FIRST RESULTS FROM THE MODULAR TOTAL ABSORPTION SPECTROMETER AT THE HRIBF*

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A Modular Total Absorption Spectrometer (MTAS) has been recently constructed and commissioned at the Holifield Radioactive Ion Beam Facility (HRIBF) at the Oak Ridge National Laboratory (ORNL). The main goal of MTAS is to determine the true beta-decay feeding and following gamma radiation pattern for the decays of fission products. In this contribution, we would like to present the results of the measurement of ^{86}Br. The preliminary analysis yields an average energy of emitted γ-radiation of about 4110 keV. It represents an increase of over 26% (850 keV), when compared to the average EM energies deduced using the ENSDF database.

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1. Introduction

Total Absorption Spectroscopy is a unique technique that measures γ-radiation with the very high efficiency. This makes it a perfect device for observing very weak γ-ray transitions. This is essential in β-decay measurements, in which many weak β transitions followed by cascades of γ-ray

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transitions occur. These weak and often high energy γ-ray transitions are usually missed in experiments employing high energy resolution, but low efficiency detectors. This results in overestimating the β-feeding of low-lying excited levels thus leading to underestimation of the average electromagnetic (EM) energy released in decays [1].

The average EM energy is an important observable for the calculation of the decay heat released during the operation of a nuclear reactor as well as after its shutdown. The comparisons between experimental data and calculations of the decay heat release show a significant difference for neutron-induced fission of ^{239}Pu [2]. This difference is believed to be related to the existence of weak β-transitions followed by γ-radiation in the decay of ^{239}Pu fission products. Therefore, a total absorption spectrometer should be used to study β-feeding and γ-radiation in the decays of isotopes abundantly produced in the fission process [3, 4].

In this contribution, we present preliminary results on ^{86}Br β-decay, which was not measured previously with a total absorption spectrometer. The decay of ^{86}Br was identified as one of the most important studies for the analysis of decay heat in nuclear reactors by the OECD's Assessment of Fission Product Decay Data for Decay Heat Calculations [2].

2. Experimental set-up

The first on-line Modular Total Absorption Spectrometer (MTAS) experiment was performed in January 2012 using a mass separator on-line to the Tandem accelerator, at the Holifield Radioactive Ion Beam Facility (HRIBF) at the Oak Ridge National Laboratory.

The 40 MeV proton beam hit a ^{238}UC$_x$ target, inducing fission. Fission products were extracted from the ion source, and then mass-separated by means of a magnetic field. Radioactive samples were implanted into mylar tape and periodically transported into the center of MTAS. A tape cycle was chosen to minimize the impact of daughter activity and short-lived contamination in the spectrum.

2.1. Modular Total Absorption Spectrometer

MTAS consists of 19 hexagonal shaped NaI(Tl) crystals (Fig. 1). Each crystal is 21″ (533 mm) long and approximately 8″ (203 mm) in diameter placed in a honeycomb-like structure. All NaI(Tl) crystals are encapsulated by four layers of housing made out of teflon, silicon putty, thin steel sheet and carbon fiber. Two 5″ (127 mm) diameter photo-multiplier tubes for light collection were mounted on both sides of the outer 18 crystals. The MTAS central detector has two sets of six 1″ (25.4 mm) diameter photo-multiplier tubes.

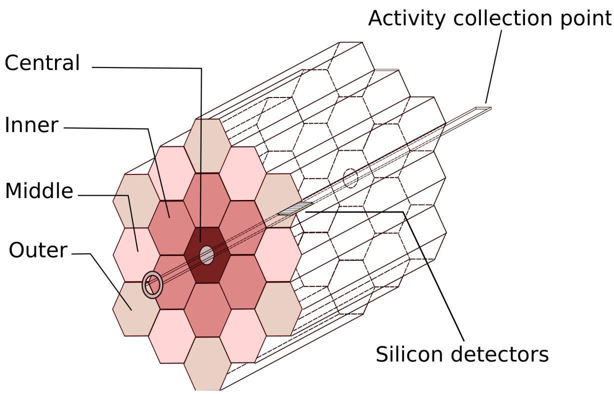

Fig. 1. A schematic view of MTAS array.

The central module has a cylindrical 2.5″ (63.5 mm) diameter hole drilled through, with an aluminium tube inside. Two 1-mm thick silicon strip electron detectors were placed inside. Signals from these Si-detectors are used as β-triggers and define the location of the radioactive sample after each tape transport.

MTAS was surrounded by lead and paraffin shielding to reduce the background. The entire shielding weighs over 5000 kg.

MTAS efficiency for full-energy γ deposition approaches nearly 80% for 1 MeV and 70% for 5 MeV γ-ray transitions. The total efficiency for γ-absorption is in the range of 96–98% (see Fig. 2).

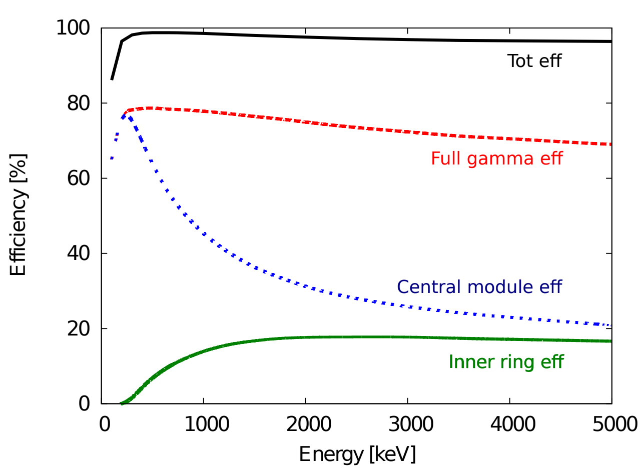

Fig. 2. Simulated efficiencies of MTAS array. Black line represents total efficiency corresponding to full-energy or partial-energy detection. Long-dashed/red, short-dashed/dark blue and grey/green lines represent full-energy γ efficiency for the whole MTAS detector, its Central module and its Inner ring, respectively.

3. Data evaluation

3.1. Geant simulation

The analysis of total absorption spectroscopy data is based on reconstruction the β-decay feeding pattern, using experimental data and the response matrix of the detector. The detector response function enables the simulation an energy spectrum for the decay of any radioactive source. Comparison of the experimental spectrum and the simulated one verifies the decay scheme.

In order to reproduce the response function of the MTAS detector, a Monte Carlo simulation code was developed by means of the Geant4 toolkit. Proper modelling of the detector response requires accurate reconstruction of its geometry and inclusion of all important physical processes occurring in MTAS during the radiation absorption. For this purpose, over 200 volumes and more then 10 types of materials were developed. Light production in the scintillator detectors was included in the simulation code based on the work of Cano-Ott *et al.* [5].

To verify the simulation code, the comparison of measured and simulated spectra was made for a set of calibration sources. The following sources were used: ^{54}Mn, ^{65}Zn, ^{137}Cs and ^{60}Co. Figure 3 presents a comparison of experimental and simulated spectra for ^{60}Co decay. A good agreement between calculated and measured spectra has been achieved.

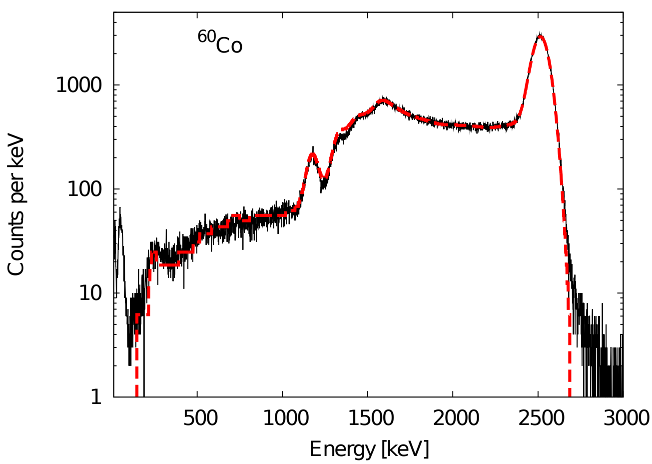

Fig. 3. Measured MTAS spectrum (solid/black line) of ^{60}Co decay together with a simulated spectrum (dashed/red line).

In order to obtain a response matrix of the detector, the simulations of monoenergetic β and γ particles emitted from the center of the detector, at various energies, were performed. The current database includes the response function for γ ray and β energies from 20 to 10 000 keV at 20 keV interval.

3.2. Spectrum reconstruction

To reconstruct the measured spectrum using the MTAS response functions all possible γ and β transitions have to be taken into account. The easiest way to do that is to define a sum over all populated levels from the response of β transitions convoluted with the response function of each level, see Eq. (1)

$$S = \sum_j I_j^\beta \beta_j \otimes L_j \,, \tag{1}$$

where I_j^β is the intensity of β transition feeding level j. The sum of all β intensities has to be normalized to 1. β_j is a response to β transitions calculated from the responses of monoenergetic electrons from a distribution defined by assuming only allowed β-decay transitions [6].

The L_j term is a simulated MTAS response function for all transitions occurring during j level de-excitation. L_j can be calculated iteratively from the responses of low-lying levels and appropriate γ-ray transitions, see Eq. (2)

$$L_j = \sum_i I_{ij}^g g_{ij} \otimes L_i \,. \tag{2}$$

I_{ij}^g is the intensity of transitions emitted from level j and feeding level i. Intensities assigned to a particular level have to be normalized ($\sum_i I_{ij}^g = 1$). Response of the ground level (L_0) is a spectrum with one count in the zero channel. Transitions emitted from each level (g_{ij}) consist of γ-ray transitions, conversion electrons and X-rays [5].

3.3. β-feeding calculation

The β-feeding function is based on the comparison between experimental and simulated spectra. If the beta decay scheme is complete, the simulation based on the decay scheme given in the ENSDF database should reproduce the experimental spectrum. Otherwise, changes to the β-feeding intensities are made to minimize χ^2 by the simplex method [7].

If this procedure does not reproduce the experimental spectra, a group of new pseudo-levels is added to the level scheme starting from a particular excitation energy ($E_{\rm min}$) in the daughter nucleus up to the Q_β value in intervals of approximately 40 keV. Under energy $E_{\rm min}$, the existing decay scheme is considered to be complete. The spin of each pseudo-level is randomly chosen from the spins possible after allowed Gamow–Teller β-transitions. It is assumed that the pseudo-levels de-excite through all the levels present in the decay scheme, however only M1, E1 and E2 transitions are taken into account. Other transitions are considered to have a marginal influence on the intensity budget.

The β-feedings of the pseudo-levels are recursively adjusted by means of the simplex method until good agreement between simulated and experimental spectrum has been achieved. The intensity of the ground-state to ground-state transition is derived from the low-energy part of the MTAS spectrum which consists mainly of signals from β particles emitted in the decay to the ground state.

The average electromagnetic energy per decay is calculated from Eq. (3)

$$\overline{E}_{\mathrm{EM}} = \sum_j I_j^{\beta} L_j^{\mathrm{EM}}\,, \tag{3}$$

where I_j^{β} is the β-feeding intensity for the j transition feeding j level, and L_j^{EM} is the electromagnetic energy emitted during j level de-excitation.

4. Preliminary results

During the experiment, the decays of over 20 fission products including ^{86}Br have been measured with MTAS. The results of a preliminary analysis of ^{86}Br decay are presented in Fig. 4.

The ^{86}Br β-decay scheme contains 22 known γ-ray transition from 14 excited levels according to the recent ENSDF evaluation [8]. Sixty five pseudo-levels starting from 5 MeV excitation energy in the ^{86}Kr daughter and 487 γ-ray transitions were added to the decay scheme to reconstruct the experimental spectrum.

The average electromagnetic (EM) energy released in the decay of ^{86}Br was obtained as 4110 keV, 26% larger than the value of 3260 keV deduced from the ENSDF database. Error analysis will be performed to complete the evaluation. However, based on current estimates, the relative error in average EM energy will not exceed 10%.

Figure 5 presents cumulative β-feeding as a function of the excitation energy of ^{86}Kr after ^{86}Br β decay. As expected, there is a shift of the β-feeding from low energy states to the higher energy. Also the ground-state to ground-state β transition intensity was increased, as can be seen at the low-energy part of the spectrum given in Fig. 4.

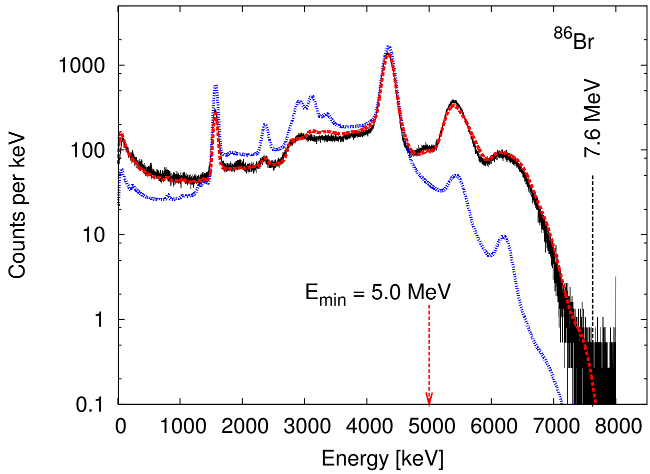

Fig. 4. Experimental MTAS spectrum (solid/black line) for ^{86}Br decay together with a simulated spectrum based on ENSDF (dotted/blue line) and our preliminary data evaluation (dashed/red line). Black and grey/red arrows indicate Q_β energy and the lowest energy of introduced pseudo-level, respectively.

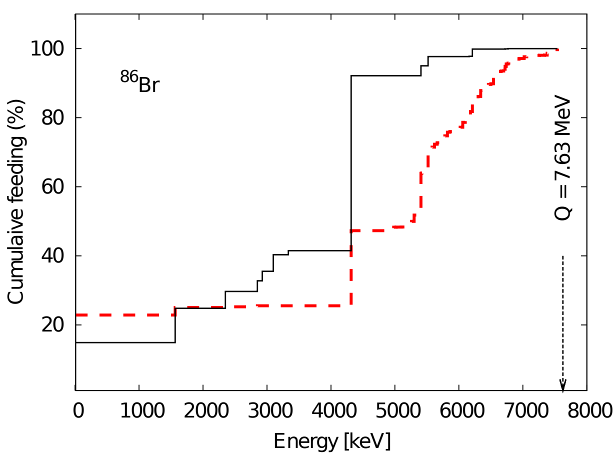

Fig. 5. Cumulative feeding of ^{86}Kr states after ^{86}Br β-decay. Solid/black line follows ENSDF data. Dashed/red line corresponds to the preliminary MTAS data evaluation.

5. Conclusion

The new Modular Total Absorption Spectrometer has been constructed and characterized at the HRIBF (ORNL, Oak Ridge). The first measurement campaign was performed in January 2012. The decays of over twenty fission products, including seven decays of highest priority, as established by the evaluation of the OECD's Nuclear Energy Agency [2] have been measured. Preliminary analysis of ^{86}Br activity, listed among the highest priority decays, yields an average EM energy of about 4110 keV. This is an increase over 26% (850 keV), in comparison to the average EM energies presented in the ENSDF database. The impact of the new results in decay heat calculations will be examined in the course of further analysis.

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