Photoactivation analysis of ores using short-lived radionuclides

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(Received April 13, 2006)

The results of applying photoactivation for express nondestructive multicomponent analysis of apatite-nepheline, gold-bearing, polymetal and lapparite ores are presented. The experiments were carried out at the IPP microtron with bremsstrahlung of 10 to 15 MeV electrons. A setup with mixed gamma-neutron field was built and used for elemental analysis. Experiments show that such elements as P, Al, Ti, Ba, and Ce can be determined with a sensitivity of approximately 0.1 mass% and a statistical error of a few percents. These values are wholly satisfactory to modern demands of industry. Gold and silver can be successfully determined in gold-bearing ores on the level of grams per ton.

Introduction

Activation analysis is usually applied to the analysis of small amounts of impurity or trace elements in various objects. We consider the problem of the determination a large amount of elements in ores and rocks with high accuracy and speed. In particular, this problem appears in the evaluation of the quality of multicomponent ores in the mining-geological industry, and in exploration and prospecting for mineral deposits. The rapid analysis of the quality of ores is of great interest for the industry. For instance, the main component of apatite nepheline ore is P$_2$O$_5$ and its concentration varies in apatite deposit at Kolsky peninsula (Russia) in the range 8–22%. Efficient extraction of useful components from the ore depends to a large extent on prompt determination of the phosphorus concentration.

Rocks and ores are difficult to analyze due to the complexity of their elemental content, inhomogeneous distribution of some elements in the bulk, and the large range of concentrations of elements to be analyzed (10$^1$ to 10$^{-8}$ mass%). In addition, it is important that the main rock-forming elements have threshold energies of photoactivation that are higher than the corresponding energies for ore elements of interest. For example, carbon has the threshold energy for the ($\gamma$,n)-reaction of 18.7 MeV, oxygen – 15.5 MeV, Na – 12.5 MeV, Mg – 16.6 MeV, Al – 13 MeV, Si – 17.2 MeV, K – 17.2 MeV, Ca – 10.1 MeV, Ti – 13.2 MeV, Fe – 13.8 MeV.

Actually, a lot of physical, physical-chemical, chemical methods are applied for the analysis of ores and rocks. Among these are optical spectroscopy, various XRF instrumentation, and neutron activation analysis.

The main problem in the analysis of geological samples is the requirement to have representative samples. The rather high nonuniformity of the distribution of elements in minerals leads to the need to analyze probes of large mass (tens or even thousands of grams). It follows from this peculiarity of geological objects that the methods applied for analyzing them have to be characterized by rather deep penetrability of the probing radiation. Such commonly used methods as X-ray spectroscopy or radiometric techniques indicate the surface content of elements only (this especially relates to elements with small Z). The common approach to overcome this difficulty is powdering of samples up to 200 mesh and mixing well. Actually, in such an approach, the time of analysis may be tens of hours or even some days.

Photoactivation analysis (PAA) performed at an electron accelerators make it possible to overcome this problem because the penetrability of high-energy photons in ores reaches 30 cm. The main advantages of photonuclear methods in the analysis of rocks and ores is that it is possible to avoid radioactivation of matrices in the determination of ore-forming elements by varying the end-point energy of bremsstrahlung. Besides, in photonuclear reactions, as a rule we obtain short-lived radionuclides. These features of photoactivation analysis make it possible to perform rapid analysis of ores and minerals of samples of large mass.

Experimental

We report the results of applying PAA for rapid analysis of gold-bearing, apatite-nepheline, polymetal and lapparite ores. These ores are of great interest for geology and industry because they contain gold and silver (gold-bearing ores), phosphorous, aluminum, titanium (apatite nepheline ores), niobium, and rare elements (lapparites).

Experiments were carried out using external electron beam from the IPP microtron with variable electron energy from 7 to 15 MeV and average current of 25 µA. As the bremsstrahlung target, a 3 mm thick tungsten plate was used. Irradiation of samples gives rise to produce radionuclides that are mainly due to ($\gamma$,n), ($\gamma$,p), and ($\gamma$,f) reactions.

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The samples that placed in a polyethylene container (15 cm³ in volume) were irradiated for a short time by bremsstrahlung, and were transported to and from the irradiated facility by a pneumatic post. To provide uniform irradiation of the sample the following construction of the irradiation setup was used. The radiation chamber consists of an outer thin-walled aluminum tube and an inner cylinder which rotates freely about it. The inner cylinder has tangentially directed channels to allow passage of air and a drainage hole at the end. When the container hits the cylinder, the force of the air flowing through causes the container to begin to rotate together with the cylinder at about 5 rev/s. The container is induced to rotate by an externally mounted hermetically sealed contact which is activated when the small permanent magnet mounted on the inner cylinder passes by.

The transportation time of a sample from the irradiation setup to the γ-detector was approximately 3 seconds. Gamma-ray spectrum measurements were performed with a Ge(Li) detector of 15 cm³ volume with 5% efficiency for 300 keV gamma-rays and with a resolution of 6 keV. Gamma-ray spectra were processed using a multichannel analyzer.

Analysis of gold-bearing ores

The abundance of gold and silver in nature is severely limited. The mean (Clarke) contents of these elements are 5·10⁻⁷ and 1·10⁻⁵ mass%, correspondingly. Large-scale analyses, needed for geology and industry, require high-accuracy and rapid analytical methods for determining these elements.

The rapid variant of PAA is based on the use of short-lived isomers of gold and silver ¹⁹⁷ᵐAu, ¹⁰⁷ᵐAg, and ¹⁰⁹ᵐAg, which are formed as a result of inelastic scattering of gamma quanta on corresponding stable isotopes. The half-life of ¹⁹⁷ᵐAu is 7.3 seconds, and the energies of the emitted γ-rays are 70, 130 and 277 keV. The most intensive are 130 and 277 keV, which are used for gold identification. The nuclear characteristics of silver isomers (¹⁰⁷ᵐAg –44 s, 94 keV, ¹⁰⁹ᵐAg –39 s, 88 keV) are close in value and are not usually separated in the data processing.

PAA of gold-bearing ores was successfully used for rapid analysis in Magadan region (Russia) and in Uzbekistan with the use of a linac and a microtron as a source of bremsstrahlung, and with a NaI(Tl)-detecting system. We present here the results of detecting induced gamma-radioactivity with a Ge(Li)-detector at different bremsstrahlung end-point and for various types of gold-bearing ores to show the influence of the matrix content on the detection limit of PAA. It is necessary to note also that the existence in the gold-bearing ores of other elements is of great importance. For example, Ba and Hf are paragenetically connected in some fields with Au and are geochemical criteria in ores searches. Se and Ba, together with other sulphides, are often present in gold-bearing ores and are the products of secondary geochemical processing of a source.

In accordance with their mineralogical composition, samples can be divided into two groups: “simple” in which silica and aluminosilicates with an insufficient content of sulphides and oxides of different metals are the main part of the matrix, and “complex” where metal sulphides and oxides are at a considerable (1–10%) level. Figures 1 and 2 display gamma-spectra from two typical representatives of these groups. Irradiations were performed at three electron energies: 8, 10, and 15 MeV.

Photopeaks of Ag (90 keV), Se (162 keV), Hf (217 keV) and Au (279 keV) are distinctly seen in the spectra. It is also seen that in a complex sample, even at the 8 MeV energy, the photopeaks from Au and Ag are on a Compton background from the Ba γ-line that decreases the sensitivity of the analysis.

Figure 3 shows the dependence of sensitivity of determination of Au concentration with end-point of bremsstrahlung for simple (1) and complex (2) gold-bearing ores. It is seen that for simple ores the detection limit is 0.2 grams/ton for Au. For silver, the detection limit equals 10 grams/ton.

Fig. 1. Gamma-ray spectrum of the simple gold-bearing sample analyzed at the bremsstrahlung end point energies 8 (1), 10 (2) and 15 MeV (3)