INDEPENDENT METHOD OF Au DETERMINATION IN DIFFERENT MATERIALS BY APPLYING $(\gamma,\gamma')^m$-REACTION

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Represented results are based on using the ^{197}Au$(\gamma,\gamma')^{197m}$Au reaction, the experimental integrated cross sections of the isomer excitation through activation levels and the Monte-Carlo simulation method. It is shown that applying of high-current electron accelerator (EA) with energy 8.7 MeV permits to determine the threshold of gold definition in nature and technological materials $\geq$ 0.1 g/t.

1. INTRODUCTION

Gold is a less-common metal. So it is necessary to apply very susceptible methods having a high precision, efficiency and output for Au concentration to be found in geological samples at searching, exploration and mining. As an example of a successful decision of this problem it can be pointed applying the gamma-activation techniques for gold presence analysis in different materials. Two the most important reactions used in these techniques are (γ,n) and (γ,γ') ones.

For example the reaction ^{197}Au$(\gamma,n)^{196}$Au (half-life of the ^{196}Au $T_{1/2}$ = 6.8 days) was used to determine Au content in the Ukraine ores [1]. Activation was realized by bremsstrahlung photons. For this purpose, it was used the linear EA (LEA) with an energy range of $16 \ldots 25\,MeV$, a mean current $I_e = 0.1 \ldots 1\,mA$ and an irradiation time (t_{irr}) from 1 hour up to days. The threshold of Au determination $0.03 \ldots 0.1\,g/t$ is gotten for the most of ores researched except the samples having high content of Mn ($\geq$10%) where the threshold is below than 1-1.5 g/t. A mass of the probes irradiated is $M_{pr} \leq 500\,g$. The cooling time (t_c) is generally $5 \ldots 10\,hours$. Output of the method is $\sim$ 50 samples per shift.

First experimental investigations [2-5], using bremsstruhlung photons and ^{197}Au$(\gamma,\gamma')^{197m}$Au reaction for elemental analysis of geological probes, have showed principal opportunity of using high-current EA for express-analysis of natural and technological materials for Au determination. Soon on a base of the laboratory investigation results was worked out and introduced into industrial exploitation photonuclear analytical complex "AURA" [5]. For analysis is used LEA with energy $E_e = 8\,MeV$. Sample weight is 500 g. Diameter of mixture particles are 1 mm. Time of irradiation, cooling and measuring (t_m) – 15, 3 and 15 s respectively. Detecting system consists of two monocrystalls NaJ(Tl) $150 \times 100\,mm$. Above mentioned parameters provide the definition threshold of Au $0.2 \ldots 0.5\,g/t$ in ores of different composition. Project complex productivity is 400 analyses per shift.

In 1992 the systematic investigation into the photoexcitation of isomers over wide mass ranges, including Au, was studied [6] with the bremsstrahlung facility at the superconducting Darmstadt LEA. Excitation functions were measured for the $(\gamma,\gamma')^m$ reactions populating the metastable states for energies of $2 \ldots 7\,MeV$ and the important intermediate states were identified. Simultaneously it was defined integrated cross sections $(\sigma\Gamma)^j_{iso}$ of isomer ^{197m}Au excitation through intermediate (activated) levels E_j. Obtained experimental data gave the opportunity to calculate a number of activated Au nuclei by new, independent method.

In the present work, it is considered the possibility of applying the high-current EA for the express-analysis of Au content in nature and technological materials. Calculations were made taking into account existing experimental and theoretical investigation results, measured energy of the activated levels and integrated cross sections of isomer excitation and achievements in optimization region.

2. WORKING PARAMETERS SELECTION

As it is known [5], a yield of the $(\gamma,\gamma')^m$ reaction increase with growing of the endpoint photon energy ($E_{\gamma max}$). So increasing of activated nuclei number in $(\gamma,\gamma')^m$ reaction can be obtained by increasing of electron energy. However maximum E_e must be lower than the neutron emission threshold (ε_n) of the exit and dividing foils, converter, target and protection materials as neutron appearance lead to a formation of by-products radionuclides. Comparison of the ε_n

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of different materials shows that the energy of electrons can be increased up to 8 (9) MeV, using natural Ti (Al, Cu) for exit and dividing foils and Au (Cu) for converter.

A proposed arrangement of activation equipment on the EA exit is represented by Fig.1. Notations of Fig. 1: 1, 4 - are Ti foils, every one is of 50 μm; 2 - is two water layers, every is of 1.5 mm; 3 - Au converter; 5 - air layer of 5 mm; 6 - gold- containing mixture; 7 - Al container; 8 - polyethylene cover.

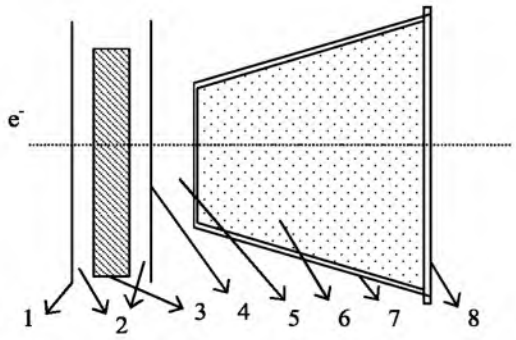

Fig.1. *Block - scheme of the activation equipment*

Whereas the neutron emission threshold of Au is large as against traditional metals Ta and W, the gold disk is proposed to use as the converter. Based on simulation by the GEANT3.21 package, it was found that an optimal Au converter thickness for proposed kinematics is 1 mm.

Selectivity that is ability of the analysis to separate nuclei of a desired element from rest sample nuclei plays an important role for founding optimal conditions in carrying out the activation analysis. On the Fig.2 [7], it is shown the dependence of a specific selectivity of gold determination in quartz ore on a endpoint energy $E_{\gamma max}$ of a bremsstrahlung spectrum for four values of irradiation time. Curves 1-4 correspond $t_{irr} = T_{1/2}, 3T_{1/2}, 5T_{1/2}, 7T_{1/2}$.

It can be seen the most specific selectivity is observed at $E_{\gamma max} = 8.5$ MeV for any t_{irr} and increases with decreasing irradiation time. So this optimal energy is taken as the base in this work. However, as electrons lose $\sim$ 200 keV passing an EA exit foil and a water layer, the initial beam energy is increased up to 8.7 MeV in further estimates.

The container in which broken muck is to be charged consists of a straight truncated Al cone with the wall thickness of 1 mm and the polyethylene cover of 1 mm thickness. The cone height 40 mm, small r and large R of cone radiuses are equal 20 and 35 mm respectively. Internal volume of the container is 97.39 cm^3 at the above mentioned parameters. A density of the milled sample is 0.8 g/cm^3 (mean value of the gold-bearing ore [8]), the weight of a container content is 77.92 g and the Au mass is 0.779×10^{-4} g under condition that its content in mixture is 1 ppm. At above conditions number of Au nuclei per cm^2 is $N_T = 0.979\times10^{16}$. Required calculations are carried out with the following parameters. The electron energy 8.7 MeV, the mean current is 1 mA, half-live of ^{197m}Au 7.73 s, detected γ-line $E_\gamma = 279.0$ keV. Irradiated time $t_{irr} = 16$ s, cooling time $t_c = 3$ s and measuring time $t_m = 19$ s.

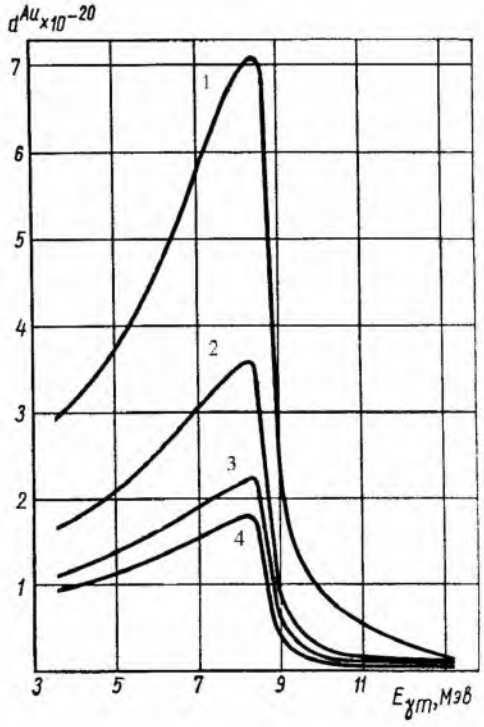

Fig.2. *The dependence of a specific selectivity on a endpoint energy $E_{\gamma max}$*

3. ISOMER PHOTOACTIVATION

Different investigations (see cites in [9]) have confirmed that direct resonant excitation of the isomeric levels is negligible because of very small width. The population of isomers by (γ,γ')-reactions at excitation energies below the photoneutron threshold proceeds by the process depicted schematically in Fig.3.

The figure identifies the relevant parameters including the natural width of the intermediate level Γ, which is the sum of all partial widths for transitions on all levels, lying between the excited and ground state and the branching ratios b_0 and b_{iso} for decay from the intermediate state directly to the ground state and by unknown cascade to the isomer.

Number of activated Au nuclei N_f is found by the expression of a normalized reaction $(\gamma,\gamma')^m$ yield [9]:

$$Y = \frac{N_f}{N_T N_e} = \sum_j (\sigma\Gamma)^j_{iso} N(E_j, E_{\gamma max}), \quad (1)$$

where N_e is a number of electrons passed through the converter, $(\sigma\Gamma)^j{}_{iso}$ is an integrated cross section of isomer excitation by the intermediate j level, $N(E_j,E_{\gamma max})$ is the photon field spectral intensity in solitary energy interval at energy $E_{\gamma max}$.

E_j - denote the activation level energy. The quantity $N(E_j,E_{\gamma max})$ is calculated for individual geometry of irradiation by code GEANT3.21.

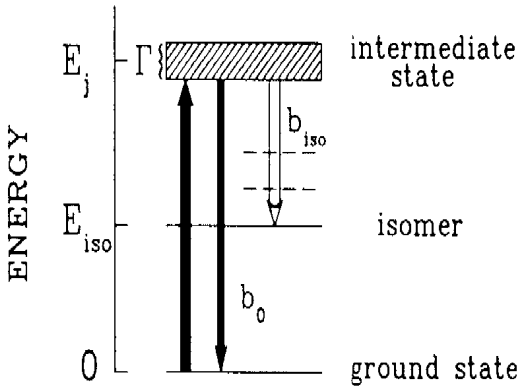

Fig.3. *Resonant photoexcitation mechanism by which an isomer at energy of E_{iso} is populated through the hatched states*

Estimation of N_f is fulfilled with the statistics of 10^6, step size $10\,keV$ and the experimental integrated cross sections [6] given by Table. These cross sections give the most contribution to gold isomer population. According to Eq. (1), number of activated nuclei in studied reaction ^{197}Au(γ,γ')^{197m}Au is 7822.

Intermediate levels and integrated cross sections

E_j, MeV	$(\sigma\Gamma)^j_{iso}$, $(10^{-29}\,cm^2\cdot keV)$
1.70±0.30	70±30
2.50±0.10	500±50
3.20±0.15	4500±500
4.20±0.20	20000±4000

4. DETECTOR EFFICIENCY

To estimate number of γ-quanta with $E_\gamma = 279.0\,keV$ registered really by the γ-spectrometer it is need to introduce corrections for efficiency of the system. The crystal NaJ(Tl) $150\times100\,mm$ is considered as the detector system. In the case, when only events with total energy absorption are taken into account, total efficiency is [10]:

$$\varepsilon_{tot} = \varepsilon_{geom}\cdot\varepsilon_{absorp}\cdot\varepsilon_{sample}\cdot\varepsilon_{event}\,. \qquad (2)$$

ε_{geom} is fraction of total number of photons, flying out in 4π angle, reaching a detector. ε_{geom} is calculated by the Monte-Carlo simulation method. The surface of crystal NaJ(Tl) is at $3\,mm$ distance from the probe surface. According to the calculation, 34.7% of γ-quanta with $E_\gamma = 279.0\,keV$ reach the crystal of $150\,mm$ diameter.

ε_{absorp} takes into account of influence of intermediate materials that absorb a part of irradiation before photons reach the detector.

$$\varepsilon_{absorp} = exp(-\sum\mu_i\cdot\rho_i\cdot x_i), \qquad (3)$$

where μ_i-, ρ_i- and x_i are the mass absorption coefficient, density and thickness of i-th intermediate material, respectively. To reach the detector a photon with energy of 279.0 keV is to go through the Al foil of 1 mm thickness, 1 mm polyethylene and 1mm air layer. Coefficients of mass weakening and materials density were found in [11].

ε_{sample} is a part of γ-quanta irradiated by the sample. According to [10]:

$$\varepsilon_{sample} = \frac{1-exp(-\mu\cdot\rho\cdot x)}{\mu\cdot\rho\cdot x}, \qquad (4)$$

where μ-, ρ- and x are the mass absorption coefficient, density and thickness of material, respectively.

In this work, it is considered that the mixture of γ-emitting element with sample matrix elements is uniform and homogeneous enough with respect to composition and density. Besides particles emitting photons are small, so self-attenuation inside a single particle is negligibly small. These requirements secure the linear attenuation coefficient has a single value in a large enough range and it can be used for calculation. Estimation of ε_{sample} is fulfilled with the mass absorption coefficient and density taken for the SiO_2 chemical compound [11].

ε_{event} is a probability that γ-quant reaching detector contributes a pulse to a total absorption peak. This correction was obtained from expression (41) [12] for a spot radiation source, disposed at the axis of crystal.

5. DISCUSSION

Known number of activated nuclei N_f permits to estimate number of real photons registered by the γ-spectrometer within a photopeak area (S_γ). Upon that it is need to introduce corrections [7] taking account of nuclei decay during irradiation time $1-\exp(-\lambda\cdot t_{irr}) = 0.76$, cooling time $\exp(-\lambda\cdot t_c) = 0.76$ and measuring time $1-\exp(-\lambda\cdot t_m) = 0.82$. Besides, it is necessary to introduce a correction connected with intensity of γ-line $I_\gamma = 0.71$, to take into account the detector total efficiency $\varepsilon_{tot} = 0.144$ and the dependence on the decay constant $\lambda = 0.693/T_{1/2} = 0.0897$.

Thus, in above mentioned geometry of equipment arrangement and EA parameters, number of photons with energy $279.0\,keV$, registered by the detector after finishing irradiation of sample with gold concentration $1\,ppm$, is equal 4740. Obtained number of γ-quanta indicates that applying EA with energy $8.7\,MeV$ permits for one exposure of 16 s duration at $1\,mA$ current to determine the threshold of gold definition $\geq 0.1\,g/t$ ($S_\gamma \sim 474$). If it is need one can extract the Au concentration $\sim 1\,g/t$ in sample reducing current up to $50\,\mu A$ ($S_\gamma \sim 237$). Obtained results are in agreement with data [4,5].

The threshold of Au definition can be made better if to increase a probe mass up to $500\,g$, set up two NaJ(Tl) detectors on the opposite sides of sample or apply semiconductor detector from high purity germanium with a large sensitive volume.

6. CONCLUSIONS

The represented results are based on using the ^{197}Au(γ,γ')^{197m}Au reaction, the experimental integrated cross sections of the isomer excitation through activated levels and the Monte-Carlo simulation method. It is shown that applying of high-current EA with energy 8.7 MeV permits for one exposure of 16 s duration to determine the threshold of gold definition $\geq$ 0.1 g/t. Simulation results can be usefull for elaboration and creation of an Industrial Photonuclear Activation Complex in Ukraine.

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НЕЗАВИСИМЫЙ МЕТОД ОПРЕДЕЛЕНИЯ Au В РАЗЛИЧНЫХ МАТЕРИАЛАХ С ПРИМЕНЕНИЕМ $(\gamma,\gamma')^m$- РЕАКЦИИ

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Представленные результаты основаны на использовании ^{197}Au(γ,γ')^{197m}Au-реакции, экспериментальных интегральных сечений возбуждения изомеров через уровни активации и моделирования методом Монте-Карло. Показано, что применение мощного ускорителя электронов с энергией 8.7 МэВ обеспечивает предел обнаружения золота в природных и технологических материалах $\geq$ 0.1 г/т.

НЕЗАЛЕЖНИЙ МЕТОД ВИЗНАЧЕННЯ Au У РІЗНИХ МАТЕРІАЛАХ З ВИКОРИСТАННЯМ $(\gamma,\gamma')^m$- РЕАКЦІЇ

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Представлені результати засновані на використанні ^{197}Au(γ,γ')^{197m}Au-реакції, експериментальних інтегральних перерізів збудження ізомерів через рівні активації і моделювання методом Монте-Карло. Показано, що застосування могутнього прискорювача електронів з енергією 8.7 МеВ забезпечує межу виявлення золота у природних і технологічних матеріалах $\geq$ 0.1 г/т.