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STUDIES OF PHOTONUCLEAR REACTIONS AND PHOTON ACTIVATION ANALYSIS IN THE GIANT DIPOLE RESONANCE REGION USING MICROTRONS

TRAN DUC THIEP, NGUYEN VAN DO, NGUYEN KHAC THI,
TRUONG THI AN AND NGUYEN NGOC SON

Institute of Physics and Electronics, Vietnamese Academy of Science and Technology

Abstract. *Mictrorons are accelerators of electrons and are simultaneous sources of bremsstrahlung photon flux and fission neutrons. In 1982, a microtron of seventeen trajectories Microtron MT - 17 was put into operation at the National Institute of Physics of Vietnam. Though very modest, microtons are very useful for developing countries such as Vietnam in both fundamental and applied physics research. During the recent years by using the above mentioned MT - 17 and microtrons from other institutes we have carried out different investigations. In this report we present some results obtained in the studies of photonuclear reactions and photon activation analysis in the giant dipole resonance region.*

I. INTRODUCTION

Microtrons is an accelerator of electrons working by cyclotron principle in modified way. In microtron electrons entered the acceleartion camera not in central part of the magnetic fields as in cyclotron but at its end where the resonator is placed. In this case the electrons after each circle enter the resonator in the accelerating phase. The radius of the electron trajectory varies after acceleration in the resonator and approaches the radius of the site of location of the resonator. From the last trajectory electrons are deflected out of microtron. Electrons can be accelerated up to 50 -100 MeV in micotron. Microtrons can work in continuous as well as pulse regime. The advantages of microtron are easy deflection of electron beam, small energy fluctuation and sufficently high intensity at low energy.

Our microtron MT - 17 has seventeen trajectories and electrons up to 15 MeV. It is simultaneous source of 15 MeV bremsstrahlung photons and fission neutrons. As microtron MT - 17 is very limited in energy therefore in many cases we used microtrons MT -22 and MT - 25 of the Joint Institute for Nuclear Research, Dubna, Russia to carry out investigations in a wide energy range (the maximum bremsstrahlung energy can be varied stepwise from 10 to 22 MeV and 10 to 25 MeV for MT - 22 and MT - 25 respectively).

Two measuring systems were used for the experiments. At the Institute of Physics, Hanoi, samples were measured by a spectroscopic system consisting of 62 cm^3 coaxial HPGe detector (ORTEC) with a resolution of 2.1 keV at 1332 keV gamma line of Co^{60}, a spectroscopic amplifier (CANBERRA mode 2001) and a 4096 channel analyzer (mode ND- 66B, Nucl. Dat. Inc) coupled with a PDP 11/23 computer for data processing. At the Joint Institute for Nuclear Research, Dubna the measuring system consisted of a 45 cm^3 Ge (Li) detector, a NOKIA spectroscopic amplifier, a 4096 channel analyzer (NOKIA, model LP - 4069). In case of short lived isotopes of interest, the samples were transported from the measuring site to the irradiation site and vice versa by a pneumatic transfer system with minimum transfer time of 2s. Recently the multipurpose coincidence spectrometer for experimental nuclear physics has been established at our Institute. This is suitable for many kinds of experiments with and without using coincidence technique.

On the basis of the above mentioned facilities we have carried out different photonuclear reactions as well as photon activation analysises in the giant dipole resonance region.

II. STUDIES OF PHOTONUCLEAR REACTIONS

Photonuclear reactions seem to be favourable for investigation of the nuclear structure as in the case electromagnetic interaction is well known and the theoretical consideration is simplified. Due to the missing Coulomb barrier compound states of low excitation energy are easily populated. The number of open channels is reduced compared with particle induced reactions. The main advantage, however, is the spin selectivity of the excitation The giant electric dipole resonance dominates the photoabsoption cross section in the most important energy range. Additionally, microtrons are high intense photon sources therefore they are suitable for the studies of photonuclear reactions where the reaction cross sections are small. For the above mentioned reasons we have concentrated our attention to the studies of photonuclear reactions,namely photofission, photonuclear reactions, forming isomeric states and determination of the integrated cross section.

1. Photofission

Photofission represent a powerful tool for investigating the double - humped fission barrier of actinide nuclei. The spin selectivity of the electromagnetic interaction leads to the favoured excitation of a few specific fission channels. In the fission the mass and charge distributions are ones of the most interesting observable characteristics as their parameters can be related to the dynamics of the fission process. For these reasons we have concentrated our investigations in these characteristics.

Our first studies on photofissions of Pu^{242} and U^{235}were started in 1982 at the Joint Institute for Nuclear Research, Dubna, Russia by using microtron MT - 22 and are published in [4,5]. By that time the data on the mass distribution for Pu^{242} have not been published. We have determined the postneutron product yields for the photofission of Pu^{242} with 18.1$\pm$ 0.2 MeV and 20.7$\pm$ 0.3 MeV bremstrahlungs by catcherfoil technique described in [1$\div$3]. The target of 3$\pm$ 0.3 mg of dioxide plutonium enriched to 94.7% Pu^{242} was prepared on a 70 μm thick aluminium disk of 55 mm diameter. The active layer had a diameter of 20 mm. The catcherfoil which consisted of 0.1 mm thick very pure aluminium foil (purity 99.99%) was placed at a distance of 1 mm from the Pu^{242} source. The target was irradiated for 5 hours and 6 hours with 18.1 and 20.7 MeV bremsstrahlungs, respectively. The relative cumulative yields for the fission fragments were determined from successive measurements of fission gamma spectra from the cathcherfoil by method presented in [1]. The relative total yields for a given mass chain were obtained from the relative comutative yields in the same way show in [1] by making correction with the expression:

$$P(Z) = \frac{1}{\sqrt{\pi c}} \exp\left[\frac{(-Z - Z_p)^2}{c}\right]$$

with c=0.80±0.14 and Z_p is most probable charge in mass chains.

The absolute product yields were obtained by normalizating to 200% the area under the total mass distribution. We have obtained for the Pu^{242} photofission 25 mass chains. The results are shown in Fig. 1. We can see that there is a weak fine structure in mass region 133-135 due to the close neutron shell N=82. The investigation of the U^{235} photofission with 18 MeV bremsstrahlung was carried out with the same technique as for Pu^{242} and 34 product yields were obtained [4]. The target of 30 mg dioxide uranium, enriched to 97% U^{235} was used in the experiment. Our results are in good agreement to that of Thierens [2].

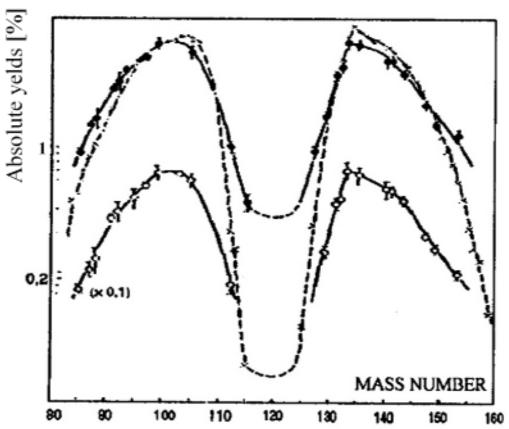

Fig. 1. Mass distribution for photofission of Pu^{242} with 20.7 (•) and 18.1 MeV (o) bremsstrahlung, (x) Fission of Pu^{241} with thermal newtrons.

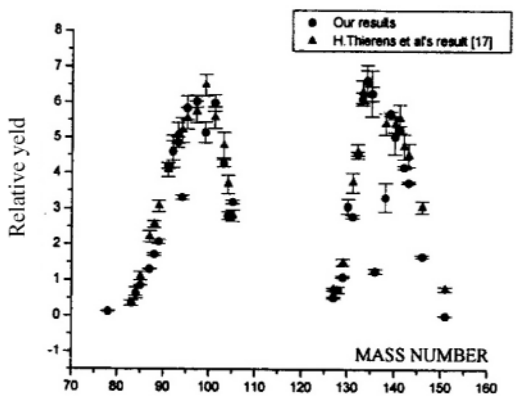

Fig. 2. Mass distribution for photofission of U^{238} with 15 MeV bremsstrahlung.

The photofission of Th^{232} and U^{238} with 15 MeV bremsstrahlung were performed at the Institute of Physics and presented in [6,7]. The Uranium target enriched to 99.6% U^{238} was prepared on a 0.5 mm thick high pure aluminium disk of 20 mm in diameter and for Thorium the target was a pure Th^{232} sample with density of 15 mg/cm^2 wrapped in a thin layer of lapsan on 1mm thick pure aluminium disk. the targets after irradiations were measured with direct gamma spectroscopic technique without any chemical separation. For U^{238} the cumulative yields for 44 mass chains have been determined. A fine structure in the mass regions 133-135 and 140-142 was observed and our results are compared to those of D. De Frenne et al [3]. For Th^{232} 38 cumulative mass yields have been established. Our results are compared to that obtained by other groups and fine structure is exhibited.

For example, the mass distributions for photofission of U^{238} with 15 MeV bremsstrahlung is shown in Fig. 2.

As it is known in fission the independent isomeric yield ratios are measure of the primary fission fragment angular momentum. Isomeric yield ratio can be determined if the isomeric pairs are screened product, i. e, if the products can be formed only directly in fission reaction. Up to now the studies on the independent isomeric yield ratios in the photofission of U^{238} are limited. In our investigation [9] we succeeded in determining the isomeric yield ratios for following pairs Sb^{128m}- Sb^{128g}, I^{132m}-I^{132g} and Xe^{135m}-Xe^{135g}.

Besides, we have studied the charge distribution for the photofission of U^{238} with 15 MeV bremsstrahlung. The data on independent yields for any photofission system available in the literature are very scarce. The charge distributions for mass chains 95, 97,99,128, 130, 131, 132, 134, 135, 138, 140 and 141were investigated. We deduced from cumulative (or independent) yields the most probable charges Z_p for 7 other mass chains based on two methods namely the unchanged charge distribution and the emperical relation. Our results were reported in [8]. We have also developed the statistical model to predict independent fission yields and pairing effects which are in good agreement with experimental data [10].

2. Isomeric ratios

Isomeric ratios furnish valuable information about the level structure of nuclei and the nuclear reaction mechanism involved. By fitting the theoretical calculated isomeric ratios to the experimental ones, it is possible to obtain information about the spin dependence of the nuclear level density, in particular, the spin cut - off parameter σ and the level density parameter a. We have determined the experimental isomeric ratios for different kinds of photonuclear reactions as (γ, n), (γ, p), (γ, np) in the energy region from 15 to 25 MeV i.e in the giant dipole resonance region with error about 5÷10%[11÷14]. Some results on (γ, n) photonulear reactions are shown in Table 1.

Table 1. *The isomeric ratios of investigated reactions*

Nuclear	spin state		Isomeric ratio/bremsstrahlung energy [MeV]					
Reaction	high	low	15	16	18	20	22	25
$Nd^{142}(\gamma,n)Nd^{141m,g}$	$1/2^-$	$3/2^+$	0.022	0.045	0.049	0.052		
$Sn^{144}(\gamma,n)$ $Sn^{143m,g}$	$11/2^-$	$3/2^+$	0.031	0.039	0.043	0.044		
$Zr^{90}(\gamma,n)$ $Zr^{89m,g}$	$9/2^+$	$1/2^-$		0.7	0.75	0.92		
$Pd^{110}(\gamma,n)$ $Pd^{109m,g}$	$11/2^-$	$5/2^+$	0.060	0.062	0.068	0.072		
$Sb^{121}(\gamma,n)$ $Sb^{120m,g}$	8^-	1^+	0.018		0.052	0.062	0.062	
$Sb^{123}(\gamma,n)$ $Sb^{122m,g}$	8^-	2^-	0.014		0.015	0.019	0.019	0.038
$Sr^{86}(\gamma,n)$ $Sn^{85m,g}$	$9/2^+$	$1/2^-$			0.565	0.505	0.590	0.541

We used the method proposed by Huizenga and Vandenbosh [15] in a modified way for the special case of photonuclear reactions to consider our experimental results. Our studies of (γ, n) photouclear reactions led to the following interesting information.

• The isomeric ratio in (γ, n) photonuclear reactions vary insignificantly with bremsstrahlung energies. This fact is due to the small momentum effect of photon.

• Up to about 19 - 20 MeV the statistical model can be used for interpretation of isomeric ratios or in this energy range the statistical model is applicable for the description of photonuclear reactions and equilibrium is a dominant one. In the higher energy region pre - equilibrium and direct processes should be taken into account.

• There is a general systematic trend in analysis of isomeric ratios in products of photonuclear reactions namely the linear correlation between spin cut - off parameter SCOP and center of spin of isomeric pairs COS when the isomeric is in order of unity.

3. Integrated Cross Section

Studies of integrated cross section of photonuclear reactions with bremsstrahlung furnish important information on the nuclear reaction mechanism and the nuclear structure as well as provide valuable nuclear data for different applications [16]. On the other hand the data for integrated cross sections could contribute to estimating the sensitivity of photon and photoneutron activation methods and to shielding accelerator facilities for radiation protection. For this aim some first results have been obtained for 15 MeV bremsstrahlung of MT - 17 are shown in Table 2. In our work the integrated cross sections have been determined by relative method by comparing with the data for $Cu^{65}(\gamma, n)Cu^{64}$ reaction [17].

Table 2. *Integrated croos - section of investigated nuclei*

Nuclear reaction	Integrate cross-section [MeV.mb]	Nuclear reaction	Integrate cross-section [MeV.mb]
$Sb^{121}(\gamma, n)\ Sb^{121,g}$	5.1 ± 0.3	$Sn^{118}(\gamma, n)\ Sn^{117m}$	10.05 ± 0.8
$Sb^{121}(\gamma, n)\ Sb^{120m}$	675.4 ± 26.6	$Sn^{124}(\gamma, n)\ Sn^{123m}$	934.7 ± 18.7
$Sb^{123}(\gamma, n)\ Sb^{122g}$	5.1 ± 0.3	$Sn^{112}(\gamma, n)\ Sn^{111}$	168.7 ± 16.8
$Sb^{123}(\gamma, n)\ Sb^{122m}$	12.8 ± 0.4	$Ni^{58}(\gamma, n)\ Ni^{57}$	62.1 ± 4.3
$Sr^{86}(\gamma, n)\ Sr^{85g}$	384.7 ± 14.8	$Cd^{107}(\gamma, n)\ Cd^{106}$	364.8 ± 14.7

III. PHOTON ACTIVATION ANALYSIS

The photon activation analysis method using microtrons in the giant dipole resonance region is method of high sensivity, selectivity, presentativity and weak activation of the matrix. During recent years on the basis of this method we have carried out element analysis in samples of different origins.

1. Analysis of proteint content in rice [18]

As is known in rice the protein content is proportional to the nitrogen content. Therefore by determining the nitrogen concentration it is possible to establish the protein content. The analysis of nitrogen was performed by detecting annihilation 511 keV gamma ray of N^{13} produced by $N^{14}(\gamma, n)N^{13}$ reaction. The measuring system consisted of two scintillation detector NaI (Tl) working in coincidence regime. The rice samples were irradiated by 18 MeV bremsstrahlung produced from Microtron MT - 22. Under optimum conditions (1 min irradiation time, 20 min cooling time and 1 min measurememt time) about 45 samples can be analysed for one hour with the sensitivity of about 1 ppm.

2. Photon Activation Analysis of Sn, W, Au, Cu and Ni [19, 20]

In photon activation analysis, the principal photonuclear reaction used is (γ, n). The sensitivity of the analysis is about 10 to 0.1 ppm for a large number of elements. Specially, this method is successfully used in routine analysis of Sn, W, Cu and Ni in geological samples using the following photonuclear reactions:

Sn^{124} (γ, n) Sn^{123m} ($T_{1/2}$=39.5min, Eγ=159 keV)
W^{186} (γ, n) W^{185m} ($T_{1/2}$=1.6min, Eγ=174 keV)
Au^{197} (γ, n) Au^{196} ($T_{1/2}$=6.18 d, Eγ=3.557 keV)
Cu^{65} (γ, n) Cu^{64} ($T_{1/2}$=12.7 h, Eγ=1345 keV)
Ni^{58} (γ, n) Ni^{57} ($T_{1/2}$=36.6 h, $E\gamma = 1377$ keV)

Usually, in the analysis relative method was used. However, in specific conditions the following modified classical methods such as cumulative method, the internal standard method and standard addition method have also been applied. In order to improve the accuracy of the analysis, counting losses and fine interferences corrections have also been taken in to account.

3. Mixed gamma - neutron activation analysis of rate earth elements [21]

Neutron and photon activation analysis can be considered as effective analytical methods for rare earth elements (REE). Separate irradiation with either neutrons or photon seems to be laborious and time - consuming. So it is worthwhile to investigate the simultaneous analysis of REE by irradiation in a mixed neutron - gamma field. The latter is easily available at microtron. The contents of La, Tb, Ho, Lu were determined only via (n,γ) - reactions. For Y, Ce, Nd and Dy irradiation a mixed flux is preferable due to the (γ,n) reaction. With the application of this method only one irradiation is needed for the determination of all REE. Besides REE, several elements such as Nb, Zr, etc that are not convenient for determination by thermal neutron irradiation can be easily analysed simultaneously.

4. Photon Activation Analysis using gamma rays [22]

We have considered the possibilities of using soft gamma rays for element analysis by the photon activation method. It is shown that in many cases the detection of soft gamma rays (low energy) makes the photon activation method more accurate and sensitive in comparison to hard gamma rays (high energy). Typocal results are presented in Table 3. This fact is due to lower background and cleaner spectrum measured for low energy gamma rays.

Table 3. *Comparison of the sensitivities*

Element	Nuclide used	Haft life	Energy [keV] Intensity[%]	Sensitivity [ppm]	Energy [keV] Intensity[%]	Sensitivity [ppm]
Sn	Sn^{117}	14 d	25.1 (50)	1	157.4 (65)	10
Cs	Cs^{132}	6.47 d	29.7 (40)	0.2	667.6 (100)	1
Ag	Ag^{106}	24 min	21.2 (39)	0.1		
		Ag^{110}	24s		658	20

5. Photon Activation for Multielement analysis [23]

Table 4. *Number of elements analysed in one irradiation*

Cooling time	Measurement time	Element determined
15 min	5 - 10 min	Cl, Nd, Sr, Zn
3-5 h	0.5 h	Sn, Cs, Sm, Ta, Th, Pb, Ba, Mo, Ca, Fe, Zr, Cu, Mg, Sbm Tim Ni, Cd, Ybm Sr
6-7 h up to 15 d	1-2 h 1-2 h	U, Au, As, Rb, Nb W Ce, Nam Co, Mo, Y, Cr, Zn

By using both thick and thin detectors with high energy resolution for detecting hard and soft gamma rays, we have show that under the optimum analysis condition (15 or 18 MeV bremsstrahlung irradiation with average electron beam 15 μA for 4-5 hours) about 40 elements in a sample can be analysed for one irradiation with the sensitivity from 0.1 to 100 ppm as describe in Table 4.

IV. CONCLUSION

In conclusion we would like to say that microtrons are very effective facilities for nuclear research. By using them both fundamental and applied physics can be performed.

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