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Neutron Resonance Capture Analysis (NRCA) is presently being developed at the Frank Laboratory of Neutron Physics (FLNP) to determine the elemental composition of samples. The NRCA is a non-destructive method that allows measuring objects' bulk composition. The procedure is based on detecting neutron resonances in radiative capture and the measurement of the yield of reaction products in these resonances. The experiments are carried out at the Intense REsonance Neutron source (IREN). In this study, we applied the NRCA to investigate an archaeological object provided by the Museum and Exhibition Complex (MVK) "Volokolamsk Kremlin". The object was a women's Old Believer cross (second half of the 17th century) found in the Moscow region, Volokolamsk district, the village of Chubarovo.

Keywords: neutron resonance capture analysis, non-destructive neutron analysis, time-of-flight (TOF) technique.

Introduction

The neutron resonance capture analysis allows determining samples elemental and isotopic composition of samples without any destruction and preparation. The use of this method results in their negligible residual activation. All this makes it promising for the study of artefacts. The method is applied for various studies at different institutes and sources, such as the GELINA pulsed neutron source of the Institute of Reference Materials and Measurements of the Joint Research Center (Gel, Belgium) [1], ISIS pulsed neutron and muon source in the United Kingdom [2] and the J-PARC pulsed neutron source in Japan [3]. At present, the NRCA is also used at the Frank Laboratory of Neutron Physics [4-6]. The method is based on using of a pulsed neutron source and time-of-flight (TOF) technique. The investigations are carried out at the intense resonance neutron source IREN [7, 8]. A multi-section liquid scintillation detector (210l) was developed at FLNP JINR and is used for prompt gamma-ray detection [9]. To date, the low-lying resonance parameters have been determined for almost all stable nuclei [10, 11].In addition, the set of resonance energies and parameters do not coincide at all for any pair of isotopes. The energy positions of resonances give information about an object's isotopic and elemental composition. In addition, one can calculate the number of nuclei of an element or isotope using the area under the resonances.

In this paper, the application of the NRCA to determine the elemental composition of an object is presented. This analysis was carried out for an artefact handed over by the Museum and Exhibition Complex (MVK) "Volokolamsk Kremlin" in the framework of the concluded cooperation agreement. Twenty-eight fragments of pottery and 25 metal finds were transferred for research. The investigations of artefacts by various methods and scientific analysis will make it possible to identify the centers of handicraft production; clarify the dating of finds and technology of their manufacture.One of the artefacts was the women's Old Believer cross (Figure 1). The place of finding it is the Moscow region, Volokolamsk district, the village of Chubarovo.

Figure 1. The women's Old Believer cross (second half of the 17th century) from the Moscow region, Volokolamsk district, village of Chubarovo.

Experiment

The sample was irradiated with neutrons at the resonance neutron source IREN to obtain time-of-flight spectra of (n, γ) reactions. The main part of the IREN facility is the linear electron accelerator LUE-200 with a non-multiplying neutron-producing target of Wf(90 %)+Ni+Fe -alloy [7, 8]. The parameters of the facility were as follows: average electron energy $\sim$ 60 MeV, peak current $\sim$ 1.5 A, electron pulse width $\sim$ 100 ns, and repetition rate of 25 Hz. The total neutron yield was about 3×10^{11} sec^{-1}. The measurements were carried out at a 58.6-m flight path of IREN beamline 3. A large liquid scintillation detector was used for detection of γ-rays [9]. The sample was placed inside the detector. To obtain time-of-flight spectra, an 8 independent-input multistop time-to-digital converter (TDC) developed at FLNP JINR was used. The START signal is generated by the synchronizer of the IREN facility, the STOP signals come from the detectors. During measurements, the power of the neutron source was constantly monitored. An SNM-17 neutron counter was used as a monitor. The generated signal from the counter was also fed to one of the detector inputs of the TDC. Further, during processing, the sum of the monitor counts practically over the entire time spectrum was used. The measurements of the sample lasted about 38 h. The resonance energies were determined by the following formula:

$$E = \frac{5227L^2}{t^2} \tag{1}$$

where t - time of flight (µs), L - flight path length (m), E - neutron kinetic energy (eV). The resonances of copper, silver, and zinc were identified in the time-of-flight spectrum (Figure 2) based on the values of resonance energies in [10].
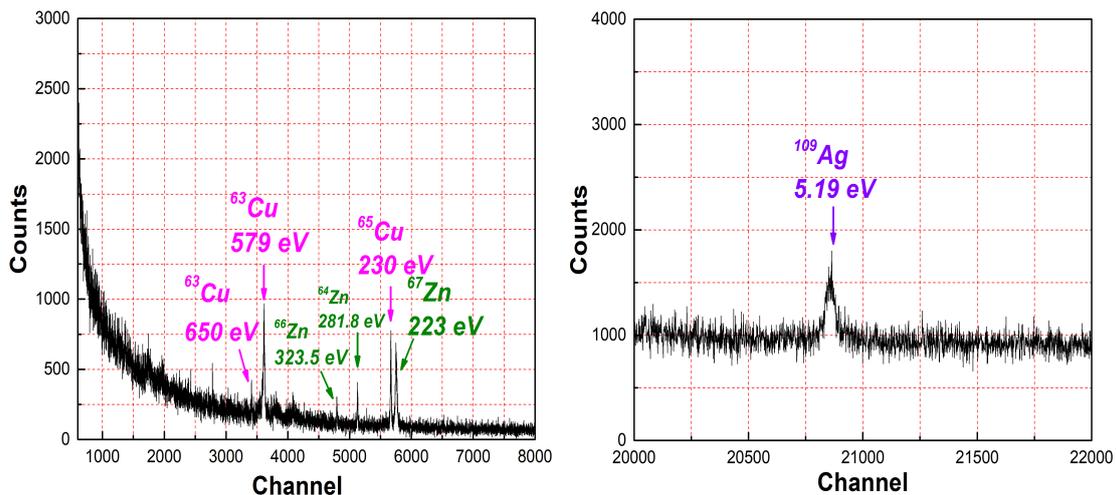

Figure 2. Part of time-of-flight spectrum of (n, γ) reactions obtained in measurements on the Old Believer cross material. The time channel width from 0 to 20000 channels is 0.05 µs, from 20000 to 22000 is 1 µs.

The measurements with standard samples of copper, silver and zinc were made in addition to the measurement with the investigated sample (Figure 3, 4, 5).
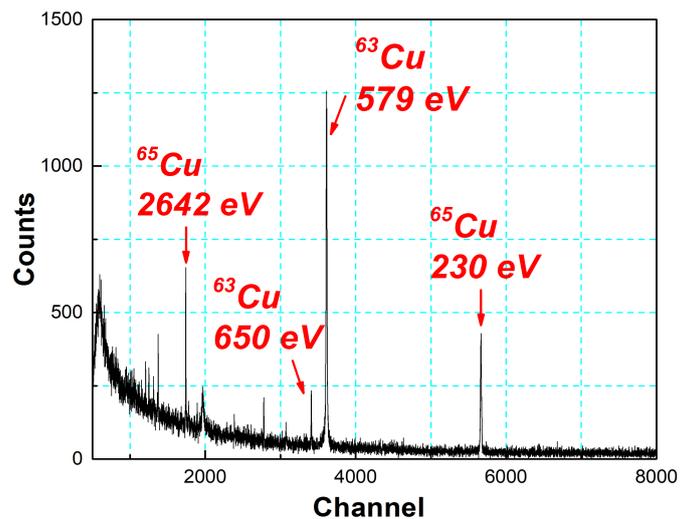
Figure 3. Part of time-of-flight spectrum of (n, γ) reactions of copper standard sample. The time channel width is 0.05 μs.
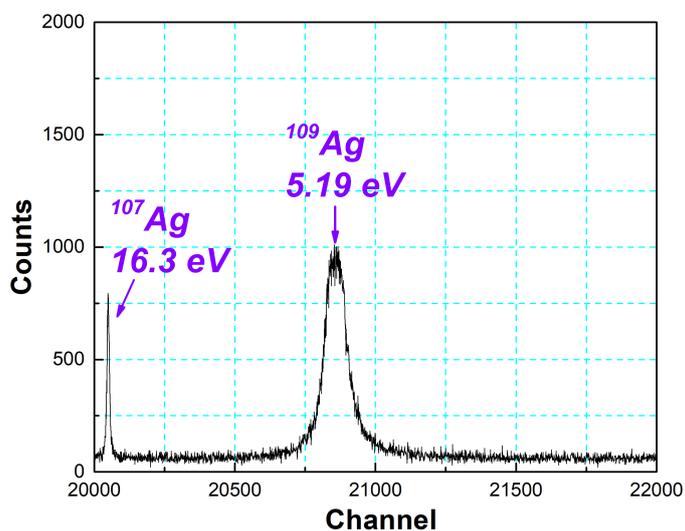

Figure 4. Part of time-of-flight spectrum of (n, γ) reactions of silver standard sample. The time channel width is 1 μs.
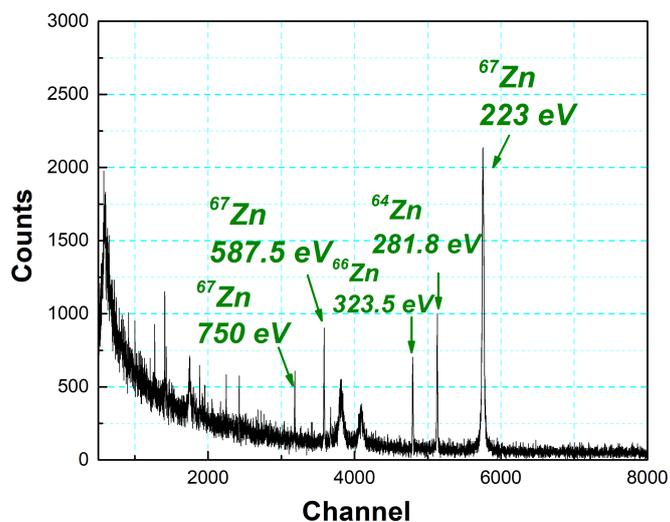

Figure 5. Part of time-of-flight spectrum of (n, γ) reactions of zinc standard sample. The time channel width is 0.05 μs.

Data analysis and results

The number of element nuclei in the sample was determined by measuring the yield of gamma-rays in resonances. Two resonances of zinc, one resonance of copper, and one resonance of silver were selected during the experimental data analysis. During the passage through the object, the neutron flux will decrease in resonances due to capture and scattering [12]. This is known as the self-shielding and multiple-scattering effects for which the experimental number of counts should be corrected. In our case, $n\sigma(E_0) \lesssim 1$ (thin sample), here n is the number of isotope nuclei per unit area and $\sigma(E)$ is the total cross-section at this energy, we consider that the multiple-scattering effect is negligible. The self-shielding can be taken into account by using the Doppler-broadened resonance function and the following equations, which were described in [6]. The sum of detector counts in a resonance is:

$$\sum N = f(E_0)St\varepsilon_\gamma \frac{\Gamma_\gamma}{\Gamma}A \qquad (2)$$

where, $f(E_0)$ is the neutron flux density at the resonance energy E_0, S is the sample area, t is the measurement time, ε_γ is the detection efficiency of the detector for radiative capture, $\Gamma\gamma$, Γ are the radiative and total resonance widths.

$$A = \int_{E1}^{E2} [1 - T(E)]dE \qquad (3)$$

is the resonance area on the transmission curve, where $E1$, $E2$ are the initial and final values of the energy range near the resonance.

$$T(E) = e^{-n\sigma(E)} \qquad (4)$$

is the energy dependence of the neutron transmission through the sample; $\sigma(E)$ is the total cross section at this energy with Doppler broadening, n is the number of isotope nuclei per unit area. The value of A was determined for the sample under study from experimental data using the following formula:

$$A_x = \frac{\sum N_x M_s S_s}{\sum N_s M_x S_x} A_s \qquad (5)$$

where, $\sum N_x, \sum N_s$ – counts under the resonance peak of the investigated and standard samples, S_x, S_s – the area of the investigated and standard samples. M_x, M_s – the number of monitor counts during the measurement of the investigated and standard samples. We used a program written according to the algorithm given in [13] to calculate the values of A_s (resonance area on the transmission curve of the standard sample) and n_x (the number of isotope nuclei per unit area of the investigated sample). This procedure is schematically shown in (Figure 6). The As value was calculated using the known resonance parameters and n_s parameters for the standard sample (step 1 in Figure 6). The n_x value was determined from the A_x value of the investigated sample (step 2 in Figure 6). The analysis results are presented in Table 1.
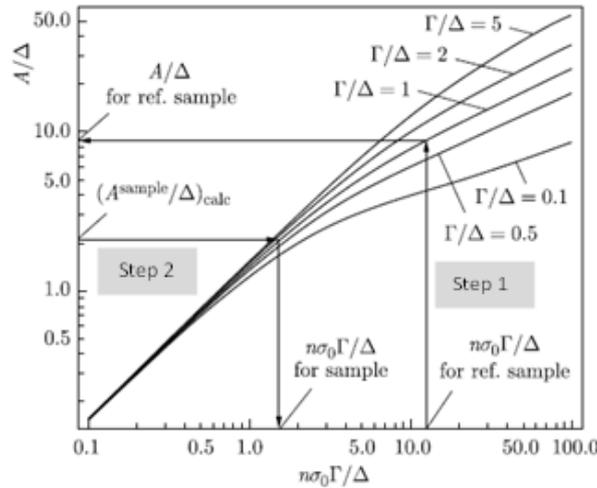
Figure 6. Dependence of value A on the number of nuclei and resonance parameters taking into account $\triangle$ (Doppler effect) [13].

Table 1.
The results of measurements of the women's Old Believer cross by the NRCA method.

	Element	Mass, g	Weight, %
1	Cu	18.3 ± 1.0	58.3 ± 3.2
2	Zn	8.542 ± 0.062	27.21 ± 0.19
3	Ag	0.02712 ± 0.00057	0.0863 ± 0.0018

At present, the NRCA has limitations related to both the neutron flux intensity at the IREN facility, which is not yet operating within its design parameters, and the matrix of elements in samples under study. In addition, this method has low sensitivity to the lighter elements than iron and to those whose atomic mass is close to magic mass numbers (bismuth, lead, etc.). Therefore, it is planned to carry out additional X-ray fluorescence (XRF) measurements.

Conclusion

This paper presents the results of the NRCA study of the women's Old Believer cross found in the Moscow region, Volokolamsk district, village of Chubarovo. The weight of the artefact is 31.4 g. According to the results of the analysis, the sum of the masses of all determining elements agrees with the mass of the cross within the margin of error. Furthermore, additional measurements will be performed to improve the result. The elemental composition may help identify the place of manufacture and the object's origin. Most likely, it should be compared to both the local and the Pomor casting (Vygovsky Monastery), casting of the Transfiguration Monastery in Moscow, casting of Old Believers of Vychegda.

Acknowledgments

References

[1] H. Postma and P. Schillebeeckx, Neutron Methods for Archaeology and Cultural Heritage (Springer, Cham, 2017) 235-283. [*CrossRef*]

[2] G. Gorini, Ancient Charm Collaboration et al., Il Nuovo Cimento C **30** (2007) 47-58. [*CrossRef*]

[3] T. Kai et al., Physics Procedia **88** (2017) 306-313. [*CrossRef*]

[4] N.V. Bazhazhina et al., Physics of Particles and Nuclei Letters **12** (2015) 578-583. [*CrossRef*]

[5] D.N. Grozdanov et al., Physics of Particles and Nuclei Letters **15** (2018) 537-540. [*CrossRef*]

[6] P.V. Sedyshev et al., Physics of Particles and Nuclei Letters **17** (2020) 389-400. [*CrossRef*]

[7] O.V. Belikov et al., Journal of Physics: Conference Series **205** (2010) 012053. [*CrossRef*]

[8] E. Sriyono et al., Journal of Physics: Conference Series **1175** (2019) 012024. [*CrossRef*]

[9] H. Maletsky et al., Detector of neutrons and gamma rays for work in the field of neutron spectroscopy, Communication of JINR 13-6609. [*Google Scholar*]

[10] S.F. Mughabghab, Neutron Cross Sections: Neutron Resonance Parameters and Thermal Cross Sections (Academic Press, New York, 1984). [*Google Scholar*]

[11] S.I. Sukhoruchkin, Z.N. Soroko and V.V. Deriglazov, Low Energy Neutron Physics: Vol. I/16B of Landolt-Bornstein (Springer Verlag, Berlin, 1998). [*Google Scholar*]

[12] H. Postma et al., AIP Conference Proceedings **769** (2005) 1393. [*CrossRef*]

[13] V.N. Efimov and I.I. Shelontsev, Calculation for graphs of determining the parameters of neutron resonances by the transmission method, Communications of the JINR P-641 (1961) 1-19. [*Google Scholar*]