The remodelling outline of the neutron irradiation facility of the Kyoto University research reactor mainly for neutron capture therapy

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Abstract. The Heavy Water Thermal Neutron Facility of the Kyoto University Research Reactor (KUR, full power: 5 MW) was wholly updated in March 1996 mainly for neutron capture therapy (NCT). The performance as a neutron irradiation facility was improved using the epi-thermal neutron moderator of the aluminum-heavy water mixture ($AI/D_20 = 80/20$ in volume percent), the neutron energy spectrum shifter of heavy water whose thickness changed from 0 cm to 60 cm, and the thermal neutron filters of I mm-thick cadmium and 6.4 mm-thick boral plates. The clinical irradiation utilisation under the fill-power continuous KUR operation was realised employing both the Radiation Shielding System, and the Remote Carrying System for a patient. The safety and utility of the facility were improved due to the Safety Observation System. The KUR Advanced Irradiation System for NCT was organised.

1. INTRODUCTION

The Heavy Water Thermal Neutron Facility at the Kyoto University Research Reactor (KUR, full power: 5 MW) had been constantly utilised for several research fields such as physics, engineering, biology, medics, etc. since the first KUR criticality in June 1964 [1,2]. Fundamental study for neutron capture therapy (NOT) has been continued since 1970 at the KUR. For the physical and engineering research field, the following main results were obtained: (i) design and development of a clinical thermal-neutron irradiation field with low level gamma ray contamination [2], (ii) development of thermal-neutron shielding material using ⁶LiF with low secondary gamma ray generation [3], (iii) establishment of a measurement method for ¹⁰B concentration in tissue by (n, *r*) Prompt Gamma ray Analysis (PGA) [4], (iv) cell-level estimation method of absorbed dose for NCT [5], etc.. These subjects were strongly connected with the thermal neutron irradiation technique and the estimation of ¹⁰B absorbed dose in tissue, and were indispensable for the NCT study not only for the fundamental research but also for the clinical trial.

A clinical irradiation for NOT at the facility was carried out in May 1974 for the first time, and it has been regularly performed from February 1990. By November 1995, just before the remodelling, sixty-one clinical trials were carried out for about six years [6]. In the NOT clinical irradiation at the pre-updated facility, the startup and shutdown of the reactor was needed for setting of the patient, etc.. On these experiences, the update of the facility was requested by the clinical irradiation staffs and the users in the other research fields, so as to enable (1) the utilisation under the full-power continuous KUR operation for the increase of the opportunity for NOT clinical irradiation, (2) the utilisation of epi-thermal neutrons to improve the irradiation effectiveness for deep-seated tumour, and (3) the clinical irradiation concurrently with the experiments in the other research fields, etc. [7].

Moreover, the following problems of the old facility had been pointed out; (i) difficulty of the handling for the routine maintenance and checkup, and for the irregular damage, and (ii) risk of the leakage of the cooling light water for the heavy water tank, because of its structure that the heavy water tank was settled on the reactor tank with an 0-ring of about 2 m diameter by thirty-six stainless-steel bolts and the primary cooling water flew through a narrow channel between the both tanks. In regard to these matters, the necessity of the fundamental reconstruction of the whole facility became remarkably recognised, from the viewpoint of the safety and stability of the facility and its usage.

2. CONCEPTION FOR THE REMODELLING

The old facility had a high performance as a thermal neutron irradiation field. On the basis of keeping the advantages of the old facility, the main design subjects for the updated facility, Heavy Water Neutron Irradiation Facility (HWNIF), were chosen as described in the following sections. The design studies were performed mainly by simulation calculations using transport calculation codes for neutrons and gamma rays. The transport calculation codes of "ANISN" and "DOT 3.5" were used for one dimensional and two dimensional simulations, respectively. The proprieties of the simulation calculation codes and the calculation processes were confirmed from the comparison between the measured data and calculation results for the old facility [8].

2.1. Improvement in safety for the facility

From the standpoint of the improvement of the facility safety, the following subjects were carried out.

- (1) The heavy water tank was separated from the reactor tank. A cooling system for the heavy water tank was newly settled on the tank. Due to this separation, an air gap was formed between the both tanks. In order to hold argon gas activated in the air gap, "Activated Argon Attenuation System" was installed.
- (2) In order to protect the side surface of the reactor tank facing to the facility, and secure the primary cooling water channel for the reactor tank and the thermal shield, a cooling water jacket was newly settled on the reactor tank by welding.
- (3) The whole of the HWNIF, from the heavy water tank to the outer lead layer, was made as one component. Exceptionally, the outer lead layer can be independently removed for the maintenance of the cadmium and boral filters, which were installed on the core-side of the outer lead layer.

2.2. Advanced neutron utilization for NOT

In the viewpoint of the advanced utilisation of neutrons in a biomedical field, three kinds of neutron irradiation fields, (i) mainly thermal neutrons, (ii) mix of thermal and epithermal neutrons, and (iii) epi-thermal neutrons, were studied according to the following subjects;

- (1) the epi-thermal neutron moderator,
- (2) the neutron energy spectrum shifter and the thermal neutron filter,
- (3) the reflector element of the reactor core,
- (4) the cooling water thickness between the reactor tank and the heavy water tank,
- (5) absorbed dose distribution in a human body under epi-thermal neutron irradiation, and
- (6) the clinical collimators for thermal and epi-thermal neutron irradiation.

2.3. NCT clinical irradiation under the full-power continuous operation

In order to utilize the KUR for NOT clinical irradiation under the continuous operation at the full-power of 5 MW, it is necessary to secure the condition that persons can work in the irradiation room under the KUR operation. A design criterion of the Radiation Shielding System was that a total dose equivalent rate of neutrons and gamma rays was less than 100 μ Sv/hr at the working area in the irradiation room. Moreover, in the viewpoint of the reduction of the working time in the irradiation room, the Remote Carrying System for a patient was

produced. A clinical collimator is settled on this system, then the positioning for the patient are possible outside of the irradiation room previously to a clinical irradiation.

2.4. The design goals of the HWNIF

The design goals for the HWNIF were set as listed below. The numerical values are proper for a free-in-air condition with no irradiated sample at the normal irradiation position, which corresponds to the central point of the bismuth layer surface. The neutron energy regions for thermal, epi-thermal and fast neutrons are defined to be below 0.6eV, from 0.6eV to 10keV and over 10keV, respectively.

- (1) Thermal neutron irradiation field: Thermal neutron flux (fluence rate) is more than 3 x 10^9 n/cm²/s, and the cadmium ratio for gold activation foil (thickness of gold foil is 50 μ m, and thickness of cadmium cover is 0.7 mm) is more than 1,000. The ratio of the incident gamma ray dose to thermal neutron fluence is less than 3 x 10^{-11} cGy/(n/cm²). For the incident fast neutron dose, the incident ratio to thermal neutron fluence is less than 1 x 10–11 cGy/(n/cm²).
- (2) Mixed irradiation field of thermal and epi-thermal neutrons: Thermal neutron flux is more than $3 \ge 1 \sim n/cm^2/s$, and epi-thermal neutron flux is more than $3 \ge 108 n/cm^2/s$. For the incident gamma ray dose, the ratio is almost the same as that for the thermal neutron irradiation field.
- (3) Epi-thermal neutron irradiation field: Epi-thermal neutron flux is more than 3 x 108 $nlcm^2/s$. The ratios of the incident gamma ray dose and fast neutron dose to epi-thermal neutron fluence are less than 3 x 10'~ cGy/(n/cm²) and 1 x i0~'⁰ cGy/(nlcin²), respectively.

3. THE HEAVY WATER NEUTRON IRRADIATION FACILITY

Figure 1 shows the outline of the HWNIF. The epi-thermal neutron moderator, the neutron energy spectrum shifter, the thermal neutron filters and the bismuth layer were installed in order from the core side. As the heavy water tank was settled not connectedly with the reactor tank, it can be removed together with the polyethylene layer and the lead layer outside of the tank. The cooling water jackets of 1 cm thickness are attached both to the reactor tank and the heavy water tank. The jacket plates are welded in order to avoid the cooling water leakage. The inner lead layer of 10 cm thickness and the outer lead layer of 20 cm thickness were settled for the gamma ray shielding. This inner lead layer is an effective shielding against the gamma rays from the core side under the maintenance work. Between the inner and outer lead layers, a polyethylene layer is inserted as a supplementary shield against fast neutrons.

3.1. The epi-thermal neutron moderator and the neutron energy spectrum shifter

The installation position of the epi-thermal neutron moderator was restricted to be the inside of the heavy water tank adjacent to the reactor core. From the design study results on priority of the safety and stability, the moderator was decided to be 80%/20% in the mixing volume-ratio of aluminum and heavy water, 60 cm in diameter, and 66 cm in thickness. The periodic structure of 20 mm-thick aluminum plates and 5 mm-thick heavy water gaps was decided, on the expectation of the heat removal by natural-convection. That is because (1) we had much experience for the utilisation and handling of aluminum and heavy water at the old facility, and (2) these materials had been considered as a moderator in some plans for the epi-thermal neutron irradiation field [9,10].

The neutron energy spectrum shifter was decided to be installed on the outside of the epithermal neutron moderator in the heavy water tank. The spectrum shifter is comprised of three shifter tanks of almost 70 cm diameter, whose thickness are 10, 20 and 30 cm in order from the reactor core side. The supply and drain of heavy water are possible independently for the respective shifter tanks. By the combination of "full" and "empty" of heavy water in the three shifter tanks, the total thickness of the heavy water layer can be controlled from 0 to 60 cm in 10 cm increments. The water shutter is a cylinder of 60 cm diameter and 30.5 cm thickness, and it is surrounded by the bismuth neutron scatterer of 5 cm thickness. The tanks of the spectrum shifter and the water shutter are made with 10 mm-thick aluminum plate.

3.2. The thermal neutron filters and the bismuth layer

The installation of two kinds of the thermal neutron filters, the cadmium filter and the boral filter, were decided. These energy characteristics for neutron absorption are different especially in the energy range from thermal neutrons to epi-thermal neutrons. The installation space for the neutron filters is 10 cm thick including the casing, due to those driving mechanisms. In order from the core side, the boral filter and the cadmium filter are arranged. The respective filter thickness are 1 mm and 6.4 mm. The cadmium filter is sandwiched with a 1 mm-thick aluminum plate (for the core-side) and a 5 mm-thick aluminum plate, for the increase in the mechanical strength. The filters can cover the area of 70 cm diameter for the fully-close case, according as the core-side surface of the bismuth layer is 60 cm diameter. The apertures of the both filters can be adjusted continuously from 0 to 62 cm.

The center of the bismuth layer are removable for a rectangle part of 25 cm x 25 cm in square and 5 cm in thickness, and a convex part of 20 cm diameter for 5 cm thickness and 15 cm diameter for 13.4 cm thickness, from the irradiation room side. So, four kinds of the bismuth thickness such as 0, 5, 18.4 and 23.4 cm can be selected at the bismuth center.

3.3. The irradiation modes and the basic irradiation characteristics

Table I shows the measured values of the irradiation characteristics for several irradiation modes at the normal irradiation position [11]. The "irradiation mode" means a condition of the facility-side, such as open or close of the cadmium and boral filters, full or empty of heavy water in the neutron energy spectrum shifter and heavy water shutter tanks, and the thickness of the center part of the bismuth layer. The first and second characters in the symbol defining irradiation mode represent the open or close conditions of the cadmium and boral filters. The character "0" means the filter "opened (not full-closed)", and the character "C" and "B" mean the cadmium and boral filters "full-closed", respectively. The four numbers represent the conditions of the tanks of the heavy water shutter and the spectrum shifter, in order from the irradiation-room side. The number "0" and "1" mean "empty" and "full", respectively. The last character represents the condition of the center thickness of the bismuth layer. The characters "E", "G", "F" and "H" mean the thickness of 0, 5~ 18.4 and 23.4 cm, respectively. Usually, the bismuth layer thickness is 18.4 cm, namely, in the "F" condition

3.4. Stability of the KUR operation

The stability as to the power and reactivity of the KUR was experimentally confirmed for the influences of the drain-supply of heavy water in the tanks of spectrum shifter and the heavy water shutter, and the open-close of the thermal neutron filters under the full-power continuous operation at 5 MW. In the estimation about these influences on the KUR stability, the changes and change rates for the linear power "Lin-N", the logarithm power "Log-N", the period monitor, the safety power channels and thermal power, were monitored.

The influences were observed inconsiderably for the spectrum shifter and the heavy water shutter, but hardly for the open-close of the thermal neutron filters. Also, the influences



FIG. 1. Outline of the updated Heavy Water Neutron Irradiation Facility.

Irradiation	D ₂ 0	Cadmium	Thermal neutron	Epi-thermal neutron	Gamma ray dose
mode	thickness	ratio	flux	flux*	equivalent Rate
	(cm)		$(n/cm^2/s)$	$(n/cin^2/s)$	(cSv/hr)
00-1111-F	90	790	1.6E+08	4.1E+05	10
00-0111-F	60	700	5.9E+08	1.7E+06	40
00-0110-F	50	650	7.7E+08	2.4E+06	50
00-0101-F	40	400	1.OE+09	5.1E+06	60
00 - 0011-F	30	160	2.OE+09	2.5E+07	100
00-0010-F	20	51	2.3E+09	9,3E+07	110
00-0001-F	10	22	3.3E+09	3.2E+08	180
00-0000-F	0	9.4	5.OE+09	1.2E+09	330
00-0000-F	0	Almost 1	Not estimable	1.1 E+09	60
OB-0000-F	0	Almost I	Not estimable	4.OE+08	50

Table V. Measured values of the neutron fluxes and gamma ray dose equivalent rates at the bismuth surface during the full-power (5MW) KUR operation.

Measurements were carried out using the "irradiation rail device".

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Neutron fluxes were estimated with gold activation foils, and gamma ray doses were measured with TLD (BeO). *It is assumed that the epi-thermal neutrons have a pure l/E spectrum.

of the control rod positions and the accumulated operation time from the reactor startup to a condition change were not observed. For the heavy water drain-supply in the tanks of the spectrum shifter and the heavy water shutter under the full-power operation, the following results were mainly obtained;

- (1) Two safety power channels change about 0.02 MW (below 0.5%),
- (2) Thermal powers estimated from both the primary and secondary coolant systems change about 0.04 MW (below 1%),
- (3) These changes are minus for the supply and plus for the drain, and
- (4) These changes are observed during the supply or drain within a few minutes.

Incidentally, the KUR is controlled for the Lin-N signals to be almost constant, within 1%. It was confirmed that the control stability and safety of the KUR are maintained by the condition change of the HWNIF under the full-power continuous operation.

4. OUTLINE OF THE ADVANCED CLINICAL IRRADIATION SYSTEM FOR NCT

Figure 2 shows the layout of the Advanced Clinical Irradiation System supporting NOT. This system consists mainly of (i) the Radiation Shielding System, (ii) the Irradiation Room and the Entrance Shield Door, (iii) the Remote Carrying System and the Medical Treatment Room for a patient, and (iv) the Safety Observation System. Additionally, the Irradiation Rail Device is provided for basic experiments.

4.1. The radiation shielding system

For the efficient radiation shielding at the mixed field of neutrons and gamma rays, it is general to investigate on the following three divisions; (i) epi-thermal and fast neutrons, (ii) thermal neutrons, and (iii) gamma rays. Fast neutrons with the average energy of 2 MeV generated from the reactor core are efficiently shielded due to the absorption by boron-10, cadmium etc. after the moderation and thermalization.



FIG.2. Layout of the KUR Advanced Clinical Irradiation System.

The gamma rays yielded due to the neutron capture, together with the primary gamma rays from the reactor core, are shielded by high-density materials such as lead, bismuth, etc. On the bases of these concepts, the Radiation Shielding System was investigated.

The radiation shielding system consists of (I) the heavy water shutter and the neutron energy spectrum shifter against fast neutrons, (2) the thermal neutron filters of cadmium and boral against thermal neutrons, and (3) the Beam Shutter, the irradiation room and the entrance shield door against neutrons and gamma rays. For the water shutter, light water was thought to be chosen in the conceptual study [11]. However, the available space for the water shutter was decreased to be about 30 cm. The shutter material was changed from light water to heavy water, in the viewpoint of the simplification of the water drain-and-supply system In order to compensate the insufficient radiation shield against fast neutrons, a Beam Shutter was installed outside of the bismuth layer. The beam shutter has a multi-layer structure consisting of iron, lead, polyethylene, borated-polyethylene. The whole thickness of the Beam Shutter is 74 cm, which is the maximum size in order to install it in the pit space for the radiation shield of the old facility.

The open-and-close operations of these shutters and doors can be done by remote control, and it takes about five minutes in maximum to fully open or close. As workers and researchers enter the irradiation room under full-power continuous KUR operation, the means of the reduction of the induced-radiation especially from activated aluminum were taken prudently.

4.2. The irradiation room and the entrance shield door

The outline of the updated irradiation room is shown in Fig. 3. For the updated irradiation room, both the inner width and height are 2.4m, the depth is 3.6m for the installation of the remote carrying system. The entrance shield door is sliding-door type and the maximum open size is 2.2m. The door has a multi-layer structure consisting of iron, polyethylene and borated-polyethylene, whose total thickness is 1.2 m, for the improvement of the shielding performance against epi-thermal and fast neutrons. The inside of the irradiation room is overall covered with 1 cm-thick borated-polyethylene, in order to reduce the activation of the structure materials.

Six experimental tunnels are cut through the heavy concrete blocks of the irradiation room; two vertically through the ceiling block and four horizontally through the right and left blocks, are cut through. Additionally, four small tunnels are holed for cables, etc., horizontally through the right and left blocks. The irradiation rail device can be set through one of the horizontal experimental tunnels. The monitor lines for a patient, such as anaesthesia hose, etc., the signal lines for monitor televisions, the other lines for experiment, etc., can be drawn out from the irradiation room through a pit under the entrance shield door.

4.3. Dose distributions of the irradiation room under the full-power continuous operation

In order to estimate the exposure dose for the utilisation under the continuous operation, the dose rate distributions inside and around the irradiation room were measured under the full-power operations of 5 MW for the condition of the spectrum shifter and the heavy water shutter tanks "full-filled", the boral filter "full-close", the Beam Shutter "close" and Entrance Shield Door "full-open". A rem-counter and an ionised chamber were used for the dose measurements of neutrons and gamma rays, respectively. The measured dose equivalent rates are shown in Fig. 3. At the 180 cm height from the floor level, where the Beam Shutter does not reach, the doses were higher. At the 90 cm height near the center axis, which corresponded to the normal working area (3 m distant from the bismuth layer surface), the total dose equivalent rate of neutrons and gamma rays was almost 250 μ Sv/hr. This value is larger than the design criterion of 100 μ Sv/hr, due to the addition of the scattered component from the non-shielded areas due to the Beam Shutter. The admittance time per a week will be limited within four hours.

Employing the remote carrying system together with the radiation shielding system, the setting and positioning for a patient is possible at the outside of the irradiation room, and a patient can be carried into the irradiation room by the remote patient carrier, under the full-power continuous operation.



TOP VIEW



SIDE VIEW

FIG.3. Measured dose distribution of neutrons and gamma rays inside and outside of the irradiation room employing the Radiation Shielding Systems under the full-power (5MW) KUR operation.

The carrier moves from the medical treatment room into the irradiation room along the rails. A medical treatment room is settled adjacent to the irradiation room. For the countermeasure against falling bacteria, a bactericidal air-conditioning system is attached on the ceiling in this room. A driving motor for the remote patient carrier is settled in the small pit at the center part under the irradiation room floor-level. The carrier can be remotely moved by electrical power about 90 cm in the irradiation room.

A clinical collimator system and a manual X-Y table are settled on the carrier. A clinical bed with position-control mechanism for up-down and rotation is put on the X-Y table. Then, the positioning to the collimator aperture is easily possible by laser pointers attached on the medical treatment room. Three kinds of the collimator are provided for thermal, mixed and epi-thermal neutron irradiation. Those maximum aperture sizes are 190 mm. These collimators are used together with the inner collimators for several use conditions. The maximum size of irradiated sample treatable by this remote carrying system, is 200 cm in width, 180 cm in height and 2 t in weight.

4.4. The safety observation system

The operation conditions for the HWNIF are always under concentrated observation by the safety observation system. In the standing points of radiation-exposure protection for workers and safety for a patient, the following two interlocks are set. (1) The "open" operation of the entrance shield door is interlocked for the conditions of the beam shutter "close", the heavy water shutter and the spectrum shifter tanks "full", during the KUR power larger than 10 kW. (2) The carrier cannot be moved from the waiting position for the condition of the beam shutter "not open". On the contrary, the beam shutter cannot be closed, for the condition that the carrier is at the irradiation position.

5. CONCLUSION

We completed this remodelling works on the basis of the knowledge for the maintenance and repair of the old facility, the experiences of the NCT clinical irradiation and the basic experiments in many research fields, etc.. In the updated HWNIF of the KUR, the neutron irradiation with various energy spectra, such as mixed irradiation of thermal and epithermal neutrons, solo-irradiation of epi-thermal neutrons, are possible using the thermal neutron filters together with the neutron energy spectrum shifter. The utility and application of the facility became remarkably improved for NCT clinical irradiation.

The first NCT clinical irradiation at the HWNIF was performed for a brain tumour with the thermal neutron irradiation mode in November 1996. Fourteen NCT clinical irradiation; four with thermal neutron irradiation mode "00-0011-F" and ten with the mixed neutron irradiation mode of thermal and epi-thermal neutrons "00-0000-F"; thirteen for brain tumour and one for melanoma, were already performed as of June 1999. Solo-irradiation of epi-thermal neutrons is planning to start in near future. From now on, we will promote the more effective utilisation and application of this facility on the basis of the facility safety.

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