VIII-II-1. Project Research

Project 2

Study on Nuclear Data for MA and LLFP Nuclides

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OBJECTIVE and RESEARCH SUBJECTS:

The main objective of this research project is to improve the accuracy of nuclear data (cross sections) for the design of innovative reactors through the experimental research for the MA and LLFP nuclides. This project consists of six research subjects from five institutions, as follows.

<u>20P2-1</u>: A Study on Neutron-Capture Reactions by High-Energy Gamma-ray Spectroscopy (H. Harada et al., JAEA)

<u>20P2-2</u>: Study on Neutron Capture Cross Sections of MA and LLFP Nuclei using Various Detectors (J. Hori et al., Kyoto Univ.)

<u>20P2-3</u>: MLPPAC(Multi-Layer Parallel Plate Avalanche Counter) for Measurements of Fission Cross-sections of Minor Actinides (T. Ohtsuki et al., Tohoku Univ.)

<u>20P2-4</u>: Fractionation of Stable Strontium Isotopes in a Precipitation Reaction (T. Fujii et al., Kyoto Univ.)

<u>20P2-5</u>: Study on Nondestructive Analysis and Neutron Behavior Using Photonuclear Reaction (K. Ishibashi, et al., Kyushu Univ.)

<u>20P2-6</u>: Study on Neutron Energy Spectrum Measurements for Pulsed Neutron Source (K. Nakajima et al., Kyoto Univ.)

MAIN RESULTS and CONTENTS of this REPORT:

<u>H. Harada *et al.* (20P2-1)</u> have studied the neutron capture prompt gamma-rays of ²³⁸U by using a 4π Ge spectrometer system consisting of 20 Ge crystals surrounded with large BGO detectors at the KURRI electron linac facility. The neutron resonance dependence of the prompt gamma-ray spectrum was quantitatively deduced for the energy region from thermal to 37 eV. The data shows significant changes of prominent prompt gamma-ray yields among the resonances in the investigated energy region. The obtained information on quantitative decay properties will be used to study the accuracy of data analysis method on neutron capture cross sections.

<u>J. Hori *et al.* (20P2-2)</u> have measured capture gamma rays from the 110-eV resonance of 93 Zr and the 301-eV resonance of 96 Zr with a 4 π Ge spectrometer to check the

capability of discriminating the capture events caused by different isotopes. Through the analysis of the data, it is expected that the backgrounds due to impurities in the samples can be eliminated by applying the gates on the 919-keV ground-state transition for ⁹³Zr and three primary transitions for ⁹⁶Zr, respectively.

T. Ohtsuki *et al.* (20P2-3) have rebuilt MLPPAC without light elements and tested it using the lead slowing-down spectrometer at KURRI. In the measurements, 241 Am was used as a target sample and 235 U was used as a reference one. The results show a good agreement with the other experimental data and the evaluated values of JENDL-3.3, which suggests the successful use of the detector.

<u>T. Fujii *et al.* (20P2-4)</u> have studied quantum chemical calculations of hydrated Sr(II) complexes. Equilibrium geometry and hydration enthalpy of $Sr(H_2O)_8^{2+}$ were studied. The calculation results agreed with the literature values. The reduced partition function ratios of Sr(II) species were also calculated in order to know the magnitude of isotope fractionation of Sr.

<u>K. Ishibashi *et al.* (20P2-5)</u> examined the feasibility of nondestructive analysis of "difficult to measure" nuclei, such as 238 U, in a nuclear waste using an electron linac. They have measured the average photonuclear reaction cross sections for 238 U and 93 Nb at 26 and 32 MeV of photon energies. As a result, the existing data at SAC-LAY and Lawrence Livermore have same tendencies but 40 - 60 % larger values than those of this experiment.

<u>K. Nakajima *et al.* (20P2-6)</u> have investigated the techniques to measure the neutron energy spectrum of the pulsed neutron source such as a source using the electron linear accelerator. The following three techniques were investigated in this study: (1) Multi-foil activation method, (2) Time-of-Flight (TOF) method, and (3) Neutron counter measurement. Those three techniques have some drawback and advantage, respectively, for the application to the pulsed neutron. For the fast energy region, the neutron counter measurement, using the proton recoil proportional counter would be suitable. For the thermal to epithermal range, the TOF method would give the good results. For the intermediate energy range, multi-foil activation method would be available, if the proper foils are used.

PR2-1 Study of Neutron-Capture Reactions by High-Energy Gamma-ray Spectroscopy

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INTRODUCTION: Accurate neutron capture cross sections of minor actinides and fission products are required for the study of an innovative nuclear system. To satisfy the demand, a 4π Ge spectrometer has been developed, and utilized for the neutron capture cross section measurements using the time-of-flight (TOF) technique. In the process of deducing the capture cross sections, the knowledge of neutron resonance dependence of γ -ray spectrum is required. To study the dependence, we have utilized the spectrometer in the measurement of the neutron capture cross section of 238 U.

EXPERIMENTS and RESULTS: The 4π Ge spectrometer system consisting of two cluster type Ge detectors (7 Ge crystals each) surrounded by BGO anti-coincidence shields and of 6 coaxial Ge detectors was set at the 10 m neutron flight path of the electron linear accelerator of KURRI. A natural Uranium sample (11.05g) was set in the center of the spectrometer system. The linac was operated with electron energy of 30MeV, an average beam current of 64μ A, a repetition rate of 50Hz and a pulse width of 3.0 μ s.

Fig.1 shows a TOF spectrum for the U sample measured by the spectrometer, and γ ray spectra gated by the neutron resonances at 6.67, 20.87 and 36.68eV are shown in fig.2.

Fig.3 shows ratios of γ yields gated by the resonances at



Fig. 1. The TOF spectrum measured by the 4π Ge spectrometer for a U sample



Fig. 2 The γ ray spectra measured by the 4π Ge spectrometer for a ²³⁸U sample gated by low energy resonances. The energies of each γ peak with symbols are a:4060.3keV, b:3982.6keV, c:3583.0keV, d:3567keV, e:3541.3keV, f:3481.7keV and g:3197.2keV, respectively.

6.67, 20.87, 36.68eV and a thermal energy region, which are represented by

$$Ratio_{i} = \frac{Y_{i}}{\sum_{n=a}^{g} Y_{n}} , \qquad (1)$$

where *i* is peak symbol in fig.2 and *Y* is the yield of γ peak.

The neutron resonance dependence of γ ray spectrum for 238 U is shown clearly. Using the deduced information, the neutron capture cross sections will be improved.



Fig. 3. Ratios of emitted γ rays for each neutron resonance, where the Peak IDs correspond to the symbols of γ ray peaks in fig.2. The values of Library (Thermal) are referred to database of National Nuclear Data Center in Brookhaven National Laboratory.

Present study is the result of "Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system" entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

採択課題番号 20P2-1 高エネルギーガンマ線分光法による中性子捕獲反応の研究 プロジェクト (原子力機構・基礎工学)原田秀郎、大島真澄、小泉光生、古高和禎、藤 暢輔、北谷文人、 中村詔司、木村 敦、金 政浩、後神進史(東工大・原子炉)井頭政之、水本元治、片渕竜也 (京大・原子炉)堀 順一

Study on Neutron Capture Cross Secitons of MA and LLFP Nuclei using Various Detectors

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INTRODUCTION: Accurate neutron capture cross section data of minor actinide (MA) and long-lived fission products (LLFPs) are required for the study of transmutation system. However, the present accuracy of experimental data is insufficient. In the case of LLFPs such as 93 Zr and 107 Pd, this is because capture samples are ordinarily prepared through chemical process of fission products and contains their stable isotopes which have large effects on measurement. A promising way to eliminate backgrounds due to impurities is to measure discrete capture γ rays using Ge detectors with a high energy resolution. In the present study, we have measured capture γ rays from enriched 93 Zr and 96 Zr samples with a 4 π Ge spectrometer.

EXPERIMENTS and RESULTS: The measurements have been performed by the neutron TOF method using an electron linear accelerator (linac) at the KURRI. The 4π Ge spectrometer was made up with 2 sets of so called cluster-type Ge detectors consisting of 7 Ge crystals and 6 sets of coaxial Ge detectors. The cluster-type Ge detectors were surrounded with large BGO detectors for Compton suppression. The events from 20 Ge detectors were stored in the list mode.

The sample of 93 Zr, which was made of oxide powder (93 ZrO₂) of 3.31 g, was purchased from Oak Ridge National Laboratory. The sample is packed in an aluminum disk container. The isotopic purity of 93 Zr is about 19 %, and the stable isotopes of ${}^{90, 91, 96}$ Zr as well as the radioactive impurity of 125 Sb are also contained. Therefore, a 15 mm thick lead was set on the surface of each detector and the discrimination level was set at about 700 keV not to store the events of strong decay γ -rays from 125 Sb. The sample of 96 Zr, which was made of oxide powder (96 ZrO₂) of 1.23 g, was packed in an aluminum disk container. The isotopic purity of 96 Zr is about 58 % and the stable isotopes of ${}^{90, 91, 92, 94}$ Zr are also contained. In the case of 96 Zr sample, the discrimination level was set at

about 100 keV.

The linac was operated with electron energy of 30 MeV, a repetition rate of 100 Hz, an average beam current of 30 μ A and pulse width of 100 ns.

The neutron capture γ -ray pulse-height (PH) spectra corresponding to the 110-eV resonance of 93 Zr and the 301-eV resonance of 96 Zr were obtained for the first time as shown in Figs. 1 and 2. In the case of 93 Zr, the strong transition from the 919-keV to the ground state and two cascade transitions to the 919-keV state were observed. In the case of 96 Zr, the strong primary transitions from the capture state to the 1103-keV, 3550-keV, and ground states were observed. It is expected that backgrounds due to impurities in the sample can be eliminated by applying gates on the 919-keV ground-state transition for 93 Zr and three primary transitions for 96 Zr, respectively.



Fig. 1. Observed capture γ -ray PH spectrum corresponding to the 110-eV resonance region of ⁹³Zr.



Fig. 2. Observed capture γ -ray PH spectra corresponding to the 301-eV and off resonance regions of 96 Zr.

Present study is the result of "Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system" entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

採択課題番号 20P2-2 各種検出器を用いた MA、LLFP 核種の中性子捕獲断面積の研究 プロジェクト (京大・原子炉) 堀 順一、八島 浩(原子力機構・基礎工学) 大島真澄、原田秀郎、小泉光生、 古高和禎、藤 暢輔、北谷文人、中村詔司、木村 敦、金 政浩、後神進史 (東工大・原子炉) 井頭政之、水本元治、片渕竜也(北大・工学研究科) 木野幸一、鬼柳善明

PR2-3 MLPPAC for Measurements of Fission Cross-sections of Minor Actinides

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INTRODUCTION: In order to measure the neutron-induced fission cross sections for minor actinides, a multi-layered parallel plate avalanche counter (MLPPAC) was developed. The MLPPAC has two sets of four electrodes, two anodes and two cathodes, and a back-to-back sample holder between the sets of the electrodes.

In the recent experiment, MLPPAC was tested using KULS. Then, it was found that the neutron spectrum was distorted by hydrogen-compound materials in MLPPAC. Therefore, MLPPAC was rebuilt without the light elements and tested by the measurement of the cross section for the 241 Am(n,f) reaction. In this report, we show that for 241 Am(n,f).

EXPERIMENTS: The sample of ²⁴¹Am and the reference of ²³⁵U were prepared by the filtration method[1] where the hydroxide precipitation of the sample was filtered on an alumina membrane filter. The numbers of atoms in each sample was measured by the α -spectrometry, and estimated to be $(1.38\pm0.01)\times10^{16}$ for ²⁴¹Am and $(1.18\pm0.02)\times10^{16}$ for ²³⁵U.

Figure1 shows a two-dimensional plot of the pulse height spectra of the first and second anodes taken with the fission of ²⁴¹Am sample. Projections onto x and y axis are also shown by the red lines. The black lines represent the pulse height spectra for all events including the single events. By taking a coincidence between the first and second anodes, much clearer discrimination between fission fragments and α particles are achieved in this system.

RESULTS: The ²³⁵U(n,f) reaction is not appropriate for the measurement of the excitation function due to its resonance peaks. Therefore, the excitation function of the ²⁴¹Am(n,f) reaction was relatively obtained to that of ¹⁰B(n, α) reaction. The relative cross section was normalized to its absolute value measured with ²³⁵U(n,f).

The cross section for the 241 Am(n,f) reaction is shown in Fig.2. The solid line is the evaluation of JENDL-3.3 that is broadened according to the energy resolution of KULS. The result of Ref.[2] is also shown for comparison. Our data are in good agreement with the previous result and the JENDL evaluation. It suggests the successful use of MLPPAC.



Fig. 1. Two-dimensional plot of the pulse height spectra for the first and second anodes. Thick lines are the projections onto x and y axis, respectively. Thin lines are the pulse height spectra for all events.



Fig. 2. The cross section for the ²⁴¹Am(n,f) reaction. Previously obtained data and the JENDL-3.3 evaluation are also shown. The result of the present work shows a good agreement with them.

Present study is the result of "Study on nuclear data by using a high intensity pulsed neutron source for advanced nuclear system" entrusted to Hokkaido University by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

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採択課題番号 20P2-3 マイナーアクチノイド核の中性子誘起核分裂の研究 (東北大院・理)廣瀬健太郎、大槻 勤、岩佐直仁、柴崎義信、結城秀行 (京大・原子炉)堀 順一、高宮幸一、八島 浩、柴田誠一(原子力機構)西尾勝久

プロジェクト

PR2-4

Ab Initio Calculations of Hydrated Ions of Sr(II)

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INTRODUCTION: Strontium 90 (90 Sr) is known as one of fission products, and it has a large fission yield. In order to know the chemical behavior of Sr complexes, we study the highly precise isotopic analysis of Sr [1]. The analytical technique will be helpful to understand the fractionation mechanism of Sr isotopes in environmental samples and so on. In parallel with the analytical studies, quantum chemical calculations are performed to check the validity of analytical results. The equilibrium constant of the isotope exchange reaction can be theoretically obtained as the reduced partition function ratio (RPFR) of isotopologs. The RPFR value is usually calculated through harmonic vibration analysis. In this study, we report some *ab initio* calculations of hydrated Sr²⁺ complexes.

COMPUTATIONAL DATAILS: The orbital geometries and vibrational frequencies of hydrated Sr(II) complexes were calculated by using the conventional Hartree-Fock (HF) approximation and the density functional theory (DFT) as implemented by the Gaussian03 code [2]. The DFT method employed here is a hybrid density functional consisting of Becke's three-parameter non-local hybrid exchange potential (B3) with Lee-Yang-and Parr (LYP) non-local functionals (B3LYP). The 6-31++G** basis set was chosen for H and O, and LanL2DZ was chosen for Sr. The former is an all-electron basis, while the latter is an effective core potential (ECP) basis.

RESULTS: The hydrated Sr^{2+} species is generally thought to be present as eight-coordinated $Sr(H_2O)_8^{2+}$. The optimized geometry is shown in Fig. 1, the structure was converged to be the square antiprism with 2.69 Å Sr-O bond length. A structural study on hydrated strontium has been performed by a combination of the extended X-ray absorption fine structure (EXAFS) spectrometry and molecular dynamics (MD).[3] Our result was similar to the reported value, 2.63Å.



Fig. 1. Equilibrium geometry of $[Sr(H_2O)_8]^{2+}$.

Then, we examined the hydration enthalpy of
$$Sr(H_2O)_8^{2+}$$

$$-\Delta H^{\circ}_{\rm hyd} = -\Delta E_{\rm b} + \Delta E_{\rm sol} + n\Delta H_{\rm vap} + \Delta nRT - \Delta E(\rm Cp) - \Delta E_{\rm zp} + \Delta E_{\rm rel} + \Delta E_{\rm geom} \quad (1)$$

In order to relate the calculated quantities to the experimental hydration enthalpy (ΔH°_{hyd}) at 298 K, correction terms were considered in the same way of a report [4]. The ΔH°_{hyd} value obtained is shown in Fig.2. The hydration enthalpy of metal cations has been determined by thermochemical methods and these literature values are shown together for comparison. The ΔH°_{hyd} value obtained agreed well with the literature value.



Fig. 2. Hydration enthalpy of $Sr(H_2O)_8^{2+}$. X-axis showed some molecular cavity calculation codes [2].

The isotope enrichment factor due to this effect can be evaluated from the logarithm of reduced partition function ratio, $\ln(s/s')f$.[5] The $\ln(s/s')f$ values of hydrated Sr(II) species were estimated by quantum chemical calculations (Table 1). These values are useful to estimate the degree of isotope fractionation.

Table 1. $\ln(s/s')f$ (‰) of hydrated Sr²⁺ species for an isotope pair, ⁸⁶Sr-⁸⁸Sr.

	HF	B3LYP
$[Sr(H_2O)_8]^{2+}$	1.424	1.416
$[Sr(OH)(H_2O)_7]^+$	1.540	1.420
$[Sr(OH)_2(H_2O)_6]$	1.593	1.508

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採択課題番号 20P2-4 核データの信頼性向上にむけた放射化学的サンプル調整法 プロジェクト 及びサンプル中の不純物分析に関する研究

(京大・原子炉)藤井俊行、福谷 哲、窪田卓見、八島 浩、堀 順一

Study on Nondestructive Analysis and Neutron Behavior Using Photonuclear Reaction

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INTRODUCTION: The activity density evaluation is essential to dispose the radiological waste including the transuranium and fission products. Indirect methods like the scaling factor method have been applied for quantitative evaluation of "difficult to measure" nucleus. The methods, however, require the destructive analysis of many samples. Measurements of gamma rays generated by photonuclear reactions to a "difficult to measure" nucleus is proposed to evaluate activity densities by "nondestructive analysis". The photon-induced cross section data for some nuclei were measured at SACLAY [1], Lawrence Livermore [2] and evaluated by IAEA [3]. The ²³⁸U is one of nuclear fuel elements. The average (γ, n) cross section for radioactive isotopes ²³⁸U were measured and compared with the evaluated data. The same kind of (γ, n) cross sections for stable isotopes ⁹³Nb were also measured.

EXPERIMENTS: The experiment was performed at the electron linear accelerator facility in Kyoto University Research Reactor Institute. Figure 1 stands for the experimental arrangement. A plate of 238 U was adopted as radioactive isotope samples. A foil of 93 Nb was used as stable isotope sample. Bare gold foils were also set in front and back of each sample to obtain the neutron flux in front and back of the iodine sample. All samples were irradiated with high energy bremsstrahlung X ray beam for 30 minutes. The X ray beam was generated by bombarding a thin Pt target with 26 and 32 MeV electrons. The electron beam had the energy distributions as plotted in Fig. 2. The gamma rays from each sample were measured for 5 minutes by a Ge detector after the irradiation and cooling.

RESULTS: The photon flux and energy distribution is essential to know the amplitude of (γ, n) reaction cross section. The photon flux and energy distribution at the sample position were calculated with the EGS 5 code [4] using the electron energy distributions and normalized by the activity of ¹⁹⁶Au produced by the ¹⁹⁷Au(γ , n)¹⁹⁶Au reaction.

The yields of photonuclear reaction products were obtained by counting the gamma-rays from the products. The average photonuclear reaction cross section results are shown in Fig. 3. The figure shows that the existing cross section data are 40 - 60 % larger than our experimental values for both 238 U and 93 Nb samples at both electron energies.



Fig. 1. Experimental setup.



Fig. 2. Electron beam intensity.



Fig. 3. The (γ, n) average cross sections for ²³⁸U and ⁹³Nb.

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採択課題番号 20P2-5 長寿命放射性廃棄物の光核反応に関する研究 (九大院・工)石橋健二、有馬秀彦、執行信寛、AKML ラーマン、加藤邦男 (京大・原子炉)堀 順一、中島 健 プロジェクト

PR2-6 Study on Neutron Energy Spectrum Measurements for Pulsed Neutron Source

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INTRODUCTION: The pulsed neutron source using the electron linear accelerator at Research Reactor Institute of Kyoto University is widely used for nuclear science researches including nuclear data (neutron reaction cross sections) measurements. It is important to understand the characteristics of the pulsed source, such as neutron spectra, for the proper use of the source. We have investigated the techniques to measure the neutron spectra in this study, and evaluated their applicability to the measurement of neutron spectra of pulsed neutron sources. The following three techniques were studied: (1) Multi-foil activation method, (2) Time-of-Flight (TOF) method, and (3) Neutron counter measurement.

(1) MULTI-FOIL ACTIVATION METHOD: This method is widely used to obtain the neutron spectra at the irradiation fields in thermal and fast nuclear reactors, and in accelerators. In this method, several kinds of foils, which have different activation cross sections, are irradiated, and their activities are measured. Then, the neutron spectrum is analyzed using the measured activities. To obtain the differential data (neutron spectrum) from the integrated data (activities), the "unfolding" technique is employed. In general, this method requires neither wide space nor special equipment, and the wide range of neutron energy can be measured. However, for the high energy resolution measurement, it is very difficult because it measures only energy-integrated data.

(2) TIME-OF-FLIGHT(TOF) METHOD: This method measures the time of neutron flight from the pulsed neutron source to a neutron detector located at a fixed distance from the source. Since the flight time is



Fig. 1. Layout of Time-of Flight experiment at KURRI-LINAC.

proportional to the inverse of neutron speed, the energy of detected neutrons can be obtained easily. At the LINAC, the neutron spectrum leaked from the water moderator has been often measured. Figure 1 show a layout of TOF measurement. In this case, the B-10 sample was located at the end of flight tube (12.7m-length), and the gamma-ray of neutron capture reactions in the sample was detected by a multiple-BGO detector. The measured neutron spectrum is shown in Fig.2.



Fig. 2. Neutron Spectrum measured by TOF Method.

This method can be obtained the differential data directly. However, as shown in the figure, the range of measurement is limited to about 1keV. This is due to the distortion of signal by the gamma burst generated when the pulsed neutron produced. To avoid this effect, the longer distance of neutron flight path is effective, but, in that case, the count rate of neutron decreases and hence the error of measurement increases.

(3) NEUTRON COUNTER MEASUREMENT: In this method, several kinds of neutron counters are used. For the thermal energy region, the He-3 proportional counter with neutron moderator is used. By changing the thickness of moderator, the different energy of thermal neutrons are detected. For the fast energy region, proton recoil counters, of which the pulse height is proportional to the energy, are generally used. For the both measurements, calibration of the counters with the standard spectrum field is necessary, and some kind of unfolding process is required.

Those three techniques have some drawback and advantage, respectively, for the application to the pulsed neutron. For the fast energy region, the neutron counter measurement, using the proton recoil proportional counter would be suitable. For the thermal to epithermal range, the TOF method would give the good results. For the intermediate energy range, multi-foil activation method would be available, if the proper foils are used.

採択課題番号 20P2-6 パルス状中性子スペクトル測定に関する基礎研究 (京大・原子炉)中島 健、堀 順一 プロジェクト