

# EXPERIMENTAL STUDY OF TRITIUM ACTIVITY CONCENTRATION IN COOLING WATER OF HEAVY ION RESEARCH FACILITY IN LANZHOU

by

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Cooling water of Heavy Ion Research Facility in Lanzhou is the media of choice for absorbing heat generated by a multitude of accelerator components. During the operation, tritium is produced by the secondary neutrons in the accelerator loop of accelerator cooling water, which is uniformly distributed in the water system in the form of hydrogen tritium oxide. The purpose of this study is to establish a rigorous method to measure the low levels of tritium in the accelerator cooling water system. The cooling water samples of Sector Focusing Cyclotron and Separated Sector Cyclotron of Heavy Ion Research Facility in Lanzhou, as well as the natural levels of tritium, were measured successfully, using Quantulus1220 ultra-low-level background liquid scintillation spectrometer. The electrolytic enrichment method was employed to measure the tritium activity concentrations of the cooling water samples. Moreover, the minimum detectable activity concentration and the counting rate efficiency curve of the liquid scintillation spectrometer were obtained to ensure reliability. In this work, we provide a reference to establish the related standards and specifications for monitoring the tritium levels in heavy-ion accelerator cooling water system.

*Key words: tritium, activity concentration, cooling water, accelerator*

## INTRODUCTION

Tritium ( $^3\text{H}$ ) is a radioactive isotope with a physical half-life of 4500 days [1]. It is created in nature by cosmic rays interacting with the atmosphere, nuclear explosion, and nuclear facility operations, such as nuclear power plants and particle accelerators.

The Heavy Ion Research Facility in Lanzhou (HIRFL) consists of the Sector Focusing Cyclotron (SFC), the Separated Sector Cyclotron (SSC), and the cooler storage ring (CSR) [2]. As SSC runs in conjunction with SFC, the maximum energy of  $^{12}\text{C}^{4+}$  is 1200 MeV, and the corresponding current is 60  $\mu\text{A}$ , while the maximum energy of  $^{209}\text{Bi}^{31+}$  is 1985.5 MeV, and the corresponding current is 45 nA [3].

The cooling water of HIRFL is the media of choice for absorbing heat generated by a multitude of accelerator components. A flow diagram of the cooling water system of SFC and SSC in HIRFL is shown in fig. 1. It consists of two loops, which are the second-

ary loop and the accelerator loop. During the operation of the accelerator, tritium was produced by the secondary neutrons in the accelerator loop. Thus, the tritium concentration in cooling water might be higher than the natural levels in the vicinity of HIRFL. Tritium enters the environment usually as a replacement for one of the hydrogen atoms in water, which is uniformly distributed into the water system with the most probable form of Hydrogen Tritium Oxide [4, 5], which can be observed in a local radioactive impact. However, there is no standard method to guide the measurement of tritium in the accelerator cooling water system. It is necessary to carry out an experimental study on the activity concentration of tritium in accelerator cooling water at the Lanzhou heavy ion research facility.

The purpose of this experimental study is to establish a rigorous method to measure the low-level of tritium in accelerator cooling water. The research was performed in four steps: Design and machining of electrolytic enrichment device, Electrolytic enrichment, Measurement, and Analysis. The results show that the tritium concentration in SFC and SSC of

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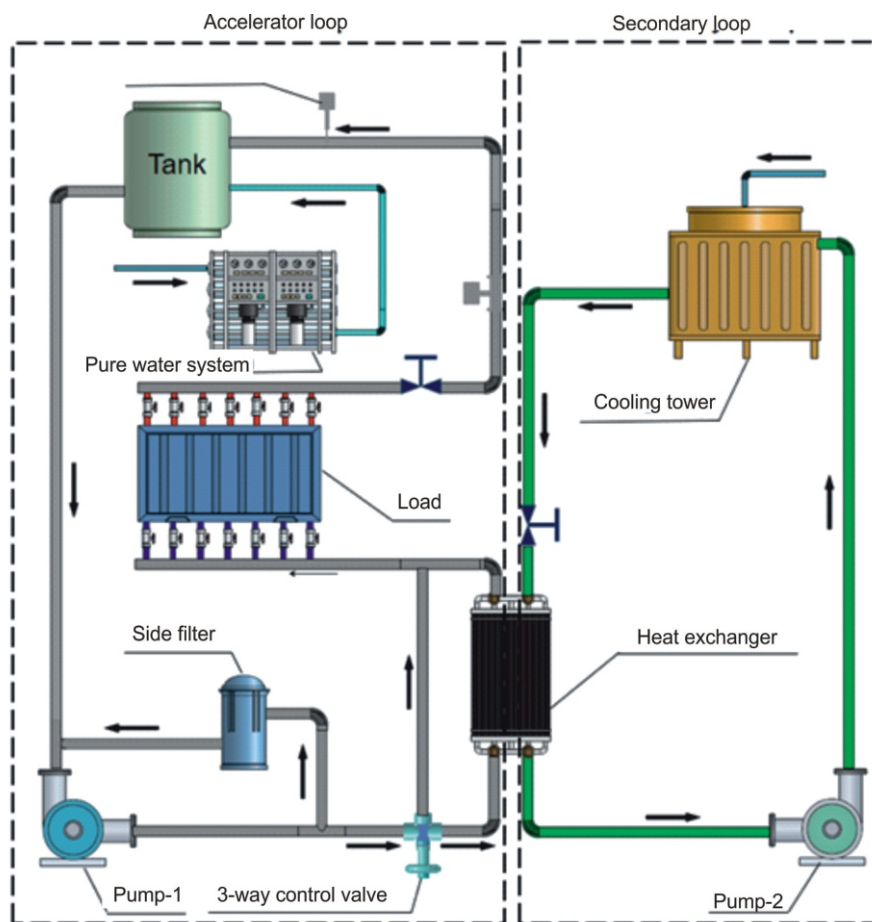


Figure 1. Flow diagram of the cooling water system of the SSC and SFC in HIRFL

HIRFL were 3.268 and 3.293 times higher than that in the background sample, respectively. According to the results, appropriate advice will be provided for monitoring the activity concentration of tritium in accelerator cooling water.

## EXPERIMENTAL PROCEDURE

Tritium is a soft beta-emitter radionuclide with a maximum energy of 18.6 keV. Based on the radioactivity, Tritium can be observed by measuring the electrons produced by the decay of tritium or by radiometric measurements. The common methods for low-level counting of tritium are either using gas-proportional counting or liquid scintillation counting [6, 7]. Considering the low-level counting rate, the Quantulus1220 ultra-low-level background liquid scintillation spectrometer produced by Perkin Elmer was employed.

### Design and machining of electrolytic enrichment device

Due to the low-level counting rate in the water samples, it is necessary to improve the tritium concen-

tration by electrolytic enrichment [8, 9]. According to GB12375-90 *Analytical method of tritium in Water* [10], a tritium electrolytic enrichment device in accelerator cooling water was developed. The main unit of the electrolytic enrichment device is shown in fig. 2. Auxiliary devices include the power supply system and the cooling system. The anode is a nickel (purity: 99.9 %) tube and the cathode is a mild steel tube. The anode and cathode are connected to Teflon insulation sealed spacer.

The anode has a vent, through which the gases produced by electrolysis can enter the liquid condensation tube. The water vapor, as well as oxygen and hydrogen gases produced by electrolysis, condensed in the condensing tube, and the condensed water returns to the electrolytic cell, which could reduce the water loss and improve the electrolytic recovery of tritium.

The outside of the cathode is designed as a double-layer structure. The interlayer is connected with anhydrous ethanol, which is connected with the cooling circulator to transfer the heat generated by electrolysis and relieve the electrochemical corrosion caused by the rise of electrolyte temperature. The temperature of anhydrous ethanol is 5 °C. The electrolytic enrichment device is supplied by a stabilized DC power supply with a working voltage of 3 V.

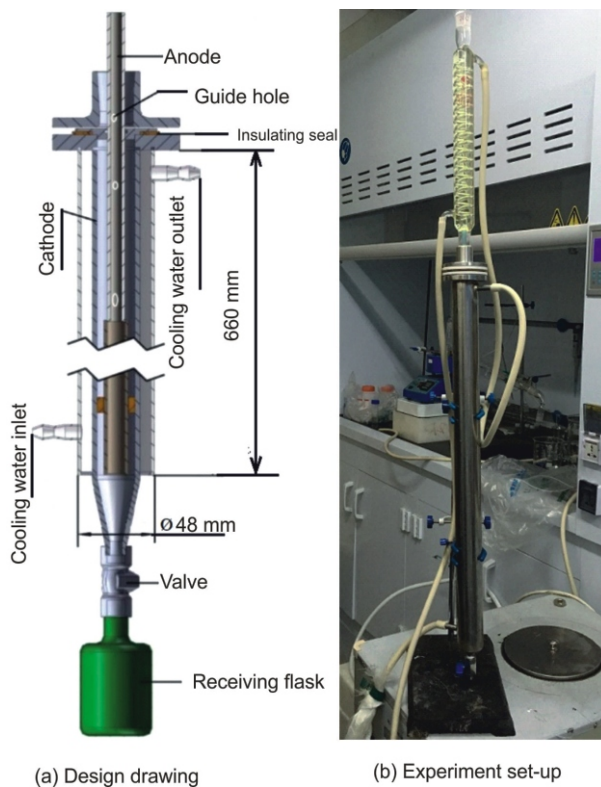


Figure 2. Tritium electrolytic enrichment device

## Electrolytic enrichment

### *Cleaning electrolytic cell*

Before the experiment, a concentrated hydrochloric acid (50 %) and a sodium hydroxide solution (10 %) are configured to remove oil and dust impurities in the electrolytic cell. The cell was first cleaned with concentrated hydrochloric acid, and then with sodium hydroxide, and finally washed with anhydrous ethanol. After the electrolysis of each sample, the electrolytic cell should be cleaned in time. The cleaning steps involve triple rinsing with tap water, ultrapure water, and anhydrous ethanol.

### *Sampling*

The cooling water used in the accelerator loop is ultra-pure water. The 500 ml cooling water from SFC and SSC were collected after the side filter, filtered by the purified resin, and the samples are labeled as  $S_1$  and  $S_2$ , respectively. Another 500 ml ultra-pure water was collected from the return pipe of the accelerator cooling water system and labeled as  $B_0$ .

### *Electrolytic enrichment*

Three groups of samples ( $S_1$ ,  $S_2$ , and  $B_0$ ) were decomposed by electrolysis, respectively. For each sample, 2.5 g of NaOH was added to 250 ml of the previously distilled water. The NaOH is an alkaline me-

dium favorable to electric conduction and hence the reaction of water electrolysis is performed. When the sample solution volume was reduced to 8 ml, the anode disengaged from the liquid level and the electrolysis stopped. The total time of electrolysis of one sample was about 140 hours.

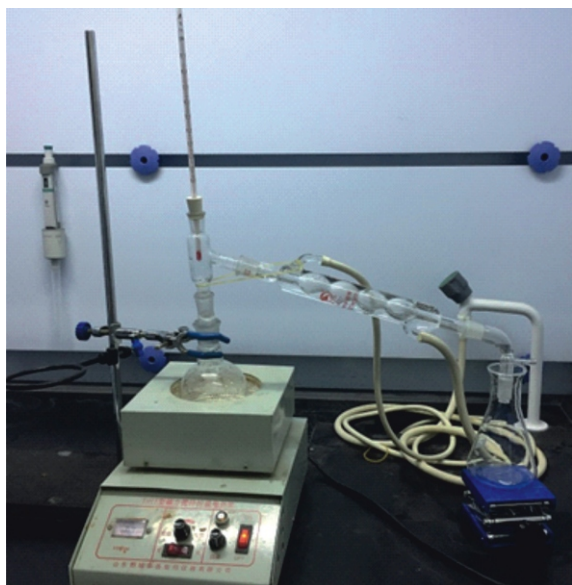
### *Neutralization distillation*

To reduce the quenching effect during the measurement using a liquid scintillation spectrometer, the residual electrolyte was neutralized and distilled with  $\text{CO}_2$  to reduce the amount of NaOH in the samples. The residual electrolyte treatment apparatus is shown in fig. 3.

## Measurement

For each sample, 8 mL of the residual electrolyte was collected after neutralization and distillation. The diluted residual electrolyte was transferred into the liquid flash measuring bottle, together with 12 ml of scintillating cocktail (Ultima Gold TM), and the solution was thoroughly mixed. The mixed sample was stored in dark for 24 hours and then measured with the Quantulus 1220 liquid scintillation spectrometer.

The concentration ratio of tritium after electrolysis is equal to the ratio of the activity concentration of tritium in the concentrated solution of electrolysis to the initial activity concentration of the sample. The quenching correction is the ratio method [11].



**Figure 3. Electrolysis residual liquid distillation apparatus**

#### Counting efficiency of liquid scintillation spectrometer

To obtain the activity concentration of the measured sample, it is necessary to measure the relationship between the counting rate and decay rate of samples with different activity concentrations. The counting efficiency  $E$  of the liquid scintillator measuring instrument is also measured in this stage [12].

In this experiment, the efficiency calibration of ultra-low background liquid scintillation spectrometer was conducted by the ratio method using eq. (1)

$$E = \frac{N_s}{A_0} \frac{N_b}{100\%} \quad (1)$$

where  $N_s$  is the counting rate of standard tritium solution, in pulses per second,  $N_b$  – the counting rate of the black sample, in pulses per second, and  $A_0$  – the activity of standard tritium solution. The decay per minute (DPM) of tritium in each sample was then obtained using the following eq. (2).

$$DPM = \frac{N_d}{E} \frac{N_b}{100\%} \quad (2)$$

where  $N_d$  is the counting rate of the sample, in pulses per minute, and  $N_b$  – the counting rate of the black sample, in pulses per minute.

The sample activity is obtained by eq. (3)

$$A = \frac{DPM}{60} \frac{V_1}{V_2} \frac{1}{T} \frac{1}{V_3} \quad (3)$$

where  $A$  [ $\text{BqL}^{-1}$ ] is the activity concentration of tritium in the sample,  $V_1$  [mL] – a volume of diluted samples.  $V_2$  [mL] – a volume of diluted samples used for measurement.  $V_3$  [L] – the concentration volume of sam-

ples, and  $T$  – the concentration multiple which is obtained by eq. (4).

$$T = \frac{A_s}{A} \quad (4)$$

where  $A_s$  is the activity concentration of tritium after electrolysis, [ $\text{BqL}^{-1}$ ].

Since the sample was treated by electrolysis concentration and distillation, the electrolysis concentration multiple of tritium and the collection efficiency of distillation should be considered when calculating the activity concentration of tritium in the sample [13]. The SS is an 8 ml electrolyte residual liquid obtained by electrolytic distillation of 250 ml  $N_2$  with an activity concentration of 100  $\text{BqL}^{-1}$ . The activity concentration of SS is 48.50  $\text{BqL}^{-1}$ . It can be obtained by considering the measured value of SS, counting efficiency curve, and distillation loss correction.

The minimum detectable activity concentration is calculated by eq. (5).

$$L_D = \frac{4.65\sqrt{N_b/T_b}}{60E\eta V} \quad (5)$$

where  $N_b$  is the background counting rate,  $T_b$  [min] – the background sample counting time.  $E$  [%] – the sample counting efficiency.  $V$  [L] – the sample volume.

## RESULTS ANALYSIS

The volumes and dilution ratio of each residual electrolyte after distillation are shown in tab. 1.

Standard tritium solutions with different activity concentrations were prepared based on the initial activity concentration of 15.40  $\text{Bqmg}^{-1}$  (The production date is April 21, 2017). The specific values are shown in tab. 2.

The efficiency curve of the ultra-low-level background liquid scintillation spectrometer is shown in fig. 4. The  $Y$  is DPM and  $X$  is activity concentration.

The measured background sample volume is 8 ml, and the measurement time of the background sample is 1 minute. The counting efficiency of the background sample is 36.73 %. The minimum detectable activity concentration is 0.32  $\text{BqL}^{-1}$ . Thus, the counting rate of the background sample is 1.4223 DPM. Table 3 shows the activity concentration of  $^3\text{H}$  in cooling water in SFC and SSC.

According to the analysis that has been aforementioned, the background activity concentration of tritium in the accelerator cooling water is 0.41 ± 0.02  $\text{BqL}^{-1}$ , the activity concentrations of tritium in  $S_1$  and  $S_2$  is 1.34 ± 0.04  $\text{BqL}^{-1}$  and 1.35 ± 0.04  $\text{BqL}^{-1}$ , respectively.

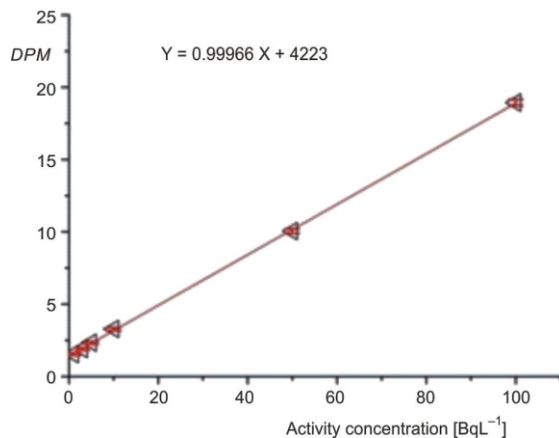
For the accelerator, due to the manufacturing error of the main magnet in the cyclotron and space charge effect, some particles will sense incorrect trans-

**Table 1. The volumes of each sample after distillation**

Sample	The volumes after distillation [mL]	Dilution ratio
$B_0$	2.00	4.00
$S_1$	5.60	1.43
$S_2$	7.00	1.41
SS	7.90	1.01

**Table 2. Standard tritium with different activity concentrations**

Sample	The activity concentration [ $\text{BqL}^{-1}$ ]	Measured value [Bq]	Error [%]
$N_1$	555.74	$1.720 \cdot 10^0$	0.451
$N_2$	94.97	$3.158 \cdot 10^{-1}$	1.103
$N_3$	9.50	$5.467 \cdot 10^{-2}$	3.291
$N_4$	4.75	$3.883 \cdot 10^{-2}$	4.243
$N_5$	2.85	$3.183 \cdot 10^{-2}$	4.934
$N_6$	0.95	$2.583 \cdot 10^{-2}$	5.803
$N_7$	0.66	$2.567 \cdot 10^{-2}$	5.833
$N_8$	0.47	$2.500 \cdot 10^{-2}$	6.142
$N_9$	0.28	$2.317 \cdot 10^{-2}$	6.340
$N_{10}$	0.09	$2.183 \cdot 10^{-2}$	6.631
$N_{11}$	0.05	$2.450 \cdot 10^{-2}$	6.075
$N_{12}$	47.48	$1.675 \cdot 10^{-1}$	1.591

**Figure 4. Counting rate efficiency curve of Quantulus1220****Table 3. Activity concentration of  $^3\text{H}$  in cooling water in SFC and SSC**

Sample	Measured value [Bq]	Activity concentration of tritium [ $\text{BqL}^{-1}$ ]	Error %
$S_1$	$6.517 \cdot 10^{-2}$	1.34	2.915
$S_2$	$6.550 \cdot 10^{-2}$	1.35	2.904
$B_0$	$2.367 \cdot 10^{-2}$	0.41	3.891

verse and longitudinal focusing magnetic field and lost on the vacuum chamber wall along the beam transmission path finally. The lost particles interact with the surrounding material to produce secondary neutrons, which cause the activation of water. It can be seen that the activity concentration of tritium in SFC and SSC

cooling water were about 3.3 times the background sample since the cooling water running time of SFC and SSC is the same, about 3 months, each.

## CONCLUSIONS

An electrolytic enrichment method to measure the tritium activity concentrations in accelerator cooling water has been developed based on the experiment using the cooling water of HIRFL. The minimum detectable activity concentration and the counting rate efficiency curve of Quantulus1220 ultra-low-level background liquid scintillation spectrometer were measured. The activity concentrations obtained when the counting rate is over the minimum detectable activity concentration are reliable within the uncertainty. Thus, not only cooling water samples of SFC and SSC but also natural levels of tritium have been measured successfully.

This result can be provided as a concept for estimating the induced radioactivity in accelerator cooling water. Also, it provides a reference to establish the related standards and specifications for monitoring the activity concentration of tritium in particle accelerator cooling water.

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## AUTHORS' CONTRIBUTIONS

The idea for the study was put forward by C. Xu and Y. Su. The samples were prepared by C. Xu, Y. Wang, and T. Zhu. The measurements were carried out by C. Xu and Y. Wang. The discussion was carried by C. Xu, H. Wang, J. Zuo, and J. Han. The manuscript and figures were prepared by C. Xu.

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**ЕКСПЕРИМЕНТАЛНО ПРОУЧАВАЊЕ КОНЦЕНТРАЦИЈЕ  
АКТИВНОСТИ ТРИЦИЈУМА У ВОДИ ЗА ХЛАЂЕЊЕ ТЕШКОЈОНСКОГ  
ИСТРАЖИВАЧКОГ ПОСТРОЈЕЊА У ЛАНЦОУ**

Расхладна вода из постројења за истраживање тешких јона у Ланцоу, одабрани је медијум за апсорпцију топлоте генерисане мноштвом компоненти акцелератора. Током рада, трицијум се производи секундарним неутронима у акцелераторској петљи са водом за хлађење акцелератора, који се равномерно распоређује у води система у облику водоник-трицијумоксида. Сврха ове студије је да успостави прецизну методу за мерење ниског нивоа трицијума у систему за хлађење акцелератора водом. Узорци расхладне воде секторског фокусирајућег циклотрона и издвојеног секторског циклотрона тешкојонског истраживачког постројења у Ланцоу, као и природни ниови трицијума, успешно су мерени помоћу Quantulus1220 – течног сцинтилационог спектрометра ултра-ниског позадинског зрачења. Метода електролитичког обогаћивања коришћена је за мерење концентрација активности трицијума у узорцима расхладне воде. Штавише, како би се осигурала поузданост, добијена је минимална концентрација активности која се може уочити као и крива ефикасности брзине бројања течног сцинтилационог спектрометра. Овим радом обезбеђујемо референцу за успостављање сродних стандарда и спецификација за праћење нивоа трицијума у систему за хлађење водом акцелератора тешких јона.

*Кључне речи:* трицијум, концентрација активности, расхлада вода, акцелератор