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BULK ANALYSIS METHOD OF GOLD DETERMINATION IN ORES USING EPITHERMAL NEUTRONS OF ELECTRON ACCELERATOR MICROTRON MT-22

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Экспрессная методика определения золота в рудах с использованием резонансных нейтронов электронного ускорителя микротрона МТ-22

Описана экспрессная методика определения золота в рудах с использованием инструментального нейтронно-активационного анализа. Порошковые (100–200 меш) образцы были облучены в экране из кадмия толщиной 1 мм в каналах фотонейтронного источника — микротрона МТ-22 Центра ядерных исследований Монгольского государственного университета (Улан-Батор). Получены пределы определения 0,1 мг/кг Au в образцах массой 30–50 г при облучениях в течение 1–2 ч.

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Bulk Analysis Method of Gold Determination in Ores Using Epithermal Neutrons of Electron Accelerator Microtron MT-22

Bulk analysis method of gold determination in ores by Instrumental Neutron Activation Analysis (INAA) is described. The powder (100–200 mesh) samples were irradiated in Cd foils of 1 mm thick with photo-neutrons at the Microtron MT-22 of the Nuclear Research Center, Mongolian State University (Ulaanbaatar). The sensitivity of 0.1 mg/kg Au can be obtained using 30–50 g samples and irradiation time of 1–2 h.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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INTRODUCTION

The studies of gold deposits and its processing required a fast and efficient analytical methods for determination of gold and other major or minor elements in ores. The determination of major and minor elements has provided important information about mineral resources, geochemistry data and apart from ecological consequence of heavy metals and toxic elements gives also a possibility to improve reliability of prospecting deposits.

The fire assay method is one of the analytical techniques, with detection limit of 0.2 mg/kg in samples of 50 g weight [1, 2]. The fire assay method in combination with AAS and ICP techniques can be used for determination of gold content.

Recently, the instrumental analytical methods such as activation analysis with low power ($\sim$20 kW) research reactor [3], particle accelerators [4] and isotope neutron sources, with neutron fluxes from 10E + 8 to 10E + 11 neutrons/($\mathrm{cm}^2 \cdot \mathrm{s}$) are applied for gold analysis. Detection limit of the nuclear methods can be in the range of 0.01–0.1 mg/kg for representative samples of 30–60 g [5–6].

The cyclic electron accelerator — Microtron MT-22, which was installed in 1994 at the Nuclear Research Center, Mongolian National University [7] has been widely used as a nuclear analytical setup for element analysis including gold. The electron cyclic accelerators have some advantages over other nuclear physical equipment: low cost, easy of operation and low requirements for radiation protection [8].

Some ores samples consisting of several kinds of deposits were used to study the applicability of the accelerator MT-22 (all samples are collected by A. Karivai and G. Ganchimeg) for determination of gold.

Gold ores reference materials, such as USZ 21–98 (1.06 $\pm$ 0.16 mg/kg); USZ 23–98 (3.28 $\pm$ 0.19 mg/kg) and USZ 31–2000 (10.72 $\pm$ 0.81 mg/kg) were supplied by Central Geological Laboratory and were used as quality control materials. These reference materials were approved by Mongolian National Agency of Standardization. The samples with variable matrix were analyzed, which allowed one to estimate detection limit of gold and some other elements in real samples.

Average concentrations of major and minor elements are given below:

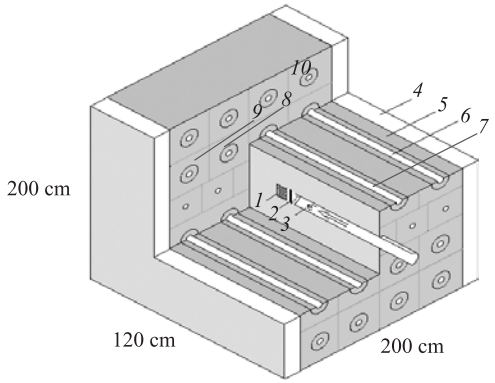
Na,%	Cu,%	Fe, %	Zn,%	As, %	Sb, %
from $n^* \cdot 10\mathrm{E}-2$ up to 3.0	from $n^* \cdot 10\mathrm{E}-3$ up to 23.0	from $n^* \cdot 10\mathrm{E}-1$ up to 19.0	from $n^* \cdot 10\mathrm{E}-3$ up to 12.0	from $n^* \cdot 10\mathrm{E}-2$ up to 6.0	from $n^* \cdot 10\mathrm{E}-3$ up to 0.6

* n = 1–9.

1. MATERIAL AND METHODS

The ores samples of 30–60 g weight were packed in polyethylene capsules ($\oslash$ = 7.3 cm, h = 1.3 cm) and placed inside irradiation channels of photo-neutron source – 2.0 × 2.0 × 1.2 m graphite cube of the Microtron MT-22.

In the center of the graphite cube of the Microtron MT-22 a metallic uranium rod of 530 g weight in Al capsule was used as a target. Photo-neutrons were emitted in $^{238}\mathrm{U}(\gamma, n)$ reaction by irradiation with electrons having energy up to 22 MeV and the beam current of 10–15 μA. All gold ores samples and reference materials were irradiated together at horizontal irradiation channels of the graphite cube in Cd cover as shown in the Figure.



Sample irradiation channels in the graphite cube. *1* — ^{238}U, 530 g; *2* — 2-mm Ta foil; *3* — electron beam; *4* — graphite; *7, 8, 9, 10* — irradiation channels

The R_{Cd} ratios were as follows:

Channel number	Irradiation channel-1	Irradiation channel-2	Irradiation channel-3	Irradiation channel-4
R_{Cd} for Au	1.87	1.93	2.0	1.94

The low ratio R_{Cd} for gold (1.9–2.0) gives possibility of decreasing Compton radiation background in the region of 411.8 keV analytical peak and analytical peaks of trace elements (Na, Sc, Cr, K excluding Sb, As). Consequently, the total activity of samples was decreased but not that of 411.8 keV peak of gold. Experimentally optimized irradiation, cooling and measurement times for gold analysis were t_{irr} = 1–4 h, t_{cool} = 10–24 h and t_{mes} = 20 min, respectively.

The counting rate was determined using the following equation [1, 8]:

$$A = D\varepsilon\frac{am}{M}N_A\Phi\sigma[1-\exp\left(-0.693\,t_{\mathrm{I}}/T_{1/2}\right)]\exp\left(-0.693\,t_{\mathrm{d}}\,T_{1/2}\right), \qquad (1)$$

where A is the measured counting rate at a time $t_{\rm d}$, counts per second; D is the branching ratio for the emission being measured; ε — the detector efficiency of the measuring system; a — isotopic abundance of considered isotope; m — mass of target element; M — molar mass of target element; N_A = (6.02 1.0E + 23) is Avogadro number; Φ — the neutron flux, n (cm^{-2} s^{-1}); σ — the neutron absorption cross section of the particular reaction with the target isotope at a given incident neutron energy, cm^2; $t_{\rm I}$ — irradiation time; $t_{\rm d}$ — decay time from the termination of the irradiation to the start of counting; $T_{1/2}$ — the half-life of the product nucleus being measured.

Some nuclear data for the reaction of gold with neutrons and bremsstrahlung on Microtron are given in Table 1.

Table 1. Nuclear data for the reactions of gold with neutrons and bremsstrahlung on Microtron

Neutron energy	Nuclear reaction	Neutron cross-section, b	Half-life	Gamma-ray energies, MeV
Thermal neutron (0.025 eV)	^{197}Au (n,γ) ^{198}Au	98.8	64.8 h	0.4118 (0.95)
Resonance neutron $E_n > 0.025$ eV	^{197}Au (n,γ) ^{198}Au	1553	64.8 h	0.4118 (0.95)
Bremsstrahlung	^{197}Au (γ,n) ^{196}Au	0.0081	6.18 d	0.333(0.24); 0.356 (0.936)

2. RESULTS AND DISCUSSION

The distribution of neutrons in channels was measured using gold monitors, which were placed between samples and reference materials. The monitors were prepared by dropping with pipette 300 μg Au on filter paper.

The self-absorption of 411.8 keV energy peak of ^{198}Au in samples was calculated according to the exponential absorption equation for sample with thickness x as follows:

$$I = I_0(K_0 - 1)/(\ln K_0), \tag{2}$$

where K_0 is $\exp(-\mu x)$; μ is linear mass absorption coefficient; I_0 is intensity of radiation at $x = 0$; I is measured intensity of radiation at thickness x.

The K_0 coefficient is determined by measuring irradiated gold's 411.8 keV analytical peak intensity using various samples of different matrices. Results of measurements showing that the absorption of the 411.8 keV energy peak intensity varies from 0.5% in quartz gold ore up to 5% in sulphite base metallic ores.

The detection limit (DL) for gold is determined using gold reference materials irradiated by mixed (thermal and epithermal) and epithermal neutrons using the following equation:

$$\mathrm{DL} = C_{\mathrm{Au}}\, x \left(\frac{3\sqrt{S_{\mathrm{bg}}}}{S_{\mathrm{Au}}} \right), \tag{3}$$

where S_{bg} — background under the analytical line; S_{Au} — analytical peak area of ^{198}Au; C_{Au} — concentration of gold in reference material.

The activities of irradiated samples were measured using gamma spectrometer with HPGe detector GC-3020 and PC analyzer S-100 (CANBERRA). Energy resolution of the detector was 1.9–2.2 keV for 1332 keV line of ^{60}Co.

Table 2. Comparison of results of neutron activation analysis for gold determination with chemical analysis. Concentration range of gold in samples of 0.8–10.0 mg/kg. Method of analysis: Instrumental Neutron Activation Analysis (INAA)

No.	Code of samples and reference materials	Results of CGL, mg/kg	Results of INAA NRC, mg/kg	Coefficient of variance	Acceptable error, %
1	651	3.31	4.18	23.23	62
2	652	7.65	9.0	24.52	62
3	653	3.03	3.65	18.83	62
4	654	6.8	8.12	17.09	62
5	655	3.4	4.3	23.52	62
6	335	4.12	4.18	1.46	62
7	332	4.95	7.02	33.41	62
8	5	7.35	9.76	28.37	62
9	346	8.6	11.35	27.57	62
10	350	7.2	9.5	27.54	62
11	353	3.98	4.53	12.93	62
12	373	3.74	3.55	5.23	62
13	380	8.68	10.59	19.82	62
14	382	5.6	5.28	5.88	62
15	6543	6.8	9.01	27.96	62
16	350	7.2	9.63	28.88	62
17	373	3.74	2.49	40.13	62
18	380	8.68	11.93	31.54	62
19	382	5.6	5.3	5.5	62
20	USZ31-2000	10.05	9.24	8.4	62
21	USZ21-98	0.79	1.28	47.34	62
22	B7/2	5.92	7.0	16.72	62
23	USZ23-98	3.28	3.46	5.34	62

The measuring time for samples — 20 min and for monitors — 5 min, respectively. For quantitative determination of gold content in samples 411.8 keV analytical peak of ^{198}Au was used.

The detection limit of gold achieved in this condition was in the range from 0.1 to 0.3 mg/kg, which is adequate for prospecting and exploitation of gold deposit purposes. In these experiments, the irradiation of samples in Cd should be used for bulk analysis method of gold, while for determination of other elements together with Au the samples should be irradiated without Cd cover.

Results of comparison analysis of gold ores samples by INAA and the fire assay methods are shown in Table 2.

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