ICANS-XIV
14th Meeting of the International Collaboration on
Advanced Neutron Sources
June 14-19, 1998
Starved Rock Lodge, Utica, Illinois

Behavior of ^{7}Be in the Moderator Cooling System at the Pulsed Neutron Source, KENS

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ABSTRACT

We measured the radioactivity of ^{7}Be and ^{3}H in the cooling system of the ambient water moderator at KENS in order to obtain some knowledge about the behavior of ^{7}Be in the cooling water.

1. Introduction

At high-power pulsed-neutron facilities, some difficult operational problems arise in water cooling systems: ^{7}Be produced by the spallation reaction of oxygen nuclei in water with high-energy hadrons tends to coat the inside of the piping of the cooling systems. This leads to quite high radiation levels around the piping. The dose rate around the piping can be as high as 500 μSv/h at the ISIS Facility at Rutherford Appleton Laboratory, for instance [1]. At a higher-power neutron source, the dose rates will be much higher, and, therefore, the cooling system must be designed based on the radiation safety from ^{7}Be. It has been reported that the coating rate of ^{7}Be onto the container depends on the hydrogen exponent (pH) of water as well as the materials of the containers [2]. In the design of cooling systems in high-power facilities, it is important to have some knowledge about such behavior of ^{7}Be in cooling systems. The moderator cooling system at the pulsed spallation neutron source, KENS, at High Energy Accelerator Research Organization (KEK) is ideal for such investigations, because it is a simple cooling system where the flux of neutrons can be easily estimated. We have investigated the radioactivity of ^{7}Be and ^{3}H in the moderator cooling system to obtain some knowledge about the behavior of ^{7}Be.

2. Experimental

Figure 1 shows the target-moderator-reflector assembly at KENS. The proton beam with 500 MeV

Key words: ^{7}Be, activation of cooling water, ambient water moderator

hits the target with a cross section of 7.7×5.6 cm^2 and a length of 12 cm. The target consists of four tantalum blocks, and there are cooling water channels between the blocks perpendicular to the proton beam. The moderator of ambient water (H_2O) with 10×10×5 cm^3 is located under the target, and a neutron beam is provided from the moderator surface with the cross section of 10×10 cm^2 to the spectrometers. The water is supplied from its cooling circuit to the moderator (Fig.1). The cooling circuit consists of a pump, a surge tank, an ion-exchanger, a filter and a radiator. The pumping rate of the pump is 50 cm^3/s, and the water is supplied with a flow rate of 5 cm^3/s and 45 cm^3/s to the moderator and the radiator, respectively. The cooling water from the moderator and that from the radiator merge at the surge tank. The total volume of the water in the circuit is 2.5×10^4 cm^3. During the KENS machine cycle, Cycle 97-8 (9:00 13 Feb. 1998 - 9:00 23 Feb. 1998), the target was hit by 2.4×10^{19} protons. After the cycle, sampling of water in the moderator cooling system was performed from the sampling port located at the bottom of the surge tank, at t=0, 10, 20, 30 minutes, 1,2,3,6,9 hours, 1,2,3 and 7 days, where t is the time from the end of the cycle (9:00 23 Feb. 1998). The radioactivity and pH of each sample water were measured. The background radioactivity was also measured before the Cycle 97-8.

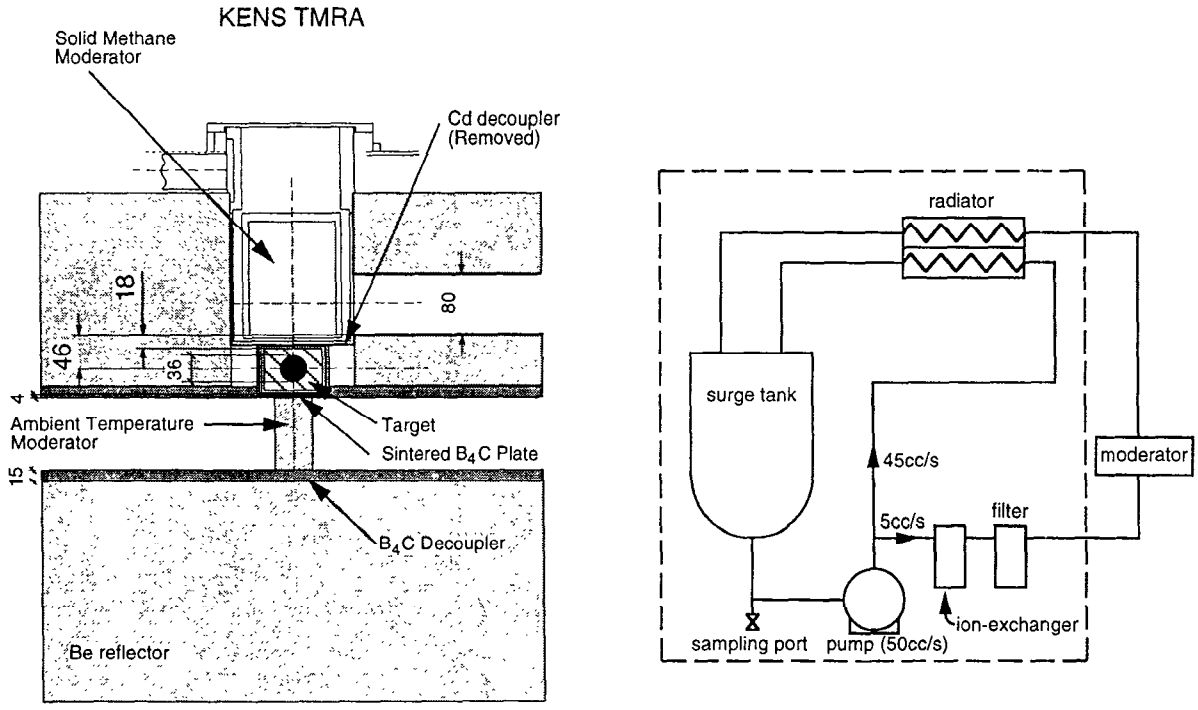

Fig.1 Target-moderator-reflector assembly at KENS (left) and schematic diagram of the cooling circuit of the water moderator (right).

3. Neutronics calculation and production rate

A neutronics calculation was performed by using LCS, in order to obtain the neutron flux in the moderator with the geometrical configuration shown in Fig.1. The flux is the averaged value over the moderator volume. Since the averaged proton current during the cycle was 4.5 μA, the neutron

flux with high energies greater than 20MeV was calculated to be $\phi_{HE}=2.0\times10^{10}$ cm^{-2}s^{-1}. From the energy spectrum of the neutron flux ($\phi(E)$) and the excitation functions ($\sigma(E)$) [3-5], the averaged production cross section for neutron energies greater than 20MeV was calculated as follows:

$$\langle\sigma\rangle = \int_{20MeV}^{500MeV}\sigma(E)\phi(E)dE \Big/ \int_{20MeV}^{500MeV}\phi(E)dE \quad (1)$$

and was obtained to be 12.2 mb and 7.6 mb for ^{3}H and ^{7}Be, respectively. The calculated flux of the thermal neutrons was well fitted to a Maxwellian centered at 27 meV, and the Maxwellian integral was obtained to be $\phi_{th}=6.7\times10^{10}$ cm^{-2}s^{-1} for 4.5 μA.

4. Detected activity in water samples

First, the detected activities of ^{3}H and ^{7}Be before and just after the cycle are summarized in Table I. The detected activities (A_{det}) in the moderator are the values at the end of the cycles, where the decay of the background activity is corrected. The activity (A_o) produced in the moderator during the cycle can be estimated from the neutron flux and the averaged production cross section described above. The detected activity of ^{3}H almost follows the estimation; however, that of ^{7}Be is only about 0.1% of the estimation. Next, the t-dependence of the detected activity ($A_D(t)$) of ^{7}Be and ^{3}H in the moderator are plotted in Fig.2. $A_D(t)$ is the detected activity at t, the sampling time from the beam stop, where the decay of the nucleus is corrected to the value at t=0, but the background activity measured before the cycle is not corrected. Although $A_D(t)$ of ^{3}H is independent of t, that of ^{7}Be decreases with increasing t as well fitted to the following formula:

$$A_D(t) = A_D(0)\ \{a\exp(-t/\tau_1)+(1-a)\exp(-t/\tau_2)\}. \quad (2)$$

The following parameters were obtained by the fitting: $\tau_1=2.1\times10^4$s, $\tau_2=5.1\times10^5$s and a=0.53. $A_D(0)$ was detected to be 24.0 Bq/cm^3. Although the measured the pH were scattered (this may have depended on the condition of measuring pH: pH decreases when CO_2 in the air is dissolved in water), pH was roughly 7.5.

Table I. The detected activities of ^{3}H and ^{7}Be before and after the cycle. The detected activities (A_{det}) in the moderator are the values at the end of the cycles, where the decay of the background activity is corrected. The activity (A_o) produced in the moderator during the cycle can be estimated from the neutron flux and the averaged production cross section.

RI	activity (Bq/cm^3)		A_{det} (Bq/cm^3)	A_o (Bq/cm^3)	A_{det}/A_o
	before cycle	after cycle			
^{3}H	1580	1860	280	248	1.13
^{7}Be	11.8	24.0	13.7	1.23×10^4	0.0011

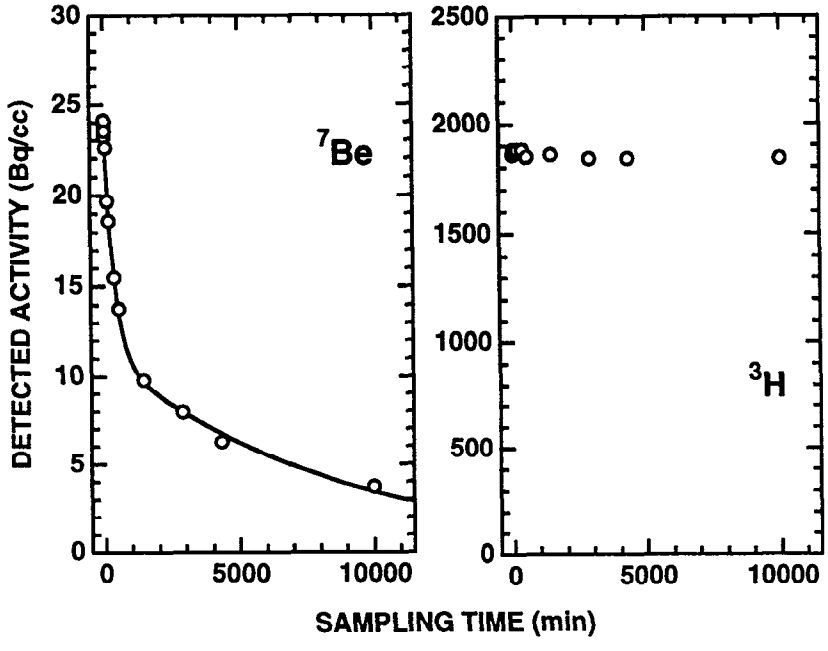

Fig.2 Sampling-time dependence of detected activity of ^{3}H and ^{7}Be. The solid line is a fitted curve (see text).

In the moderator, A_{det} of ^{3}H almost follows the estimation, and $A_D(t)$ of ^{3}H is independent of t. This indicates that the produced ^{3}H homogeneously exists in water of the circuit. On the other hand, A_{det} of ^{7}Be is about 0.1% of the estimation, and $A_D(t)$ depends on t. This suggests that the produced ^{7}Be is trapped inside of the circuit. It is noted that the measured pH was greater than 7, and therefore, the condition for coating of ^{7}Be onto the inside walls of the circuit was realized [2]. We try here to evaluate the detected activity of ^{7}Be by using the time constants obtained above. Since the obtained time constants are much less than the duration of the cycle (irradiation time), the background activity measured before the cycle need not be considered. At t=0, the ratio of the detected activity to A_0 is presented by

$$A_D(0)/A_0 = (1-c)A/A_0 + cA'/A_0, \tag{3}$$

where A is the activity estimated from the obtained time constants and A' is the activity due to a process other than that detected at present. A/A_0 is given by

$$A/A_0=\{\Sigma_i a_i(\lambda_0/\lambda_i)(1-\exp(-\lambda_i T))\}/(1-\exp(-\lambda_0 T)), \tag{4}$$

with the duration of the cycle (T), the decay constant of ^{7}Be ($\lambda_0=1.5\times10^{-7}s^{-1}$), $\lambda_i=\lambda_0+\tau_i^{-1}$ (i=1,2), $a_1=a$ and $a_2=1-a$. A/A_0 was calculated to be 0.26 and $A_D(0)/A_0$ 0.002. A' is negligibly small, because no other process described by eq. (2) was detected; eventually, c was obtained to be 0.99. The trapped activity is given by $(1-c)(1-A/A_0)$ and $c(1-A'/A_0)$ due to the presently-detected process and processes other than the present detection, and was estimated to be 0.006 and 0.992, respectively. This indicates that an overwhelming amount of ^{7}Be due to processes other than the

present detection is trapped in the cooling water circuit. Assuming that A' has a similar functional form to eq. (4) with the time constant τ', the upper limit of τ' can be estimated from $A_D(0)/A_0 > cA'/A_0$, and is obtained to be $\tau' < 1.6\times10^3$s ($\tau' < 10$s, if $A_D(0)$ in the inequality is replaced by its experimental error). This result suggests that almost all of the produced ^{7}Be is trapped with a very short time constant.

5. Distribution of ^{7}Be in the cooling circuit

In order to detect the activity due to processes other than the present detection, which should be almost A_0, the activity of a part of the piping was directly measured. A_0 was calculated to be 12 kBq/cm^3, *i.e.*, the total activity in all the circuit was 300 MBq. The sample of the piping was 6 cm of length and 0.8 cm of inside diameter, and the activity due to ^{7}Be inside the sample piping was measured to be 1.5 kBq 133 days after stopping the beam. The activity was estimated to be 8.5 kBq at the beam stop by correcting the decay of ^{7}Be. This activity is an integrated one for operation over about ten years. It is estimated that 60% of the detected activity is the production during the cycle of 10 days. Therefore, the activity of 5.1 kBq is the contribution to the cycle. In the following we discuss the contribution to the cycle. Since the surface area of the inside of all parts in the cooling circuit (except for the elements of the ion-exchanger and the filter) is 4.3×10^4 cm^2, the total activity and the averaged activity over the cooling circuit can be estimated to be 15 MBq and 0.6 kBq/cm^3, respectively, if ^{7}Be uniformly coats the inside of the cooling circuit. In order to know the distribution of ^{7}Be in the cooling circuit, the intensity of γ-ray was measured from outside parts of the circuit, except for the moderator, as listed in Table II. The measurement was performed by using a NaI detector, and the intensity is of the integrated energy spectrum including the peak of 477 keV of γ-ray from ^{7}Be. From the result, assuming the concentration of ^{7}Be in the ion-exchanger to be 10 times as large as that in the other parts, the total activity and the averaged activity over the cooling circuit can be estimated to be 41 MBq and 1.6 kBq/cm^3, respectively; this

Table II. The intensity of γ-ray at the outside parts of the cooling circuit measured by a NaI detector. The intensity (γ-ray counts) is of the integrated energy spectrum including the peak of 477 keV of γ-ray from ^{7}Be.

part of cooling circuit		γ-ray counts (10^4min^{-1})
ion-exchanger	upper part	213
	middle part	119
	lower part	62
filter	upper part	44
	lower part	44
surge tank	upper part	24
	middle part	37
	lower part	30
radiator		13
piping near surge tank		16
piping	circuit shielding outside	2
outside wall	circuit shielding outside	1

is still less than A_0. Since the activity from the ion-exchanger is very huge, the background for the other parts increases. If the concentration in the ion-exchanger is 100 times as large as that in the other parts, the total activity and the averaged activity can be estimated to be 300 MBq and 12 kBq/cm^3, respectively; this reproduces A_0. Although a quantitative discussion cannot be made further based on this γ-ray measurement on the outside parts of the circuit, this indicates the detection of a part of the activity due to the process other than the present detection in the sampling of water. Considering the short time constant of processes other than the present detection, it is also possible that a large amount of the produced ^{7}Be is trapped in the moderator, or near there. To elucidate the distribution of ^{7}Be in the circuit, a direct measurement of the activity from ^{7}Be in the inside moderator is also required, for instance. It is noted that ^{7}Be has a very large absorption cross section (σ_a=3.9×10^4 b) for thermal neutrons [6]; however, $\phi_{th}\sigma_a$ (=2.5×10^{-9}s^{-1}) is expected to be smaller than λ_0, τ_i^{-1} and τ'^{-1}. Therefore, the disappearance due to absorption is not significant compared with the trapping process.

6. Summary

In summary, we performed measurements of the activity of ^{7}Be and ^{3}H in the moderator cooling system at KENS in order to obtain some knowledge about the behavior of ^{7}Be in the cooling water. The detected activity of ^{3}H almost follows the estimated production, and the activity of ^{3}H in the sampling is independent of the sampling time. This indicates that the produced ^{3}H homogeneously exists in the water of the circuit. On the other hand, the detected activity of ^{7}Be is about 0.1% of the estimated production, and the activity in the sampling depends on the sampling time. This suggests that the produced ^{7}Be is trapped inside of the circuit. The sampling-time dependence of the activity shows two time constants with 5.1×10^{5}s and 2.1×10^{4}s shorter than the decay of ^{7}Be, and also suggests that almost all of the produced ^{7}Be is trapped due to processes other than the present detection with a time constant shorter than ~10^{3}s. To elucidate the distribution of ^{7}Be in the cooling circuit, further investigations are required.

Acknowledgement

We would like to thank Prof. Y. Nagai and Dr. M. Numajiri for discussing the cross sections, and S. Ishihama and T. Kunifuda for technical assistance for measuring radioactivity of the samples.

References

[1] T. Broome, Proc. ICANS-XII (Rutherford Appleton Laboratory, RAL94-025, 1994) T-20.
[2] K. Katoh et al., KEK Internal 81-12 (1982), in Japanese. The coating rate of ^{7}Be onto the container increases with increasing pH at a basic side, and the coating rate onto glass is larger than that onto acrylic acid resin.
[3] A. Yu. Konobeyev and Yu. A. Korovin, Nucl. Instr. Meth. Phys. Res. B 82 (1993) 103, and references therein.
[4] S. Shibata et al., Radiochimica Acta 75 (1996) 1.
[5] S. M. Read and V. E. Viola, Jr., Atomic Data and Nuclear Data Tables 31 (1984) 359, and references therein.
[6] P. E. Koehler et al., Phys. Rev. C 37 (1988) 917.