SOME PECULIARITY OF THE PHOTON ACTIVATION ANALYSIS BY BREMSSTRAHLUNG GAMMA-RAY

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1. INTRODUCTION

The photon activation analysis was first performed with the help of radionuclides as an activating radiation source in the beginning of the 1950's and still nowadays is the frequently applied one (see, for example, [1]). After the advent of electron accelerators (betatron, linac and microtron) the high energy bremsstrahlung γ-rays with continuum spectrum produced by these machines have been used for photon activation analysis. In addition, sometimes quasimonoenergetic annihilation photon beams are used for photo-nuclear reaction study [2].

In the activation analysis both of absolute and relative methods are acceptable. The absolute method demands that the flux of bombarding particle or γ-rays, which cause the activation, should be beforehand determined. The determination of flux is possible for quasimonoenergetic γ-rays using the reference nucleus sample with known cross section in the required energy range. However, it is difficult to determine the flux of the wide continuum spectrum bremsstrahlung γ-rays. In this case for activation analysis to use the relative method is more easier and convenient than absolute one. In the relative method of activation analysis the reference isotopes are also used. If the same isotopes are used for the reference and determination purposes, serious problem will not be occurred in the calculation procedure of measured data. But in the case of use of the different isotopes for the reference and element content determination purposes some difficult takes place in connection with different threshold energy and cross sections of the photo-nuclear reactions for these isotopes. On the other hand, in practice suitable isotopes for reference element are not always available.

In this paper for photon activation analysis by bremsstrahlung γ-rays we suggest a correction factor which takes into account difference in the threshold energy and cross sections for the reference and analysing isotopes.

2. THEORETICAL

2.1. The Correction Factor

During the irradiation by γ-rays, the variation of the number of radioactive nuclei N(t) at moment t can be expressed as follows:

$$\frac{dN(t)}{dt} = \sigma(E_\gamma) n \phi(E_\gamma) - \lambda N(t) . \qquad (1)$$

Here: $\sigma(E_\gamma)$ is the photo nuclear reaction cross section; E_γ is the γ-ray energy; n is the number of target nucleus; $\phi(E_\gamma)$ is the γ-ray flux; and λ is the decay constant of the formed radioactive

nucleus. By resolving Eq.(1) for the case of continuum spectrum bremsstrahlung photon activation, the following expression for the number of radioactive nuclei is obtained:

$$N(t_i) = \frac{n}{\lambda}(1 - e^{-\lambda t_i}) \int_{E_{th}}^{E_{\max}} \sigma(E_\gamma)\phi(E_\gamma)dE_\gamma \ , \tag{2}$$

where: t_i is the irradiation time; E_{th} is the threshold energy of the photo nuclear reaction and E_{max} is the maximum energy (end-point energy) of the bremsstrahlung γ-rays. The number of target nucleus is determined by

$$n = \frac{mN_A\theta}{A} \ . \tag{3}$$

Here: m is the target mass; N_A is the Avogadro's number; A is the atomic mass and θ is the isotope abundance for pure metal sample.

So, taking into account the irradiation, cooling and measurement times, from (2) and (3) the number of detected γ-rays is expressed as follows:

$$S = \frac{mN_A\theta\varepsilon I}{A\lambda}(1 - e^{-\lambda t_i})e^{-\lambda t_d}(1 - e^{-\lambda t_m}) \times \int_{E_{th}}^{E_{\max}} \sigma(E_\gamma)\phi(E_\gamma)dE_\gamma \ , \tag{4}$$

where: S is the area under photopeak measured by gamma detector; ε is the detector efficiency; I is the gamma ray intensity; t_d and t_m are the cooling and measurement times, respectively.

The so called integral effective cross section of the regarded nuclear reaction can be defined [1] from (4) as follows:

$$\sigma_{eff} = \int_{E_{th}}^{E_{\max}} \sigma(E_\gamma) f(E_\gamma)dE_\gamma \ , \tag{5}$$

where: $f(E_\gamma)$ is the normalized differential spectrum of the bremsstrahlung γ-rays which is determined from the following expression

$$\int_{E=0}^{E_{\max}} \phi(E_\gamma)dE_\gamma = \varphi = \varphi \int_{E=0}^{E_{\max}} f(E_\gamma)dE_\gamma \ . \tag{6}$$

Eq.(6) means that

$$\int_{E=0}^{E_{\max}} f(E_\gamma)dE_\gamma = 1 \ . \tag{7}$$

Using these definitions the integral in (2) can be expressed as:

$$\int_{E_{th}}^{E_{\max}} \sigma(E_\gamma)\phi(E_\gamma)dE_\gamma = \sigma_{eff}\varphi \ , \tag{8}$$

where: φ is the integral flux density.

Then, photopeak areas for the reference and sample isotopes, respectively, are expressed from (4) and (8) as follows:

$$S_1 = \frac{m_1N_A\theta_1\varepsilon_1I_1}{A_1\lambda_1}\varphi\sigma_{eff\,1}(1 - e^{-\lambda_1 t_{i1}})e^{-\lambda_1 t_{d1}}(1 - e^{-\lambda_1 t_{m1}}) \tag{9}$$

and

$$S_2 = \frac{m_2N_A\theta_2\varepsilon_2I_2}{A_2\lambda_2}\varphi\sigma_{eff\,2}(1 - e^{-\lambda_2 t_{i2}})e^{-\lambda_2 t_{d2}}(1 - e^{-\lambda_2 t_{m2}}). \tag{10}$$

Here:

$$\sigma_{eff\,1} = \int_{E_{th\,1}}^{E_{\max}} \sigma_1(E_\gamma) f(E_\gamma)dE_\gamma \tag{11}$$

and

$$\sigma_{eff\,2} = \int_{E_{th\,2}}^{E_{\max}} \sigma_2(E_\gamma) f(E_\gamma) dE_\gamma \ . \qquad (12)$$

The sample mass (or element content in the sample) is obtained from (9) and (10) as:

$$m_2 = \left(\frac{S_2 A_2 \lambda_2}{S_1 A_1 \lambda_1} \right) \left(\frac{\theta_1 \varepsilon_1 I_1}{\theta_2 \varepsilon_2 I_2} \right) \left(\frac{\sigma_{eff\,1}}{\sigma_{eff\,2}} \right) \times \frac{(1-e^{-\lambda_1 t_{i1}}) e^{-\lambda_1 t_{d1}} (1-e^{-\lambda_1 t_{m1}})}{(1-e^{-\lambda_2 t_{i2}}) e^{-\lambda_2 t_{d2}} (1-e^{-\lambda_2 t_{m2}})} m_1 . \qquad (13)$$

The general formula (13) can be used to obtain the element content in the sample in the case of use of the different isotopes for the reference and analysing element.

In the partial case, when the same isotope is used for reference and analysing purposes, may be gotten following very simple formula from (13):

$$m_2 = \left(\frac{S_2}{S_1} \right) \frac{e^{-\lambda_1 t_{d1}}}{e^{-\lambda_2 t_{d2}}} m_1 , \qquad (14)$$

where we assume $t_{i1}=t_{i2}$ and $t_{m1}=t_{m2}$.

All parameters, with the exception of σ_{eff}, in (13) are immediately measured and taken from the compilation or calculated. As to the integral effective cross section σ_{eff}, we introduce the correction factor which is expressed by

$$K = \frac{\sigma_{eff\,1}}{\sigma_{eff\,2}} = \frac{\int_{E_{th1}}^{E_{\max}} \sigma_1(E_\gamma) f(E_\gamma) dE_\gamma}{\int_{E_{th\,2}}^{E_{\max}} \sigma_2(E_\gamma) f(E_\gamma) dE_\gamma} \ . \qquad (15)$$

Then, the formula (13) can be written in the following form:

$$m_2 = \left(\frac{S_2 A_2 \lambda_2}{S_1 A_1 \lambda_1} \right) \left(\frac{\theta_1 \varepsilon_1 I_1}{\theta_2 \varepsilon_2 I_2} \right) \times \frac{(1-e^{-\lambda_1 t_{i1}}) e^{-\lambda_1 t_{d1}} (1-e^{-\lambda_1 t_{m1}})}{(1-e^{-\lambda_2 t_{i1}}) e^{-\lambda_2 t_{d2}} (1-e^{-\lambda_2 t_{m2}})} m_1 K \qquad (16)$$

In Ref.[3,4] the bremsstrahlung γ-rays spectrum $f(E_\gamma)$ and the photo-nuclear reaction cross section $\sigma(E_\gamma)$ were separately integrated as independent two integrals. We consider this method is incorrect and suggest a new method to calculate the integrals in (15). The bremsstrahlung γ-ray spectrum $f(E_\gamma)$ is difficult to measure in the wide energy range of *0* to end-point energy E_{max}. So, to obtain the correction factor *K*, the Schiff formula for the bremsstrahlung γ-ray energy spectrum and the Lorentz formula for the giant resonance of photo-nuclear reaction can be used.

2.2. The Schiff Formula

Schiff obtained [5] the energy distribution cross section formula of the radiation from fast electrons in very thin targets by integration of the Bethe-Heitler differential bremsstrahlung cross section [6] over the scattered electron angles and the angles of the radiation:

$$\sigma(E_0, E_\gamma) = \frac{2Z^2 r_e^2}{137 E_\gamma} \left\{ \left(1 + \frac{E^2}{E_0^2} - \frac{2E}{3E_0} \right) \left(\ln a + 1 - \frac{2}{b} \tan^{-1} b \right) + \frac{E}{E_0} \left[\frac{2}{b^2} \ln(1+b^2) + \frac{4(2-b^2)}{3b^3} \tan^{-1} b - \frac{8}{3b^2} + \frac{2}{9} \right] \right\} \qquad (17)$$

where:
$$b = \frac{2E_0 EZ^{1/3}}{111\mu E_\gamma} \tag{18}$$

and
$$\frac{1}{a} = \left(\frac{\mu E_\gamma}{2E_0 E}\right)^2 + \left(\frac{Z^{1/3}}{111}\right)^2 . \tag{19}$$

Here: $\mu = mc^2$ is the rest energy of an electron; E_0 is the incident electron energy; E is the scattered electron energy: $E_\gamma = E_0 - E$; Z is the scattering atom number and r_e is the electron radius that is determined by
$$r_e = \frac{e^2}{mc^2} . \tag{20}$$

In Ref.[7] the normalized differential energy spectrum of the bremsstrahlung γ-rays is obtained as following
$$f(E_\gamma) \propto \frac{1}{16} \times \frac{137E_\gamma}{2Z^2 r_e^2} \sigma(E_0, E_\gamma) . \tag{21}$$

So, from (17) and (21) can be obtained the following formula:
$$f(E_\gamma) = \left\{\left(1 + \frac{E^2}{E_0^2} - \frac{2E}{3E_0}\right)\left(\ln a + 1 - \frac{2}{b}\tan^{-1} b\right) + \frac{E}{E_0}\left[\frac{2}{b^2}\ln(1+b^2) + \frac{4(2-b^2)}{3b^3}\tan^{-1} b - \frac{8}{3b^2} + \frac{2}{9}\right]\right\}. \tag{22}$$

Our calculated results of the bremsstrahlung γ-rays spectrum by Schiff formula (22) for Z=74 (W target) and E_0=30 MeV are compared with Tsipenyuk [8] and Burmistenko [9] curves in Fig.1 to verify the calculation method. It is seen that our results, except the Tsipenyuk curve for the low energy region of the bremsstrahlung γ-rays, are in good agreement with these curves. This fact means that our calculation method for the bremsstrahlung γ-rays spectrum is correct. Some difference between our and Tsipenyuk results in the low energy part of γ-ray spectrum curves is may be caused by that our calculation for angle integrated case and Tsipenyuk result for the forward direction, only.

However, the reason of this discrepancy should be thoroughly considered in future.

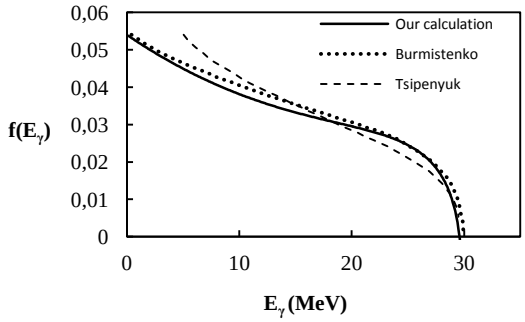

Fig.1. Bremsstrahlung γ-rays spectrum calculated by Schiff formula for Z=74 and E_0=30

2.3. The Lorentz-shaped resonance cross section

The energy dependence of the giant resonance absorption cross section for medium and heavy nuclei has often been approximated by a Lorentz-shaped resonance formula:

$$\sigma(E_\gamma) = \frac{\sigma_m \Gamma^2 E_\gamma^2}{(E_m^2 - E_\gamma^2)^2 + \Gamma^2 E_\gamma^2} \quad . \tag{23}$$

Here: σ_m is the maximum cross section at the energy E_m of the γ-ray; Γ is the total energy width of the giant resonance at the half maximum.

For deformed nuclei, the giant resonance splits into two main peaks. For light nuclei, the giant resonance shows considerably fine structure related mainly to the properties of individual nucleon motion, but also to collective surface vibrations (see, for example, [10]).

The comparison of experimental [11] and calculated by Lorentz formula cross sections is shown in Fig.2, as an example, for the ^{197}Au(γ,n)^{196}Au reaction. The Lorentz formula parameters were taken from [11-13]. It is seen that our calculated (γ,n) cross section curve is satisfactorily in agreement with the experimental data. Similar results were obtained for other investigated isotopes.

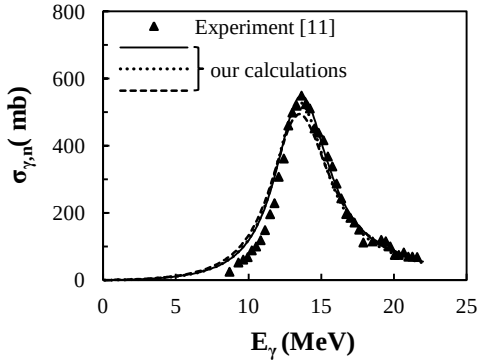

Fig.2. Experimental and calculated by Lorentz formula cross sections for the ^{197}Au(γ,n)^{196}Au reaction. The solid, dotted and dashed curves are the cross sections for which the Lorentz parameters were taken from [11], [12] and [13], respectively.

3. EXPERIMENTAL

3.1. The gamma-ray source

The bremsstrahlung gamma-rays were produced by the cyclic accelerator Microtron MT-22 of the Nuclear Research Center, National University of Mongolia, using the Ta 2 mm thick target for 22 MeV electron beam.

For absorbtion of the electrons gone through Ta target, the Al 25mm thick shield was used.

3.2. Samples

Samples from natural pure metal thin disks of Cu, Au and Mo were irradiated by bremsstrahlung γ-rays on the electron accelerator Microtron MT-22. Arrangement of the samples is shown in Fig.3. Characteristics of the sample isotopes are given in Table.1

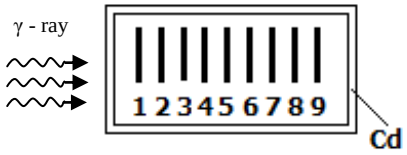

Fig.3. The arrangement of the samples. 1, 3, 5, 7 and 9 are the Cu; 2 and 4 are the Au; 6 and 8 are the Mo.

Thickness of the samples was around 0.2 mm and the diameter was ~1cm. All tight-parked metal foils were covered by 2mm thick Cd for shielding from the thermal neutron background. A distance between the front of Cd-cover and the Al-absorber was 8 mm.

Table.1. Characteristics of the sample isotopes

№	m (g)	Sample Isotope	θ (%)	Produced Isotope	$T_{1/2}$ (h)	E_{th} (MeV)	E_γ (MeV)	I_γ (%)
1	0.0296	^{65}Cu	30.9	^{64}Cu	12.7	9.9	1345.8	0.48
3	0.0283							
5	0.0284							
6	0.0300							
8	0.0280							
10	0.0294							
12	0.0284							
14	0.0287							
2	0.1442	^{197}Au	100	^{196}Au	148.4	8.1	332.9	22.9
4	0.3409						355.7	86.9
11	0.0817	^{100}Mo	9.63	^{99}Mo	65.9	8.3	140.5	4.52
13	0.0877						739.4	12.1
							777.8	4.36

3.3. Irradiation and Measurement

Metal foil samples were irradiated by bremsstrahlung γ-rays with $E_{max} \approx 22$ MeV at the electron accelerator Microtron. Irradiation time was 130 min. The electron beam current was ~ 6μA. Cu - foils were used as a reference material and for monitoring of gamma-ray flux. Au and Mo foils were studied to determine a sample mass by relative method. After irradiation of the metal foils the γ-spectra were measured by 47 cm^3 HP-Ge detector with 2 keV energy resolution at the photopeak of 1332 keV for ^{60}Co. The detector efficiency curve, which was determined by ^{252}Eu-source and was used in our calculations, is shown in Fig.4. Measurement time was 600 sec for irradiated metal foils and 1000 sec for background.

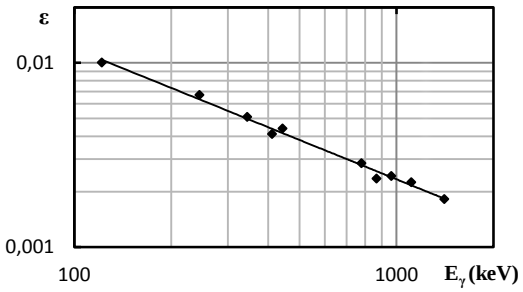

Fig.4. Detector efficiency

3.4. Half-life determination

Half-lives of all studied isotopes were determined using the different gamma-peaks for each isotope to test our experimental methods. Experimental half-lives of the all isotopes were in good agreement with reference data given in Table.1. This fact means that our experimental method is correct and suitable to use in this measurement.

4. RESULTS AND DISCUSSIONS

4.1. The correction factor calculations.

The correction factor, which expressed by the formula (15), is determined by two methods: experimental and theoretical.

In the case of the experimental correction factor, experimental data of (γ,n) cross sections [15] for the reference and sample isotopes are used. In the experimental correction factor we were forced to use the Schiff formula of the bremsstrahlung gamma-ray spectrum in connection with difficulty of exact measurement of flux for wide continuum energy spectrum gamma-rays. So, to obtain the experimental correction factor, the bremsstrahlung gamma-ray energy range from 0 to the end point divided by step 0.2 MeV and instead integral in (15) was taken a sum.

As to theoretical correction factor, the Schiff formula for the bremsstrahlung gamma-ray spectrum (22) and the Lorentz formula for the giant resonance cross section (23) are used. The Lorentz formula parameters were taken from [11-16]. In this case, the Schiff and Lorentz formulas are firstly multiplied and after that the integrals in Eq.(15) are calculated by using the MatLab programming. Calculated correction factors for studied isotopes are given in Table. 3 and 4 (see subsection 4.2).

4.2. Experimental Results and Discussions

Firstly, mass of a Cu-foil was determined relatively with Cu-reference by simple formula (14) to test our mass determination method (see Table.2). It is seen that real mass and determined by simple formula (14) mass are in good agreement with uncertainty $\Delta m \leq 3\%$. This result again shows that our method is correct. Mass of the Au and Mo samples were determined by formula (16) using the peak area from measured gamma-spectra, the calculated correction factors and data given in Table.1. Mass m_1 and other data of Cu-foil, which was close contact with given sample, are used for determination mass m_2 in formula (16) to reduce an uncertainty from gamma-ray flux difference for reference material and samples.

Table. 2. Mass determination results for Cu foil

Sample	E (keV)	K	Real mass (g)	Determined mass (g)	Relative uncertainty (%)
Cu-3	1345.8	1	0.0283	0.0291	2.82
Cu-5	1345.8		0.0284	-	

Table. 3. Mass determination results for experimental correction factor

Sample	E_γ (keV)	K	Real mass (g)	Determi-ned mass (g)	Relati-ve uncer-tainty (%)
Au-4	355.7	8.15	0.3409	0.3238	5.01
Cu-3	1345.8		0.0283	-	
Au-2	355.7	8.15	0.1442	0.1545	7.14
Cu-3	1345.8		0.0283	-	
Mo-11	739.4	2.14	0.0817	0.0753	7.83
Cu-12	1345.8		0.0284	-	
Mo-13	739.4	2.14	0.0877	0.0810	7.63
Cu-14	1345.8		0.0287	-	

Table. 4. Mass determination results for theoretical correction factor

Sample	E_γ (keV)	K	Real mass (g)	Determi-ned mass (g)	Relati-ve uncer-tainty (%)
Au-4	332.9	6.49	0.3409	0.3365	1.29
Cu-3	1345.8		0.0283	-	
Au-2	332.9	6.49	0.1442	0.1614	11.92
Cu-3	1345.8		0.0283	-	
Mo-11	739.4	1.77	0.0817	0.0911	11.51
Cu-12	1345.8		0.0284	-	
Mo-13	739.4	1.77	0.0877	0.0979	11.63
Cu-14	1345.8		0.0287	-	

Mass determination results of all samples are given in Table. 3 and 4 for the experimental and theoretical correction factors, respectively.

Relative uncertainties of the mass determination were obtained in comparison with real sample mass and were given in Table. 3 and 4, also. In the both cases of the correction factors the mass was determined for Au and Mo foils with uncertainties $\Delta m \leq 12\%$. In addition, the mass determination results for two different calculation methods of the correction factor are satisfactorily in agreement between themselves. It is possible to conclude that results for Au and Mo-foils are acceptable, however some effort to reduce uncertainty is needed.

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