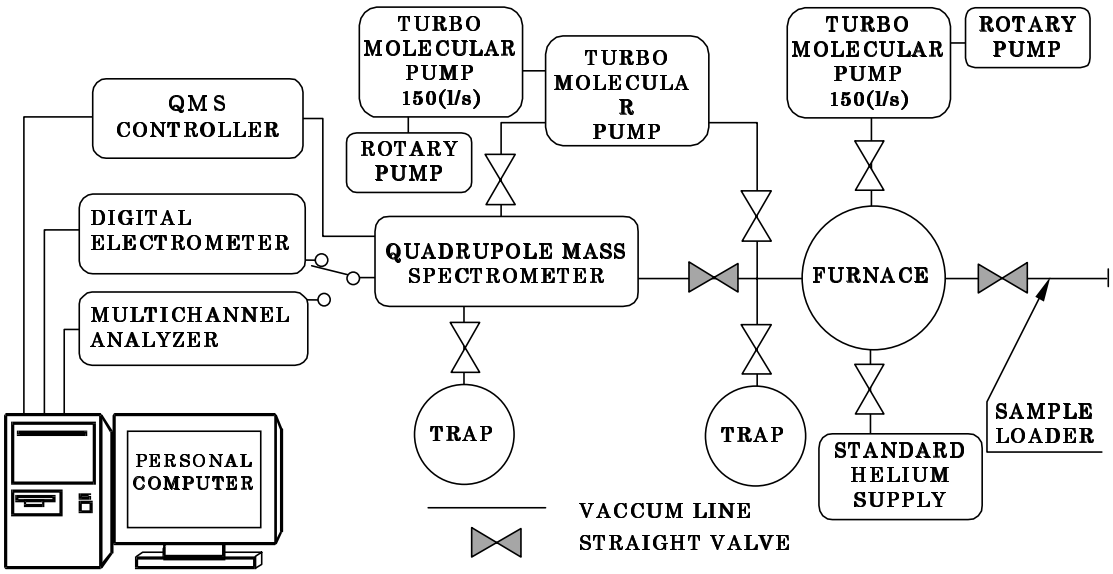

Fig.1 A block diagram of the Helium Atoms Measurement System

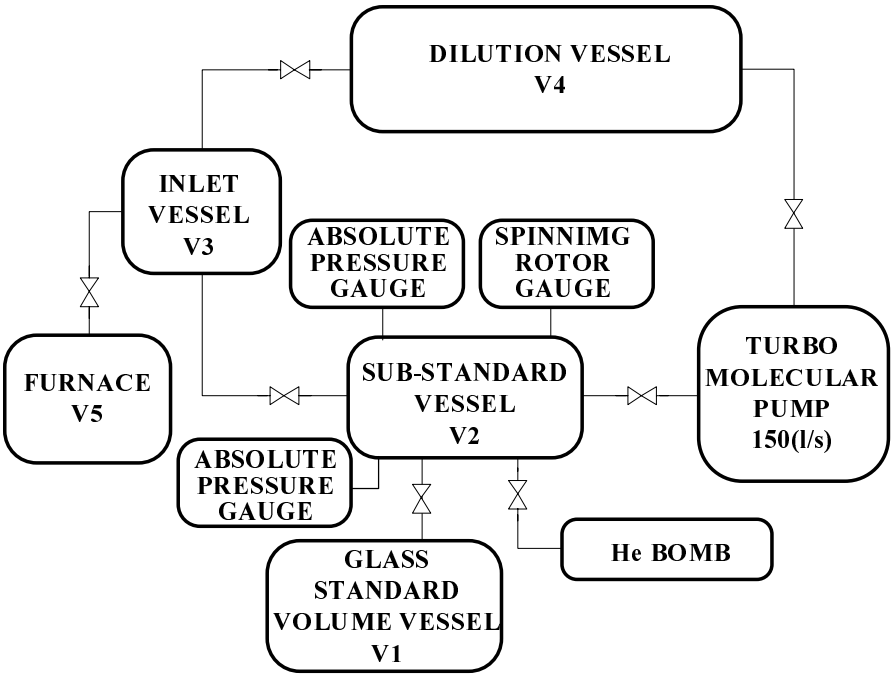

Fig.2 A block diagram of Standard He Gas Supply

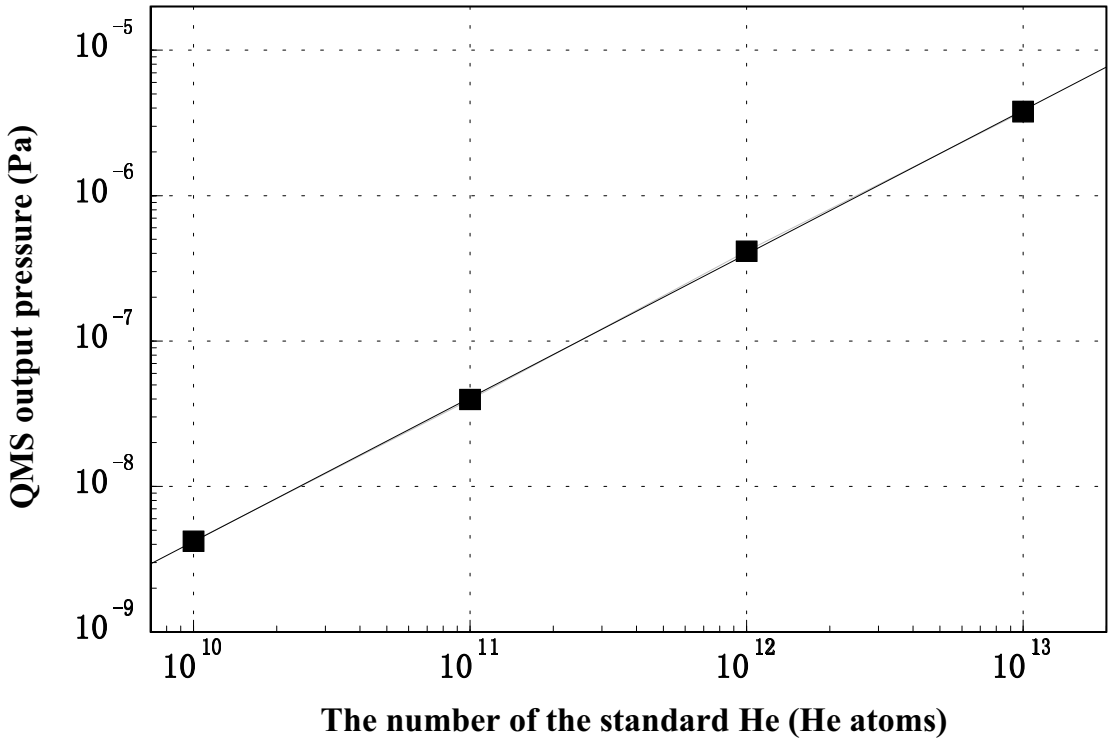

Fig.3 The results of standard He measurement in the range of 1×10^{10}～1×10^{13} He atoms

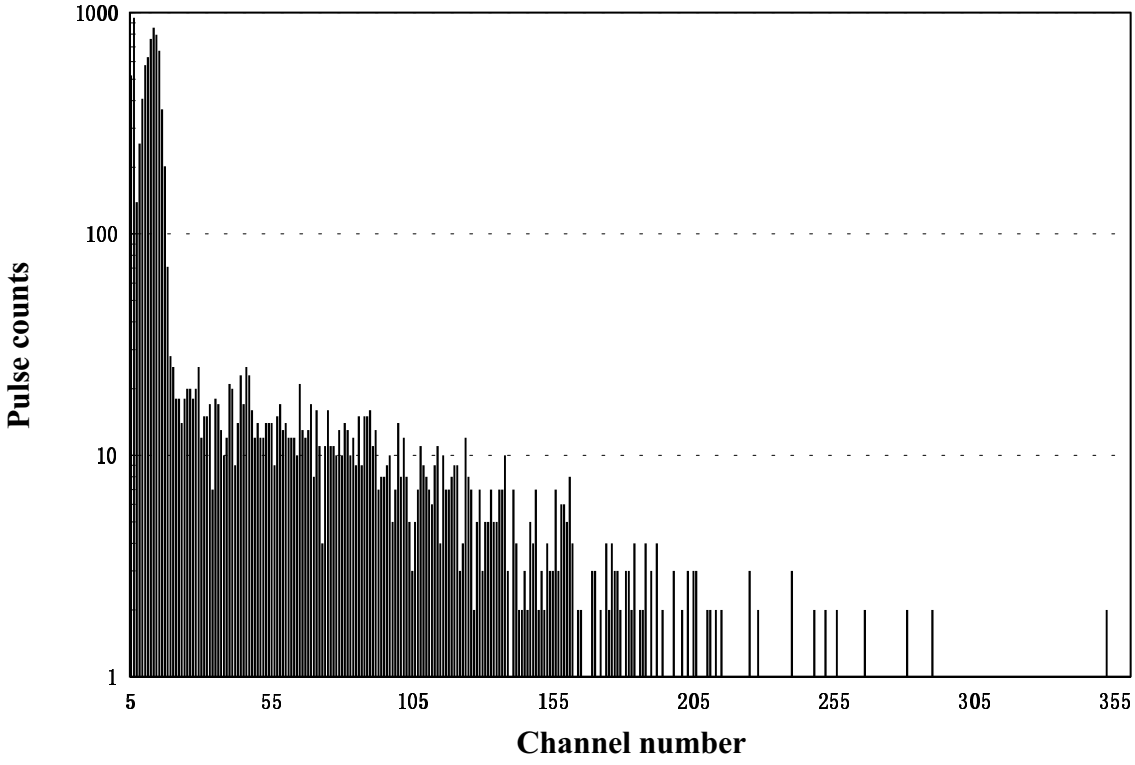

Fig.4 The result of a background measurement in the pulse counting mode