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EXPERIMENT AND ANALYSIS OF TRITIUM IN PRIMARY-LOOP OF HTR-10[1]

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ABSTRACT

Tritium is one of important nuclides in the primary loop of High-Temperature Gas-cooled Reactor. Tritium collecting device are well designed especially for HTR-10. Helium sample is introduced into this device, while tritium in the sample is collected and systematic measured. The results reveal the relationship tritium activity with power level, temperature, and etc.

1. INTRODUCTION

High purity helium is employed as primary loop coolant of 10MW high temperature gas-cooled reactor (HTR-10). It is necessary to measure the impurities, especially radioactive impurities in the primary loop coolant to prevent environmental radioactive contamination. One of the radionuclides to be measured is tritium (^{3}H).

Tritium is a pure β radionuclide, its half-life period is 12.323 years (1 year = 365.25 days), and average energy of β is 5.685keV, while the maximum energy of β is 18.582keV. Radioactivity of tritium is 3.5575×10^{14}Bq/g (9615Ci) per gram, and releases heat 0.324W/g. Tritium decays by β converted into ^{3}He:

$$^3\mathrm{H} \rightarrow \beta^- + {}^3\mathrm{He} + \bar{\nu}_e + 18.582\mathrm{keV}\ (\bar{\nu}_e\text{:anti-neutrino})$$

From the point of view of radiation protection, tritium belongs to the radionuclides of low toxicity but difficult to protect. It can penetrate the shell of the metal pipe at high temperatures. Therefore, tritium is the important radioactive contamination in the primary and secondary loop of HTR-10. Tritium is produced from the fission of ^{235}U, nuclear reaction of neutron with ^{7}Li impurities of graphite reflection layer and structures, as well as the nuclear reaction of neutron with ^{3}He in the primary loop coolant. Therefore it is necessary to measure the tritium activity in the primary loop coolant.

2. SELECTION OF TRITIUM MEASUREMENT

Tritium measurement is divided into three categories:

1. Bubbling method. For low levels of tritium, bubbling is employed to collect tritium and produce samples frequently, and then the samples are measured by a liquid scintillation spectrometer. For HTO, the gas sample is led into the bottle filled with distilled water or glycol. Most of the water vapor in the sample processes the isotope exchange with the water in bubbling apparatus. After a sufficient time interval, the total tritium activity in the water is measured by liquid scintillation counting. Based on sample gas flow rate and collection time, the total volume sample gas is calculated and the tritium activity in the sample gas. For HT, the sample gas is led into a catalytic oxidation unit and then the bubbler, finally also the liquid scintillation spectrometer. The advantage of this method is high sensitivity with low cost, but it is not suitable for real-time measurement.

2. Ionization chamber method. This method is employed to measure the tritium activity in gas samples for real-time measurements. Because of the low energy of tritium β, the gas sample must be directly introduced into the ionization chamber. Since the number of particle ions generated by each tritium β is relatively small, it do not measure a single pulse but measure the current which is based on the results of proportional to the

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current and activity of tritium. The approximate rule of thumb is: tritium produced by ionization current is 1μA/3.7×10^{10}Bq (1μA/Ci). Imagine that ionization chamber is 1 liter and tritium activity is 3.7×10^4 Bq / m^3 (1μCi / m^3), the ionization current is about 1fA, which need a very sensitive electrometer and stable and with a related electronics to achieve this sensitivity. 3.7×10^{4}Bq / m^3 (1μCi/ m^3) of tritium activity is the practical lower limit of these method.

3. Proportional counting method. Sample gas is mixed with some argon - methane gas and introduced into the proportional counter. The number of ion pairs produced by the strong electric field around the positive pole gas is amplified and increased sufficiently to produce the charge pulse, which can be detected. The number of pulses (with the appropriate energy) is proportional to the activity of tritium. By using the multi-wire chambers, pulse shapes or pulse height screening effect, it is possible to identify electronic tritium β-rays and α-rays or γ-rays produced. Theoretically, the proportional counter is sensitively than ionization chamber. However, it is very difficult to achieve such high sensitivity in practice. Table1 shows the comparisons of three ^{3}H measurement methods.

Table1 Comparisons of three ^{3}H measurement methods

^{3}H measurement method	Sensitivity	Response time	Pattern	Implementation difficulty	Expense
Bubbling method	High	Slow	Offline	Easy	Low
Ionization chamber method	Low	Fast	Online	Hard	High
Proportional counting method	Medium	Fast	Online	Very hard	Very high

The calculated value of total tritium activity in HTR-10 primary loop is 5.7×10^{9}Bq, the volume of primary loop helium under standard conditions is approximately 2000m^3, and then the tritium activity is approximately 2.85×10^{6}Bq/m^3. Any of three methods above can be employed. However, in consideration of economic factors combined with the HTR-10 characteristics, accuracy and real-time requirements, the first method is employed: Using bubbling tritium gas samples to produce liquid sample, and then liquid scintillation is employed to measure the tritium activity.

3. COLLECTION SOLUTION

The upper half of Fig.1 shows helium purification system diagram of HTR-10 (lower right branch is gas sampling collection device). It works as follows: electric isolation valves on both the left and right sides serve as boundary of primary loop and the helium purification system. Before operating the helium purification system, the electric isolation valves and valves of helium purification system must be open, and purification loop flow must be confirmed. Then diaphragm compressor starts, the flow is established and then the electric heater starts working and heat helium to the rated temperature 250℃ oxidation-reduction reaction happens on copper oxide bed:

$$H_2 + CuO \rightarrow Cu + H_2O$$

$$CO + CuO \rightarrow Cu + CO_2$$

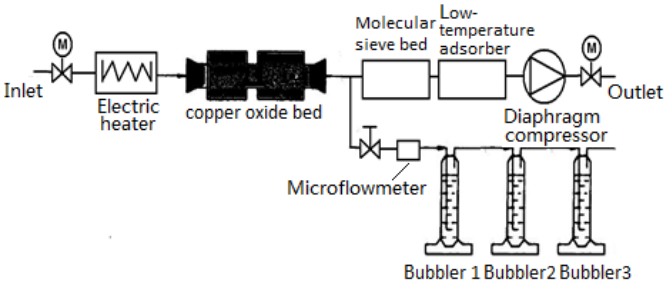

Fig.1 HTR-10 primary loop tritium collecting test schematic diagram

H_2, HT and CO in helium are oxidized to H_2O, HTO and CO_2. While, based on the results of the gas sampling and analysis system, copper oxide bed oxidation efficiency is very close to 100%. After the copper oxide bed, there is no H_2, HT and CO in the helium stream, and only H_2O, HTO and CO_2. Subsequent molecular sieve bed is employed for adsorbing the fission gas, ^{41}Ar, CH_4, steam and CO_2. Finally the low-temperature absorber, which employs activated carbon as an adsorption medium, liquid nitrogen as a cold source, all impurities (including residual fission gases, ^{41}Ar, CH_4, water vapor, CO_2 and N_2, etc.) in helium flow has not yet been cleared are adsorbed. According to the data of operation experience of German AVR of and results from gas sampling and analysis system, helium purification system for various impurities removal efficiency is about 100%. The flow is about 5% of the whole primary loop per hour, and, whole primary loop helium could be purge in approximately 20 hours.

The gas sampling port in this experiment is set in the export of copper oxide bed, as shown in Figure 1. Manual gas control valve is employed to control the flow rate to be collected, the micro flow meter is connected as an indicator, and the sample gas passes through three a bubbler. Finally, the gas discharge into the negative pressure ventilation system. The whole tritium collection devices are in a certain negative pressure, and will not contaminate the laboratory and testing personnel.

In order to improve the collection efficiency of the bubbler, in this experiment, following measures are adopted:

1) Using high precision manual valve to control sample flow rate;

2) Using high precision micro flow meter (The range is 0-130ml / min, and measurement accuracy is 5 ml/min) to measure the sample flow rate;

3) Sample flow rate control at 100 ml / min and remained stable;

4) Using three bubblers;

5) Each bubbler were cleaned with pure deionized water for ten times, it is confirmed that no radioactive contamination, and each bubbler is filled with pure deionized water 1 liter (accurate measured).

6) Inner diameter 7mm of the glass tube inserted into the sampling medium is selected and outlet is under water level of 130mm, a distance of 2mm above the bottom;

7) The sampling time is set to 20 hours, the sample gas is 120 liter;

8) Employing dew point transmitter to monitor the water vapor content of the sample gas before entering bubbler;

9) Using the thermometer to monitor the ambient temperature.

The last two are used to correct the significant changes of water vapor content in the sample gas, which may affect the deionized water bubbler mass.

4. COLLECTION SOLUTION

This experiments has been process for two times.

The first collection experiment is after 33days 3.5MW stable operation. The primary loop pressure is 1.2MPa, the total amount of helium is about 1350m^3, collection time is 20hours, the average flow rate of the sample gas is 81ml/min, and total sample gas collected is 97.5 liter. Wallac 1220 Quantulus™ ultra-low background liquid scintillation spectrometer is employed to measure the tritium activity of liquid samples. The results are as follows: Tritium activity in liquid sample from the bubbler1 is 50380Bq/liter, tritium activity in liquid sample from the bubbler2 is 314Bq/liter, while, tritium activity in liquid sample from the bubbler3 is 295Bq/liter. Obviously, the bubbler1 collection efficiency is more than 99%. Then the tritium activity of primary helium calculated: 5.22×10^5Bq/m^3, a total activity tritium of the primary loop is 7.05×10^8Bq.

The second collection experiment is in the case of the 3.1MW normal operation of the reactor. Helium is added to primary loop before operation, and the pressure rise from 1.2MPa to1.6MPa. The total helium is about 1800m^3, To test the memory effects of tritium, bubbler1 and bubbler3 are reversed during the collection, the sample gas will be followed by the number 3,2,1 bubbling device, collection time 20 hours, an average flow rate of the sample gas 90ml / min total collected sample gas 108.0 liter. The results are as follows: Tritium activity in liquid samples from bubbler1 is 558Bq/liter, the tritium activity in liquid sample from bubbler2 is 461Bq/liter, while tritium activity in liquid sample from bubbler3 is 38889Bq/liter. Bubbler2 collection efficiency is still higher than 98%, although the bubbler has been repeatedly washing (washing with pure deionized water 10 times or more), the memory effect tritium remains. Then the tritium activity of primary helium calculated: 3.67×10^5Bq/m^3, a total activity tritium of the primary loop is 6.61×10^8Bq.

5. EXPERIMENT RESULTS ANALYSIS

Compared two results (7.05×10^8 Bq and 6.61×10^8 Bq) with the theoretical value (5.7×10^9Bq) of the total tritium activity of primary loop, it can be seen that experiment results is less than theoretical value nearly an order of magnitude lower. But it does not demonstrate the theoretical calculations serious bias, the main reasons are as follows:

1) The theoretical value is in the more conservative parameter selection. In order to ensure the safety of the reactor and the actual impact on the surrounding environment, the parameters are chosen to leave some margin. This is the rules that the reactor design should follow;

2) HTR-10 power running time is relatively short, the rate of accumulation and release of tritium does not reach equilibrium level;

3) Helium purification system long-term stable operation, has effectively remove a certain amount of tritium in the primary loop coolant.

6. CONCLUSIONS

So far, although the measurements results of the HTR-10 primary loop coolant tritium activity is still significantly rough, and some measurement error remains. It still needs to be further accurate experiments to be processed. However, this two results still show that the theoretical value of a loop HTR-10 total tritium activity is reliable. According to the data obtained, it can be estimated roughly that total amount of tritium leaking to the outside of a loop every day: the leakage rate measured primary coolant is about 7‰, assuming tritium and helium have same leak rate, the tritium released into the environment every day from HTR-10 is 4.78×10^6Bq (average of two tritium activity results), which is much lower than "10MW high temperature gas-cooled reactor final safety analysis report".

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