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CARBON DETECTION USING AN HPGE DETECTOR

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ABSTRACT

Fast neutron activation analysis has been applied to bulk carbon detection. DT-neutron generator emitting 14.1 MeV neutrons was used as a neutron source. Prompt 4.44 MeV gammas from the inelastic neutron scattering on carbon were detected using a 30% HPGe semiconductor gamma spectrometer shielded from the primary neutron beam. The sensitivity of the experimental setup is discussed.

Keywords: Activation analysis; Fast neutrons; Gamma spectrometry.

1 INTRODUCTION

Fast neutron activation analysis for carbon content determination using neutron generators is a well-known technique and is used for about two decades [1]. ^{12}C nuclei are excited in inelastic fast neutron scattering (neutron threshold energy E_{th} = 4.8 MeV) to the excited 2^+ state at an energy of 4.44 MeV, decaying by gamma emission to the ^{12}C ground state. The full width at half maximum (FWHM) of the 2^+ state Γ = 10.8(3) meV [2]. The half-life T of the 2^+ state can be calculated from $T = \ln 2\ h/(2\pi\Gamma) = 4.22(12).\ 10^{-14}$ s (h is the Planck constant). The half-life is shorter than the stopping time of the recoiled excited ^{12}C nuclei. Therefore, they decay in flight resulting in the Doppler broadening of the 4.44 MeV gamma peak [3]. The maximum initial kinetic energy of the excited ^{12}C nuclei corresponding to backscattered neutrons can be calculated from the conservation laws of energy and linear momentum:

$$T_n = E_C + T_n' + T_C' \quad \text{and} \quad p_n = -p_n' + p_C' , \qquad (1)$$

where T_n [MeV] is the initial neutron kinetic energy, E_C = 4.44 MeV is the ^{12}C excitation energy, T_n' [MeV] is the backscattered neutron kinetic energy, T_C' [MeV] is the recoiled ^{12}C kinetic energy, p_n [MeV/c] is the initial neutron linear momentum, p_n' [MeV/c] is the backscattered neutron linear momentum, and p_C' [MeV/c] is the recoiled ^{12}C linear momentum.

The maximum Doppler shifted energy E_{max} of the emitted gammas can be obtained from the formula:

$$E_{max} = E_C \frac{1}{1-\beta} , \qquad \beta = \frac{p_C' c}{m_C c^2} , \qquad (2)$$

where $m_C c^2$ [MeV] is the carbon rest energy and c [m/s] is the speed of light in vacuum. For T_n = 14.10 MeV we get T_n' = 6.36 MeV from (1) and E_{max} = 4.55 MeV from (2). From a similar consideration we get for the minimum Doppler shifted energy E_{min} = 4.33 MeV.

The measured 4.44 MeV gamma intensity is proportional to the number of ^{12}C nuclei that represent 98.93(8) % of natural carbon [2], thus enabling us to determine the carbon content of the investigated sample. The gamma intensity of the 4.44 MeV carbon line is usually measured by scintillation gamma spectrometers (NaI(Tl) or BGO). In the present paper, we report results obtained with an HPGe spectrometer that provides a better energy resolution (typically 30 times better than a NaI(Tl) spectrometer [4]).

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2 EXPERIMENTAL SETUP

14.1 MeV neutrons were generated in the deuterium-tritium fusion (DT) in the MP320 neutron generator (NG) at the Laboratory for Neutron Activation Analysis and Gamma Spectrometry at VSB – Technical University of Ostrava, Czech Republic. Operating parameters of the NG are the accelerating voltage U = 80 kV and the beam current I = 60 μA in the continuous regime. The neutron emission angle represents approx. 90° with respect to the direction of the accelerated deuterons impinging on the NG target. In this regime, the NG emits approx. $Y = 10^8$ neutrons/s into the 4π solid angle (nominal neutron emission declared by the manufacturer [5]). The HPGe spectrometer (CG3018, Canberra) was positioned at a distance of r = 50 cm from the NG target area. The neutron flux ϕ at this position can be estimated from the formula $\phi = Y/(4\pi r^2) = 3180$ n/cm^2/s.

To reduce the background, detector dead time, and Ge crystal damage, it is necessary to shield the detector from the primary neutron beam. It is important that our detector is the n-type one that is more neutron damage resistant than the p-type [4]. Since we want to detect carbon, it is beneficial to use shielding materials with lower carbon content. We chose steel with a carbon content smaller than 0.5%, copper and lead. The thicknesses of steel, copper and lead represent 10 cm each. The primary neutron beam attenuation effect of the proposed shielding was checked by the copper foil activation method [6,7].

In the copper foil activation measurements, copper foils of a diameter of 2.5 cm and a thickness of 0.78 mm were irradiated by both unshielded and shielded neutron beams, and the annihilation photons accompanying decay of ^{62}Cu produced in the fast neutron reaction ^{63}Cu(n,2n) were measured using a 3” x 3” NaI(Tl) gamma spectrometer. The ratio of the number of detected annihilation photons for the shielded and the unshielded foil corrected for irradiation, transport and measurement times gives the shielding attenuation factor f. In our case f = 22 (5).

The non-destructive method of carbon content determination was tested for a graphite sample placed in an aluminium container (diameter 8 cm, height 7 cm, wall thickness 1.5 mm) and positioned on the endcap of the HPGe spectrometer. For a schematic view of the experimental setup, see Fig. 1.

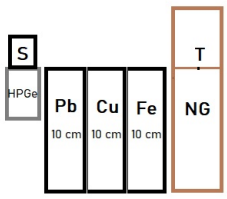

Figure 1. *A schematic view of the experimental setup with the neutron generator (NG), its target (T) as a center of the neutron emission zone, three shielding materials, the HPGe spectrometer and the sample container*

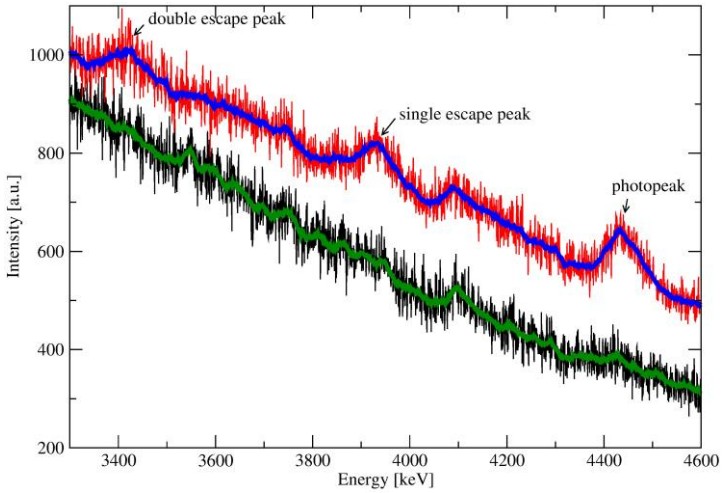

Figure 2. *Spectra of the irradiated graphite sample (raw spectrum: red, spectrum averaged over 30 channels: blue) and background (raw spectrum: black, spectrum averaged over 30 channels: green); spectra are artificially shifted to make their differences visible*

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3 RESULTS

The aluminium container filled with graphite (mass 172.41 (3) g) was irradiated by 14.1 MeV neutrons for 2400 s. The gamma spectrum measured by the HPGe spectrometer was analyzed using the software SpectraLine 1.6 (Baltic Scientific Instruments). The aluminium container was then left empty, and a background spectrum was accumulated under the same conditions for a longer time of 3210 s to improve the statistics. In Fig. 2, both background and graphite spectra are shown. It can be clearly seen that the carbon photopeak at 4.44 MeV is Doppler broadened as expected. Its width equals 0.09 MeV in agreement with the expected maximum Doppler broadening of about 0.2 MeV, which is much larger than full widths at half maximum (FWHM) in this energy region (about 25 keV). To reduce the fluctuations in raw spectra, it is worth averaging values in each channel over 30 neighbouring channels (see Fig. 2). There are also two other carbon peaks visible in the irradiated spectrum, namely single escape and double escape peaks at 3.93 MeV and 3.42 MeV, respectively.

The sensitivity of the carbon content determination can be found from the total background corrected area of the 4.44 MeV carbon peak. The measurement results in a value of 10760 (220) pulses for the carbon mass of 172.41 (3) g, i.e., 62.4 (13) pulses/g.

The minimum determination limit *MDL* can be calculated from the formula [8]:

$$MDL = \frac{m}{NC} 3.3\sqrt{B} \qquad (3)$$

where NC = 10760 and B = 132280 are the background-subtracted net counts for the graphite sample of mass m = 172.41 and the background counts under the 4.44 MeV peak, respectively. In our experimental setup, we get *MDL* = 20 g.

To detect the carbon content in an unknown sample, it is necessary to fill the aluminium container with the same volume of material and irradiate it in the same geometry as the graphite sample. To check the stability of the neutron flux, it is recommended to simultaneously irradiate the copper foils and measure the number of annihilation photons using the 3” x 3” NaI(Tl) gamma spectrometer.

We can define flux correction factors, C_{fe} and C_{fx}, for the graphite etalon and the unknown sample, respectively:

$$C_{fe} = \frac{S_e}{[1-exp(-ln2/T_f \times t_{ie})]exp(-ln2/T_f \times t_{tfe})[1-exp(-ln2/T_f \times t_{mfe})]}, \qquad (4)$$

$$C_{fx} = \frac{S_x}{[1-exp(-ln2/T_f \times t_{ix})]exp(-ln2/T_f \times t_{tfx})[1-exp(-ln2/T_f \times t_{mfx})]}, \qquad (5)$$

where S_e and S_x are numbers of the photons detected in the annihilation photopeak for the copper foils used in the irradiation of the graphite etalon and the unknown sample, respectively; t_{ie}, t_{tfe}, and t_{mfe} are the irradiation, transport, and measurement times for the foil used in the irradiation of the graphite etalon; t_{ix}, t_{tfx}, and t_{mfx} are the irradiation, transport, and measurement times for the foil used in the irradiation of the unknown sample, and T_f = 9.67 min is the ^{62}Cu half-life.

The carbon mass in the unknown sample can be found from:

$$m_{Cx} = m_{Ce} \frac{S_x t_{ie} C_{fe}}{S_e t_{ix} C_{fx}}, \qquad (6)$$

where m_{Cx} is the carbon mass in the unknown sample, m_{Ce} is the carbon mass in the graphite etalon, S_x is the background corrected number of photons detected in the 4.44 MeV carbon peak during the irradiation of the unknown sample, and S_e is the same quantity for the graphite etalon.

4 CONCLUSIONS

Inelastic fast neutron scattering of 14.1 MeV neutrons emitted by the MP320 neutron generator was investigated. The resulting Doppler broadened 4.44 MeV gamma peak was detected by an HPGe semiconductor gamma spectrometer. The detection limit (minimum determination limit) for carbon was found to be 20 g for the neutron flux at the irradiation site of 3180 n/cm^2/s. The method can be used to determine the carbon content in different samples (explosives, drugs, other organic materials, or organic minerals).

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