Determination of the boron content in polyethylene samples using the reactor Orphée

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Abstract

The boron content of two unknown types of polyethylene has been determined relative to a known reference type. Samples of polyethylene, including a known boron-less one, were irradiated with thermal neutrons at the reactor Orphée at Saclay in France. Prompt gamma rays were measured with a $CeBr_3$ detector and the intensity of the 478 keV line from the $^{10}B(n,\alpha_1\gamma)^7Li^*$ reaction was extracted for each sample.

Introduction

Borated polyethylene is widely used as neutron shielding material. It is commercially available, usually with quantities of natural boron up to several percent. The isotopic composition of natural boron is 19.8% of ^{10}B and 80.2% of ^{11}B. Neutrons are moderated by elastic scattering from the hydrogen nuclei present in polyethylene and are subsequently absorbed by ^{10}B through the $^{10}B(n,\alpha)$ reaction. With a probability of 0.937, the ^7Li nucleus is left in its first excited state by the $^{10}B(n,\alpha_1\gamma)^7Li^*$ reaction, with a cross section of about 3600 b at 25.3 meV. The excited $^7Li^*$ and decays to its ground state by emitting a 478 keV gamma ray. The other possibility, with a probability of 0.063, is that the ^7Li nucleus is left in its ground state via the $^{10}B(n,\alpha_0)$ reaction, with a cross section of about 240 b at 25.3 meV). No gamma radiation is emitted in this case.

The neutron time-of-flight facility n_TOF at CERN uses neutron shielding at various places along the two neutron beam lines. Sizeable quantities of two types of

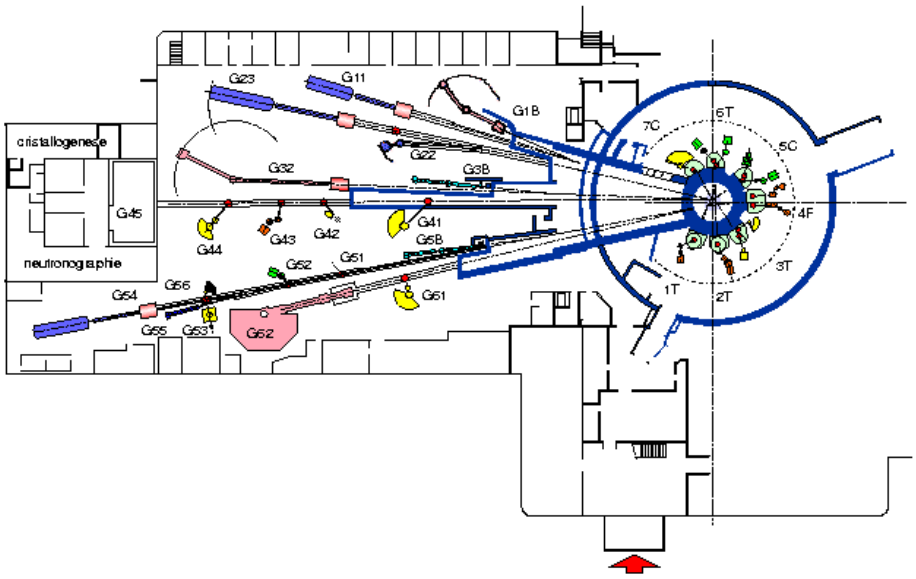

Figure 1: The layout of the neutron beam lines of the reactor Orphée [1, 4]. Experimental area G4-3 was used for the boron determination experiment.

polyethylene were localized in a storage area, but with unknown characteristics, in particular the possible boron content.

In order to determine the boron content of the two unknown types of polyethylene relative to a known reference sample of borated polyethylene, an experiment was carried at the reactor Orphée [3] of the Laboratoire Léon Brillouin in Saclay, France. The goal was to detect the intensity of the 478 keV gamma ray emitted after neutron absorption by ^{10}B present in the polyethylene, relative to a known standard.

Samples of polyethylene, including a reference sample with the same dimensions, were placed in the same position in a subthermal neutron beam. The emitted prompt gamma rays during irradiation were measured with a portable $CeBr_3$ detector. The measured intensity of the 478 keV gamma ray was then used to determine the boron content relative to the reference sample, similar as for example in ref. [2].

1 The measurement setup

The reactor Orphée [1, 3] is operated by the Laboratoire Léon Brillouin, a research institute jointly governed by the Commissariat à l'Energie Atomique et aux Energies Alternatives (CEA) and the Centre National de la Recherche Scientifique (CNRS). The reactor is a 14 MW light-water moderated reactor with a compact core of highly enriched ^{235}U providing a flux of up to 3×10^{14} ther-

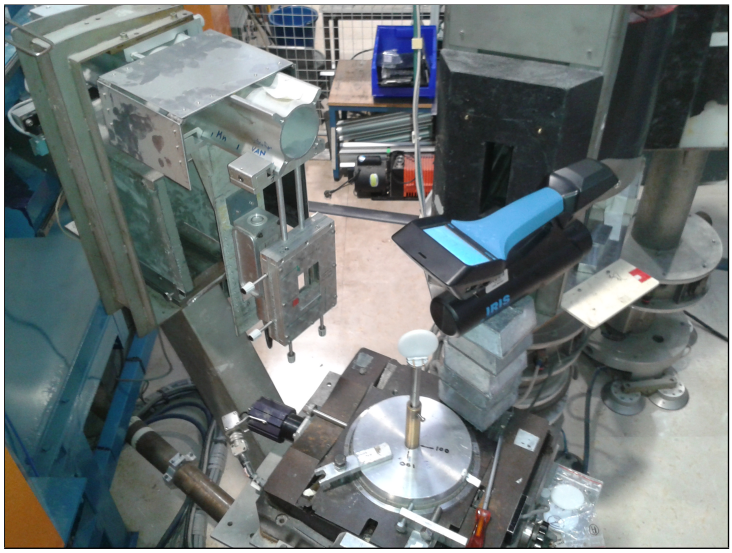

Figure 2: One of the tested setups for the measurement. A white polyethylene sample is positioned in the neutron beam, shape by the final borated aluminum collimator. For the final measurement, the distance of the standalone Iris $CeBr_3$ detector system was with respect to the sample was increased.

mal neutrons$\cdot$cm$^{-2}\cdot$s^{-1}, in the surrounding heavy water reflector tank. Neutron beams from 9 horizontal neutron beam lines are pointing tangentially towards the core, and from 6 curved neutron guides, are routinely used to study a large variety of solid state physics cases by neutron scattering for wavelengths ranging from 0.7 to 15 Å. The layout of the beam lines [1, 3] is shown in figure 1. Even if neutron-induced gamma-ray spectroscopy is not part of the usual activities at Orphée, it was possible to use one of the scattering setups which could be used with minor modifications.

We have used experimental area G4-3 with the neutron beam deviated with a monochromator from neutron guide G4, resulting in a beam of monochromatic neutrons with a wavelength of 0.236 nm (14.7 meV) and a neutron flux of about 1.5×10^6 n$\cdot$cm$^{-2}\cdot$s^{-1} over an area of approximately 3×3 cm^2. The neutron beam was shaped with the help of collimators made of boral, aluminium with a high concentration of boron carbide. The polyethylene samples were placed in the neutron beam using an aluminum sample holder, allowing to reproduce the geometrical position of each sample.

Different configurations were tried changing the positions of the $CeBr_3$ detector in order to minimize the activation of the crystal due to sample-scattered neutrons and to optimize the count-rate from the sample-emitted gamma rays in the detector. In figure 2 one of the tested setups is shown. In the setup used for the final measurements, the detector was placed further away from the sample.

2 Polyethylene samples

Several disc-shaped samples with a diameter of 60 mm and a thickness of 5 mm were produced for each of the 4 types of polyethylene: two types containing an unknown quantity of boron, labelled ”A” and ”B”, a reference sample ”R” containing a known quantity of boron, and a sample of standard polyethylene with zero boron content labelled ”Z”. The aspect of the reference sample R is white, softer than the other samples, and with visible, small dark particles in the material. The reference polyethylene sample was made from bulk material purchased from John Caunt Scientific Ltd., containing 5% of natural boron.

The unknown type A and zero boron type Z polyethylene both have a clear white color and feel very smooth. The type B has a black color and is smooth as well. For each sample type the mass was measured and the density calculated as reported in table 2.

Table 1: Properties of the different polyethylene samples, all of diameter 60 mm and thickness 5 mm. The uncertainty on the density is mainly due to the uncertainty of the thickness and is estimated as less than 2%.

label	color	mass (g)	density (g/cm^3)
R	white	14.55	1.03
A	white	13.79	0.98
B	black	13.42	0.95
Z	white	13.33	0.94

3 Gamma-ray detector and calibration

For the gamma-ray spectroscopy we have used a 2x2 inch $CeBr_3$ detector system Iris from Itech Instruments [5], which combines an on-board multi-channel analyzer (MCA), screen and high voltage supply into a portable device. The device was kindly put at our disposal for the measurements by the radioprotection unit of Orphée. Given the expected gamma-ray spectra, the resolution of the detector was sufficient for our purposes.

The MCA registered gamma-rays up to about 2.4 MeV into 1024 channels. In order to characterize the detector, it was brought in the vicinity of a strong ^{192}Ir source. This source has several gamma ray rays, including the triplet at 296.0, 308.5 and 316.5 keV, a peak at 468.1 keV, and the triplet at 588.6, 604.4 and 612.5 keV. These energies cover the full energy range of interest for the present measurement. In figure 3 the measured spectrum of this source is shown. We fitted the three regions with closely lying peaks separately, while for each region fixing the known intensities and assuming a constant efficiency over the multiplet. An

example of the fit of the first region and the resulting energy calibration is also shown in figure 3. This energy calibration was found to be in agreement with a linear calibration using the single 662 keV gamma ray of a ^{137}Cs source, which was used for the final measurements.

4 Measurement results

We measured the spectra of each of the 4 types of polyethylene, A, B, R and Z, as well as a spectrum with an empty sample position with beam and the ambient background without beam. Each spectrum was taken with an irradiation time of 180 seconds. The reactor power and hence the neutron flux was considered constant over the measurements. The resulting measured spectra are shown in figure 4.

We see that the 478 keV gamma ray is also present in the measurement with the sample without boron (Z), due to the use of boral in the beam collimation. The 478 keV is has the same magnitude for the samples B and Z, from which we can already conclude that sample B does not contain boron. From this figure can also be observed that the intensity of the 559 keV peak and several other gamma rays, reduces when the the boron content in the sample is higher. This may be explained by neutron-induced reactions elsewhere in the experimental area by neutrons from the beam that are not absorbed by the boron, but scattered from the sample due to the high hydrogen content in polyethylene. In the same way, the intensity of the 511 keV annihilation peak is lower when the boron contents is higher.

As an order of magnitude, the transmission for the 5% boron-loaded sample is about 0.3 due to boron only, where the main interaction occurs through the (n,α) reaction, and another transmission factor of 0.3 due to hydrogen, reacting mainly by (n,n) scattering, but also with a tiny part (1%) through (n,γ) creating gamma rays of 2223 MeV, visible in figure 4.

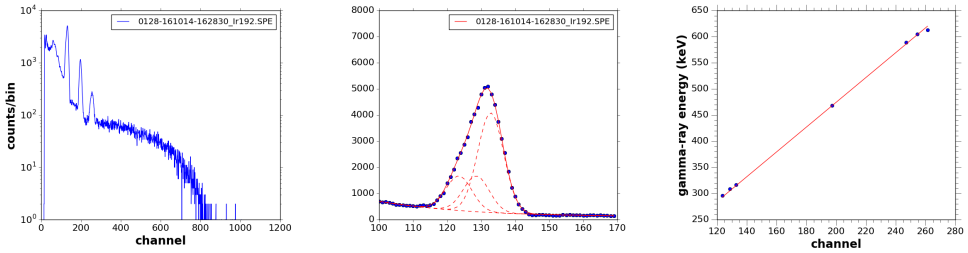

Figure 3: The measured spectrum of the ^{192}Ir source (left panel), a zoom on the first triplet together with the fitted Gaussians with a fixed ratio of intensities (middle panel), and the resulting channel to energy calibration (right panel).

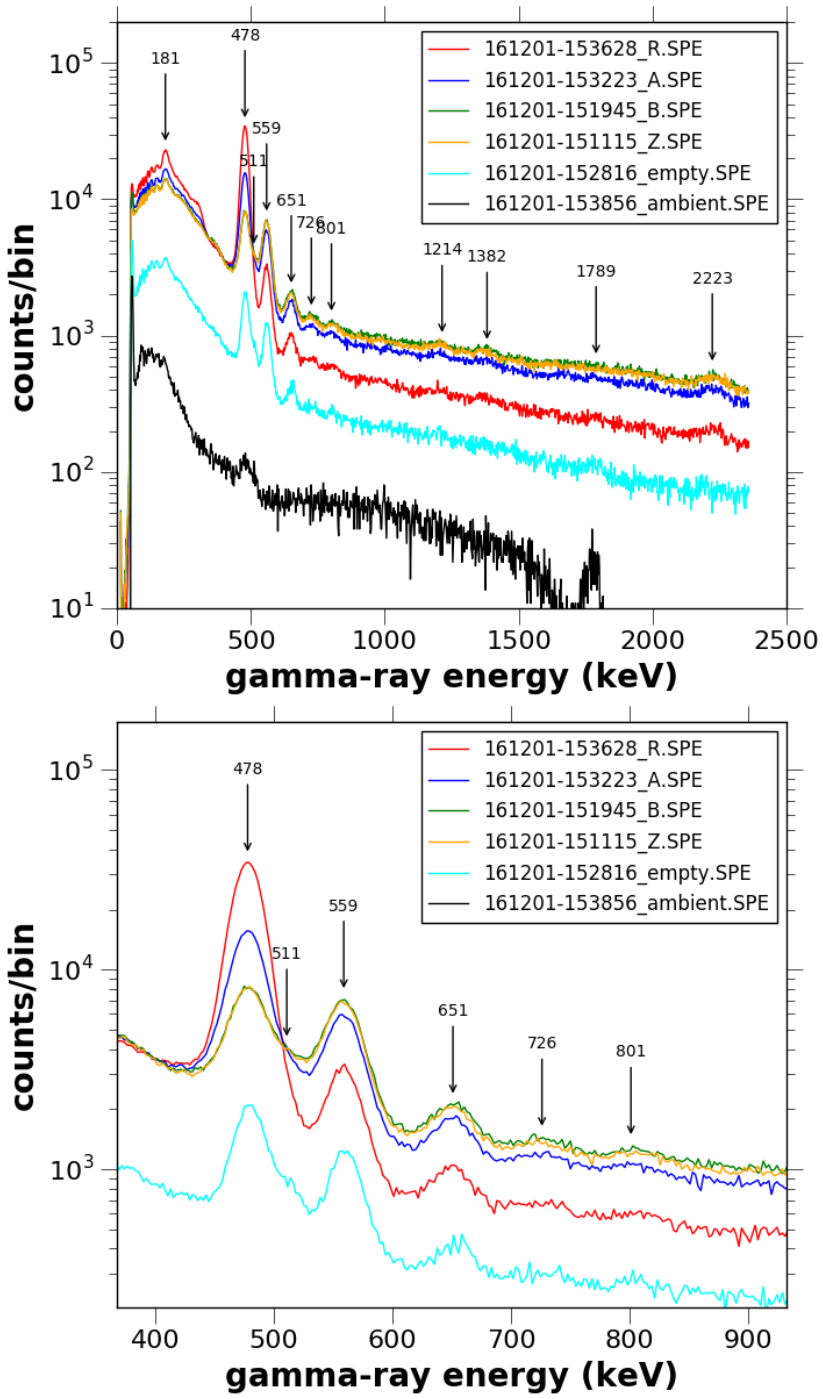

Figure 4: The measured spectra of all samples, corresponding to an irradiation time of 180 seconds, shown as the full energy region (upper panel) and a zoom in the region containing the 478 keV peak from boron (lower panel).

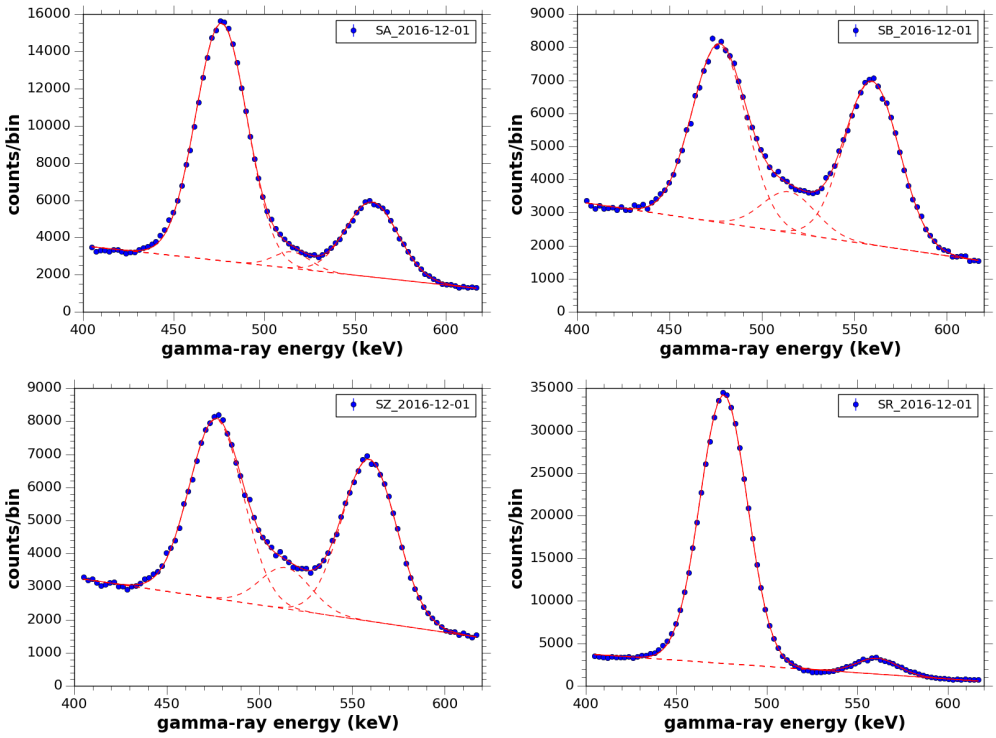

Figure 5: A zoom of the measured spectra in the region of the 478 keV peak together with a fit of three gamma-rays from which the peak areas are determined, for each of the four samples A, B, Z, and R.

From sample A the boron contents has to be determined relative to the reference sample R from the intensity of the 478 keV peak. This can be done either by fitting the subtracted spectra "A-Z" and "R-Z", or by fitting the intensities I_A, I_Z, and I_R of the 478 keV peak separately to obtain the ratio f as:

$$f = \frac{I_A - I_Z}{I_R - I_Z} \tag{1}$$

where the ratio f represents the fraction of boron relative to the known reference sample. In figure 5 the data and the fit with three Gaussian peak profiles are shown. From the peak areas the ratio f_A for sample A can be calculated resulting in

$$f_A = 0.305 \pm 0.010 \tag{2}$$

The quoted uncertainty is only due to the results of the fit, taking into account the correlation of the uncertainty on I_Z in f. We already concluded that $f_B = 0$.

Conclusion

From the present measurements we have determined the ratio f of boron content from two unknown samples relative to a known reference sample.

Table 2: The ratio f of boron in the unknown samples A and B, relative to the reference sample R. Assuming a content of 5% in the reference sample, the nominal boron content is also given.

sample	ratio f	nominal boron content (%)
A	0.305 ± 0.010	1.5
B	0.0	0

The present measurement was performed with a portable $CeBr_3$ detector. Assuming a boron content of 5% in the reference sample, we determined a boron concentration of 1.5% in sample A and 0% in sample B.

References

[1] `http://www-llb.cea.fr`
[2] T. Matsumoto and O. Aizawa, *Appl. Radiat. Isot.* **41** (1990) 897-903
[3] Ch. Alba-Simionesco, A. Menelle and J.-P. Visticot, *Neutron News* **22** (2011) 10-13
[4] P. Monceau and M. Alba, *Neutron News* **14** (2003) 10-12
[5] `http://itech-instruments.com`