TWENTY YEARS OPERATING OF FLEROV NUCLEAR REACTION LABORATORY MICROTRON MT-17 IN VIETNAM

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Abstract: Mictrorons are accelerators of electrons and a simultaneous sources of bremsstrahlung photon flux and fission neutrons. I 1982, a microtron of seventeen trajectories Microtron MT – 17 of FLNR wa put into operation at the National Institute of Physics of Vietnam. Thoug very modest, microtrons are very useful for developing countries such a Vietnam in both fundamental and applied physics research. During the rece twenty years by using the above mentioned Microtron MT – 17 an microtrons from other institutes we have carried out different investigation In this report we present some results obtained in the studies of photonucle reactions and photon activation analysis in the giant dipole resonance region.

I. INTRODUCTION.

FLNR microtron MT – 17 has seventeen trajectories and accelerat electrons up to 15 MeV and works in pulse regime with pulse width of 3 and frequency of 400Hz. It is simultaneous source of 15 MeV bremstrahlu photons and fission neutrons. As microtron MT- 17 is very limited therefo in many cases we used microtrons MT – 22 and MT – 25 of the Joint Institu for Nuclear Research, Dubna, Russia to carry out investigations in a wi energy range (the maximum bremstrahlung energy can be varied stepwi from 10 to 22 MeV and 10 to 25 MeV for MT – 22 and MT – respectively).

MT - 17 has technical parameters the following:

- Maximum energy of accelerated electrons, MeV 15
- Range of change of energy, MeV 10-1
- Number of orbits 17
- Beam intensity of electrons, μA 20
- Flow of fast neutrons, 1/s. 10^{11}
- Fluence of thermal neutrons, $1/cm^2.s$ 10^{8}
- Fluence of resonance neutrons, $1/cm^2.s$ 10^{7}
- Working vacuum, Torr 10^{-6}

- Power consumption, KW 20
- Frequency of high - frequency generator 2790 MHz
- Pulse power of the Magnetron, MW 1.6
- Frequency of modulations, Hz 100,200,400
- Pulse duration of modulations, μs 3
- External diameter of the chamber, mm 1200
- Internal diameter of the chamber, mm 720
- Cooling Water, air

Two measuring systems were used for the experiments. At the Institute of Physics, Ha noi, 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 – 4096). 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. This system consists of polyethylene pipes and two compressors Taiphon - 5. Switching on and off the compressors is realized by a control system.

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.

II.1. Photofission.

Photofission represents a powerful tool for investigating the double – humped fission barrier of actinide nuclei, especially in the case of studies on shape isomer, formed by photonuclear reactions. 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 distribution are ones of the most interesting observables in 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 were photofissions of Pu^{242} and U^{235} started in 1982 at the Joint Institute for Nuclear Research, Dubna, Russia by using microtron MT – 22 and are published in [3,4]. 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 ± 0.2 MeV and 2.7 ± 0.3 MeV bremstrahlungs by catcherfoil technique discribed in [1,2]. The target of 3 ± 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 bremstrahlungs respectively. The relative comulative yields for the fission fragments were determined from successive measurements of fission gamma spectra from the catcherfoil by method presented in [1]. The relative total yields for a given mass chain were obtained from the relative comulative yields by making correction with the expression for charge distribution.

The absolute product yields were obtained by normalization 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 bremstrahlung 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. In order to avoid thermal neutron induced fission of U^{235} the target was packed by Cd envelope of 0.5

mm thickness and fast neutron induced fission is less than 0.2 % can be neglected.

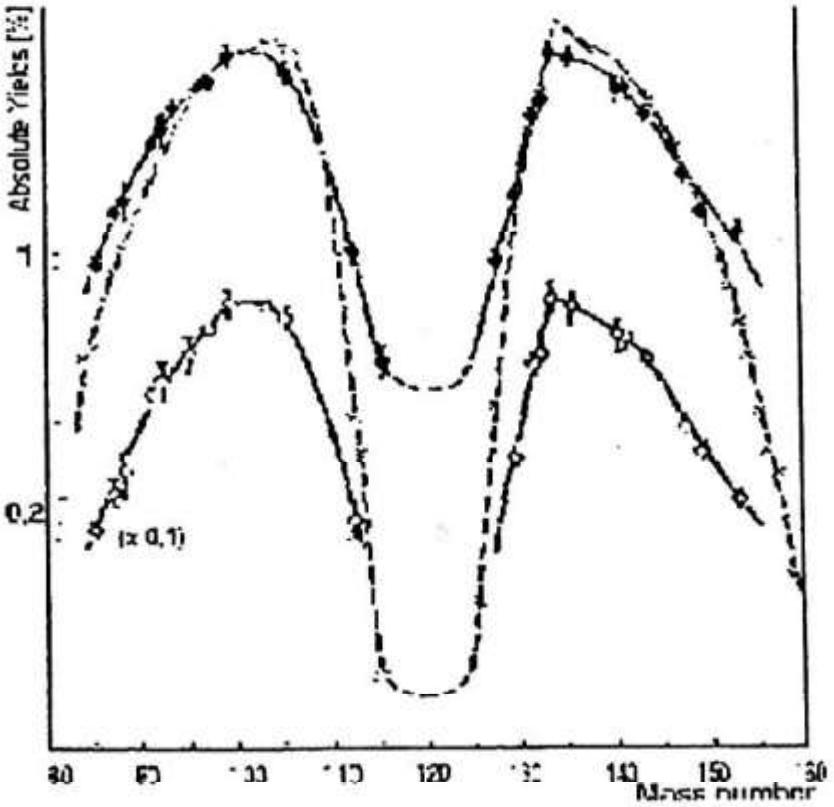

Fig. 1. *Mass Distribution for photofission of Pu^{242} with 20.7 (•) and 18.1 MeV(o) bremsstrahlungs, (x) Fission of Pu^{241} with thermal neutrons.*

The photofissions of Th^{232} and U^{238} with 15 MeV bremstrahlung were performed at the Institute of Physics and presented in [5,6]. The target of U^{238} was an amount of 18 mg U_3O_8 enriched to 99.6 % U^{238} prepared on a 0.5 mm thick high pure aluminium disk of 20 mm in diameter and for Th^{232} target was a pure Th sample having diameter of 20 mm, thickness of 0.5 mm and density of 15 mg/cm^2 wrapped in a thin layer of lapsan on 1 mm thick pure aluminium disk. The target 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 [2]. 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. The mass distributions for

photofission of U^{235} with 18 MeV bremstrahlung, of U^{238} and Th^{232} with 15 MeV bremstrahlung are shown in fig 2, 3 and 4.

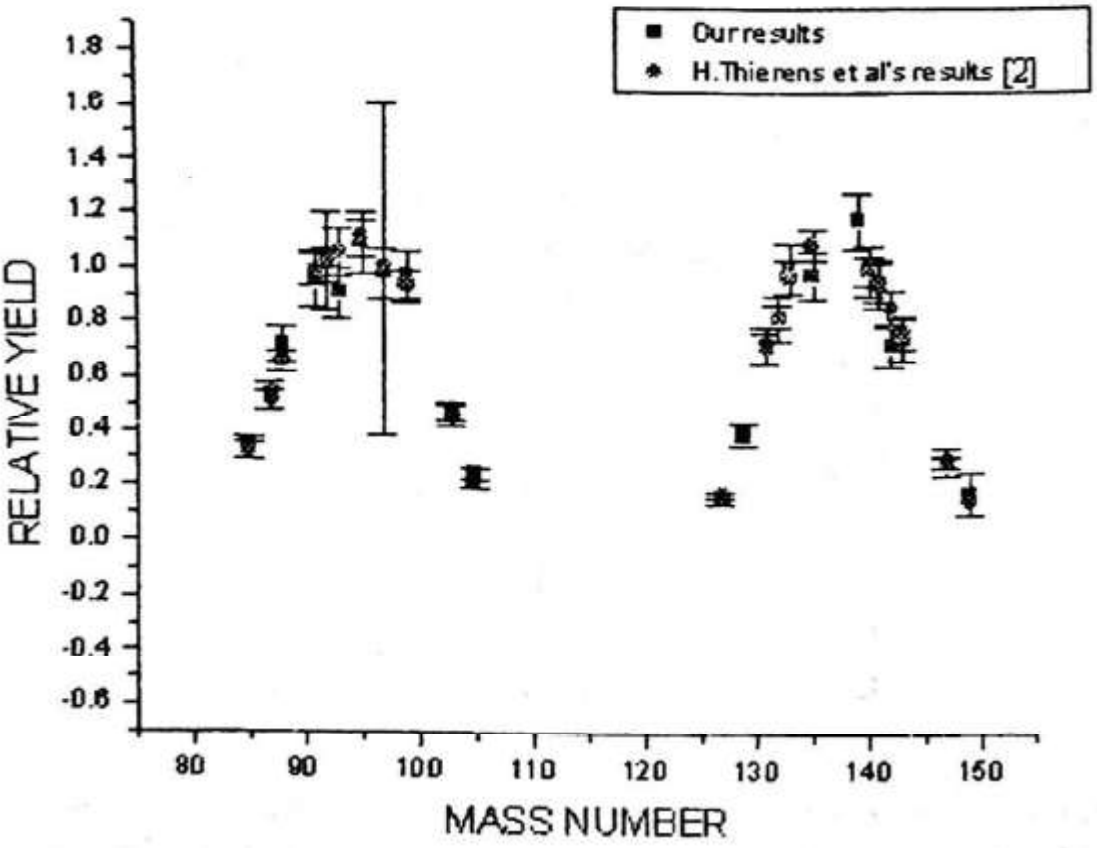

Fig.2. Mass distribution for photofission of U^{235} with 18 MeV bremstrahlung

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 [8] 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 bremstrahlung. Most of the experimental data on photofission charge distribution were obtained from independent yields which were determined using chemical separation or by direct gamma spectroscopic method. In pratice there are very small number of mass chain where it has

been possible to determine the independent yields of more than one isobar. The data on independent yields for any photofission system available in the literature are very scarce. In the case of U^{238} photofission the Z_p values lie too far from the β - stability line so that in a given mass chain, the isotopes produced with the highest probability have very short haft - lives, making the measurements of these gamma spectra very difficult. We compromise by assuming that information about charge distribution can be obtained from cumulative yield for products lying far enough away from the line of beta - stability. The charge distributions for mass chain 95, 97, 99, 128, 130, 131, 132, 134, 135, 138, 140 and 141 were 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 [7]. We have also developed the statistical model to predict independent fission yields and pairing effects which are in good agreement with experimental data [9].

II.2. Isomeric Ratios.

In nuclear reaction, the isomeric ratios are directly connected to the relative population of an isomeric state and an unstable ground state. They furnish valuable information about the level structure of nuclei and the nuclear reaction mechanism involved. On the other hand, the isomeric and metastable states are formed in the same nuclei and are measured in the same experimental conditions, so the isomeric ratios can be determined with high accuracy. 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 [10÷16]. Some results are shown in table 1. In our studies the isomeric ratios is defined as the ratio of the cross – section for the production of high spin state to that of low spin state. In case of the photonuclear reactions with bremstrahlung this ratio is defined as:

$$R = \int_{E_{th}^{m}}^{E_{m}} N\phi(E)\sigma_{m}(E)dE \Big/ \int_{E_{th}^{g}}^{E_{m}} N\phi(E)\sigma_{g}(E)dE$$

where N is number of target nucleus, $\phi(E)$ – bremstrahlung photon flux with endpoint E_m and E_{th}^m, σ_m and E_{th}^g, σ_g are the threshold energy and the cross- section for the isomeric and ground states respectively

Table1. *The isomeric ratios of investigated reactions*

Nuclear Reaction	Spin state		Isomeric ratio / bremstrahlung energy [MeV]					
	High	low	15	16	18	20	22	25
$Nd^{142}(\gamma,n)Nd^{141m,g}$	11/2⁻	3/2⁺	0.022 ±0.002	0.045 ±0.004	0.049 ±0.004	0.052 ±0.004		
$Sn^{144}(\gamma,n)Sn^{143m,g}$	11/2⁻	3/2⁺	0.031 ±0.003	0.039 ±0.003	0.043 ±0.003	0.044 ±0.003		
$Zr^{90}(\gamma,n)Zr^{89m,g}$	9/2⁺	1/2⁻		0.7 ±0.04	0.75 ±0.05	0.92 ±0.06		
$Pd^{110}(\gamma,n)Pd^{109m,g}$	11/2⁻	5/2⁺	0.060 ±0.005	0.062 ±0.005	0.068 ±0.005	0.072 ±0.006		
$Sb^{121}(\gamma,n)Sb^{120m,g}$	8⁻	1⁺	0.018 ±0.001		0.052 ±0.007	0.062 ±0.009	0.062 ±0.005	
$Sb^{123}(\gamma,n)Sb^{122m,g}$	8⁻	2⁻	0.014 ±0.001		0.015 ±0.001	0.019 ±0.004	0.019 ±0.002	0.038 ±0.002
$Sr^{86}(\gamma,n)Sn^{85m,g}$	9/2⁺	1/2⁻			0.565 ±0.055	0.505 ±0.176	0.590 ±0.090	0.541 ±0.018
$Sn^{118}(\gamma,p)Sn^{117m,g}$	9/2⁺	1/2⁻				3.686 ±0.387		
$Sn^{119}(\gamma,np)Sn^{117m,g}$	9/2⁺	1/2⁻				3.890 ±0.540		

We have carried out consideration of our results with the statistical model. In this model the level density of excited nuclei as a function of spin J and excitation energy E is represented by the known level density formular:

$$\rho(J,E) = \rho(E)\rho(J) \sim E^{-n} \exp\left[2(aE)^{1/2}\right](2J+1)\exp\left(-J(J+1)/2\sigma^2\right) \quad (1)$$

where n = 0, 5/4, 2. The SCOP σ is related to the moment of inertia θ and the thermodynamic temperature t by

$$\sigma^2 = \frac{\theta t}{\hbar^2} \quad (2)$$

We used the method proposed by Huizenga and Vandenbosh [17] in a modified way for the special case of photonuclear reactions. Here a short summary of the most important steps and assumptions of the method is presented.

Starting with a target nucleus with spin J_0, the absorption of an E_1 γ - ray leads to excited states in the first compound system with spin $J_c = J_0$, $J_0 = \pm 1$. For $J_0=0$ only states with $J_c = 1$ are excited. The relative occupation probability is proportional to the number of magnetic substates,

$$P(J_c) \sim 2J_c + 1 \tag{3}$$

The spin distribution in the first compound system is changed by successive evaporation of are one or more neutrons. The neutron transition probability from a state with spin J_i to a state with spin J_f is given by

$$P(J_f) \sim \rho(J_f) P(J_i) \sum_{S=\left|J_f - 1/2\right|}^{\left|J_f + 1/2\right|} \sum_{l=|S-J_i|}^{|S+J_i|} T_l(E_n) \tag{4}$$

where $T_l(E_n)$ is the penetrability for a neutron with angular momentum l and kinetic energy E_n. The evaporation energy E_n is replaced by an average value $\overline{E_n}$, assumed to be $\overline{E_n} = t$

with t calculated from

$$U = at^2 - t \tag{5}$$

where U is the excitation energy and t – the thermodynamic temperature

- If the remaining excitation energy is below the particle threshold is supposed that the residual nucleus deexcitates predominantly by E1 γ -ray emision with an average of

$$E_\gamma = 4\left(\frac{U}{a} - \frac{5}{a^2}\right)^{1/2} \tag{6}$$

where U is the residual excitation energy. The γ-ray probability to states with spin J_f is assumed to be

$$P(J_f) \sim \sum_{J_i} P(J_i)\rho(J_f) \tag{7}$$

- The γ-cascade is continued until the residual energy is smaller than the "γ-ray cut – off" energy [18]. The following γ-ray, the "deciding gamma", for which an arbitrary multipolarity is admitted now, feeds that state to which the transition has the lowest multipolarity. If the residual energy remains within the "γ-ray cut – off" region, partly a further E1 γ-ray is emitted and partly the "deciding gamma" is emitted as described above

- The isomeric ratio is calculated by

$$R = \frac{\sum_{J_f=\cos}^{\infty} P(J_f)}{\sum_{J_f=0}^{\cos} P(J_f)} \qquad (8)$$

where $\cos = \frac{J_h + J_l}{2}$, J_l and J_h are the low and high spins of the isomeric pair.

Our calculation procedure [19] is similar to that from [20].

We have to note that in the Huizenga – Vandenbosh method, the calculated spin distribution for different SCOP values after the neutron evaporation and E1 γ -ray deexcitation are nearly symmetric with the maximum value at about (SCOP –1/2). From formula (8) we can see that when the isomeric ratio is in order of unity, the COP and deduced SCOP are naturally correlated. Small deviations of the experimental isomeric ratio from unity cause a small shift to higher (R>1) or smaller (R < 1) SCOP values, i.e a small spread of the SCOP around the COS. By using this method we have established the nuclear level density function for a number of excited nuclei [21].

Our studies of (γ,n) photonuclear reactions led to the following interesting information.

- The isomeric ratio in (γ,n) photonuclear reactions vary insignificantly with bremstrahlung 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 discription of photonuclear reactions and equilibrium is a dominant one. In the higher energy region pre – equilibrium and direct processes should be taken into account (For example see fig 5).
- There is a general systematic trend in analysis of isomeric ratios in products of photonuclear reactions namely the linear correlation between SCOP and COS of isomeric pair (see fig 6).

II.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 application [22]. On the other hand as is known in the photon activation method based on (γ,n) reaction, the integrated cross –section is directly connected to the analysis sensivity and the neutron number emitted from the irradiated samples. Therefore 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.

Table 2. *Integrated cross – section of investigated nuclear*

Nuclear Reaction	Integrated cross– section [MeV.mb]	Nuclear Reaction	Integrated cross– section [MeV.mb]
$Sb^{121}(\gamma,n)Sb^{120,g}$	5.1 ± 0.3	$Sn^{118}(\gamma,n)Sn^{117m}$	10.5 ± 0.8
$Sb^{121}(\gamma,n)Sb^{120m}$	675.4 ± 26.6	$Sn^{124}(\gamma,n)Sn^{123m}$	934.7 ± 18.7
$Sb^{123}(\gamma,n)Sb^{122g}$	902.8 ± 27.1	$Sn^{112}(\gamma,n)Sn^{111}$	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
$Sr^{86}(\gamma,n)Sr^{85g}$	68.6 ± 2.1	$Pb^{204}(\gamma,n)Pb^{203}$	2142.1 ± 35.8

For this aim some first results have been obtained for 15 MeV bremstrahlung 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 [23].

III. PHOTON ACTIVATION ANALYSIS

The photon activation analysis method using microtrons in the giant dipole resonance region is method of high sensitivity, selectivity presentativity and weak activation of the matrix. Besides, for this method it is easy to be applied automatic and informatic equipments in the analytical procedure. Due to the above advantages the photon activation method has became very effective and widely applied in different fields. During recent year on the basis of this method we have carried out element analysis in samples of different origins.

II.1. Analysis of proteint content in rice

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 proteint content. The analysis of nitrogen was performed by detecting annihilation gamma ray 511 keV of N^{13} produced by $N^{14}(\gamma,n)N^{13}$ reaction. The measuring system consisted of two scintillation detectors NaI (Tl) working in coincidence regime. Under optimum conditions (1 min irradiation time, 20 min cooling time and 1 min measurement time) about 45 samples can be analysed for one hour with the sensitivity of about 1ppm [24].

III.2. Photon Activation Analysis of Sn, W, Au, Cu and Ni

In photon activation analysis, the principal photonuclear reaction used is (γ,n). Other reactions that are useful, albeit less of ten are (γ,p) and (γ,γ'). The sensitivity of the analysis is about 10 to 0.1 ppm for a large number of elements [25÷28]. Specially, this method is successfully used in routine analysis of Sn, W, Au, Cu and Ni in geological samples using the following photonuclear reactions:

$Sn^{124}(\gamma,n)Sn^{123m}$ ($T_{1/2}$ = 39.5 min, Eγ = 159 keV)
$W^{186}(\gamma,n)W^{185m}$ ($T_{1/2}$ = 1.6 min, Eγ = 174 keV)
$Au^{197}(\gamma,n)Au^{196}$ ($T_{1/2}$ = 6.18 d, Eγ = 355.7 keV)
$Cu^{65}(\gamma,n)Cu^{60}$ ($T_{1/2}$ = 12.7 h, Eγ = 1345 keV)
$Ni^{58}(\gamma,n)Ni^{57}$ ($T_{1/2}$ = 36.6 h, Eγ = 1377 keV)

Usually, in the analysis relative method was used. However, in specific conditions the following modified classical methods such as cumulative method, 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.

III.3. Mixed gamma- neutron activation analysis of rare earth elements

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. In order to obtain a mixed gamma – neutron field a cubic lucite block 30x30x30

cm^3 in size with a central hole was used. A lead $\gamma \rightarrow n$ converter with a thickness of 20 mm placed in the cube center served as a fast neutron source. 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, ect that are not convenient for determination by thermal neutron irradiation can be easily analysed simultaneously[29].

III.4. Photon Activation Analysis using soft gamma rays

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) [30]. The results are presented in table 3. This fast is due to lower back ground and cleaner spectrum measured for low energy gamma rays.

Table 3. Comparison of the sensitivities

Element	Nuclide used	Half life	Energy [keV] Intensity [%]	Sensitivity [ppm]	
				soft gamma	hard gamma
Sn	Sn^{117}	14 d	25.1 (50) 157.4 (65)	1	10
Cs	Cs^{132}	6.47 d	667.6 (100) 29.7 (40)	0.2	1
Ce	Ce^{139}	137.5 d	165.8 (81) 33.4 (9)	2	6
Sb	Sb^{120}	5.76 d	1172 (100) 89.9 (77)	0.1	0.6
Ba	Ba^{135m}	28.7 d	286 (16) 32.2 (37)	1	5
Ag	Ag^{106} Ag^{110}	24 min 24 s	21.2 (39) 658	0.1	20

III.5. Photon Activation for Multielement analysis

By using both thick and thin detectors with high energy resolution for detecting hard and soft gamma rays, we have shown that under the optimum analysis condition (15 or 18 MeV bremstrahlung 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 [31] as described in table 4

Table 4. Number of analysed elements in one irradiation

Cooling time	Measurement time	Elements 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, Sb, Ti, Ni, Cd, Yb,Sr
6-7 h	1-2 h	U, Au, As, Rb, Nb
up to 15 d	1-2 h	W, Ce, Na, Co, Mo, Y, Cr, Zn

IV. CONCLUSION AND ACKNOWLEDGEMENT.

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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