Effect of concrete buildings on the environmental neutron flux by using the reaction $^{197}Au (n,\gamma)\ ^{198}Au$ and extremely low level gamma ray spectroscopy

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Abstract The activation of gold by environmental neutrons through the reaction $^{197}Au (n,\gamma)\ ^{198}Au$ was used to study the effect of concrete buildings on the neutron flux, and to estimate the thermal neutron flux inside and outside these buildings. The results showed that three ceilings of thickness 34 g/cm^2 decrease the fast neutrons to 26% from its original value. However, the same reinforced concrete decreases the slow neturon flux to only 62% of its original value. The thermal neutron flux at 283 m from the center of Training Reactor of Kinki University, was twice higher than the environmental neutron background.

Keywords Environmental neutrons, Gold, Activation, Concrete

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1 Introduction

The vertical cut-off of geomagnetic rigidity and the change of geographical latitudes affect the distribution of the secondary cosmic radiation including neutrons. However, at sea level and vicinity points, the measured neutron data are more affected by the meteorological conditions.[1] The primary cosmic ray interacts with all materials to produce secondary cosmic neutrons. The background measurements at sea or land is made up of neutrons produced, attenuated, reflected, and absorbed by air, water, or ground.[2] The meteorological data include air pressure, overhead cloud-amounts, wind speed, precipitation, and water contents. This means that, the air-ground boundary effects on the spatial and energy distributions of the neutrons depend greatly upon the type of local surroundings.

The results of altitude dependence measurements of the environmental neutron flux by Komura et al,[3] revealed some difference between the measured neutron fluxes inside and outside the concrete buildings. This difference appeared as a decrease of the measured neutron flux inside the buildings compared with that outside at the same altitude and position. The effect of the concrete was more clearly at high altitudes than low altitudes for slow neutron flux. The advantage of this kind of experiment, i.e. effect of the concrete on neutron flux, is that the composition of the concrete can approximately be taken as that of the earth with constant moisture contents.[4]

On the other hand, one of the most important applications of the gold activation by environmental neutrons is to study the neutron flux in the environment around some strong neutron sources such as reactors, accelerators and so on. Around these sources, the neutrons contribute with a great part of radiation doses to the workers in these facilities and other surrounding faces of life.

The aim of the present article is to study the effect of concrete buildings on the environmental neutron flux inside and around these buildings and to estimate the neutron flux in the environment around the training reactor of Kinki University.

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2 Experimental

2.1 Analytical technique

The environmental neutron measurements were carried out indirectly through the radioactivity induced in pure gold by neutron activation. Because of its suitable half-life (2.7d) and high cross section (98.5±0.4 b) for thermal neutrons, the ^{198}Au was the most convenient neutron detector as a product of the ^{197}Au(n,γ)^{198}Au interaction with 100% abundance.[5] The β-decay of ^{198}Au occurs with the emission of 411.8 keV (95.56 %) γ-rays.[6] The γ-ray measurements were made using an extremely low-level gamma-ray counting system consisting of a well-type germanium detector and its electronics. Especially designed for low-level counting,[7] the components of the system are optimized for each application to provide a good energy resolution. The detector has the following characteristics: sensitive volume 344.1 cm^3; relative efficiency 64.7%; energy resolution 1.44 keV at 122 keV, 2.2 keV at 1332 keV; peak/Compton ratio 63; FWTM/FWHM 1.85; FWFM/FWHM 2.43.

To shield the spectrometer from natural radioactive background originating from cosmic rays, uranium and thorium series, and ^{40}K, all the measurements were carried out in a laboratory (underground 270 m) constructed in a tunnel of former Ogoya copper mine which was closed more than 40 years ago. Contribution of cosmic rays in the laboratory was evaluated to be 1 /200 that of the overground level, and the average radon activity values are 20 Bq/m^3. The main shield of the detector was constructed by using 2.5 cm thick old iron plates and 5 cm thick ordinary lead bricks. The total thickness of every shield is at least 20 cm for every side around the detector. Extremely low background lead bricks prepared from old lead was placed as inner shield to reduce γ-rays emitted from the main shield. Old lead was produced more than 200 years ago.

After neutron exposure, the targets were wrapped with cadmium sheets and brought back to the laboratory as early as possible to start γ-ray measurements. The 412 keV γ-ray from ^{198}Au was measured to calculate the number of ^{198}Au atoms produced per unit weight of gold and then converted to the neutron flux using the three-dimensional Monte Carlo Code MORSE-CG.[8]

2.2 Sampling

To study the effect of concrete buildings on the environmental neutron flux, the experiments were mainly conducted in a 3-storied concrete building of Low Level Radioactivity Laboratory (LLRL). Data were taken at each floor, as well as the roof of the building. Three targets of 50g gold grains inside 8.5 cm paraffin thickness were exposed at 1st, 2nd floor and the roof of LLRL. In addition, twelve gold targets of 20g grains were distributed at the roof and the three floors of the building. Three targets (one was bare, the second was covered with paraffin and the third was covered with 0.5-mm cadmium) were exposed at every point with at least 80-cm distance between them. The thickness of every reinforced-concrete layer was estimated to be about 34 g/cm^2.

At Kinki University Training Reactor, 25 gold sheet targets were divided into 5 groups to estimate the thermal neutron flux inside and around the reactor as follows (see Table 3 below).

The first group consisted of 4 targets (Nos. A1~A4), which were exposed inside the reactor body for one-hour operation. Two of these targets (A1, A3) were placed at the core of the reactor and the other two (A2, A4) were placed at 20 cm above the core. One target (A3, A4) at each place was covered with 0.5 mm Cd sheet to estimate the Cd ratio in the high intensity flux. The targets were cooled for 17 days before the γ-ray measurement, this was because of their high radioactivity level.

The second group included 4 targets. Three of them (Nos. 6, 11 and 8) were distributed inside the reactor room at different distances from the reactor center. The rest one (No.3) was in the reactor room but over the reactor body. This group was exposed for one hour only during the reactor operation to estimate the thermal flux around the reactor body during the operation. After 24 h cooling, the γ-ray activity of the targets was measured.

The third group consisted of four bare targets (Nos. 7, 4, 12 and 9) in addition to two targets with Cd cover (Nos.5 and 10). The targets of the third group were exposed at the same points of the second group but for one week to evaluate the average neutron flux over one week operation.

The fourth group was another five gold targets (Nos. 13, 14, 15, 16 and 17) which were distributed inside the reactor building. This group was placed to evaluate the thermal neutron flux which the workers were exposed to for one week.

The last group consisted of 6 targets (Nos. 19, 20, 18, 21, 23 and 22), which were distributed outside the reactor building until 283 m distance to estimate the thermal neutron flux of the reactor environment, and to evaluate the runaway neutrons from the reactor to the surrounding life. Two of these targets were placed together at the far point (Nos. 23 and 22). This group was exposed for one week except for one of the couple targets (No.22), which was lifted for one-month exposure to be compared with the other target.

All targets were wrapped in Cd sheets during the cooling time. The short exposure and cooling times were corrected during the calculations of the production of gold atoms (N) using the known equation (1).[9-11]

$$N=\frac{C}{\varepsilon}\bullet\frac{1}{1-\exp(-\lambda t_{\mathrm{R}})}\bullet\frac{1}{\exp(-\lambda t_{\mathrm{D}})}\bullet\frac{1}{1-\exp(-\lambda t_{\mathrm{C}})},\quad A=\lambda N \tag{1}$$

where A is the reaction rate, λ is the decay constant of the nuclide ^{198}Au, C is the peak count, ε is the peak efficiency, t_R is the irradiation (exposure) time, t_D is the decay (cooling) time before counting, and t_C is the counting (measuring) time.

3 Results and discussion

3.1 Influence of concrete buildings on the environmental neutron flux

Table 1 depicts some results of the altitude dependence of environmental neutron flux.[3] These results show that at 500 m height the slow neutron flux inside the concrete building decreased to 80% of the outside value. The ratio decreased to 70% at 1000m height, which means that the reinforced concrete is more effective for the high fluxes. On the other hand, these ratios were 80% and 79% for the fast neutrons. This may be due to that the difference between the fast neutron fluxes at these altitudes were not so high.

Table 1 Effect of concrete building on neutron fluxes at different altitudes

Altitude (m)	Slow neutron flux ($\times10^{-3}$cm$^{-2}\cdot$s^{-1})			Fast neutron flux ($\times10^{-3}$cm$^{-2}\cdot$s^{-1})		
	Outside the building	Inside the building	Reduction ratio (%)	Outside the building	Inside the building	Reduction ratio (%)
500	12.5 ± 1.7	10.0 ± 1.5	79.8	2.3 ± 0.3	2.1 ± 0.3	80.4
1000	17.1 ± 2.3	12.0 ± 1.5	70.2	3.0 ± 0.3	2.3 ± 0.5	79.3

Fig.1 depicts the results of the concrete effect experiments. The effect of reinforced concrete is clearly observed from these results. The neutron fluxes were gradually decreased from the roof to the 1st floor.

Table 2 shows the reduction of the neutron flux measured in different stores. From the table, the fast neutron flux at the 1st floor showed 36% and 26% reduction from its values at the roof in the first and second experiment, respectively. As a comprehensive estimation, the second experiment is the most interesting for discussion. Although in the fast neutrons measurement, the reduction ratio of 3rd floor / roof and 1st floor / 2nd floor is constant, about 13%, this

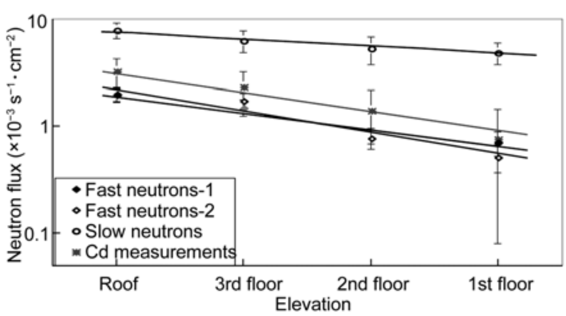

Fig.1 Effect of the concrete ceilings on the measured neutron fluxes.

ratio increased to 47 % for 2nd / 3rd floor. This high reduction can be attributed to the thermalization of fast neutrons due to the interaction or scattering with the component of the library, such as aluminum shelves, books, … and so on where the targets were placed.

Table 2 Experimental neutron fluxes measured on the roof and floors of the concrete building

Experiment No.	Exposure condition	Exposure elevation	Reduction ratio / floor	Cd ratio
I	Fast neutron flux	Roof	1.00	-
		2nd floor	0.76	-
		1st floor	0.36	-
II	Fast neutron flux	Roof	1.00	-
		3rd floor	0.86	-
		2nd floor	0.39	-
		1st floor	0.26	-
	Slow neutron flux	Roof	1.00	-
		3rd floor	0.81	-
		2nd floor	0.68	-
		1st floor	0.62	-
	Cadmium measurements	Roof	1.00	2.4
		3rd floor	0.72	2.7
		2nd floor	0.44	3.7
		1st floor	0.23	6.4

The cadmium ratio measurements had a similar behaviour like that of the fast neutrons because its value depended mainly on the epithermal flux. On the other hand, the slow neutron flux showed a different behavior. The reduction ratio of 3rd floor / roof was about 19.6% and 13% for 2nd floor / 3rd floor. This ratio decreased to 6% for 2nd floor / 1st floor. The reduction at the 3rd floor came from the concrete effect, however, in the 2nd and 1st floors values, the absorbed slow flux by concrete is compensated by the thermalization of the high energy flux by the concrete and the other components and furniture of the building.

The Cd ratio increased gradually from 2.4 at the roof to 6.4 at the 1st floor, which means that the thermal neutrons are more predominant than the epithermal at the lower floors. This effect is a result of the thermalization of fast neutrons by the concrete.

From these results, it is evident that the concrete is an effective shield for the high-energy neutrons. Three ceilings of 34 g/cm^2 thickness decreased the flux to 26% from its original value. However, the same reinforced concrete decreases the slow flux to only 62% from its original value. These results can call attention to the effect of concrete walls as a shielding around the reactors, especially those built near to the living environments.

3.2 Estimation of thermal neutron flux around the Training Reactor

The thermal neutron flux was estimated inside and around the reactor. The source of fast neutrons is Pu-Be of 3.7×10^{10} Bq. The estimated thermal flux at the core of the reactor is 1.2×10^{7} cm^{-2}·s^{-1} and at 20 cm over the core is 1.2×10^{5} cm^{-2}·s^{-1}. The fast neutron flux is 1.3×10^{6} cm^{-2}·s^{-1} at the core. The Cd ratio had been estimated to be 5~7 at the core of the reactor (Private communication).

Table 3 shows the results of one-hour exposure targets. The thermal neutron flux at the core of the reactor is 1.24×10^{7} cm^{-2}·s^{-1} and at 20 cm above the core is 5.25×10^{5} cm^{-2}·s^{-1}. The Cd ratio at the two points is 7 and 7.1, respectively. These results are in well agreement with the original data mentioned before. The slight increase of the present data is due to the sensitivity of gold to the epithermal flux. The epithermal neutrons are reflected and moderated from the fast neutron flux by the components of the reactor or the moderator. The results around the reactor body (target Nos. 6, 3, 11 and 8) showed no clear relation between the flux intensity and the distance from the reactor. This is due to the target position with respect to the neutron source. Hence, the target No.3 at 3.1m distance was placed over the reactor body. The others (6, 11 and 8) were beside the walls of the reactor body or steel door or the concrete outer wall. So, the adjacent materials may have some effect on the neutron flux. This effect is more clearly at 5.7 m target distance where the targets No. 8~10 were stocked to steel door. Hence, the thermal neutron production over steel bodies is about 11 times higher than the environmental background.[12] This is because of the marked rise of the flux at those points. The average thermal flux inside the reactor room is about 77.6 ± 3.5 cm^{-2}·s^{-1} over the reactor (target No.3), and 15.2 ± 2.3 cm^{-2}·s^{-1} around the reactor (targets No. 6, 11 and 8).

Table 3 Results of thermal neutron flux inside and around the reactor for one hour and one week operation time

Operation time	Target No.	Conditions	Distance from the reactor center (m)	Thermal neutron flux (cm^{-2}·s^{-1})	Cd ration
One hour operation	A1	Bare, inside Reactor	0	1.24×10^{7}	
	A2	Bare, inside Reactor	0.2	5.25×10^{5}	
	A3	Cd, inside Reactor	0	1.78×10^{6}	7.0
	A4	Cd, inside Reactor	0.2	7.81×10^{4}	7.1
	6	Bare, inside reactor room	1.9	13.3±1.6	
	3	Bare, inside reactor room	3.1	77.6±3.5	
	11	Bare, inside reactor room	5.2	14.6±1.3	
	8	Bare, inside reactor room	5.7	17.8±1.8	
One week operation	7	Bare, inside reactor room	1.9	0.547±0.063	
	4	Bare, inside reactor room	3.1	3.35±0.18	
	12	Bare, inside reactor room	5.2	0.629±0.040	
	9	Bare, inside reactor room	5.7	0.860±0.066	
	5	Cd, inside reactor room	3.1	0.790±0.044	4.2
	10	Cd, inside reactor room	5.7	0.197±0.032	4.4
	13	Bare, inside building	6.0	0.412±0.040	
	14	Bare, inside building	9.7	0.101±0.014	
	15	Bare, inside building	12.6	0.102±0.015	
	16	Bare, inside building	15.7	0.0406±0.0077	
	17	Bare, inside building	17.6	0.0343±0.0082	
	19	Bare, outside reactor building	12	0.0186±0.0041	
	20	Bare, outside reactor building	40	0.0213±0.0029	
	18	Bare, outside reactor building	70	0.0245±0.0043	
	21	Bare, outside reactor building	140	0.0098±0.0026	
	23	Bare, outside reactor building	283	0.0160±0.0017	
	22*	Bare, outside reactor building	283	0.0154±0.0026	

* 22: lifted for one month outside the reactor building

Also, from Table 3 it can be seen that the thermal neutron fluxes measured for one week at the same points compared with those for one-hour operation have the same behavior except that its intensity is distributed over one week exposure. The average of the slow flux intensity over one week operation are decreased to 3.4±0.2 $cm^{-2}\cdot s^{-1}$ and 0.7±0.2 $cm^{-2}\cdot s^{-1}$ over and around the reactor, respectively. These results show that the exposure for one week decreased the average flux to 4.3% and 4.5% over and around the reactor, respectively, comparing with its value during the one hour operating. The Cd ratio at 3.1m and 5.7m were 4.2 and 4.4, respectively. Comparing these ratios with those obtained inside the reactor body shows that the Cd-shield is more effective at the high flux level.

The thermal neutron flux distribution inside the reactor room is plotted together with the flux inside the building in Fig.2 to show the variation of the intensity with the distance from the reactor center. This figure shows obviously that the thermal neutron flux decreases linearly with the distance from the reactor.

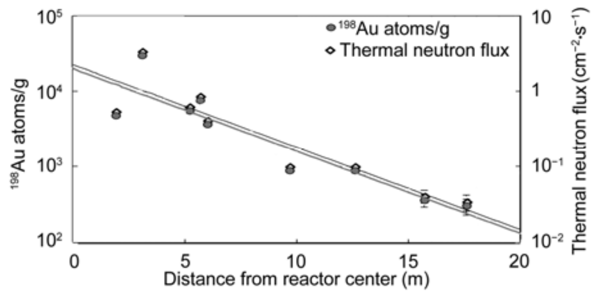

Fig.2 Variation of thermal neutron flux and production of ^{198}Au with the distance from the reactor center, inside the reactor building.

Fig.3 shows that the thermal neutron flux decreases linearly with the distance outside the reactor building and the flux is still higher than the environmental background level. The marked increase of the flux at 40 m and 70 m is due to the fact that, for the targets placed inside aluminum boxes, the production of the neutron flux is about 3 times higher than the environmental background.[12] The drop of the flux at 140 m distance is due to the effect of much concrete buildings around the workshop station, where the target was placed.

The thermal neutron flux at 283 m was (16±1.6) $\times10^{-3}$ $cm^{-2}\cdot s^{-1}$ as seen in Fig.3. This value is twice higher than the environmental background, (8.4 ± 0.6)$\times10^{-3}$ $cm^{-2}\cdot s^{-1}$.[5] Target No.22 was exposed at the same point 283 m for one-month exposure and showed nearly the same value, (15.4±2.6)$\times10^{-3}$ $cm^{-2}\cdot s^{-1}$. This result shows the stability of the environmental neutron flux over the whole time.

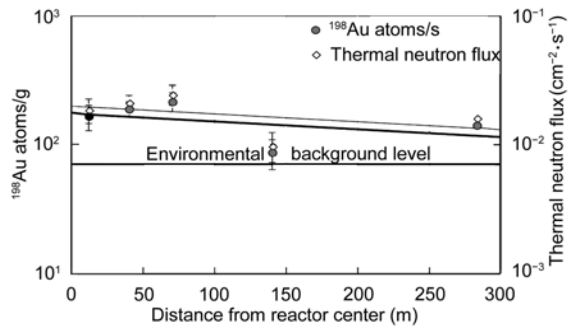

Fig.3 Variation of thermal neutron flux and production of ^{198}Au with the distance from the reactor center, outside the reactor building.

Fig.4 depicts the thermal neutron flux inside and outside the reactor building for the targets exposed for one week. From this figure it is evident that the thermal neutron flux is rapidly decrease with the increase of the distance from the reactor center. This effect is due to the interaction or absorption of the thermal neutrons with or by the air, concrete buildings, and the other surrounding materials. The figure shows obviously that the rise of the environmental neutron flux in the environment around the reactor is caused by the diffusion of the reactor neutrons.

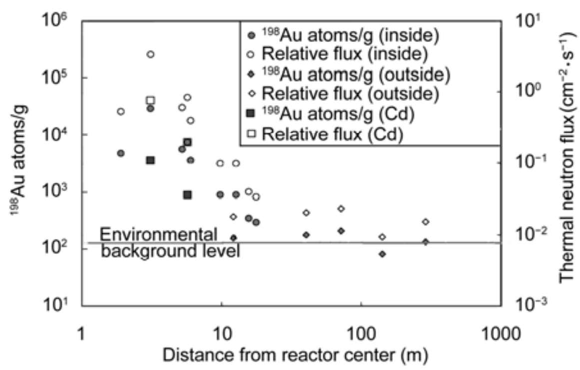

Fig.4 Variation of thermal neutron flux and production of ^{198}Au with the distance from the reactor center inside and around the reactor.

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