VIII-Ⅱ-1. Project Research

Project 10

PR10　Creation of Unique Neutron Irradiation Experiments Using B-2 Beam Hole of KUR

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Objective: New neutron irradiation apparatus had been installed in B-2 beam hole of Kyoto University Research Reactor (KUR) in 2012. Samples loaded on a carrier can be transported from a station at an experimental room to an irradiation field in the B-2 beam hole by the apparatus. The carrier can transport a sample of sizes up to 6 cm x 6 cm x 30 cm and weight up to 10 kg. And the carrier can be connected with wires or tubes to monitors or instruments located at the experimental room. Various unique neutron irradiation experiments, such as an irradiation to circulating liquid sample and online monitoring of neutron-irradiated materials, can be performed using the apparatus. In this project work, basic experiments as the first steps for applications of advanced neutron irradiation experiments in various research fields has been carried out.

Research Subjects: We have six themes in the project research. One is for measurements of fundamental data about irradiation fields, three are basic researches for advanced irradiation experiments, and two are developments for advanced experiments. The respective subjects of the research groups of this project are described as follows;
P10-1: Measurement of neutron flux at various irradiation positions in B-2 beam hole
P10-2: Basic research for application to biological experiments
P10-3: Development of irradiation method for circulating liquid sample
P10-4: Basic research for application to neutron activation analysis (NAA)
P10-5: Basic research for application to fundamental chemistry
P10-6: Development of irradiation method for biological samples
In this year period, experiments of P10-6 were not carried out because of a trouble of a sample carrier.

Results: In the measurement of irradiation characteristics in B-2 beam hole, the flux and uniformity of irradiated neutrons and the Cd ratio were measured by activation method. The flux measurements were performed at positions of 0 to 300 cm from the reactor-side edge of the B-2 beam hole at intervals of 50 cm. The wide range of neutron flux that is from 10^8 to 10^{12} n/cm^2/s was observed at 1 MW operation. The uniformity of neutron flux was measured by activation method using PET films at positions of 0 to 300 cm at intervals of 100 cm. The neutron flux was found to be uniform by IP images of irradiated PET films. The Cd ratios were measured at the positions of 100 to 120 cm with/without a polyethylene (PE) shield. The Cd ratio without the PE shield decreases as the range from the reactor-side edge. On the other side, The Cd ratio with the shield increases and seems to be saturated at the position of 15 cm from the shield surface. This means irradiating neutron spectrum is controllable by using shield materials for the purpose of each experiment.

In the basic researches for biological, NAA, chemical studies, various irradiation tests has been carried out. In the irradiation tests for biological samples, the gamma-ray dose rate has been measured by thermoluminescence detectors (TLDs) at positions of 100 to 300 cm at intervals of 50 cm because not only neutrons but also gamma rays affect biological samples strongly. And the suitable irradiation time for alpha autoradiography technique to obtain intracellular boron distributions with high position resolution is estimated by measured neutron fluxes at B-2 beam hole and irradiation results by TC-Pn.

For application to NAA experiments using B-2 beam hole, irradiation tests for large volume samples (1.0 g, 10 g and 40 g) of quartz sand in 20 mL polypropylene (PP) bottles have been carried out. After the irradiation, there found no irradiation damages to PP bottles, and radioactivities of the samples were analyzed by γ spectrometry. On the basis of obtained data, experiments on environment samples and longtime irradiation are planned.

Irradiation tests for liquid samples have been performed as the first step of chemical experiments using activated liquid samples. Irradiation damages to polyethylene bottles were checked and also irradiation test to water in the bottle was performed. In the water sample irradiation, the sample of 15 mL pure water in a PE bottle was irradiated for 2 min at the position of 150 cm. After the irradiation, radioactivity of the sample was analyzed by γ spectrometry.

For the development of irradiation method for circulating liquid samples, radiation tolerance of a PFA tube that is a main material of the liquid circulation system has been checked. PFA tubes were irradiated by neutron at the position of 0 cm for 10 min or at 200 cm for 60 min. And the tubes were also irradiated by Pn-2 irradiation hole for 1 or 10 min. After the irradiations, tensile test was performed and it was found that the dose of 7.2×10^{14} n/cm^2 does not cause serious problems on the performance of the PFA tube. Therefore, the safety for 2-hour irradiation at the position of 0 cm in B-2 beam hole is confirmed.

採択課題番号 24P10　Ｂ２照射孔を利用したユニークな中性子照射実験の創出　　プロジェクト
（京大・原子炉）高宮幸一

PR10-1 Characteristics of Neutron Irradiation Field at B-2 Beam Hole in KUR

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INTRODUCTION: Neutron irradiation experiments, such as neutron activation analysis and isotope production, have been performed using various irradiation apparatuses at research reactors. One of the most general neutron irradiation apparatus is a pneumatic transport system, which transports small capsules enclosing samples from a laboratory room to an irradiation hole near a reactor core. However, sizes and weights of samples are restricted by the transporting capacity of capsules. And furthermore, an irradiation of liquid samples is not permitted because of reactor safety problem in almost all research reactors. A new neutron irradiation apparatus has been developed at Kyoto University Reactor (KUR, 5 MW thermal power) to overcome such limitations. The apparatus transport samples loaded on a carrier from an experimental room adjacent to the reactor to irradiation fields in a horizontal beam hole (B-2) of KUR. Positions of irradiation field can be changed that realizes various neutron irradiation experiments. In this work, the neutron flux has been measured at various irradiation positions of the B-2 beam hole using the irradiation apparatus. And neutron flux measurements using a polyethylene shield have been tried to change irradiating neutron spectra for irradiation experiments by various neutron spectra.

EXPERIMENTS: The neutron flux and its Cd ratio were measured by activation method using gold wires. The flux measurements were performed at the positions of 0 to 300 cm from the reactor -side edge of the B-2 beam hole at intervals of 50 cm. The Cd ratios were measured at the positions of 100 to 120 cm with/without a polyethylene shield (5 cm × 5 cm × 20 cm). The uniformity of neutron flux was also measured by activation method using PET films [1] at the positions of 0 to 300 cm at intervals of 100 cm.

RESULTS: The results of the flux measurements depending on the range from the reactor-side edge at the B-2 beam hole are shown in Fig. 1. The wide range of neutron flux that is from 10^8 to 10^{12} n/cm^2/s was observed, which realizes various kinds of neutron irradiation experiments. The neutron flux decrease as the range from the edge increases. The decreasing trend of neutron flux changes around the range at 120 cm because the sectional shape of the beam hole changes around here. The neutron flux was found to be uniform at every position by IP images of irradiated PET films. The results of Cd ratio (Au) measurement are shown in Fig. 2. Open and closed symbols show Cd ratios (Au) with a polyethylene shield and without it, respectively, at the position of 100 to 120 cm. The Cd ratio without the PE shield decreases as the range from the reactor-side edge. On the other side, The Cd ratio of gold shielded by PE increases and seems to be saturated at the position of 15 cm from the PE surface. This means irradiating neutron spectrum becomes softer by PE shield and is controllable in the irradiation apparatus using shield materials.

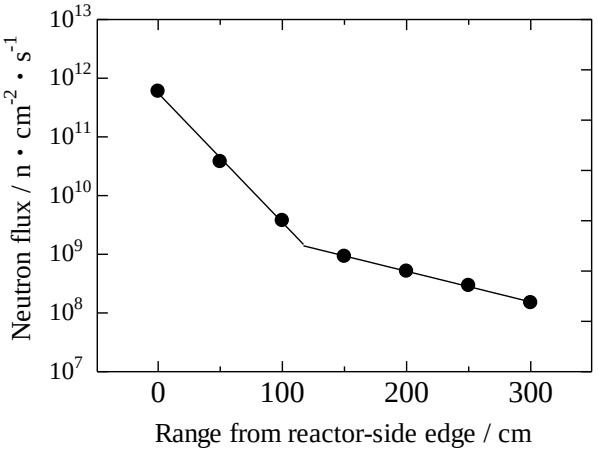

Fig. 1 Neutron flux variation at the B-2 beam hole.

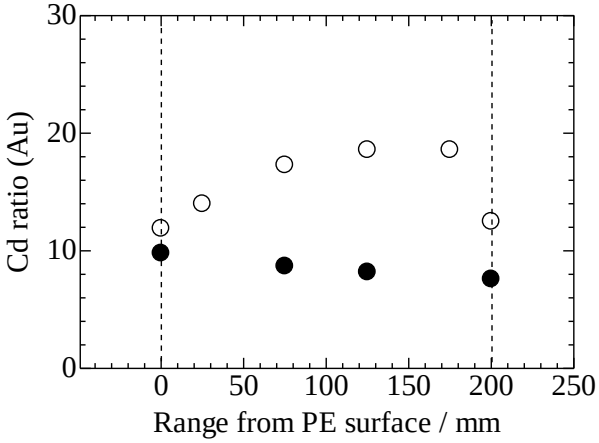

Fig. 2 Cd ratio of gold with/ without the PE shield depending on the range from the PE surface at the position of 100 cm in the B-2 beam hole.

REFERENCES:

[1] K. Takamiya, *et al*, Proc. Radiochim. Act. **1** (2011) 63-66.

採択課題番号 24P10-1　　B２照射孔における照射特性　　プロジェクト

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（京大院・工）吉田悠矢

PR10-2 Study on the Improvement of Micro-Imaging for Boron Compounds I

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INTRODUCTION: It is important to develop new boron compounds for Boron Neutron Capture Therapy (BNCT). The accumulation ratio of the tumor to the normal tissue of the boron compound which can be used now is about 8 at the maximum. For an expansion of BNCT adaptation, a boron compound with tens times accumulation ratio is desired. However, the biological effectiveness of a boron compound cannot be determined only by an accumulation ratio. Because the boron concentration measured by ICP or prompt gamma ray analysis is the average value of the group of cells with different uptake of boron compound. If cells with same boron concentration are irradiated by same neutron fluence, the biological response changes greatly with the kind of boron compounds. In order to confirm the effect of boron compound, tumor-bearing mouse administrated with boron compound and irradiated with neutrons should be bred for a long time. Then, the prediction of the characteristic of boron compound is performed by comparing with the boron micro-distribution in a cell. Alpha autoradiography is used as a method of measuring boron micro-distribution in a cell [1,2]. In this research, the high position resolution and simple measurement technique of intracellular boron distribution will be developed. Furthermore, evaluation of irradiation time using B-2 irradiation hole is curried out for alpha autoradiography.

EXPERIMENTS: In order to estimate the relationship between the boron concentration and the number of etched pits created on CR-39 track detector or neutron fluence. The sample administrated with boron compound of 2.5, 12.5, 25 ppm was installed on CR-39. The sample was irradiated by Tc pneumatic tube with thermal neutron flux of $8x10^{10}$ (n/cm^2/s) for 1 minute at KUR. After irradiation, CR-39 was etched by 6N NaOH. The etched CR-39 was observed by optical microscope.

The tissue sample administrated with boron compound of 25 ppm was also irradiated with thermal neutron. After irradiation, the tissue sample stained by Hoechst was observed by fluorescence microscope in order to identify cell nucleus and etched by 6 N NaOH. The etched CR-39 was also observed by optical microscope.

RESULTS: Fig. 1 shows the relationship between the number of etched pits in the area of $1.1x10^4$ (μm^2) and the boron concentration. The good linearity was obtained in this experiment. The neutron fluence of $4.2x10^{12}$ (cm^2) is enough to observe clearly the etched pits image. Fig. 2. shows the superimposed cell nuclei image strained by Hoechst and etched pits image. Irradiation time for tissue sample with boron concentration of 25 ppm using B-2 irradiation hole at 100cm and 50 cm is estimated to 1200 and 95 sec, respectively for alpha autoradiography.

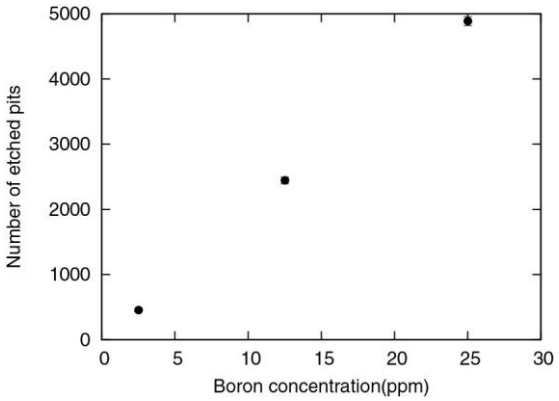

Fig.1. Relationship between the number of etched pits and the boron concentration.

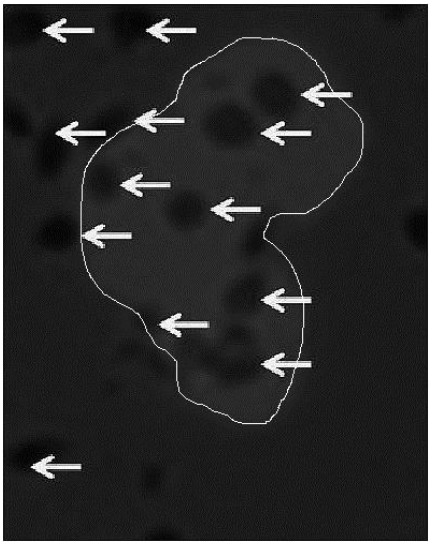

Fig.2. The superimposed image of etched pits on CR-39 and cell nucleus strained by Hoechst. Arrows and the part of enclosed white solid line indicates etched pits and cell nuclei, respectively,

REFERENCES:

[1] S. Altieri, S. Bortolussi, P. Bruschi *et. al.*, Nuclear Technology, Applied Radiation and Isotopes, **66**(2008), 1850– 1855

[2] K. Ogura, A. Yamazaki, H. Yanagie *et.al.*, Radiation Measurements, **34**(2001), 555–558

採択課題番号 24P10-2 ホウ素化合物のマイクロイメージングの高度化に関する研究 プロジェクト
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PR10-3 On-Line Production of Short Half-Life Nuclides Such as $^{142}Pr^{m}$ Using KUR

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INTRODUCTION: B-2 beam hole makes it possible to insert an experimental apparatus up to 1000 cm^3 into the Kyoto University Research Reactor (KUR) [1]. In order to prepare radioactive sources of short half-life nuclides such as $^{142}Pr^{m}$ ($T_{1/2}$ = 14.6 min [2]) in various solution, we plan to develop an on-line irradiation apparatus for solution samples at that place and to irradiate with neutrons while flowing a sample solution. There are few studies about on-line irradiation of a solution using KUR, so that, at first, we have studied radiation tolerance of the PFA tube which is a main material of the irradiation apparatus. In this work, the PFA tubes were irradiated by neutron with various flux and then subjected to tensile test to measure neutron damage.

EXPERIMENTS: Tensile specimens based on JIS K 6251 No. 8 size were prepared by stamping out from a PFA tube with an outer diameter of 10 mm and a wall thickness of 1 mm. The specimens were individually sealed with a polyethylene film and irradiated with 1 MW power using a supporting movable base emplaced at B-2 beam hole or Pn-2. The irradiation using B-2 beam hole was carried out at the most remote part of B-2 for 10 min with the fast neutron flux of 8.6×10^{10} n/cm^2/s or 200 m this side from the site for 60 min with 8.0×10^{7} n/cm^2/s. In the Pn-2 irradiation, the specimens were inserted into polyethylene capsules and irradiated for 1 min or 10 min with 1.2×10^{12} n/cm^2/s.

After the irradiation, tensile test was performed on these specimens with a crosshead speed of 5 mm/min. Experimental conditions are summarized in Table 1.

RESULTS: The tensile strength (TS) σ [MPa] and tensile elongation (TE) ε [%] are given by the following equations:

$$\sigma = \frac{F}{A} \quad , \quad \varepsilon = \frac{L_f}{L_0}$$

, where F is the maximum load [N], A is the cross-section area of the material [mm^2], L_f is the final length of the specimen, and L_0 is the initial length of the specimen. The TS and TE obtained are also listed in Table 1. Initially, the TS and TE become larger as the amount of dose increases. On the other hand, TS and TE at the dose of 7.2×10^{14} n/cm^2 is less than those of the no irradiation specimens, which is due to neutron damage on PFA samples. However, taking TS and TE values into consideration, it will be thought that even the dose of 7.2×10^{14} n/cm^2 does not cause serious problems on the performance of the PFA tube. Therefore, we conclude that the irradiation for 2 h at the most remote part of B-2 is available. At present the prototype of the on-line irradiation apparatus for solution samples is under construction.

ACKNOWLEDGMENT: The authors would like to thanks K. Miyata, technical staff of KURRI, for the helpful support on the tensile test.

REFERENCES:

[1] K. Takamiya *et al.*, in this KURRI Progress report.
[2] R. B. Firestone, in *Table of Isotopes*, 8th ed.,edited by V. S. Shirley (Wiley, New York,1996).

Table. 1. Experimental condition and results of the tensile test.

Sample name	Position	Fast neutron flux [n/cm2/s]	Irradiation time [s]	Tensile strength [MPa]	Tensile elongation [%]	dose [n/cm2]
Sample01 Sample02 Sample03	No irradiation	0.0E+00	0	23.8 ± 1.6	351.3 ± 26.3	0
Sample04 Sample05 Sample06	B-2 200 cm	8.0E+07	3600	24.2 ± 2.1	365.7 ± 41.2	2.9E+11
Sample07 Sample08 Sample09	B-2 0 cm	8.6E+10	600	27.0 ± 1.8	456.3 ± 25.4	5.2E+13
Sample10 Sample11 Sample12	Pn-2	1.2E+12	60	28.0 ± 1.7	428.0 ± 32.0	7.2E+13
Sample13 Sample14 Sample15			600	19.6 ± 0.9	313.3 ± 18.5	7.2E+14

採択課題番号 24P10-3 原子炉を用いた Pr-142 等の短半減期核種のオンライン製造　プロジェクト
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PR10-4 Neutron Irradiation of Large Volume Samples by Using the B2 Irradiation System

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INTRODUCTION: On analysis of environmental samples, representation of the sample is important and often discussed. Instrumental Neutron Activation Analysis (INAA) is useful and superior method for elemental analysis of environmental samples. But about 100mg sampling for traditional pneumatic neutron activation analysis, to keep the representation and homogeneity of samples is painful. The irradiation system for large volume samples may enable us to reduce the troublesome task of preparing analysis samples, and the system may improve the representation of samples. The purpose of this study is to develop a system of neutron irradiation for large volume samples by using the new B2 irradiation apparatus [1]. At first year of this project, we irradiated quartz sand samples which were supposed to be stable and little activation, and checked safety of the system and the method.

EXPERIMENTS: The sample was irradiated for 2 min by using the B2 facility of KURRI at a position of 150 cm from the core of the reactor. Experiments were conducted on 1MW operation, and the thermal neutron flux at the 150 cm position is 9.4×10^8 n sec^{-1} cm^{-2}. 1.0g, 10g and 40g of quartz sand were taken in 20mL vol. polypropylene bottles(Fig.1) and were was irradiated. After the irradiation, radioactivity of the sample was analyzed by γ spectrometry.

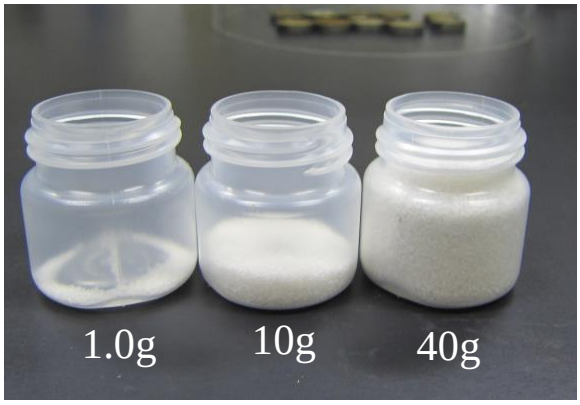

Fig.1 1.0g, 10g and 40g of quartz sand samples

RESULTS: After the irradiation, samples and propylene bottles were sound, and no damage was found. Gamma spectrum after about 90 minutes cooling time of the 10g of quartz sand sample with the propylene bottle is shown in Fig. 2. The irradiated bottle with sample had empty space, and air in the empty space was irradiated at the same time. It follows that Ar-41(1294 keV, 1.8h) which is activated Ar-40, one of air components (0.934 vol.%), is detected. The same kind of quartz sand sample was also irradiated by the orthodox pneumatic method (Pn-2, 1MW, 5.5×10^{12} n sec^{-1} cm^{-2}, 30min, 138mg) and Gamma spectrum after about 20 minutes cooling time is shown in Fig 3. A major component element of quartz sand is silicon (Si) and the γ ray peak of Si-31(1266keV, 2.6h) which was activated of Si-30 (natural abundance ratio3.1%, 0.108b) was found clearly in Fig. 3, but the γ ray peak at 1266keV was trace level in Fig.2. These are due to the difference of neutron flux and irradiation time, and possibility to eliminate interference of major matrix components is indicated. Experiments on environment samples and longtime irradiation are planned.

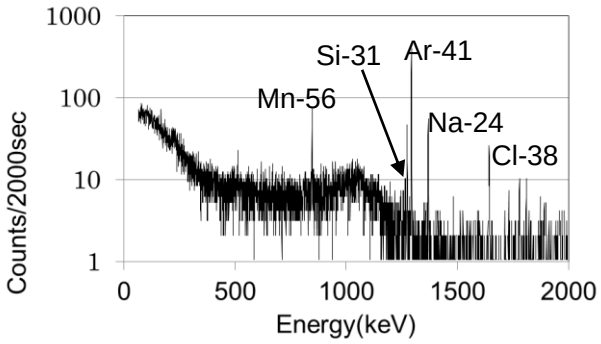

Fig. 2. Gamma spectrum after about 90 minutes cooling time of the 10g of quartz sand sample (B2)

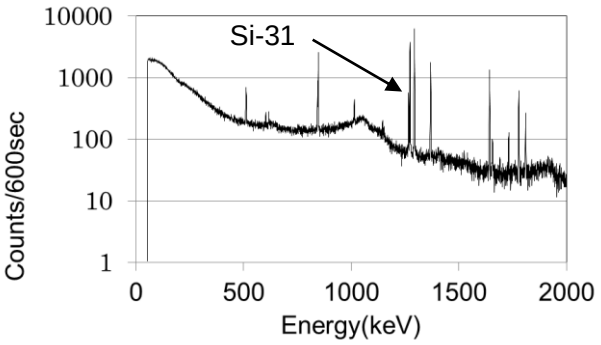

Fig. 3. Gamma spectrum after about 20 minutes cooling time of the 138mg of quartz sand sample (pn-2)

REFERENCES:

[1] K. Takamiya *et al.*, Proc. 47th KURRI Scientific Meeting (2013) 124-126.

採択課題番号 24P10-4　環境サンプル中の微量元素分析における高信頼手法の開発　プロジェクト
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PR10-5 Neutron Irradiation of Solutions by Using the B2 Irradiation System

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INTRODUCTION: Quantitative analysis of elements dissolved in environmental water is important and quick nondestructive analytical (NDA) method is useful for the purpose. Neutron activation analysis is superior to analyze trace amount of elements, but there is no facility in Japan, in which the irradiation apparatus for liquid samples is installed. Generally, trace amounts of elements in liquid samples are recovered by using ion exchange resins or by precipitation methods. Collected samples are once dried up. For nonvolatile elements, liquid samples may be directly dried up. The dried samples were then irradiated for NDA. Every method needs pretreatment of liquid samples. The irradiation apparatus for liquid samples may enable us to reduce the pretreatment processes. The purpose of the present study is to develop a system of neutron irradiation for liquid samples by using the B2 irradiation apparatus [1]. We firstly checked irradiation damage of polyethylene bottles and then tested neutron irradiations of water in the bottle.

EXPERIMENTS: The sample was irradiated for 2 min by using the B2 facility (Fig. 1) at a position of 150 cm from the core of Kyoto University Research Reactor. The thermal neutron flux at the 150 cm position is 9.4×10^8 n sec^{-1} cm^{-2}. 15 mL of pure water was taken in a polyethylene bottle and this was irradiated. Au foils were irradiated together as a neutron flux monitor. After the irradiation, radioactivity of the sample was analyzed by γ spectrometry.

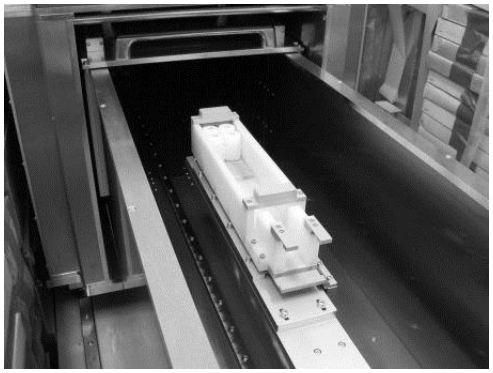

Fig. 1. B2 irradiation system [1].

RESULTS: Gamma spectra of the polyethylene bottle irradiated are shown in Fig. 2. The volume of the bottle was about 20 mL and the space was filled with air. A γ ray of 1294 keV was found in the spectrum (Fig. 2a). This is attributable to the activation of Ar in the air, that is, $^{40}Ar(n, \gamma)^{41}Ar$. The air inside the bottle was removed and analyzed again. As shown in Fig. 2b, no gamma rays were found. A γ ray of 1642 keV found is due to ^{38}Cl. A trace amount of Cl_2 in the air may have been activated. The bottles were settled in a conveyor of the B2 system by using Scotch tape. A trace contamination of Al might have been introduced from the tape. Another possibility is an impurity of the polyethylene bottle itself. Since the half-life of ^{28}Al is short (2.2 min), the γ ray of 1778 keV was quickly disappeared during the analysis (Fig. 2b).

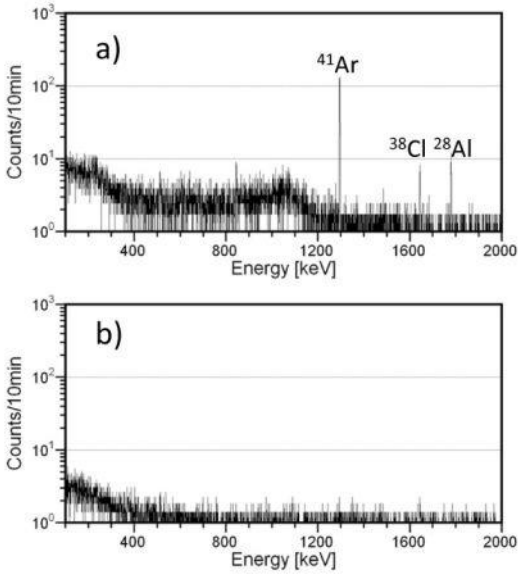

Fig. 2. Gamma spectra of irradiated polyethylene bottle. a) With air inside the bottle. b) Irradiated air was removed from the bottle.

15 mL of pure water was taken in the polyethylene bottle and this was irradiated. A few products of water were tested. Besides the γ rays shown in Fig. 2, two additional γ rays were found at 617 keV and 1461 keV. These were assigned as γ rays emitted from ^{80}Br and ^{40}K, respectively. Intensity of these γ rays was as small as that of ^{38}Cl shown in Fig. 2a.

REFERENCES:

[1] K. Takamiya *et al.*, Proc. 47th KURRI Scientific Meeting (2013) 124-126.

採択課題番号 24P10-5　　Ｂ２照射孔における溶液試料の中性子照射　　プロジェクト
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