VIII-Ⅱ-1.　Project Research

Project 10

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

K. Takamiya

Research Reactor Institute, Kyoto University

Objective: A multi-purpose neutron irradiation apparatus had been installed in B-2 beam hole of KUR. Samples loaded on a carrier can be transported from a sample station at an experimental room to an arbitrary position in 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. Connection between the carrier and monitors located at the experimental room with wires or tubes enables online neutron irradiation experiments. 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: The project research is composed by six individual subjects. 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 P10-4 and P10-6 were not carried out because of short machine time and a trouble of sample transportation. Causes of the trouble had been investigated and will be eliminated.

Results: In the measurement of irradiation characteristics in B-2 beam hole, the distribution of the neutron flux was estimated by a simulation code, PHITS, and results was compared to the experimental data. The neutron flux at the position of 100 cm was estimated by the simulation, and obtained result was comparable to the flux measured by activation method in the previous work. Variation of neutron flux in a polyethylene moderator placed in the beam hole has been measured in the previous work, and the simulation in the same irradiation condition was performed in the present work. There are larger differences between the experimental and calculated results around the center of the moderator compared to those at edges, but both results show good agreement within ~10% range. More detail simulation will be performed using accurate parameters to describe the distribution of neutron flux in the B-2 hole in the future that will realize the quantitative neutron irradiation by various spectra.

In the basic research for application to biological experiments, the neutron field of B-2 was evaluated in terms of alpha autoradiography for measurement of boron micro-distribution in a cell sample. Monte Carlo calculation code, MCNP5 was used for the calculation of the neutron spectrum at irradiation position. In order to irradiate the sample of alpha autoradiography, it is necessary to reduce the fast neutron contamination. The moderator and reflector materials such as Pb, D_2O, Bi, Al, C, AlF_3, Polyethylene, which are usually used for the BNCT irradiation field, were selected in the calculation. As a result, the fast neutron contamination increases as the thickness of moderator become thicker except for aluminum. Combination with moderator of aluminum and the reflector of polyethylene has good characteristics of fast neutron contamination.

For the development of irradiation method for circulating liquid samples, radiation tolerance of materials of the irradiation instruments has been investigated. In the present work, PTFE and ETFE tubes, which are candidate materials of the flow passage, and PEEK connector were irradiated by neutron and then measured deterioration by the irradiation. The PTFE, ETFE, and PEEK samples were inserted into polyethylene capsules and irradiated for 5 min or 20 min with Pn-2 (1 MW), and a simple bending fatigue test was performed on the irradiated samples after the irradiation. As a result, it is found that the ETFE and PEEK are appropriate materials for the on-line irradiation experiments at B-2 hole.

Neutron irradiation for solutions have been performed as fundamental test for chemical experiments using activated liquid samples. Neutron irradiations of 15 mL of pure water in polyethylene bottle were performed at B-2 beam hole, and relative intensity of neutron beam at the irradiated position was also evaluated by activation method using gold foils. After the irradiation, gamma-ray spectrometry for the irradiated samples was performed, and produced activity was identified. Relative intensities of neutron beam at various irradiated positions were evaluated from the gamma intensity of ^{198}Au. It is clear that the shielding effect of light water is quite strong.

PR10-1 Simulation of Neutron Flux Distribution in B-2 Beam Hole of KUR

K. Takamiya, Y. Yoshida[1], T. Sano, H. Yoshino, Y. Iinuma, R. Okumura and T. Ohtsuki

Research Reactor Institute, Kyoto University
[1]Graduate School of Engineering, Kyoto University

INTRODUCTION: A neutron irradiation apparatus had been developed at B-2 beam hole of Kyoto University Reactor (KUR) that enables neutron irradiation to liquid samples, large-size samples. Furthermore, online neutron irradiation experiments can be performed using the apparatus. The distribution of neutron flux in B-2 hole has been measured by activation method using gold wires in the previous work. And variation of neutron flux using a polyethylene moderator has been measured in order to prepare the neutron irradiation fields that have different neutron energy spectra. In the present work, the distribution of the neutron flux was estimated by a simulation code, PHITS[1)], and results was compared to the experimental data.

EXPERIMENTS: A gold wires were inserted into holes bored in a polyethylene block which is used as a neutron moderator in the present experiments. The size of a polyethylene block is 50 x 50 x 200 mm^3, and positions of the holes were 25, 75, 125 and 175mm from the front surface of the block. And two gold wires were placed on the front and rear surfaces additionally. The gold wires with the polyethylene moderator were irradiated by neutrons for 10 minutes at the range of 1000 mm from the reactor-side edge of the B-2 beam hole. The gamma-ray spectrometry has been performed for irradiated gold wires by a Ge-detector to determine the irradiated thermal-neutron flux.

A neutron flux distribution in the polyethylene moderator was estimated by simulation using PHITS code. JENDL-4.0 was used as the nuclear data library, and neutron spectrum led into the B-2 beam hole from the reactor core was estimated by MVP2[2)] in the present simulation.

RESULTS: The horizontal distribution of neutron flux in the B-2 beam hole calculated by PHITS is shown in Fig. 1. The reactor core is placed at the left side of the figure, and a rectangle drawn near 100 cm (z-axis) express the polyethylene moderator. The neutron flux at the position of 100 cm without the moderator was also estimated by the simulation, and obtained result was comparable to the flux measured by activation method in the previous work. Variation of the calculated thermal neutron flux at the different positions of the gold wires in the moderator is shown in Fig. 2. The thermal neutron flux determined by gamma-ray spectrometry for the irradiated gold wires is also shown by triangles in Fig. 2. There are larger differences between the experimental and calculated results around the center of the moderator compared to those at edges, but both results show good agreement within ~10% range. One of reasons for the discrepancy might be caused by inaccurate parameters of a sample carrier, which is produced with polyethylene, in the simulation calculation. More detail simulation will be performed using accurate parameters to describe the distribution of neutron flux in B-2 hole in the future that will enables the quantitative neutron irradiation with different spectra.

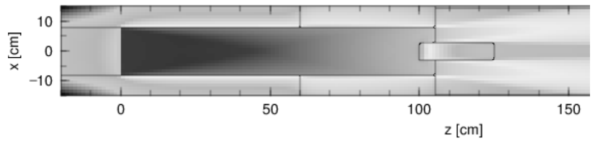

Fig. 1. Horizontal distribution of neutron flux in the B-2 hole estimated by PHITS code.

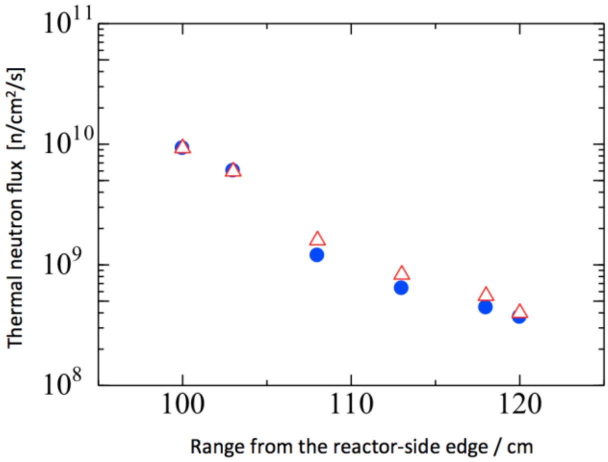

Fig. 2. Thermal neutron flux determined by the experiment (triangles) and simulation using PHITS (circles).

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[1] T. Sato *et al*. J. Nucl. Sci. Technol., **50**, 913 (2013).
[2] Y. Nagaya *et al*. Japan Atomic Energy Research Institute, 1348 (2005).
[3] K. Shibata *et al.* J. Nucl. Sci. Technol., **48**, 1 (2011).

採択課題番号 25P10-1　　　Ｂ－２照射孔の中性子スペクトル制御　　　プロジェクト
（京大・原子炉）高宮幸一、佐野忠史、吉野泰史、飯沼勇人、奥村良、大槻　勤
（京大院・工）吉田悠矢

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

H. Tanaka, K. Takamiya, T. Sano, Y. Sakurai, T. Fujii, S. Fukutani, K. Ono and Y. Kawabata

Research Reactor Institute, Kyoto University

INTRODUCTION: It is important to develop new boron compounds for Boron Neutron Capture Therapy (BNCT). 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 order to perform alpha autoradiography, neutron fluence of 10^{12} (cm^{-2}) is needed. At KURRI, there are two neutron irradiation fields for alpha autoradiography. One is the heavy water neutron irradiation facility (HWNIF) that is used for BNCT clinical studies. The thermal neutron flux of HWNIF is $1x10^{9}$(cm^{-2} s^{-1}) with the uniform field of 10 cm in diameter. The other is graphite thermal column with the thermal neutron flux of $8x10^{10}$(cm^{-2} s^{-1}). Sample size is limited to 2.5 cm in diameter. If the sample with the size of 5cm in diameter, corresponding to a whole body mouse, is irradiated, the thermal neutron irradiation field with the sufficient size and flux is necessary. B-2 port is the one of the candidates for alpha autoradiography. In this research, we evaluate the neutron field of B-2 in terms of alpha autoradiography. Furthermore, the possibilities of irradiation for live mouse sample is evaluated.

METHODS: Monte Carlo calculation code, MCNP5 was used for the calculation of the neutron spectrum at irradiation position. The measured thermal neutron flux by gold activation method was used as normalizing factor. In order to irradiate the sample of alpha autoradiography, it is necessary to reduce the fast neutron contamination. The moderator and reflector materials such as Pb, D_2O, Bi, Al, C, AlF_3, Polyethylene, which are usually used for the BNCT irradiation field, were selected in this calculation. The reflector was set behind the sample position. The ratio of fast neutron dose to thermal neutron fluence was use as figure of merit for the optimization of moderator thickness.

RESULTS: Figure1 shows the neutron spectrum at the distance from reactor core of 100 cm. Fast neutron intensity from the core is quite high to apply to alpha autoradiography or the mouse irradiation. Fig. 2 shows the relationship between fast neutron contamination and the moderator thickness. The fast neutron contamination increase as the thickness of moderator become thick except for aluminum. Combination with moderator of aluminum and the reflector of polyethylene has good characteristics of fast neutron contamination. Fig. 1 also shows the neutron spectrum using aluminum moderator with the thickness of 30cm and polyethylene reflector at the distance from core of 100 cm. Thermal neutron flux of $4x10^{8}$(cm^{-2} s^{-1}) is less than that of no usage of moderator and reflector. If the irradiation position is set to the nearest core position, thermal neutron flux is estimated to $6.4x10^{10}$(cm^{-2} s^{-1}). The fast neutron contamination is about 5 times larger than that of HWNIF. However, this neutron field can be applied to alpha autoradiography and the mouse irradiation.

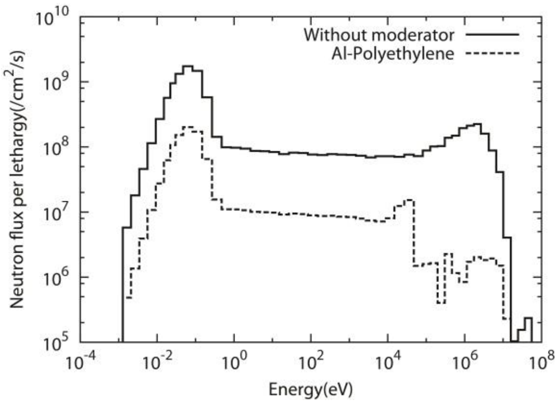

Fig.1. Neutron spectrum at the distance from the core of 100cm for no usage of moderator or the combination with aluminum and polyethylene reflector.

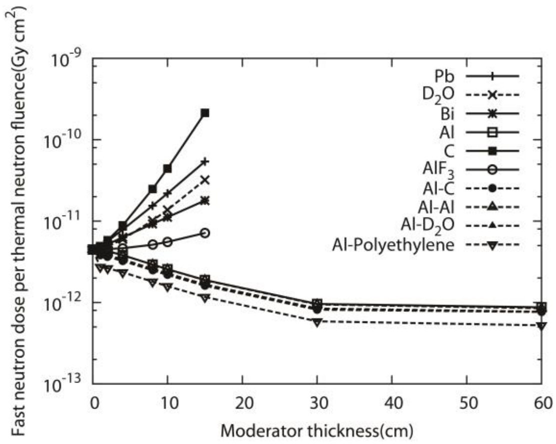

Fig.2. Relationship between the moderator thickness and fast neutron contamination in neutron beam.

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採択課題番号 25P10-2 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Ⅱ プロジェクト
（京大・原子炉）田中浩基、高宮幸一、櫻井良憲、藤井俊行、福谷哲、小野公二、川端祐司

PR10-3

Neutron Irradiation Test for the Materilas of On-Line Irradiation Apparatus at KUR B-2

H. Kikunaga, T. Ohtsuki[1] and K. Takamiya[1]

Research Center for Electron Photon Science, Tohoku University
[1]Research Reactor Institute, Kyoto University

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]. We plan to construct an on-line irradiation apparatus (Fig. 1) in this hole. The advantage in this apparatus is that short-lived isotopes produced by the neutron irradiation are able to be collected quickly and continuously as a solid, liquid or gas.

There are few studies about on-line irradiation of a solution using KUR, so that, at first, we have studied radiation tolerance of materials of the irradiation apparatus. The radiation tolerance of PFA tube, which is the material of a guard pipe for the flow passage, was investigated in last year [2]. In this work, PTFE and ETFE tubes, which are candidate materials of the flow passage, and PEEK connector were irradiated by neutron and then measured deterioration by the irradiation.

EXPERIMENTS: A PTFE tube with inner diameter of 2 mm and a wall thickness of 0.5 mm, and an ETFE tube with inner diameter of 0.25 mm and a wall thickness of 0.67 mm were used for the irradiation test. These tubes, cut to approximately 5 cm, and PEEK connectors were individually sealed with a polyethylene film. The PTFE, ETFE, and PEEK samples were inserted into polyethylene capsules and irradiated for 5 min or 20 min with Pn-2 (1 MW power).

After the irradiation, a simple bending fatigue test was performed on the irradiated samples. The test samples were fixed by holding one side, bent up to 90 degrees, and restored to the straight shape. The operations were repeatedly performed while checking whether the samples look normal.

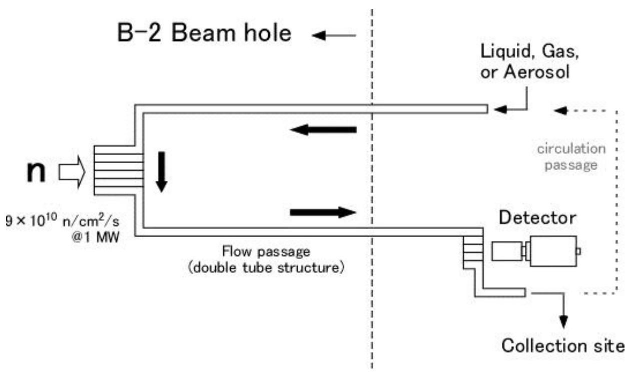

Fig. 1. Schematic diagram of an on-line irradiation apparatus at KUR B-2

RESULTS: Figure 2 shows photographic image of a PTFE (upper case) and an ETFE (lower case) tube irradiated for 20 min after the simple bending fatigue test. The PTFE tubes exhibited decreasing fracture toughness by the irradiation and were broken easily by the several bending operations even after the 5 min irradiation. On the other hand, the ETFE tubes irradiated for 20 min looked normal after more than 20 times of the bending operation. There were also no abnormalities anywhere in the irradiated PEEK connectors. From the results, it is found that the ETFE and PEEK are appropriate for the materials of the on-line irradiation apparatus.

The neutron flux of Pn-2 is approximately 14 times greater than that of the most remote part of B-2 beam hole, so that we conclude that the irradiation for 4.6 h at the B-2 beam hole is available at the shortest. The prototype of the on-line irradiation apparatus for solution samples is under construction.

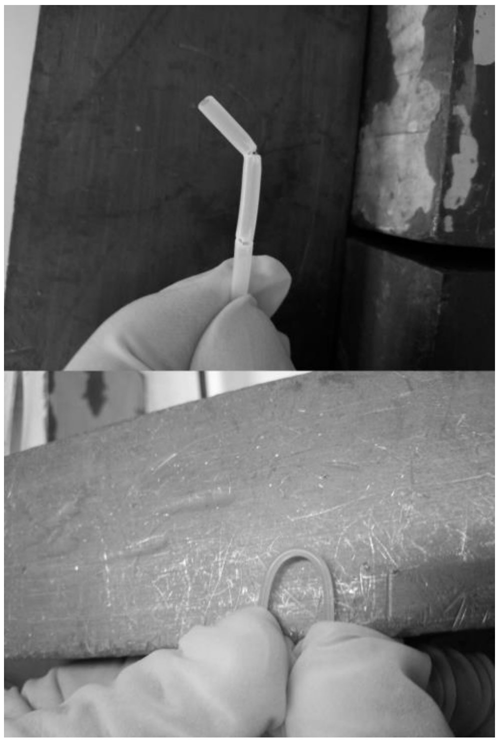

Fig. 2. Photographic image of a PTFE (upper case) and an ETFE (lower case) tube irradiated for 20 min after the simple bending fatigue test.

REFERENCES:
[1] K. Takamiya *et al.*, KURRI Progress report 2012, Project 10
[2] H. Kikunaga *et al.*, KURRI Progress report 2012, PR10-3

採択課題番号 25P10-3 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
（東北大・電子光セ）菊永　英寿（京大・原子炉）大槻　勤、高宮　幸一

PR10-4 Neutron Irradiation of Solutions by Using the B-2 Irradiation System

T. Fujii, A. Uehara, S. Fukutani, R. Okumura and K. Takamiya

Research Reactor Institute, Kyoto University

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 B-2 irradiation apparatus [1]. We tested neutron irradiations of pure water in polyethylene bottle and evaluated relative intensity of neutron beam at the irradiated position.

EXPERIMENTS: The sample was irradiated for 2 min by using the B-2 facility [2] at a position of 150 cm from the core of Kyoto University Research Reactor. The thermal neutron flux at the 150 cm position is 4.7×10^9 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.

RESULTS: Gamma spectrum of the polyethylene bottle containing 15 mL of pure water irradiated is shown in Fig. 1. The volume of the bottle was about 20 mL and the rest space was filled with air. A γ ray of 1294 keV was found in the spectrum. This is attributable to the activation of Ar in the air, that is, $^{40}Ar(n, \gamma)^{41}Ar$. A γ ray of 1642 keV found is due to ^{38}Cl. A trace amount of Cl_2 in the air and/or water may have been activated. The bottles were settled in a conveyor of the B-2 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. Gamma rays of 617 keV and 1461 keV found are due to ^{80}Br and ^{40}K, respectively. These nuclides would have been produced via neutron capture of natural Br and K isotopes in the water as trace impurities.

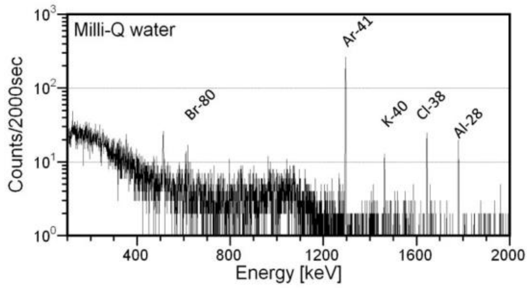

Fig. 1. Gamma spectra of irradiated polyethylene bottle containing 15 mL or pure water.

Relative intensities of neutron beam at various irradiated positions were evaluated from the gamma intensity of ^{198}Au. The result is shown in Fig. 2. It is clear that the shielding effect of light water is quite strong.

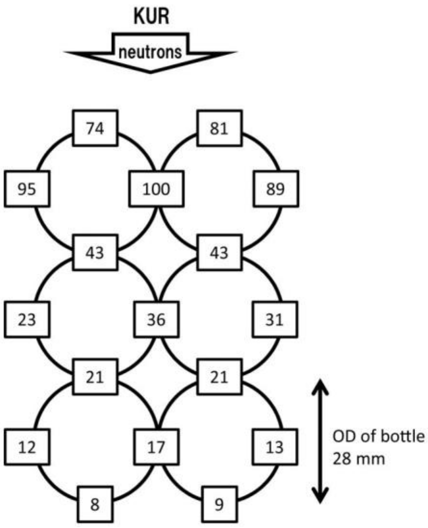

Fig. 2. Relative intensity of neutron beam estimated from gamma intensity of ^{198}Au. Each of six bottles contains 15 mL pure water.

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採択課題番号 25P10-5　　　Ｂ２照射孔における溶液試料の中性子照射　　　　　プロジェクト
（京大・原子炉）藤井俊行、上原章寛、福谷　哲、高宮幸一、奥村　良