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Abstract

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X-RAY SPECTRAL FLUORESCENCE ANALYSIS WITH A NUCLEAR SOURCE OF PRIMARY RADIATION

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In recent years, the question of the possibility of replacing an X-ray tube with an artificial radioactive radiation source has attracted interest (^{1}-^{5}). In the present work an attempt was made to carry out fluorescence X-ray spectral analysis by replacing the X-ray tube with the artificial radioactive isotope Tu^{170}. The X-ray and γ-radiation of this source was used for fluorescence excitation of the K-radiation of elements from ^{30}Zn to ^{92}U.

In view of the weak intensity of the excited fluorescent X-ray radiation, it was necessary to abandon investigation of the spectral composition of the radiation by the crystal-diffraction method and to apply a crystal-free method for investigating radiation energy, based on the use of a scintillation counter and an amplitude analyzer. The scintillation counter consisted of an NaJ(Tl) crystal ($20 \times 5 \times 1.5$ mm) and an FEU-29 photomultiplier. The gain of the linear amplifier was 500, the channel width of the single-channel amplitude analyzer was 1 V, and the resolving time was 1–2 μsec.

The samples were prepared in the form of rigid tablets by pressing a substance applied to paper together with polystyrene, which served as the backing. The consumption of substance was 100 mg for a tablet area of 6 cm^2.

The Tu^{170} source was prepared from powdered oxide Tu_2O_3, pressed into a corrugated cuvette made of aluminum foil and subjected to powerful neutron irradiation. The artificial radioactive source obtained in this way, having an initial activity of 12 Cu*, was loaded into a special container. Reliable protection from radioactive radiation was provided by the lead walls of the container, which were 4.5 cm thick. The design of the container made it possible, safely for the experimenter, to introduce samples into the container, placing them near the source in the path of the beam of its X-ray and γ-radiation.

The radiation of the Tu^{170} source comprises: 1) β-radiation with energies of 968 and 884 keV (^{6}), 2) γ-radiation with an energy of 84 keV (^{6}), 3) characteristic X-ray radiation of ytterbium and thulium (mainly Yb $K\alpha_{1,2}$, Tu $K\alpha_{1,2} \simeq 52$

keV) ([7], [8]), 4) bremsstrahlung arising upon the braking of β-particles in the substance of the source ([7]).

Initially we were oriented toward mixed excitation of the characteristic X-ray radiation of the elements both by β-particles (the primary excitation method) and by X-ray and γ-radiation (fluorescence excitation). However, the subsequent task that arose of reducing the background in every possible way in order to increase the sensitivity of the analysis forced us to settle on the fluorescence method of excitation, using only part of the radiation from the source; the β-particles were excluded from participation in the excitation of the characteristic X-ray radiation of the samples. Complete stopping of the β-particles was achieved by a system of aluminum filters placed between the thulium source and the sample under study. A considerable reduction of the background produced by radiation from the source scattered inside the container and emerging through its slit, and by X-ray radiation arising in the lead walls of the container, was achieved by the following measures: 1) careful collimation of the beam emerging from the container; 2) a rational choice of the method of preparing the samples, ensuring a reduction of scattering by the sample itself; 3) a rational choice of the material of the sample holder, ensuring better absorption of the radiation scattered at the rear wall of the container; 4) complete filtration of the β-particles.

Fig. 1. Curves of the amplitude distribution of pulses from the characteristic K-radiation of the elements Ge, Ag, Eu, Os, U, placed under different excitation conditions

The shape and height of the curve of the spectral distribution of the background when a pellet with the substance under study is introduced into the container change somewhat, but the dependence of this change on Z is insignificant. This circumstance makes it possible to carry out a graphical subtraction of the background. All the curves of the energy composition of the radiation given below are the result obtained after subtracting the background distribution curve from the experimental amplitude-distribution curve of the pulses from the fluorescent radiation of the samples.

Fig. 2. Curve characterizing the dependence on Z of the excitation efficiency of the K-radiation of elements by the X-ray and γ-radiation of a Tu^{170} source. The dashed lines indicate the boundaries separating elements that are under sharply different excitation conditions

The excitation efficiency of the characteristic radiation of different elements is determined by the energy composition of the radiation from the Tu^{170} source.

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* It should be noted that the work was carried out over the course of a year, and the results presented below were obtained with a source activity not exceeding 1.5 Cu.

Figure 3 and Figure 4

Figure 1: Figure 3 and Figure 4

In Fig. 1 are shown the lines due to the K-series of a number of elements that are under different excitation conditions:

The K-series of Ge is under unfavorable excitation conditions, since the energy of the primary radiation greatly exceeds the excitation energy (11.1 keV).

The K-series of Ag (excitation energy 25.5 keV) occupies an intermediate position between the cases of Ge and Eu.

The K-series of Eu (excitation energy 48.6 keV) is optimally excited by radiation having an energy of 52 keV; γ-radiation (84 keV) and bremsstrahlung also participate in its excitation.

The K-series of Os (excitation energy 78.1 keV) is excited by radiation with an energy of 84 keV and by bremsstrahlung, and is not excited by radiation with an energy of 52 keV.

The K-series of U (excitation energy 115 keV) is excited only by bremsstrahlung.

Fig. 3. Curves characterizing the dispersion $D(E)$ and resolving power $\Delta E/E$ of a scintillation spectrometer

Fig. 4. Curves of the amplitude distribution of pulses caused by the K-radiation of elements of the cerium group of rare earths, obtained for different contents of the sum of rare earths

Figure 2 shows a curve characterizing the dependence on Z of the excitation efficiency of the K-radiation of elements (from $Z = 30$ to $Z = 92$) by the x-ray radiation and γ-radiation of the source Tu^{170}. As is evident from this curve, the nuclear x-ray generator is most effective for elements from $Z = 50$ to $Z = 63$.

In the case where the characteristic radiation recorded by the scintillation spectrometer has an energy above 33 keV, additional maxima are observed in the pulse-amplitude distribution curve, associated with excitation by the investigated radiation of the characteristic radiation of iodine, which is part of the NaJ(Tl) crystal (*). The appearance of such "false" maxima, which do not correspond to the energy composition of the radiation emerging from the container and are associated only with the scintillator crystal, constitutes a serious hindrance in studying the energy spectrum of the investigated radiation. A possible way of eliminating these interferences is the use of scintillators with light components, such as, for example, LiF or organic crystals.

Figure 3 shows the dispersion curve of the scintillation spectrometer. In the energy region 0–100 keV this curve is a broken line consisting of two rectilinear sections. The break in the dispersion curve was also observed by other investigators [10,11], who suggest that it is connected with the properties of the NaJ(Tl) crystal. The same figure shows the resolution curve of the scintillation spectrometer, determined by the ratio of the half-width of a line to its energy. The latter has a sharp jump at the same point where there is a break in the dispersion curve.

Despite the low resolving power of the scintillation spectrometer used by us (see Fig. 1), it can nevertheless be used to solve certain analytical problems. At the same time it should be noted that the possibilities for significantly improving the resolving power of the scintillation spectrometer are far from exhausted (the use of higher-quality crystals, and also the use of a proportional counter). As one possible application of the method discussed (at the level of resolving power achieved by us), one may propose the classic example of analysis for pairs of elements Ta—Nb, Hf—Zr. The pulse-amplitude distribution curves for the mentioned pairs of elements are well resolved.

For the first practical application of the nuclear generator, we set the task of quantitatively determining the total rare earths in various minerals. Figure 4 shows a series of lines obtained on artificial mixtures with different contents of total rare earths (from 2.5 to 100%). Table 1 gives the results of analyses of several mineral samples obtained on a scintillation spectrometer with excitation of the characteristic radiation (K series) of elements of the rare-earth group by a Tu^{170} source. These results are compared with data obtained by the usual X-ray spectral fluorescence method with excitation of the characteristic radiation (L series) of the rare earths by primary radiation from an X-ray tube and by the crystal-diffraction method of studying the spectral composition of the radiation.

Table 1

Mineral	ΣTR in % (cerium group): nuclear generator and scintillation spectrometer	ΣTR in % (cerium group): X-ray tube and crystal-diffraction method
Monazite	65 ± 3	63 ± 3
Orthite	24 ± 1	26 ± 1
Unknown mineral	20 ± 1	22 ± 1

It should be noted that the pulse-amplitude distribution curves from the fluorescent radiation are characteristic for this type of mineral. This fact makes it possible to use the method for rapid diagnosis of minerals. The compactness of the apparatus and the absence of a need for large amounts of electric power (energy is expended only on powering the electronic counting apparatus) make this method suitable for use under field conditions.

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Note: Figure translations are in progress. See original paper for figures.