DETERMINATION OF ELEMENTAL COMPOSITION OF ROCKS WITH TAGGED NEUTRON METHOD

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The technology for determination of the elemental composition of rocks on the basis of the tagged neutron method is tested in the field conditions. Here we are reporting the results of experimental studies of rock samples irradiated by fast tagged neutrons with energy of 14.1 MeV. The source of the fast neutrons is a portable neutron generator with a built-in silicon alpha–detector. Characteristic gamma rays resulting from the inelastic scattering of the neutrons on nuclei of the chemical elements that make up the geological rocks and minerals are registered by gamma–detectors based on BGO crystals. A comparison of the results on the elemental composition of rocks obtained by the tagged neutron method with reference values is discussed.

1 INTRODUCTION

Detector systems based on the tagged neutrons method (TNM) [1] - [3] are widely used to detect explosives hidden in various objects of inspection [4]- [5]. By physical nature of such detectors they are used to determine the relative concentrations of carbon, oxygen and nitrogen.

In work [6] it is shown how to use the TNM to search for diamonds in kimberlite ore without destroying it. The aim of this work is the use of TNM to determine the elemental composition of rocks in the field. The method of tagged neutrons is used to analyze the spectra of the characteristic gamma - radiation produced by irradiating the object of investigation by the flow of fast neutrons with energies of 14.1 MeV produced in binary nuclear reaction

$$d+{}^{3}H\rightarrow{}^{4}He+n. \qquad (1)$$

Since the α-particle (nucleus of ^{4}He atom) and the neutron are fly

away in almost opposite directions, it is possible to determine the direction of neutron emission by registration (tagging) the α-particles with a special detector, which is a matrix of separate sensing elements (pixels). Tagged neutrons entering the object of study undergo inelastic scattering

$$n+A \rightarrow n'+A^*,\ A^* \rightarrow \gamma+A, \qquad (2)$$

resulting in an excitation of the nucleus which is removed by the emission of gamma rays with an energy spectrum characteristic of each chemical element. The registration of the characteristic γ-radiation by γ-detectors is carried in coincidences with the signal from the α-detectors. This makes possible to determine all three coordinates of the point of the sample from which γ quantum was emitted. The two coordinates are determined by the number of the pixel which was hit by the α-particle, and the third coordinate characterizing the distance from the tritium target to the point of emission of γ-quantum is calculated using the time-of-flight technique. The distance from the tritium target to the point of emission of a gamma-ray is defined as a product of time between registration of the α-particle and γ - quantum by the velocity of the neutron with the energy of 14.1 MeV, which is constant and is equal to 5 cm/ns. By measuring of the γ-spectrum from a sample consisting of several chemical elements, it is possible to decompose the γ-spectrum components and to determine the relative proportion of each element in the sample.

In the present paper we discuss the results of the first application of TNM to determine the elemental composition of rocks in the field conditions. The samples of silicate and carbonate rocks taken at Olkhon geodynamic testing ground [7] were used for the tests. The measurements were carried out at the geological base Chernorud (Irkutsk State Technical University). The main purpose of the measurements was to check the possibility to perform correct TNM analysis of the elemental composition of rocks from a relatively homogeneous groups of indicator elements. So, for the carbonate rocks it is important to separate between calcite marbles (without magnesium), dolomite marbles (with a substantial share of magnesium) and calciphyres (rock with a substantial admixture of silicates and varying ratios of calcium and magnesium). It was interesting to see how well the results of TNM are correlated with the results of the petrographic analysis carried out in the field, and how well they correlate with the results of the chemical analysis in the laboratory.

2. EXPERIMENTAL SETUP

The measurements were done using a portable detector DVIN-1 [4]. It consists of an inspection module which is connected with a cable to the operator's computer. The inspection module has dimensions of 740x510x410 mm and a weight of 40 kg. Inside it a neutron source, a

gamma detector and data acquisition electronics are placed. Power supply is taken from 220V network. Power consumption is about 300 Watts.

The neutron source used is a portable neutron generator ING-27, produced by VNIIA named after N.L.Duhov (Moscow) with a built-in silicon α-detector. The intensity of the neutron flux generated by the neutron generator (NG) was $I = 5 \cdot 10^7$ s^{-1}. The nine pixels of the silicon α-detector form a 3x3 matrix with the size of each element of 10x10 mm^2. It is positioned at a distance of 62 mm from the tritium target. The preamplifiers of the α-detector signals are mounted on the flange of NG.

The detection of the γ-rays is carried out by a single detector on the basis of BGO crystal, with a diameter of 76 mm and a thickness of 65 mm. At the γ-line of carbon (Eγ = 4.44 MeV) the energy resolution of the γ-detector is about 4.4%. The time resolution of α-γ coincidences is 3.3 ns.

The measurements took place at Olkhon geodynamic testing ground, based in the site of Chernorud (Irkutsk State Technical University). The size of the samples ranged from 20 to 50 cm, their weight is from 2 to 6 kg. No special sample preparation was performed. The measurement time for each sample was 15 min. The area of survey at the sample was approximately 9x9 cm. It is divided into 9 areas (the number of the tagged beams). The elemental composition was determined simultaneously in each area.

The chemical composition of the samples was determined by a silica analysis in the Analytical Center of the Institute of Earth's Crust (Irkutsk) by standard methods [8].

3. EXPERIMENTAL RESULTS

The gamma-ray spectrum of each sample is resolved into individual components by fitting the sum of its constituent gamma-spectra from the following elements: Si, Mg, Fe, Al, Ca, C, O. These reference spectra have been measured in advance.

For the analysis in the field conditions samples of three groups of rocks: carbonate rocks (dolomite, calcite marbles and calciphyres, 17 samples), gneisses (23 samples) and amphibolites (7 samples) were presented. The energy spectra of typical samples from selected groups of rocks are shown in Fig.1. Thus the energy spectrum of the irradiated calcite marble sample (elemental composition of $CaCO_3$) is shown in Fig.1a.

The characteristic gamma lines of oxygen shown in Fig.1a by dashed lines are clearly visible. Three oxygen peaks dominate at energies E> 5000 keV. The other three peaks are seen at E = 2742, 3089 and 3854 keV.

The characteristic spectrum of calcium shows a prominent peak at energy E = 3737 keV. The imposition of this line to the peak corresponding to the deexcitation cascade transitions for oxygen nucleus (transition

energy E = 3854 keV), creates some blurred structure of non-Gaussian shape.

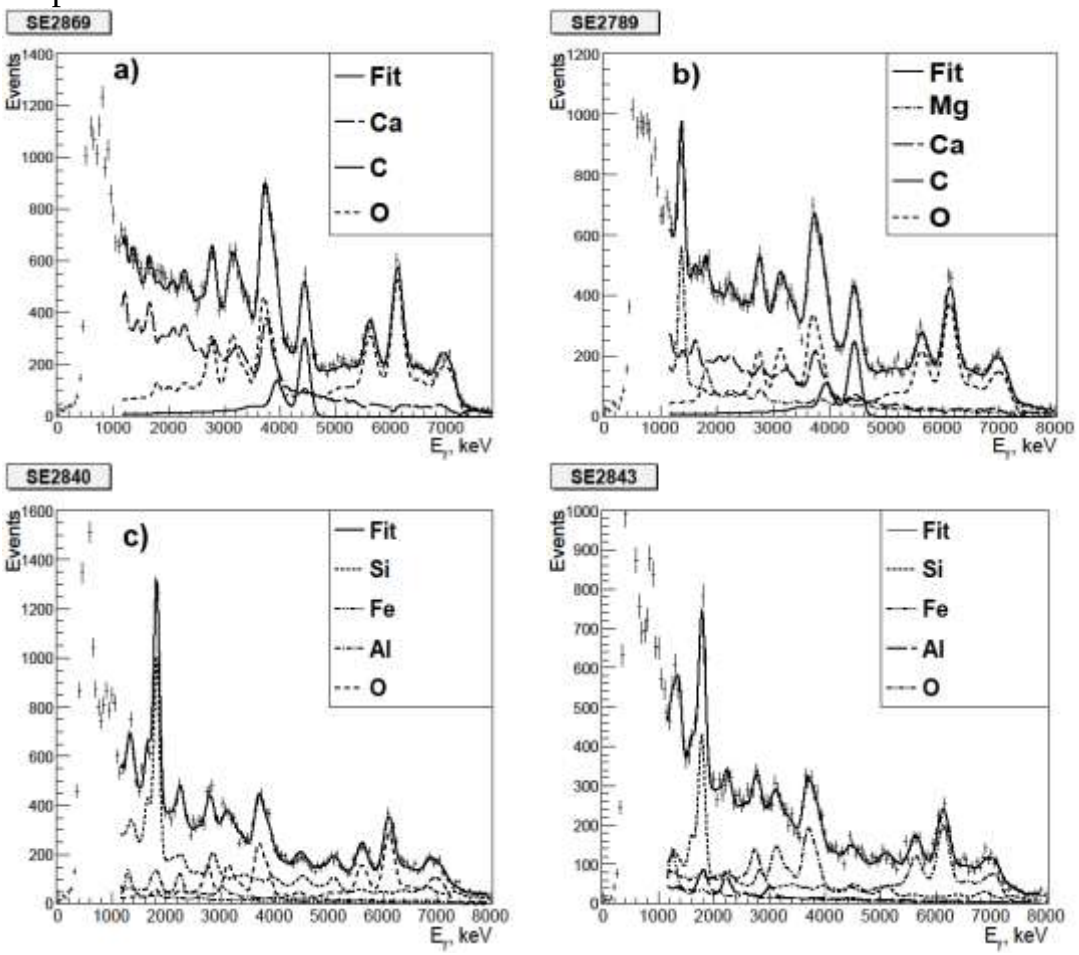

Fig. 1. The energy spectrum of gamma-rays radiation from the samples of calcite marble (a), dolomitic marble (b), gneiss (c) and amphibolite (d). The contributions of individual elements are shown. The solid line shows the result of fitting procedure.

The characteristic spectrum of carbon is the simplest - it stands out from only one line at E = 4439 keV and the peak of the escape of single annihilation gamma ray shifted to 511 keV from the main line.

These quality features of calcite are also confirmed by the quantitative results from determination of the elemental composition. Table 1 lists the mass fractions of the various elements for some samples of calcite marble obtained in the central tagged beam, which covers an area of 3x3 cm on the sample surface.

Fig. 1b shows the spectrum of a sample of dolomite marble (elemental composition $CaCO_3$ x $MgCO_3$). In addition to the characteristic lines of Ca, C and O, which are present on Fig.1a as well, a gamma-line of

magnesium at 1369 keV is clearly visible. This is reflected in the results of the determination of mass concentrations listed in Table 1. They show the correct presence of four main elements in the dolomitic marble. The mass fraction of magnesium reaches 7-10%.

Table 1. The mass fractions of the elements of various species in the samples obtained from the measurements with detector DVIN-1.

Sample	Si, %	Mg, %	Fe, %	Al, %	Ca, %	C, %	O, %
Calcite marbles							
SE2869	-	-	-	2.6±1.0	34.9±1.8	13.3±0.6	48.8±1.4
SE2801	-	-	-	-	42.3±1.3	16.1±0.7	41.6±1.2
SE2766	1.0±0.4	-	-	-	43.6±1.7	13.2±0.6	42.1±1.1
Dolomite marbles							
SE2789	-	9.6±0.6	-	2.5±1.0	27.5±2.2	14.4±0.7	45.3±1.9
SE2871	-	7.7±0.6	2.1±1.1	-	25.3±2.0	15.0±0.7	48.8±1.7
SE2842	-	8.9±0.6	-	2.0±0.8	28.3±2.1	15.9±0.8	44.6±1.7
Calciphyres							
SE2841	3.9±0.9	-	-	-	40.2±2.7	14.0±0.8	39.9±2.0
SE2803	2.9±1.0	-	-	2.2±1.4	39.6±2.5	13.9±0.8	41.4±2.0
SE2794	3.0±0.7	-	-	-	43.7±1.2	14.2±0.6	38.5±0.9
Gneisses							
SE2840	26.9±1.5	6.7±0.7	-	16.4±1.7	6.8±2.4	-	42.4±1.9
SE2870	33.1±2.3	2.8±0.8	-	13.4±2.2	-	1.2±0.8	47.1±3.0
SE2786	32.1±1.8	1.5±0.6	4.7±1.4	11.5±1.6	6.5±2.2	-	43.3±1.9
Amphibolites							
SE2843	22.9±1.9	6.0±0.9	10.3±2.1	7.8±2.0	9.8±4.7	-	42.6±3.0
SE2850	17.6±1.6	4.1±0.7	9.9±1.6	11.5±1.7	15.3±2.8	-	41.6±2.7
SE2816	21.4±1.6	4.7±0.7	11.1±1.7	10.7±1.7	12.3±2.7	0.8±0.6	39.0±2.3

Calciphyres consist from calcite or dolomite with an admixture of silicate materials. The amount of this impurity is small and amounts to 3-4% (see Table 1).

Fig. 1c shows the spectrum of a typical sample of gneiss. The spectrum shows no carbon peak at 4.44 MeV. There is also a contribution of calcium. It corresponds to the elemental composition of gneisses, which makes a major contribution of SiO_2 silica and alumina Al_2O_3. The results of determination of the mass fractions of the elements listed in Table 1 are fully consistent with this pattern.

Fig. 1d shows the spectrum of a sample of amphibolite. There is no line of carbon visible, at the same time a strong peak from the silicon and the peaks of aluminum and magnesium are present. This is consistent with

the elemental composition of amphibolite, which consists mainly of silicates and aluminosilicates, magnesium, iron and calcium. Data for the elemental composition of some samples of amphibolite according to the results obtained using DVIN-1 detector are shown in Table 1.

The spectra shown in Fig. 1, demonstrate the possibility of TNM to determine the key rock-forming elements - C, O, Si, Mg, Al, Fe, Ca.

Earlier, in works [4-5] the use of TNM was limited to determination of concentrations of carbon, nitrogen and oxygen elements only. Among these elements the largest cross section for excitation by neutrons 14.1 MeV is the line of carbon at 4439 keV for which the excitation cross section is 210 mb. For comparison, the cross section of 1779 keV line of silicon is 400 mb, and 1369 keV line of magnesium has a cross-section of 425 mb. Individual lines of iron, calcium and aluminum also have significant cross sections of excitation, which makes determination of their concentrations possible as well.

4. CONCLUSIONS

The main result of the tests is that the method of tagged neutrons correctly reproduces the main characteristics of the studied species. The measurement with the TNM correctly reproduced the elemental composition of calcite, dolomite and marble calciphyres (marbles with a mixture of silicate minerals), in calcite marbles it observed minor content of Mg and Si, in dolomitic marbles it showed significantly reduced amount of Ca and significant concentrations of Mg, in calciphyres it found increased content of Si. For the amphibolites the Si content was estimated to be around 18-20%, increased concentrations of Fe, Mg, Ca were recorded, which is characteristic of the basic rocks. In gneiss the silicon content is higher than in amphibolite (25-30% Si) with very low concentrations of Ca and Mg. TNM results correctly showed that there is no carbon presence in both groups of silicate rocks.

Thus, the results of measurements in the field have shown that the TNM based detectors can be successfully used for determination of Si, Al, Fe, Mg, Ca, C concentrations in rocks. However, it should be emphasized that the method is not an alternative to the traditional methods of analysis, and can be effectively used, if necessary, as an express determination of the composition of rocks. The use of TNM detectors in the field allows to get the result after 15-20 minutes of exposure. The rapid analysis of the composition of rocks is most demanded during determining of the places of mining (quarries, mines, tunnels) at the existing and/or explored mineral deposits.

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