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Generation of metaimages while analysis of the time series autoradiograms

V. P. Kolotov · D. S. Grozdov · N. N. Dogadkin

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Abstract To increase selectivity of activation autoradiography for the first time it has been suggested to use computer's processing of time series of autoradiography images obtained while the sample is cooling. To preserve the spatial resolution of the method, the processing must involve all coaxial pixels of the series. For visualization of the obtained results a cross section method for generation of a set of metaimages has been developed. The generated metaimages characterize the spatial distribution of pixels for half-life values lying in the selected interval. The algorithm for testing the computational data compatibility within the preselected zone has been developed. The algorithm is based on analysis of frequency distribution of half-life values for pixels array composing the sample zone under testing. The normal distribution characterizes compatible data for homogeneous part of the sample. Using the frequency analysis, a method for generation of contrasted metaimages has been developed. The method is able to distinguish zones with half-life differences of about 1 h.

Keywords Gamma-activation · Autoradiography · Image processing · Computational contrasting filter

Introduction

Visual radioanalytical methods (mainly the various variants of radiography) based on recording images have a long history. For the last 80 years, these methods have played a decisive role for the study of the distribution of radionuclides and elements (after activation or introduction of radioisotope labels) in a variety of natural and technological samples [1, 2]. The appearance of powerful local instrumental methods of analysis (X-ray spectral microanalysis, electron microscopy with EDS etc.) led to a significant displacement of radiography in the analysis of solid samples. Only in biological studies using labeled compounds is the method is still widely used. However, the potential of these methods has not been exhausted. The methods are still actively applicable for studying the radionuclides behavior in nature [3, 4], for the development of reliable methods of immobilization of radioactive wastes in stable matrices [5], for the analysis of "hot" particles [6, 7], etc. The 90th new digital methods of recording images of radionuclides have been introduced into the practice of investigations. Among these methods one can mention the image plates technique [8, 9], developed by FujiFilm, based on the effect of delayed luminescence. In the last decade, scientists are actively developing methods for the detection of labeled compounds in vivo, mainly in the study of plants. Laboratory systems such as PET (IS) [10–13] have been developed. There is a good experience with ccd cameras and micro capillary plates [14]. Other methods of nuclear radiation detection based on the latest achievements in photonics are used also.

It has been shown that gamma-activation autoradiography is an effective method for the detection of noble metals in polished thin sections of geological samples [15, 16].

By its nature, the method of autoradiography is not selective, since it is based on registration an integral dose of beta (or other) radiation of radionuclides induced in the sample during irradiation. Using of mathematical solutions implemented in the form of computer programs can provide the method of autoradiography with new features and, first of all, improved selectivity. For example, information on the distribution of radionuclides over the sample surface

V. P. Kolotov (✉) · D. S. Grozdov · N. N. Dogadkin
Vernadsky Institute of Geochemistry and Analytical Chemistry of Russian Academy of Sciences, Moscow, Russia
e-mail: vpkolotov@inbox.ru

can be determined by analysis of the registered dose dynamics during the sample cooling. It may be done by means of mathematical processing of a series of autoradiography images. This idea has been used for analysis of individual zones of the images by means of MS Excel [17] or by automatic computer pixel by pixel processing of the whole image [18].

Previously [19] the method of digital gamma-activation autoradiography has been extended for analysis of large size samples (several tens of cm^2). This novelty added the method competitive ability with respect to some non-nuclear methods of local analysis. It is worth to note, that the autoradiography method is capable to detect micro-inclusions on some depth (up to tens microns) that is a side benefit.

Usually the size of the silver halide grains (conventional nuclear film detector) is a fraction of micron. Autoradiographic images obtained by nuclear film detector can be digitized with resolution, up to 11.000 dpi. This corresponds to a spatial resolution of about 2 microns, which is comparable with the grain size that is still not reachable by new digital registration systems.

Experimental part

Samples

For autoradiographic gamma activation analysis polished thin sections of copper-nickel complex polymetallic ores of the Noril'sk deposits have been prepared. Ore sample containing artificial inclusions of some noble metals in copper-nickel ore have been made too for testing purposes.

Irradiation of the samples

Samples were irradiated at the Institute of Metallurgy and Material Science RAS by compact cyclic accelerator of electrons (microtron) with average current about 2.5–5 mA and maximum energy of bremsstrahlung about 22 MeV. Duration of irradiation was 20–90 min. The distance from tungsten converter to sample was 5–15 cm. The large size samples were irradiated with a developed device for uniform irradiation [19].

Preparation and digitization of the autoradiograms

As a nuclear radiation detector BioMax MR Film (KODAK) has been used. Calibration of the film detector response (optical density vs beta-particles fluence) was obtained by using a standard flat source of ^{60}Co. Slide scanner CanoScan 8800F was used to digitize the autoradiograms. Standard film Kodak Q-60 was used for calibration dependence of the scanner's response on optical density [15].

Software

The software package for image processing has been developed using the programming language C # (MS Visual C # Express). Program Adobe Photoshop CS2 was used to prepare a series of coaxial aligned images. MS Access database was used for storage and transmission to the processing program information concerning sample irradiation and autoradiograms acquisition.

Results and discussion

Investigation of the spatial radioactive decay dynamics of the sample by means of digital autoradiography imply using of quantitative densitometry and pixel-by-pixel computer processing of the images composing a series. Images of a series are formed while the sample exposure with film detector during the sample cooling. The task of experimenter is obtaining images of optimal optical density at constantly increasing duration of exposure, controlled by average decay rate of the induced radionuclides. Preliminary preparation of a series of autoradiography images consists of three phases: (1) coaxial alignment of autoradiography images, (2) correction of pixels luminosity for linearization of the dependence of the pixel luminosity on the optical density value, and (3) recording the detailed information on the sample irradiation and autoradiograms acquisition in the database for batch processing in an automatic mode. The details are given in Ref. [18].

Reproducibility of densitometry

The reproducibility of densitometry for quantitative autoradiography researches is a key factor determining the quality of the subsequent mathematical processing. It has been shown experimentally that the relative standard deviation of the optical density does not exceed 6 % only if all the autoradiograms of a series have been developed simultaneously in a common tank.

Algorithms and software for processing a series of images

The task of the software can be formulated as the following: for a set of coaxial pixels from a series of images it is required to estimate the apparent half-life value(s) of radionuclides mixture for each pixel. It means making a decomposition of the decay curve for one or more exponentials (depending on the number of points, the character of the decay curve, the error of experimental data, etc.). This is a known inverse problem. To solve this problem a linear least squares method has been applied. We have introduced a concept of "component", which is an individual radionuclide or mixture of radionuclides with similar or statistically indistinguishable decay constants. Due to limited number of images composing a series (usually 4–7 images) the algorithm was tuned for decomposition of the decay curve for one or two components (short-lived and long-lived ones), depending on the properties of the sampling. The algorithm uses as a logical analysis of data, and iterative mathematical processing for correction decay while acquisition.

Cross section method for generation of metaimages

To visualize the results of the images processing the program generates a series of derivative, so-called—metaimages by using cross-section method. In fact, metaimages represent the sets of monochromatic pixel whose half-life values lye in the specified interval. At the same time the program is able generate the correspondent images for visualization of residuals while LSM fitting. In the Fig. 1 a series of metaimages for one experiment is presented. The picture shows zones with uniformly increasing values of a half-life. The analysis of mapping results for ore sample allows to distinguish zones of (Pd + Pt)—micro-inclusions, with half-life within 16 ÷ 18 h (Fig. 1d–f). These zones can be identified as containing a mixture of radionuclides ^{197}Pt and ^{109}Pd ($T_{1/2} = 18.3$ and 13.7 h, respectively). X-ray microanalysis confirmed the presence of inclusions in these zones, which consist mainly of platinum (14 %) and palladium (50 %). Series of metaimages shown in Fig. 1a–c contains a zone, which can be defined as containing mainly radionuclide ^{64}Cu, presence of which is also confirmed by X-ray microanalysis. The generated metaimages are the basis for further statistical analysis of the half-life results in the interesting zones.

Statistical analysis of zones of interest

It is proposed to use frequency analysis of half-life values for pixels array composing the selected zone. Testing of the approach was performed by analysis a large area sample of ore containing the artificial inclusions of Pt and Pd. In the Fig. 2 a metaimage with pointed areas of Pt and Pd inclusions is presented. The zones of the sample selected for analysis must be pointed for program by a special image, which was named as "mask" image. The mask is a white image (the same size as the sample image) which has one or more area (s) tinted black. The "mask" image may be easily created as a layer in Photoshop. The black area points pixels of the sample to be processed. For pixels specified by a mask, the developed software forms a sampling array containing calculated half-life values. Then the program generates a frequency distribution of pixels number on half-life values. The quantization step for sampling is adjustable parameter.

It has been found that the frequency sampling for the inclusions zones may be fit by normal distribution (Fig. 3).

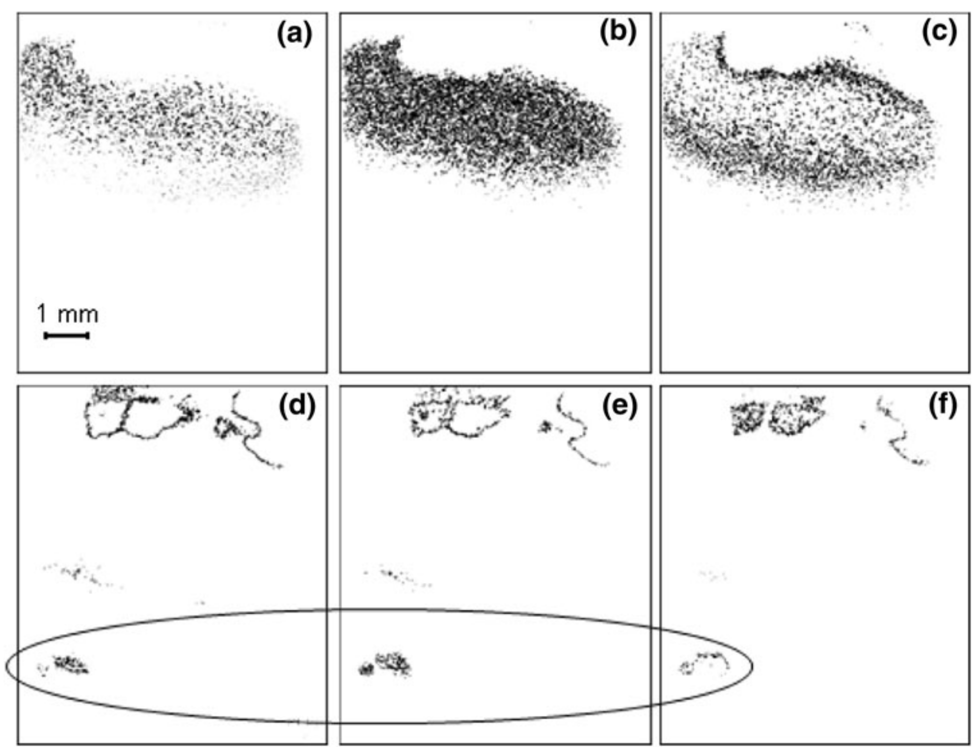

Fig. 1 Meta images of the results of processing of time series of the sample. The images **a**, **b**, and **c** correspond to half-life value of 12.3 h with increment 0.5 h; The images **d**, **e**, and **f** correspond to half-life value of 16.0 h with increment 0.5 h. Area pointed by *oval* contains inclusions. The autoradiogram of the sample is given on Fig. 4

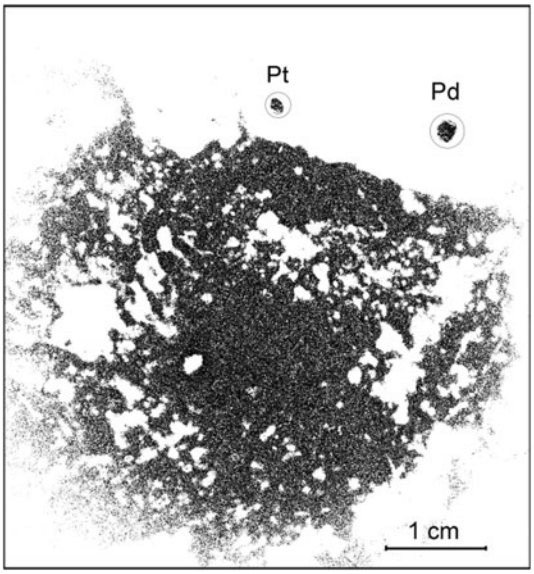

Fig. 2 Meta image obtained by cross-section method for the ore sample with artificial inclusions of Pd and Pt half life values laying in the interval (16 ± 4) h

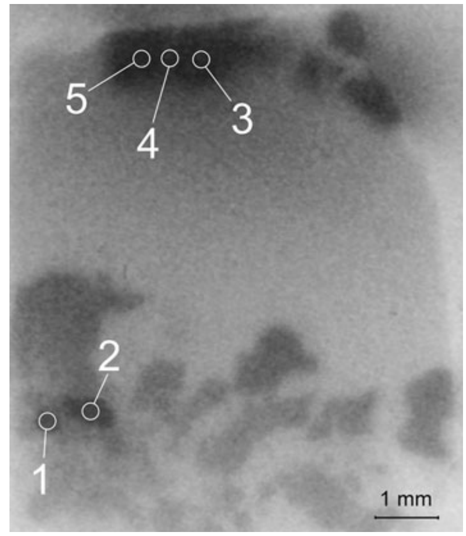

Fig. 4 Autoradiogram of the sample of polymetallic ore, with indication of zones to be statistically tested by frequency analysis

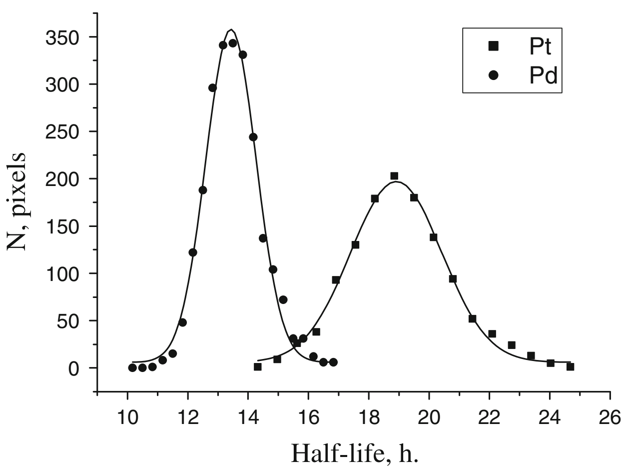

Fig. 3 Frequency distribution of half life values for zones of the sample containing artificial inclusions of Pt and Pd

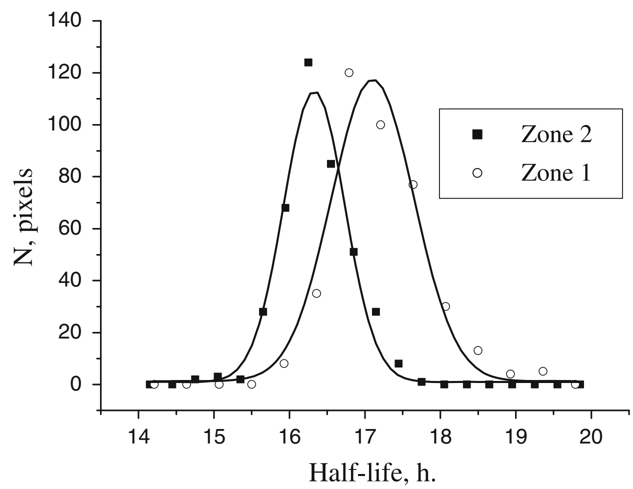

Fig. 5 Distribution of the computed half life values for *zones 1* and *2*

The analysis of the distribution allows determine half-life value for the investigated areas of the sample more accurately. Thus, calculated half-life values for areas of Pd is 13.4 ± 0,9 h and for area of Pt is 19 ± 2 that is in a good agreement with tabulated data, 13.7 and 18.3 h, respectively.

Similar experiments were performed for the sample of polymetallic ore containing a real Pt–Pd inclusions. The Fig. 4 shows the autoradiogram image of the sample with the selected zones for frequency analysis. It has been confirmed that for areas characterizing by stable results while half-life determination the frequency distribution fits by normal distribution. From Fig. 5 it follows that calculated half-life values for zones 1 and 2 of the autoradiogram are (16.5 ± 0.5) and (17.1 ± 0.6) h, respectively. Some discrepancy of the results between two areas may explained by slightly different composition. This was confirmed by X-ray microanalysis (the ratios of Pt/Pd for zones 1 and 2 are 0.26 and 0.29).

Knowing parameters of frequency distribution it is possible to estimate half-life resolution while autoradiograms series processing. Thus, according to the obtained data (Fig. 5) the resolution time is about 1 h.

For comparison, the frequency distribution of the calculated half-life values for other zones of the autoradiogram is given in the Fig. 6. One can see that the distribution can't be apparently approximated by the normal one and therefore

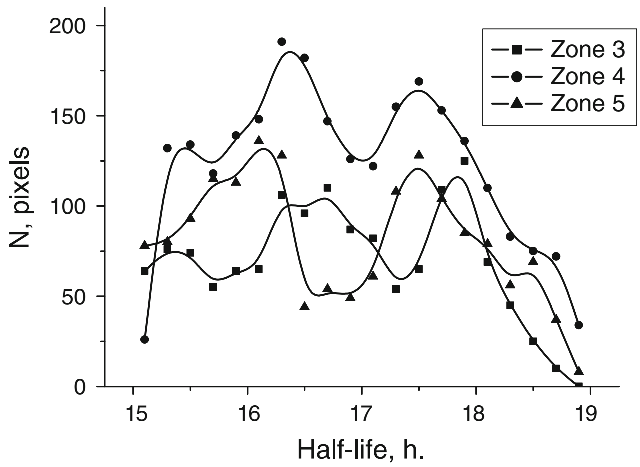

Fig. 6 Distribution of the computed half life values for *zones 3*, *4* and *5*

cannot be recognized as the inclusion's zone. This observation gives the basis for development of two-dimensional filter, allowing contrasting metaimages by means of separation of the useful zones (for example, containing inclusions) from the other ones.

Obtaining contrasted metaimages by means of two-dimensional computer's filter

The two-dimensional filter represents a square matrix with adjustable size, which scans the array of results containing half-life values for each pixel of the image. The matrix scans the image array with a preset shift in the horizontal and vertical directions. At each step data set pointed by matrix is analyzed to clarify how good the current data sampling fits normal distribution in the frames of certain boundary conditions. To increase computational efficiency of the procedure it calculates the second and third central moments of the distribution instead of application fitting algorithm. The values of these moments and their ratio allow estimate similarity of sampling under testing to normal distribution. If absolute values of these moments are less than preset limiting values (typically 0.5 and 6.0 for the second and third moments), the program assumes that the distribution of half-life values fits normal distribution. Adjustment of the preset values may control the "strength" of contrasting filter, be it more rigid or soft.

The contrasting filter works according the following scheme. On detection of normal distribution, all pixels of the current matrix are set to the peak value (half-life) of the found distribution. If the sampling of the results for matrix does not fit normal distribution all pixels are set to zero. Choosing the size of a two-dimensional filter it is possible to regulate spatial resolution of the contrasting process.

The application of the developed method for generation of the contrasted metaimage for the real ore sample proved that the developed algorithm displays quite positive effect for the selection of zones of inclusions. For comparison, in the Fig. 7a shows the metaimage generated by contrasting filter whereas Fig. 7b presents the metaimage obtained by cross-section method. One can see that the image 7a is significantly informative showing all areas definitely having useful signal, including the zones of inclusions which look more finished.

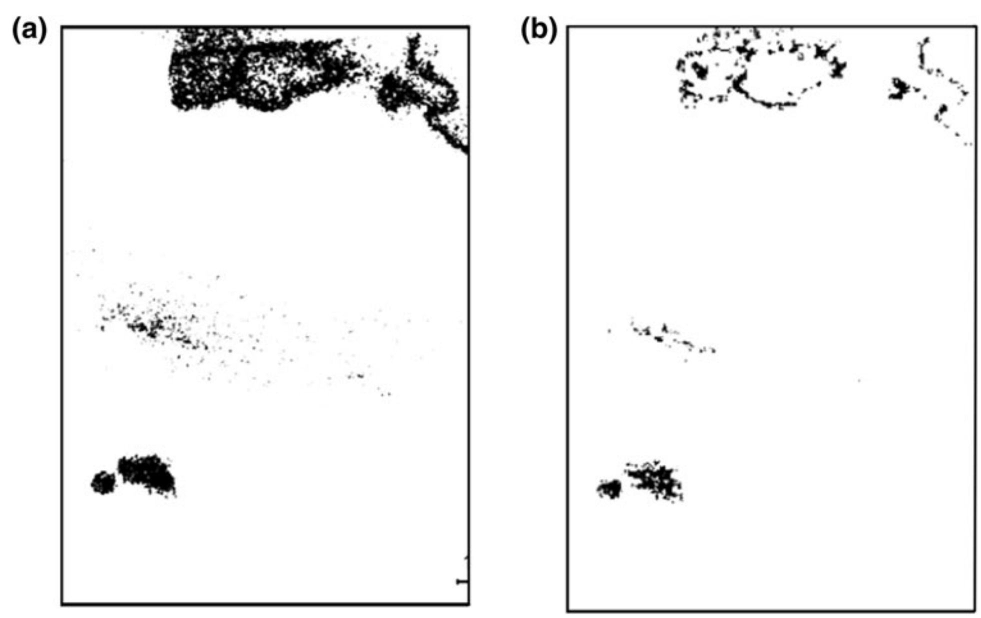

Fig. 7 Metaimages of the sample of polymetallic ore, generated by different computing methods: **a** contrasted meta image, using frequency analysis, **b** meta image obtained by cross section method for half-life interval $T_{1/2} = 17 \pm 2$ h

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