Determination of the branching ratio for the $^{209}Bi(n,\gamma)^{210}Bi$ reaction from 500 eV to 20 keV

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Abstract

Energy differential neutron capture cross section measurements have been performed to determine the branching ratio for the $^{209}Bi(n,\gamma)$ reaction. The measurements were carried out at the time-of-flight facility GELINA of the IRMM in Geel (Belgium). The capture measurements were performed at a 12 m flight path using three High-Purity Germanium detectors. The experimental set-up was optimized to reduce the prompt background due to scattered neutrons. Several γ-ray spectra corresponding to the ^{209}Bi + n resonances up to 20 keV were deduced. The results of a preliminary data analysis are given in this paper.

KEYWORDS: TOF-measurements, neutron cross section measurements, bismuth, energy dependent γ-ray branching ratio, isomeric state, ADS

1. Introduction

Future high power spallation sources may consist of liquid lead-bismuth targets. Therefore, for the operation and safety assessment of Accelerator Driven Systems the $^{209}Bi(n,\gamma)^{210m}Bi$ and $^{209}Bi(n,\gamma)^{210g}Bi$ cross-sections are important nuclear data. These two reactions contribute significantly to the short- and long-term radiotoxicity of the target via the production of ^{210}Po ($T_{1/2}$ = 138.376 d) and ^{210m}Bi ($T_{1/2} = 3.04 \times 10^6$ y), both alpha-emitters. The reaction probabilities for the population of the ground state and the meta-stable state, and the corresponding branching ratio are crucial data for operational safety reasons and long-term disposal aspects.

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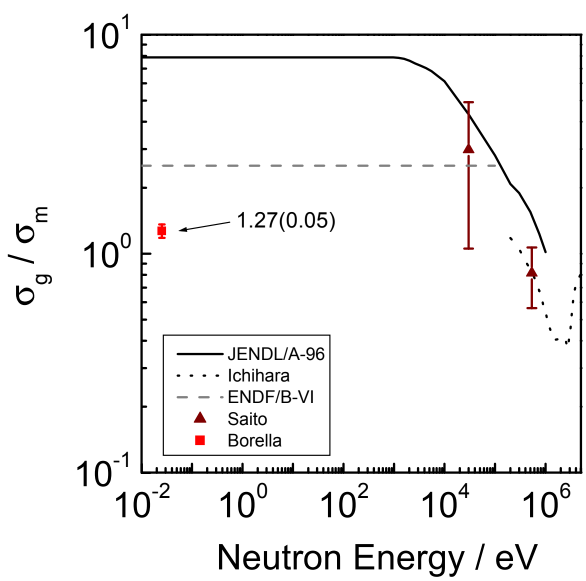
Existing data for the branching ratio are scarce. Recently the $^{209}Bi(n,\gamma)^{210m}Bi$ and $^{209}Bi(n,\gamma)^{210g}Bi$ cross sections for thermal neutrons were measured at the cold neutron beam PGAA-NIPS facility of the Budapest Neutron Centre and have been reported in Refs. [1,2]. The resulting total capture cross section and the cross sections to the ground state and the isomeric state, σ_{m+g} = 40.2 (1.2) mb, σ_g = 22.5 (1.4) and σ_m = 17.7 (0.7) mb, differ significantly from the recommended data in Refs. [3,4,5,6]. This measurement was preceded by an earlier measurement at the high flux reactor of the Institute Laue-Langevin [7].

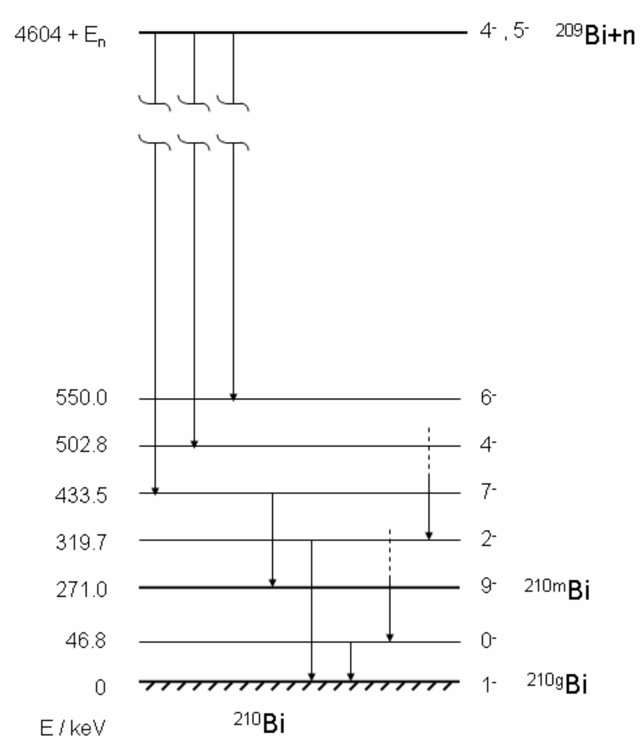
In Fig. 1 we compare these results with experimental data at 30 keV and 534 keV reported in Ref. [8] and with the evaluated data. This figure shows that the branching ratio deduced in Refs. [1,2] does not agree with the branching ratio deduced from a combination of the total capture cross section in ENDF-B/VI [3] and the cross section to the isomeric state in the activation file [5]. The results of Refs. [1,2] also disagree with the most recent evaluation performed by Ichihara and Shibata [6], based on optical model calculations adjusted to the experimental point at 534 keV [8].

Figure 1: The ratio of the cross sections for the $^{209}Bi(n,\gamma)^{210g}Bi$ and $^{209}Bi(n,\gamma)^{210m}Bi$ reactions as a function of neutron energy. The results reported in [1] are compared with the experimental data at 30 keV and 534 keV [8] and the evaluated data [3,5,6].



The existing experimental data indicate that the branching ratio is energy dependent. This hypothesis is enforced by the fact that a large contribution of the feeding of the low-energy levels is due to only few primary γ-ray transitions, which may be subject to fluctuations from one resonance to another. These transitions are shown in a simplified version of the decay scheme for the ^{209}Bi + n reaction in Fig. 2. The capture cross section to the ground state σ_g and to the isomeric state σ_m can be determined applying the method proposed by Coceva [9]. By applying this technique, σ_g and σ_m are obtained as the sum of the partial cross sections of γ–rays feeding these states. In order to study the energy dependence of the branching ratio, an experimental program has been set up to cover a wide neutron energy region with neutron time-of-flight measurements.

Figure 2: Simplified decay scheme for ^{209}Bi + n reaction.



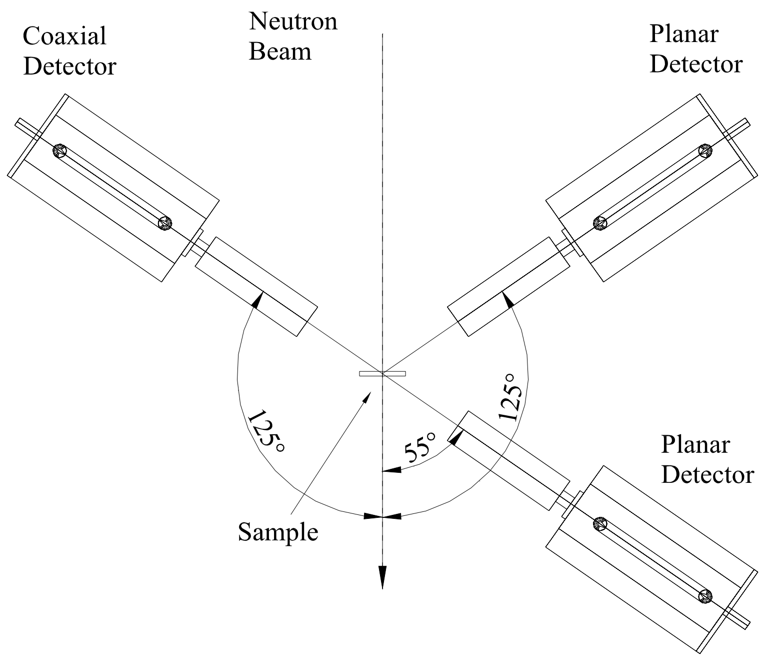
2. Experimental Conditions

Capture measurements were carried out at the neutron time-of-flight (TOF) spectrometer GELINA of the Institute for Reference Materials and Measurements (IRMM) at Geel in Belgium. The accelerator was operated at 800 Hz and 60 μA average electron current, providing electron pulses of 1 ns width with 100 MeV average electron energy. A detailed description of GELINA can be found in Ref. [10].

The measurements were carried out at a 12 m flight path of GELINA. The angle between the flight path and the direction of the electron beam was 72^{o}. The moderated neutron beam was collimated to about 80 mm in diameter at the sample position. A metallic bismuth disc sample of 81.0 mm diameter and 1.26 mm thickness (4.38×10^{-3} at/b) was positioned perpendicularly with respect to the neutron beam. To reduce the contribution of overlap neutrons a 4.16×10^{-3} at/b thick ^{10}B anti-overlap filter was used. A permanent 30 mm thick Pb filter was used to reduce the effect of the γ–flash. The choice of a relatively short flight path length assured a high neutron flux and a resolution sufficient to resolve the resonances up to 20 keV.

The γ-rays originating from the capture reaction in the sample were detected in three high-purity germanium detectors. A large volume coaxial detector was used to study the γ-ray spectra up to an energy of 8 MeV and two planar detectors were used to investigate the γ-ray spectrum in the low energy part. In Fig. 3 the position of the detectors with respect the direction of the neutron beam are given. The coaxial detector was placed at 125^o with respect to the neutron beam, while the two planar detectors were placed at 55^o and 125^o. This geometry was chosen to minimize systematic effects due to the anisotropy in the γ–ray emission. In order to optimize the experimental conditions, particular care was given to the reduction of the background by using an appropriate lead and boric acid shielding around the measurement area.

Figure 3: Top view of the experimental setup.

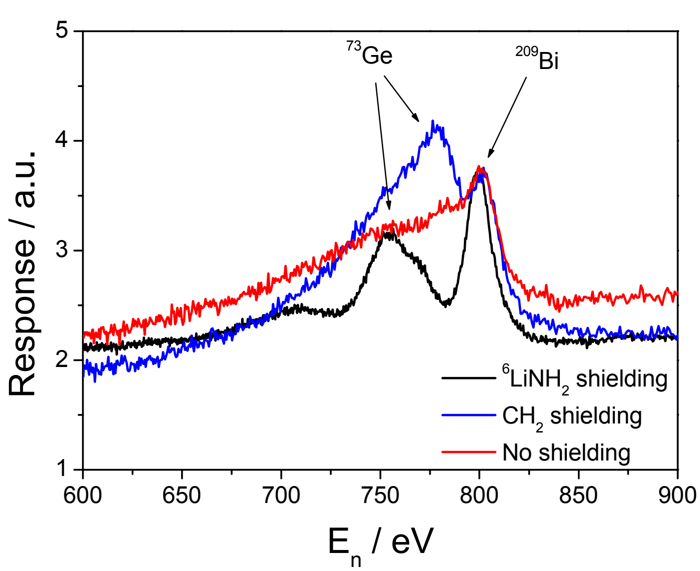


The energy calibration of the detectors was carried out using a ^{152}Eu source, the 2223 keV capture γ-ray of hydrogen, the 2614 keV γ-ray of ^{208}Tl and the 6130 keV γ-ray from the $^{13}C(\alpha,n)^{16}O^*$ reaction of a ^{238}Pu-^{13}C source. The efficiency of the detectors below 1.5 MeV was determined from the intensities of the γ-rays of a calibrated ^{152}Eu source. The efficiency up to 7.6 MeV was deduced by means of capture measurements on a ^{nat}Fe sample, using the γ-ray intensities from the 1.15 keV neutron resonance of $^{56}Fe(n,\gamma)$ reaction. To correct for the attenuation of the γ-rays in the bismuth sample, measurements with the ^{152}Eu source placed on both sides of the bismuth sample were carried out together with measurements on a Bi-Fe sample. In addition, a comparison of the response of the two planar detectors provides information about the effective γ-ray distribution in the sample.

3. Preliminary results

This kind of measurements is particularly difficult for two reasons mainly: the very low capture cross section of bismuth and the sensitivity of the germanium detectors to neutrons scattered from the sample. The reduction of the neutron sensitivity is particularly important when carrying out TOF capture measurements in nuclei where the neutron width is much higher than the capture width. In the case of ^{209}Bi the ratio Γ_n/Γ_γ is in the order of 10^3-10^4 for s-wave resonances which may induce a structured background due to the capture of scattered neutrons in Ge resonances. For example, neutron scattering in the s-wave resonance at 800 eV in ^{209}Bi induces a structure due to capture at the resonance at 750 eV in ^{73}Ge. In Fig. 4 the impact of neutrons scattered in the Bi-target on the total response of the coaxial detector is illustrated. Without any additional neutron shielding, it is almost impossible to separate the response of the 800 eV resonance of ^{209}Bi(n,γ) from the response due to capture of scattered neutrons in the Ge-detector. Fig. 4 also reveals that the background due to events from neutron scattering can be largely reduced by optimizing the neutron shielding around the detectors. To reduce this background component a 46 mm thick $^{6}LiNH_2$ cap together with a 2 mm thick LiF disc was placed in front of the coaxial detector. For each planar detector a 24.5 mm thick $^{6}Li_2CO_3$ cap and a 6 mm thick LiF disc were used. Moreover each detector was shielded with an additional 2 mm thick LiF cylinder around the aluminum detector housing.

Figure 4: Time-of-flight response of the coaxial detector in different shielding configurations.



In Fig. 5 and Fig. 6 the results of a preliminary data analysis are reported. The data were obtained from a first measurement campaign in optimum measurement conditions, for 120 hours of accelerator operation. The results in Fig. 5 and 6 show that from the data obtained in the present experimental conditions it is possible to separate, both in TOF and γ-ray energy, events resulting from the ^{209}Bi(n,γ) reaction from background events due to capture in the Ge-detector.

In Fig. 5 the γ–ray spectra, obtained with the planar detector positioned at 125°, for two different windows on the TOF are compared. There is a clear difference between the spectrum for events with a TOF-window corresponding to a 800 eV neutron energy, i.e. close to the first strong s-wave neutron resonance in ^{209}Bi, and the γ–ray spectrum for the TOF events corresponding to a neutron energy of about 850 eV. In the former the transitions at 162.2 keV and 319.7 keV of ^{210}Bi are clearly visible. These are the two most intense transitions feeding the isomeric state at 271 keV and the ground state, respectively. Since the branching ratio is mainly determined from the intensities of these transitions, it is important to separate the peaks of these transitions from the background. Fig. 5 shows also that it was possible to separate the 162.2 keV of ^{210}Bi from the 159.5 keV of ^{76}Ge(n,γ).

Figure 5: The γ-ray response of a planar detector for the 800 eV resonance of ^{209}Bi and the background contribution due to the neutron capture in Ge.

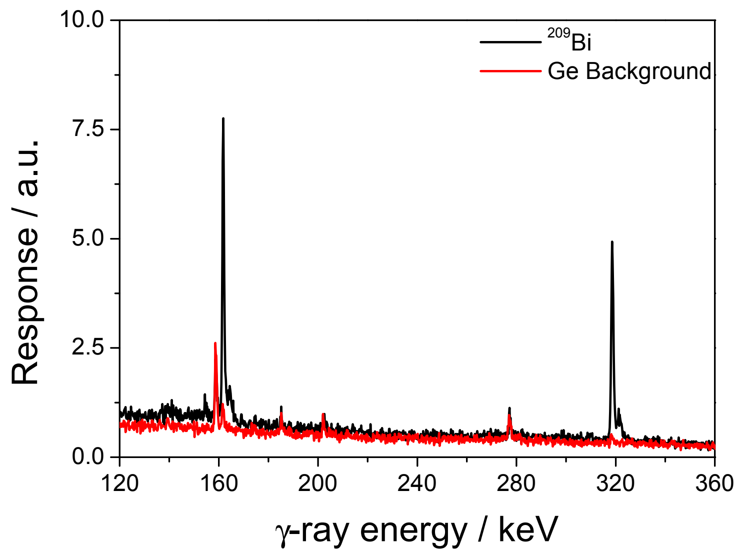
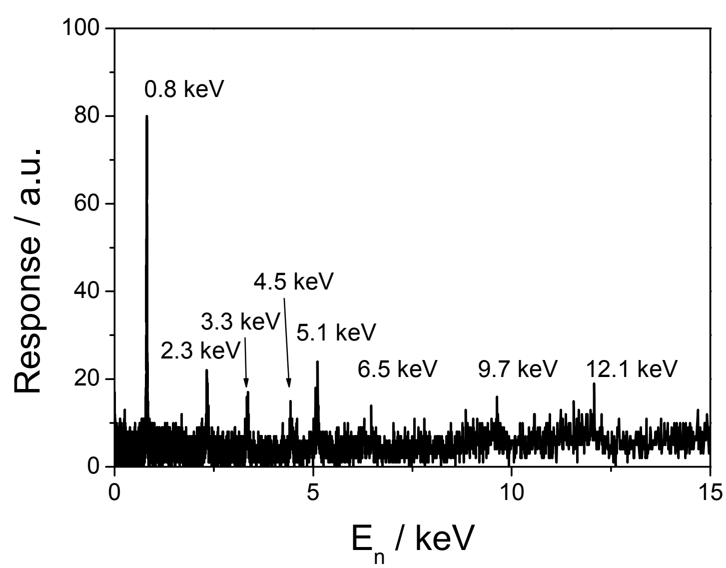


Figure 6: Time-of-flight response of a planar detector for the 320 keV γ-ray of ^{210}Bi.



In Fig. 6 the TOF response obtained by selecting events with a γ-ray energy around 320 keV is shown. This spectrum indicates that by a selection on γ-ray energy the resonances following neutron capture in ^{209}Bi can be identified up to a neutron energy of 15 keV.

In order to obtain the branching ratio as a function of neutron energy, it is necessary to work out the γ-ray spectra for the observed resonances. These spectra need to be corrected for the γ-ray efficiency and to account for the self-shielding in the sample. The branching ratio is obtained from comparison of the intensities of the transitions feeding the ground state and the isomeric state. From the data presented in Fig. 5 and 6 it is estimated that such an analysis requires a measurement campaign of at least 60 accelerator operation days.

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