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Photo- and digital-detectors for quantitative digital densitometry for distribution analysis of ores by means of gamma-activation

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Abstract A method for photo processing of nuclear film detectors, which ensures higher reproducibility (about 2 %) of densitometry has been suggested. Application of the method allows for recovering fine mineralogical texture of a complex polymetallic ore of the Norilsk deposit with time series autoradiograms after computer processing. Some problems concerning interpretation of the results are discussed. It has been shown that application of digital imaging plates as a detector demonstrates high reproducibility suitable for precise quantitative autoradiography. The developed method of digital gamma-activation autoradiography via short-lived radionuclides may be successfully used for screening analysis of large area ore's thin sections.

Keywords Activation autoradiography · Processing of time series of autoradiograms · High reproducible densitometry · Mineralogical texture · Spatial analysis of geological samples

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Introduction

Application of highly precise instrumental methods for spatial analysis in geochemistry is very promising. Among these methods, one can mention X-ray microanalysis, scanning electron microscopy (SEM), mass spectrometry with inductively coupled plasma (ICP–MS) and laser ablation, secondary ions mass spectrometry (SIMS), and others. At the same time methods for fast screening analysis of the sample are in demand. Such methods may be used for searching for zones of interest to be further analyzed by a suitable precision instrumental method. Earlier it has been shown that the computerized digital gamma activation autoradiography is a suitable method for screening analysis of polymetallic ores. The method is applicable for analysis of large size thin sections of tens cm^2 [1–3]. The specific features of the method are high spatial resolution (microns or even less) and low detection limit (μg-ng depending on element). Due to a sufficiently large path of electrons in the materials, the method is able detect a signal from a certain depth (up to hundreds of micrometers). The duration of the analysis mainly depends on the number of images to be acquired for each sample and half-life of radionuclides under measurement. It is clear that the method is more effective for registration of short-lived nuclides ($T_{1/2} = n$ hours). In this case, the measurements takes 2–3 days, with minimum of manual operations. Simultaneously one can irradiate 3–4 samples.

The main aim of the developing approach consists of increasing selectivity and extension of capabilities of the activation autoradiography method by means of an application for both the computer's assisted analysis of time series images and new digital systems for autoradiograms registration, paying special attention to the quality of the results.

Experimental

Calibration of flat samples

To study the reproducibility of the results of quantitative 2D-densitometry of three flat samples (filter papers about few cm^2 each) with different activity of ^{137}Cs have been prepared (17.2, 8.8 and 4.4 kBq/cm^2). The surface of the samples has been protected by self-adhesive film of 40 microns thickness. Autoradiographic image analysis demonstrated that the distribution of ^{137}Cs over the surface of the samples is practically uniform.

Software

Processing a series of autoradiographic images and generating meta images was performed by means of the software "Image_mapping" developed at the laboratory [2, 3].

Irradiation of ore samples

Polished thin-sections of the polymetallic ore samples were irradiated using IMET RAS microtron (2.5–5 mA average current with the maximum energy of the bremsstrahlung 22 MeV). Duration of irradiation was in the interval 20–60 min. The distance sample to tungsten converter was about 5 cm. To ensure uniform irradiation, the samples were moved in the Bremsstrahlung field using a previously developed device [1].

Obtaining of autoradiograms

Two different detectors have been used for autoradiogram acquisition. They are: conventional photo detector (BioMax MR film by Kodak) and imaging plates by PerkinElmer. In the former case, the developed image was converted in the digital form using a slide scanner CanoScan 8800F with optical resolution of 4,800 dpi. The calibration of the dependence of the scanner response on the optical density was done by means of a standard film Kodak Q-60 [2]. The autoradiograms acquired by imaging plates may be directly read in the form of digital images by a special laser scanner.

Results and discussion

The previously developed method has been designed for increasing the selectivity of gamma-activation autoradiography via relatively short-lived radionuclides. The aim is reached by means of the computer's processing of time series autoradiograms obtained during decay of the induced activities in the sample. First of all, the images composing time series should be coaxially arranged. This procedure is described in details in Ref. [2]. The following computer's processing deals with sets of decay data (optical density) for each pixel of the time series images. The software makes deconvolution of decay curve for each set of pixels for two components or, if it is impossible, for one component. The obtained half-life values are stored in the binary arrays of the same size as the size of the original autoradiograms. For visualization of the obtained half-life results, a cross-section method for generation of a set of meta images for the desired interval of half-life values has been developed [3].

It has been proved that the developed method is reliable and it is able to detect Pd–Pt inclusions in the ore. At the same time, the problem with registration of the fine mineralogical structure of polymetallic Cu-Ni ore of Norilsk deposit is still observed. Figure 1, in the left column, contains two images for copper and nickel distribution over the thin section surface obtained by SEM. The middle column (b) contains the meta images for radionuclides ^{64}Cu ($T_{½} = 12, 7$ h.) and ^{57}Ni ($T_{½} = 35, 6$ h.), obtained by the developed method. One can see that the meta images do not transmit the fine mineralogical texture of the ore sample consisting of three main minerals pentlandite, chalcopyrite, pyrrhotite which compose mosaic texture of the Norilsk area ore. The "white areas" on the meta images mean that there are no data on half-life because the program was not able deconvolute the decay curve for some pixels. The analysis of the possible reasons for the situation convinced us that reproducibility of densitometry is a key element for obtaining acceptable quantitative results. So the task of the following investigation was to find a solution to improve the quality of the radiographic results, focusing on obtaining the correct mineralogical texture of the ore sample.

Investigation of densitometry reproducibility

Photo detectors were exposed with two calibration flat samples (^{137}Cs) with different radioactivity for 10 and 20 min. Each photo detector was processed immediately after exposure with freshly prepared developer solutions strictly followed the recommended conditions for photo processing (temperature, etc.). The obtained images were digitized, with the following determination of the optical density for each sample, including background subtraction. The results for two samplings are shown in the Fig. 2a. Standard deviation for the mean optical density for both experiments is about 10 %, which is not good enough for further decay analysis of time series autoradiograms. It has been assumed that even small uncontrollable variations in development of photo detectors may significantly affect the results of densitometry.

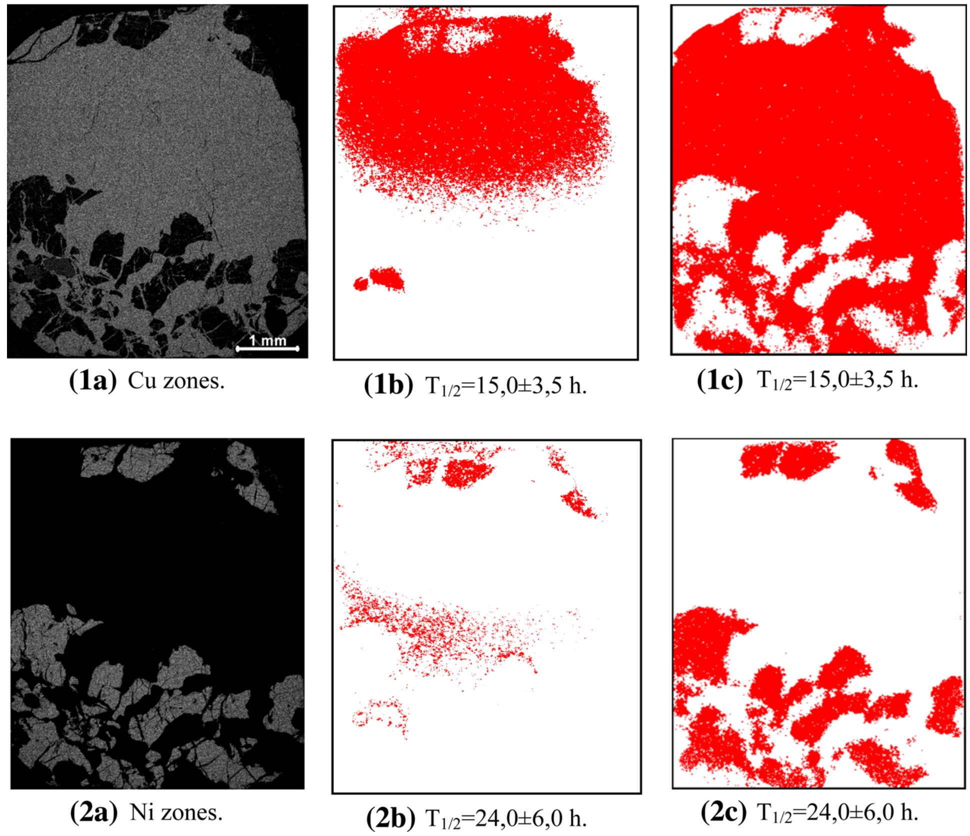

Fig. 1 The distribution map for copper (1) and nickel (2) obtained by SEM (**a**), developed earlier time series analysis method (**b**), improved time series analysis method (**c**)

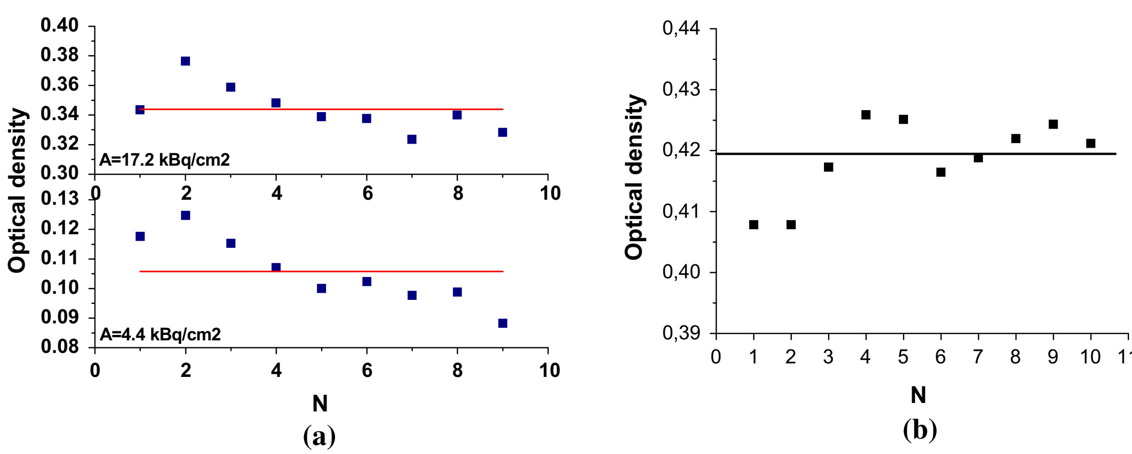

Fig. 2 Reproducibility of densitometry for autoradiography images obtained by means of different processing of nuclear photo detectors: **a** immediately after exposition **b** all detectors are developed simultaneously in the same tank

To confirm this hypothesis, another series of experiments was carried out. In this series, ten photo detectors one by one have been contacted with the flat sample of ^{137}Cs (8.8 kBq/cm^2) for 80 min. The exposed samples were left to the end of all experiments of the series. Then all exposed photo detectors were developed simultaneously in a specially designed tank without any mixing during the development process. The results are presented in Fig. 2b. Standard deviation of the mean is not higher than 2 %, which is significantly better than for the previous case.

Testing of the results by the improved method

The same polymetallic ore sample was irradiated again and a time series of autographic images were obtained using the improved method for development of the exposed photo

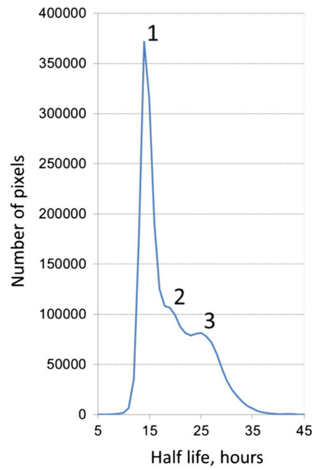

Fig. 3 Distribution of the processed pixels of the whole image on the obtained half-life values

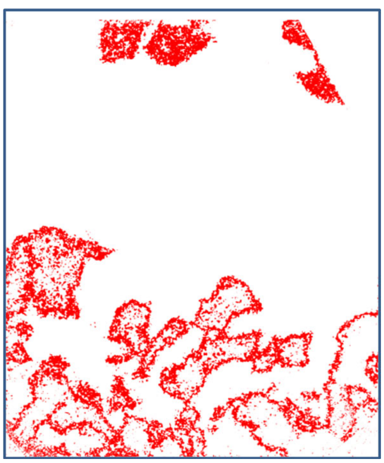

Fig. 4 Meta image corresponding to apparent half-life 18 ± 2 h

detectors. The subsequent computer processing of time series autoradiograms and generation of meta images uses the same procedures and software as it was earlier. The results for distribution of copper and nickel are presented in Fig. 1 (column c). One can see that the situation improved significantly. Mineralogical textures obtained by the developed method correspond to the texture obtained by SEM.

Figure 3 shows distribution of all processed pixels of the whole image on the computed half-life values. One can see three peaks (zones of the sample) having the following values of the apparent half-life: ~14, 18 and 25 h. The first peak may be attributed to decay of ^{64}Cu ($T_{½} = 12, 7$ h.). The last peak we have assigned to ^{57}Ni ($T_{½} = 35, 6$ h.). Copper–nickel ore of the Norilsk deposit consists of relatively fine-grained fractions of different minerals of copper, nickel and iron: chalcopyrite ($CuFeS_2$), pentlandite

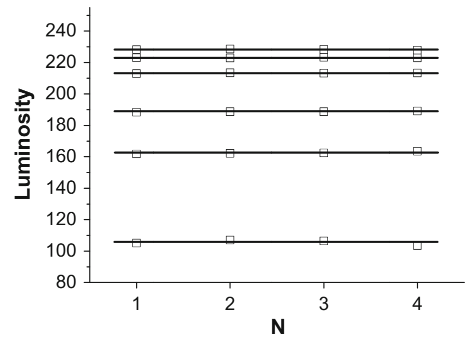

Fig. 5 Reproducibility of measurements of different luminosity (higher luminosity means lower optical density) for imaging plates

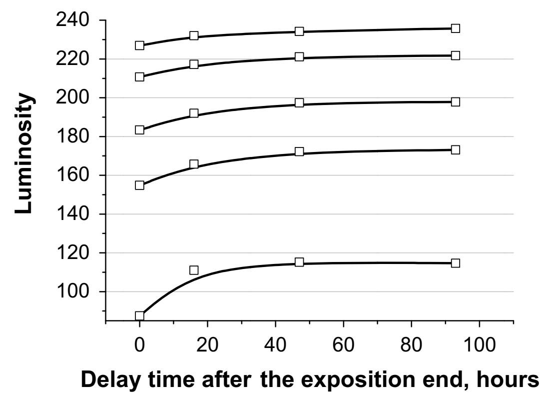

Fig. 6 Degradation of the acquired dose depending on delay time after the exposition end

($(Fe, Ni)_9S_8$) and pyrrhotite($Fe_{1-x}S_n$) [4]. We assume that difference between the apparent and the tabulated half-life values for the copper and nickel may be explained by "backlight" effect, caused by other minerals lying on some depth under a mineral located on the thin section surface. This assumption is in agreement with observation that for copper zones the apparent half-life value is slightly shifted from the tabulated one whereas for nickel zone the value of bias is greater. It may be explained by different induced activity: the activation cross section for isotope ^{65}Cu about four times higher than for isotope ^{58}Ni.

The middle small peak is artificial and associated with boundaries between minerals composing mosaics (Fig. 4). Its appearance may be explained both by error while preparation of a set coaxially arranged images and "backlight" effect.

Studying of reproducibility of luminosity measurements by means of imaging plates

The flat sample of ^{137}Cs (4.4 kBq/cm^2) has been measured 6 times for different durations (10–60 min) to acquire six different doses. The imaging plate had to be kept for 30 h in a dark place and at room temperature after the exposition before the imaging plate was scanned. The reproducibility of luminosity measurements (which is in a reverse relation to optical density) for different acquired doses is presented in Fig. 5. The obtained reproducibility is sufficient and lies in the interval 0.1–1.5 %.

The necessity to delay scanning of the exposed imaging plates is explained by degradation of optical density during 24–30 first hours after exposition (Fig. 6), which is in agreement with the results published in Ref. [5]. The obtained data confirms that the imaging plate may be successfully used for processing of series of autoradiograms for radionuclides mapping.

Conclusion

The developed method for the computer's processing of time series autoradiograms seems to be very suitable as a solution for a broad spectrum of tasks where application of spatial analysis of large area sample is needed. It will be especially effective if one of the modern digital registration systems is used as a detector, such as the quite well known imaging plates, or the recently developed digital X-ray flat panel sensors for real time image acquisition (Hamamatsu [6]).

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