Neutron activation cross sections of $^{12}$C, $^{27}$Al, $^{59}$Co, $^{64}$Cu, and $^{209}$Bi nuclides in the energy range 20MeV to 200MeV

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The neutron activation cross sections of $^{12}$C, $^{27}$Al, $^{59}$Co, $^{64}$Cu and $^{209}$Bi nuclides have been measured the quasi-monoenergetic p-Li neutron field in the energy range 20MeV to 200MeV. The irradiation experiments were performed at four cyclotron facilities of 1) INS, of Univ. of Tokyo, 2) CYRIC of Tohoku Univ., 3) TIARA of JAERI, and 4) RIKEN. Our experimental data were compared with other experimental data, the calculation data and the ENDF/B-VI high energy file data.

1. INTRODUCTION
With the increasing use of the high energy and high intensity accelerators, the radioactivities induced in the accelerator materials, shielding materials and the air and water of accelerator facility bombarded by the primary accelerating charged particles as well as the secondary neutrons have become a serious problem. Nevertheless, neutron activation cross section data in the energy range above 20MeV are very poor and no evaluated data file exists at present mainly due to very limited number of facilities having quasi-monoenergetic neutron fields. In this paper, we measured the neutron activation cross sections of $^{12}$C, $^{27}$Al, $^{59}$Co, $^{64}$Cu, and $^{209}$Bi nuclides by using the quasi-monoenergetic p-Li neutron fields at four cyclotron facilities of 1) Institute for Nuclear Study (INS), University of Tokyo [1], 2) Cyclotron and Radioisotope Center (CYRIC), Tohoku University [11], both for 20 to 40 MeV protons, 3) Takasaki Research Establishment (TIARA), Japan Atomic Energy Research Institute [21] for 40 to 90 MeV protons, and 4) Institute of Physical and Chemical Research (RIKEN) for 80 to 210MeV protons [11].

2. EXPERIMENT
The irradiation experiments were performed in the quasi-monoenergetic neutron fields produced by the $^7$Li(p,n) reaction, which have been established at three cyclotron facilities of the AVF cyclotron at INS for 20 to 40MeV, of the AVF cyclotron at TIARA for 40 to 90MeV, and of the Separate-Sector (Ring) cyclotron at RIKEN for 80 to 150MeV. The Li-targets of 2 to 10 mm thicknesses were bombarded by proton beams of 20 to 150 MeV energies which were extracted from the these cyclotrons. The neutron produced in the forward direction from the target were transported through the collimator for sample irradiation and the proton beams passed through
the target were swept out by the magnet to the beam dump at CYRIC, TIARA and RIKEN. The cross section measurements between 20 and 40MeV were performed at INS, because the neutron fluence of the CYRIC neutron field was too low for sample irradiation. The sample were placed only 10cm away from the Li target in the forward direction at INS, and the neutron spectra were measured at CYRIC where the same target configuration was prepared, because the INS neutron field has not enough space for neutron spectrometry with the TOF method. The absolute neutron fluence was determined with the PRT (Proton Recoil counter Telescope) at TIARA, and with the Li activation-method to detect the $^7$Be activity from the $^7$Li(p,n)$^7$Be reaction at CYRIC and RIKEN. Table I summarizes the physical data of irradiation samples used at INS, TIARA and RIKEN. At INS, the samples were irradiated for 4 to 13 hours, and at TIARA and RIKEN the irradiation time consisted of short irradiation time (1 to 2 hours under 120MeV, 30min above 120MeV) and long irradiation time (about 20 hours) by considering the half lives of produced nuclei. The proton beam current during irradiation was recorded by the digital current integrator to monitor fluctuations of the neutron fluence. After the irradiation, the gamma rays emitted by the irradiated samples were measured with a high purity HP-Ge detector. These samples were counted several times to identify the half-lives of the activities produced.

3. ANALYSIS
Estimation of photo-peak area of gamma-ray spectra which were measured by the HP-Ge detector was done with the computer program for the automatic analysis of gamma-ray spectrum by Komura et al. [4] or the direct counting of eye-fitting peak area only for very weak photo-peaks.

The reaction rates of radioisotopes identified from the gamma-ray spectra and the decay curves were estimated after corrected for the peak efficiency of Ge detector, the coincidence-summing effect, self-absorption effect in thick samples, and also for the beam current fluctuation during sample irradiation. The peak efficiency of the HP-Ge detector was determined by using the standard mixed gamma-ray source and the correction factor for self absorption of gamma rays in the samples were calculated by the Monte Carlo code, PEAK [5] for the INS and TIARA experiments, but for the RIKEN experiment, the peak efficiencies of the HP-Ge detector including the self-absorption of the irradiated samples were calculated simultaneously by the electron-photon cascade Monte Carlo code, EGS4[6], due to very thick (8mm) samples as seen in Table I. The coincidence-summing effect caused by the coincidence detection of two or more gamma rays in the gamma ray spectrum was corrected by using the SUMECC code [7]. Fluctuation of the neutron flux during the irradiation was treated by using the proton beam current monitor data recorded by the MCS.

The reaction rate per beam current, $R$, corrected for beam current fluctuation becomes
\[ R = \frac{\lambda \cdot C}{N \cdot \epsilon \cdot \gamma \cdot e^{-\frac{\lambda}{T_c}} \cdot (1 - e^{-\frac{\lambda}{T_m}}) \cdot \sum \{ Q_i \cdot e^{-\frac{\lambda}{T_m} \cdot (n - i) \cdot \Delta t} \}} \text{[Coulomb}^{-1} \cdot \text{atom}^{-1}] \]  

(1)

where \( \lambda \) is decay constant (sec\(^{-1}\)), \( C \) total counts of gamma-ray peak area, \( N \) number of atoms in the target (atom), \( \epsilon \) peak efficiency, \( \gamma \) branching ratio of gamma rays, \( T_c \) cooling time (sec), \( T_m \) counting time (sec) and \( Q_i \) beam current (Coulomb) for irradiation time interval \( \Delta t \) (sec).

The reaction rate given by Eq.(2) is connected to neutron cross section \( \sigma(E) \) as follows,

\[ R = \int_{E_{th}}^{E_{max}} \sigma(E) \cdot \phi(E) \cdot dE, \]  

(2)

where \( \phi(E) \) is the neutron fluence (n \cdot cm\(^{-2}\) \cdot MeV\(^{-1}\) \cdot Coulomb\(^{-1}\)), \( E_{th} \) the threshold energy and \( E_{max} \) the maximum energy of the monoenergetic peak neutrons.

The cross section at the effective energy of peak neutrons \( \sigma(E_{eff}) \) can be obtained, after corrected the contribution of low-energy tail neutrons to the obtained reaction rate as follows

\[ \sigma(E_{eff}) = \frac{R \cdot f}{\phi(E_{peak})}. \]  

(3)

The \( f \) value is the ratio of peak reaction rate from \( E_{min} \) (minimum energy of the monoenergy peak) to \( E_{max} \) and total reaction rate from \( E_{th} \) to \( E_{max} \), the peak-to-total ratio of reaction rate, which is expressed as,

\[ f = \frac{\int_{E_{th}}^{E_{max}} \sigma(E) \cdot \phi(E) \cdot dE}{\int_{E_{th}}^{E_{max}} \sigma(E) \cdot \phi(E) \cdot dE}. \]  

(4)

The \( f \) value is at first estimated from the lowest peak neutron energy by using the measured neutron flux, \( \phi(E) \) and the cross section, \( \sigma(E) \), of the evaluated high energy data files, ENDF/B-VI or experimental data complied by McLane et al. \(^{18} \). By using thus-obtained \( f \) value, the cross section at the higher peak neutron energy, \( \sigma(E_{eff}) \) can be calculated from Eq.(3).

The statistical errors of the reaction rates were about 0.5% to 30% depending on the gamma-ray peak counts of irradiated samples. As for the coincidence-summing effect, the branching ratios of \(^{202}\)Bi to \(^{198}\)Bi isotopes produced by \(^{209}\)Bi(n,8n) to \(^{209}\)Bi(n,12n) reactions are not definitely given, then we assumed to be additional 10% errors by neglecting the correction of the coincidence-summing effect. The errors of peak neutron fluences were between 4% and 15%. The error of the \( f \) value which were used for the correction of the contribution to the reaction rate due to the low-energy neutron tail was estimated to be 5% to 40% at maximum by combining the neutron flux error of 2% to 7% with the cross section error between 1% and 40% according to the variations observed for the published data.

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4. RESULTS AND DISCUSSION

The neutron activation cross sections of $^{12}$C, $^{27}$Al, $^{59}$Co, $^{64}$Cu, and $^{209}$Bi nuclide in the energy range from 20 and 150 MeV were measured by the activation method using the quasi-monoenergetic neutron field based on the $^7$Li(p,n)$^7$Be reaction. As examples, Fig.1 and Fig.6 give the cross section data of $^{12}$C(n,2n) and $^{209}$Bi(n,xn)$^{210}$Bi reactions respectively, compared with other experimental data and ENDF/B-VI high energy file data. We found that the $^{12}$C(n,2n)$^{11}$C cross section has a constant value of about 20 mbarn in the energy range above 40 MeV. The experimental cross section data of $^{59}$Co(n,xn)$^{60}$Co was compared with the calculation data by Odano\textsuperscript{[10]} and the obtained data $^{27}$Al(n,2n$\alpha$)$^{22}$Na, $^{64}$Cu(n,sp)$^{64}$Mn to $^{64}$N(n,sp)$^{65}$Ni and $^{209}$Bi(n,xn)$^{210}$Bi were compared with the ENDF/B-VI high energy file data calculated with the ALICE code\textsuperscript{[11]} by Fakahori. Our experimental results for $^{209}$Bi nuclide are the first experimental data and are generally in good agreement with the ENDF/B-VI data.

Reference

[8] National Nuclear data Center, Brookhaven National Laboratory, ENDF/B-V(1990)

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