

Development of p-Li Monoenergetic Neutron Field and Measurement of Neutron Activation Cross Sections (p-Li単色中性子場の開発と中性子放射化断面積の測定)

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論 文 内 容 要 旨

1 Introduction

Neutron activation cross section data for neutron energy above 20 MeV are very scarce and discrepant and no evaluated data files are presently available because preparing a monoenergetic neutron source for activation experiment in this energy region gives some difficulties. In this work, the intense monoenergetic neutron field produced by a ${}^7\text{Li} (p, n) {}^7\text{Be}$ reaction for irradiation was developed in order to measure the neutron activation cross sections of several enriched and natural isotopes.

Production of monoenergetic neutron field was obtained by using two types of target system, one is 2 mm thick ${}^7\text{Li}$ target backed with 12 mm thick carbon and the other one is the 12 mm thick carbon alone. The thick carbon target was used as a proton beam stopper. The true neutron spectrum was estimated by the difference of these two spectra. The same types of target system were also applied for sample irradiations.

2 Experiment

Neutron spectrum from the ${}^7\text{Li} (p, n) {}^7\text{Be}$ reaction was measured by the TOF method at the facility of Cyclotron and Radioisotope Center (CYRIC), Tohoku University, with a liquid scintillation detector NE-213 of 127 mm diameter x 127 mm long placed at about 12 m behind the target. The protons of 20, 25, 30, 35 and 40 MeV were used to bombard two targets at 0 deg, and angular distribution of produced neutrons was measured using the beam-swinger system from 0 to 125 deg.

The enriched samples of ${}^{24}\text{Mg}$, ${}^{25}\text{Mg}$, ${}^{28}\text{Si}$, ${}^{29}\text{Si}$, ${}^{54}\text{Fe}$, ${}^{56}\text{Fe}$, ${}^{63}\text{Cu}$, ${}^{65}\text{Cu}$, ${}^{64}\text{Zn}$ and ${}^{66}\text{Zn}$, and natural samples of ${}^{12}\text{C}$, ${}^{23}\text{Na}$, ${}^{27}\text{Al}$, ${}^{55}\text{Mn}$ and ${}^{197}\text{Au}$ were irradiated by using 20, 25, 30, 35 and 40 MeV proton beams extracted from the SF cyclotron at Institute for Nuclear Studay (INS), the University of Tokyo.

The proton beam intensity during irradiation was measured by digital current integrator, and a multichannel scaler to monitor the beam fluctuation during irradiation. The target cooling during irradiation was done by blowing the air into the surface of target chamber.

Since the neutron spectrum and sample irradiation were undertaken at different facilities, it is necessary to normalize the neutron spectrum obtained at CYRIC to absolute value, so that it can be used for the cross section estimation of the samples. The normalization was done by lithium activation method, based on the measurement of ${}^7\text{Be}$ yield produced by the ${}^7\text{Li}(p, n){}^7\text{Be}$ reaction, coupled with the measurement of angular distribution.

3 Analysis Method

The yield of peak neutron at 0 deg, $\phi(E_{\text{peak}})$ can be estimated by

$$\phi(E_{\text{peak}}) = \frac{N_{7\text{Li}} \left(\frac{d\sigma}{d\Omega} \right)_{\theta=0}}{\int_{4\pi} \frac{d\sigma}{d\Omega} d\Omega}, \quad (1)$$

where $N_{7\text{Be}}$ is the number of ${}^7\text{Be}$ produced in the ${}^7\text{Li}$ target during irradiation, N_p is the number of injected protons, and $\left(\frac{d\sigma}{d\Omega} \right)_{\theta=0}$ is the ${}^7\text{Li} (p, n){}^7\text{Be}$ cross section producing the peak energy neutrons at 0 deg, while $\frac{d\sigma}{d\Omega}$ is the differential cross section obtained from angular distribution measurement. Then, the absolute value of the full neutron spectrum, $\phi(E)$, during irradiation at the INS cyclotron can be obtained by using the relative neutron spectrum of wide energy region at 0 deg, $\phi_{\text{rel}}(E)$ obtained from the CYRIC TOF measurement, as follows,

$$\phi(E) = \phi_{\text{rel}}(E) \frac{\phi(E_{\text{peak}})}{\int_{\text{peak}} \phi_{\text{rel}}(E) dE}, \quad (2)$$

where $\int_{\text{peak}} \phi_{\text{rel}}(E) dE$ is the relative value of peak area of neutron spectrum.

The reaction rate of activated samples per incident proton beam current was estimated by

$$R = \frac{\lambda C}{\varepsilon_p I_\gamma (1 - e^{-\lambda t_m}) e^{-\lambda t_c} (1 - e^{-\lambda t_{ir}}) Q} \quad (3)$$

where C is the area of gamma-peak from gamma-spectrometry, ε_p the photo-peak efficiency, I_γ the branching ratio, λ the decay constant, t_m the measuring time, t_c the cooling time, t_{ir} the irradiation time, and Q the incident proton beam current.

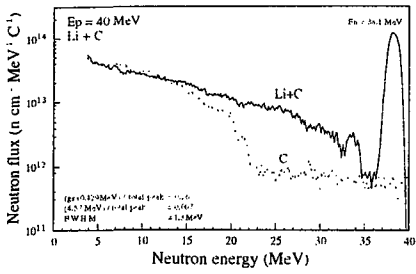
The cross section was estimated by combining the absolute value of neutron spectrum and the reaction rate with considering the low-energy continuum of neutron spectrum as,

$$\sigma(E_{\text{peak}}) = \frac{R - \int_{E_{\text{threshold}}}^{E_{\text{peak-min}}} \sigma(E) \phi(E) dE}{\int_{E_{\text{peak-min}}}^{E_{\text{peak-max}}} \phi(E) dE} \quad (4)$$

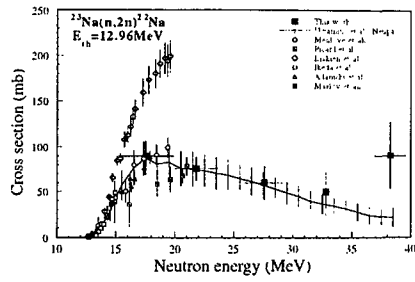
4 Results

The neutron energy spectrum obtained from bombardment of the two types of the ${}^7\text{Li} + \text{C}$ and C targets for 40 MeV protons is shown in Fig. 1. The dominant peak of ${}^7\text{Li} (p, n){}^7\text{Be}$ at the neutron energy of 38 MeV comes from the ground state and the first excited state (0.429 MeV) that can not be separated due to 1.2 MeV energy loss in the target. The smaller peak at 33 MeV neutron energy corresponds to the second excited state (4.57 MeV). The low energy continuous spectrum corresponds to the higher energy state of ${}^7\text{Be}$ and the ${}^{12}\text{C} (p, n)$ and ${}^{13}\text{C} (p, n)$ reactions. The spectrum obtained from carbon backing shows the sharp decrease at neutron energy around 22 MeV due to the Q-value of ${}^{12}\text{C} (p, n)$ reaction that is 18.1 MeV.

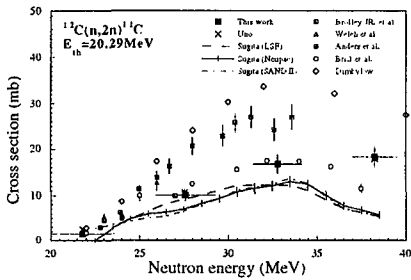
Neutron activation cross sections of fifteen nuclear reactions have been estimated. As examples, Figs. 2, 3 and 4 show the neutron activation cross sections for ${}^{12}\text{C} (n, 2n){}^{11}\text{C}$, ${}^{23}\text{Na} (n, 2n){}^{22}\text{Na}$ and ${}^{197}\text{Au} (n, 4n){}^{194}\text{Au}$ reactions, respectively. The present results of ${}^{12}\text{C} (n, 2n)$ cross section show good agreement with the data obtained by Brill et al., except for results at 38 MeV neutron energy. The same tendency is seen in Fig. 3 for ${}^{23}\text{Na} (n, 2n)$ cross section in the present data. Results of ${}^{197}\text{Au} (n, 4n) {}^{194}\text{Au}$ cross section lie in between the data measured by Uwamino et al. and the data calculated by Greenwood.



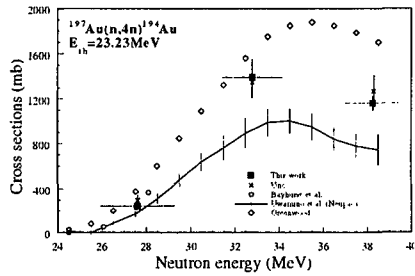
☒ 1 : Neutron energy spectra from 40 MeV protons



☒ 3 : Activation cross section of $^{23}\text{Na} (n, 2n) ^{22}\text{Na}$ reaction



☒ 2 : Activation cross section of $^{12}\text{C} (n, 2n) ^{11}\text{C}$ reaction



☒ 4 : Activation cross section of $^{197}\text{Au} (n, 4n) ^{194}\text{Au}$ reaction

審査結果の要旨

加速器利用の急速な進展に伴い、生成される中性子による放射化は作業者に対する被曝や環境への影響評価にとって極めて重要である。しかし中性子放射化断面積のデータは 20MeV 以上では単色中性子場が少ないためにほとんど存在しない。著者は、 ${}^7\text{Li} (p, n) {}^7\text{Be}$ 反応により 20MeV から 40 MeV に至る単色性に優れた中性子場を作成し、その場を用いて加速器や建屋の構造材を構成する核種の放射化断面積を求めている。本論文はその経緯を述べたもので、前編 6 章から成る。

第 1 章は序論である。

第 2 章では、p-Li 単色中性子の生成法および誘導放射能と放射化断面積の関係について概説している。2 種類のターゲット、つまり Li+C (カーボン) と Cのみ、を用いて両者の差をとる方法は、著者が新たに開発したものである。

第 3 章では、放射化断面積測定用試料の照射のために、東京大学原子核研究所サイクロトロンに作成した p-Li 単色中性子場のスペクトルを、東北大学サイクロトロン・ラジオアイソトープセンターのサイクロトロンの中性子飛行時間測定システムおよびビームスウィンガーシステムを用いて、同一のターゲットに対して行った測定について述べている。また、照射試料の放射能測定のための高純度ゲルマニウム検出器によるガンマ線スペクトロメトリーについても述べている。

第 4 章では、p-Li 中性子スペクトルと試料のガンマ線スペクトルの解析法を述べている。特に、この 2 つのサイクロトロンの p-Li 中性子場のスペクトルをつなぐために、p-Li 反応で生成した ${}^7\text{Be}$ の量をガンマ線スペクトロメトリーにより測定し、規格化する手法について詳しく述べ、放射化断面積とその誤差の評価法については新たな手法を説明している。

第 5 章が、最終的に得られた 15 種類の中性子放射化断面積をまとめた中心となる章である。20 MeV 以下での他の実験値と比べることにより、本研究の結果がよい精度を持っていることを確認し、 ${}^{12}\text{C}$, ${}^{23}\text{Na}$, ${}^{25}\text{Mg}$, ${}^{54}\text{Fe}$, ${}^{55}\text{Mn}$, ${}^{63}\text{Cu}$, ${}^{64}\text{Zn}$, ${}^{197}\text{Au}$, 各核種に対する 20MeV 以上の中性子に対する $(n, 2n)$, $(n, 3n)$ 反応を中心とする断面積データを初めて提供している。これは重要な成果である。

第 6 章は結論である。

以上要するに、本論文は加速器構造材や遮蔽材として重要な核種の 20MeV 以上の中性子放射化断面積を初めて与えたものであり、原子核工学、加速器工学の発展に寄与するところが少なくない。よって、本論文は博士 (工学) の学位論文として合格と認める。