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Decay Heat Measurements Using Total Absorption Gamma-ray Spectroscopy

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Abstract.

A knowledge of the decay heat emitted by thermal neutron-irradiated nuclear fuel is an important factor in ensuring safe reactor design and operation, spent fuel removal from the core, and subsequent storage prior to and after reprocessing, and waste disposal. Decay heat can be readily calculated from the nuclear decay properties of the fission products, actinides and their decay products as generated within the irradiated fuel. Much of the information comes from experiments performed with HPGe detectors, which often underestimate the beta feeding to states at high excitation energies. This inability to detect high-energy gamma emissions effectively results in the derivation of decay schemes that suffer from the *pandemonium effect*, although such a serious problem can be avoided through application of total absorption γ-ray spectroscopy (TAS). The beta decay of key radionuclei produced as a consequence of the neutron-induced fission of ^{235}U and ^{239}Pu are being re-assessed by means of this spectroscopic technique. A brief synopsis is given of the Valencia-Surrey (BaF_2) TAS detector, and their method of operation, calibration and spectral analysis.

1. Motivation

Commercial nuclear reactors produce roughly 1040 different nuclides, a large fraction of which are unstable. The energy released in their decays in the fuel produces heat known as decay heat. During operations in thermal fission nuclear reactors, approximately 8% of the total heat produced in the reactor is due to decay heat and this contribution must be factored into the energy production [1]. Decay heat is the sole source of heat from the fuel in the absence of fission and therefore is extremely important in reactor design, irradiated fuel operations and storage.

Decay heat has three components: heavy particles(H_{HP}), light particles(H_{LP}) and photons(H_{EM}). Heavy particles are defined as neutrons, protons, alpha particles and spontaneous fission fragments, whereas light particles are defined as electrons, positrons, Auger electrons and conversion electrons, but are sometimes referred to collectively as "betas" photons are defined as γ-rays, X-rays, bremsstrahlung and annihilation radiation [2]. Data from nuclear databases such as JEFF 3.1 [3] can be used to calculate the actinide and fission-product inventories for a specified condition of reactor operation as a function of the cooling period. The

decay heat can then be derived by summing the decay energies from the emitted heavy particles, light particles and photons weighted with the activities of the produced fission products:

$$H_{HP}(t) = \sum_{i=1}^{M} \lambda_i^T N_i(t) \bar{E}_{HP}^i \tag{1}$$

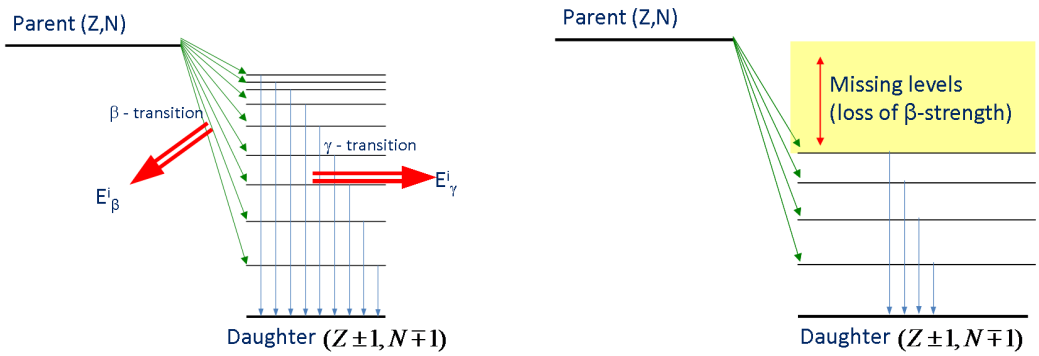
$$H_{LP}(t) = \sum_{i=1}^{M} \lambda_i^T N_i(t) \bar{E}_{LP}^i \tag{2}$$

$$H_{EM}(t) = \sum_{i=1}^{M} \lambda_i^T N_i(t) \bar{E}_{EM}^i \tag{3}$$

$\bar{E}$, λ^T and N represent the mean energy released per disintegration, the total decay constant for the nuclide and the number of the nuclides present respectively. The decay heat has also been experimentally measured by calorimetry thus providing an opportunity to test the accuracy of the nuclear datasets.

1.1. Pandemonium effect

The discrepancy between the calorimetry data and the modelled nuclear dataset is thought to be due to incorrect beta decay data (β^- and β^+) and more specifically missing beta feeding to higher-lying levels in some key nuclei (see for example Ref.[4]). The decay spectroscopy of many fission fragments was carried out using high-resolution γ-ray spectroscopy due to the simplicity of this technique. The application of high resolution γ-ray measurements to quantify the individual beta decays in some nuclei might suffer from the *pandemonium effect* resulting in the incorrect assignment of beta feeding to low-lying levels in the daughter nucleus [5]. This results in a systematic error in the deduced mean beta and gamma energies, thus affecting the decay heat calculations. Ref.[6] outlines a list of nuclear measurements that might be affected by the *pandemonium effect* and prioritises them according to their involvement in decay heat calculations for the most common nuclear fuels used in power generation.



(a) Real feeding levels (b) Detected feeding levels

Figure 1: Schematic representation of the *pandemonium effect* in beta decay.

The beta-decaying parent nucleus populates many levels in the daughter nucleus, and each decay to a different level has a precise beta-decay end point. High resolution γ-ray spectroscopy is used to measure the γ-ray intensities that both depopulate and populate these daughter

nuclear levels, and by this means individual beta feeding can be deduced from the balance of γ-ray intensities populating and depopulating each level. Decays with large Q_β values may lead to the population of a large number of levels in the daughter nucleus including some at high excitation energies (Fig.1a). The *pandemonium effect* occurs when these β-feeding distributions to the higher excitation states are erroneously derived from the γ-ray spectra (Fig.1b).

The detection of β-feeding distributions to high excitation states is impeded. At high excitation energies, a higher density of nuclear levels exist in the daughter nucleus, and the detection of β-feeding distributions is impeded. Due to the proximity of these energy levels, the β-branching to each individual level will be weaker than at lower energies. The high-excitation energy levels are open to multiple γ-decay paths to the ground state, and therefore their β-population produces many weak γ-rays that are statistically difficult to detect. These processes alone restrict the measurement of the β-feeding distributions to the high excitation states, but when combined with the low efficiency of HPGe detectors and the significant reduction of this efficiency at higher energy (> 2 MeV) the probability of detection reduces even further.

A large number of beta decay measurements have been undertaken by means of HPGe detectors before this effect was fully understood. As a result some decay schemes were thought to be complete and the β-feeding was assumed to be correct. But in reality the incorrect assignment of β-feeding as a consequence of the pandemonium effect can produce significant errors in the nuclear data sets. Fig.2 compares the measured beta strength function of ^{150}Ho obtained from the Cluster Cube (HPGe array) with that from the GSI-TAS (NaI(Tl) TAS detector)). The resulting spectra show clear evidence of the *pandemonium effect* in the HPGe data above 5 MeV, whereas the TAS measurements do not suffer from this problem. These data and spectra are taken from Ref.[7].

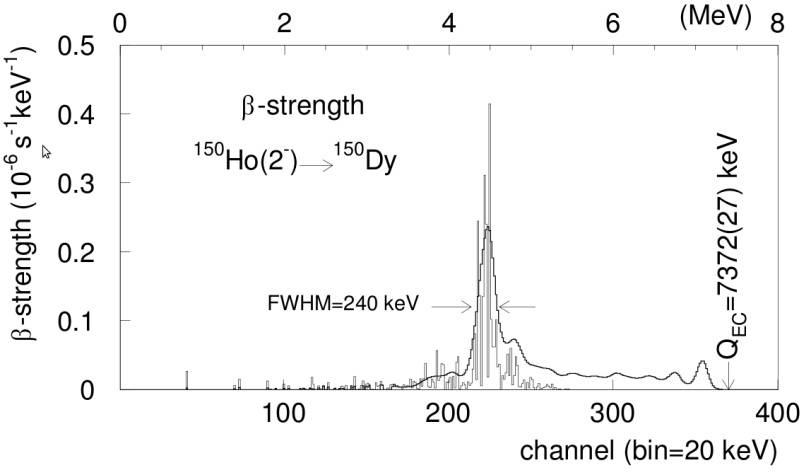


Figure 2: Beta-strength as a function of excitation energy in the daughter nucleus following the decay of the 2^- ground state in ^{150}Ho measured with the CLUSTER CUBE (depicted as a histogram) and the GSI-TAS spectrometer (continuous spectral data) [7].

2. Total Absorption Gamma-ray Spectroscopy

An ideal total absorption gamma-ray spectrometer would be 100% efficient in the detection of γ-ray radiation, cover a full 4π solid angle with good energy-resolution characteristics. TAS detectors can be made from scintillator materials (such as NaI(Tl) or BaF_2) with near 4π geometry. These relatively high-Z detection materials and the large volume of these detectors can

provide high overall detection efficiency, but with a significant deterioration in energy resolution compared with HPGe detectors. The collection of γ-rays in the TAS differs from conventional high-resolution gamma spectroscopy such as HPGe arrays, in that the TAS detector collects the full γ-ray energy cascade from a source, rather than measuring the individual γ-ray energies which make up the mutually coincident cascade.

The use of scintillators with an increased efficiency and the capability to collect full γ cascades reduces the *pandemonium effect*. Analysis of the TAS spectra is non-trivial and requires solution of equation (4):

$$d_i = \sum_j R_{ij} f_j \tag{4}$$

where d,R and f are the detector data, the response matrix and the feeding distribution respectively.

3. Experiments

The accelerator facility at the University of Jyvaskyla, Finland (JYFL), was used in November 2009 to generate eight separate radionuclides for TAS studies. Each of these nuclei had been defined as of high priority in decay heat studies [2, 6] and JYFL was selected due to the ability to produce high-purity sources. Analysis of the resulting spectral data has been divided between groups in Valencia, Nantes and Surrey.

3.1. Valencia-Surrey TAS detector

The Valencia-Surrey TAS spectrometer is a segmented barium fluoride (BaF_2) detector. The geometry of the 12 segments of BaF_2 is shown in Fig.(3) with detector dimensions of 25 cm length, 25 cm diameter with a 5 cm diameter, and 5 cm diameter longitudinal hole in the centre. Each BaF_2 crystal segment is optically insulated. Generally, BaF_2 crystals include contamination from radium due to radium and barium being chemical homologues. This contamination also gives rise to background radiation from the radioactive daughters produced within the natural decay chain of the radium isotopes. Detection of this internal radiation allows a constant gain matching between each of the PMT outputs. A planar silicon detector was placed at the centre of the TAS detector so that beta tagging could be used as a hardware trigger to reduce the random background counts in the final data.

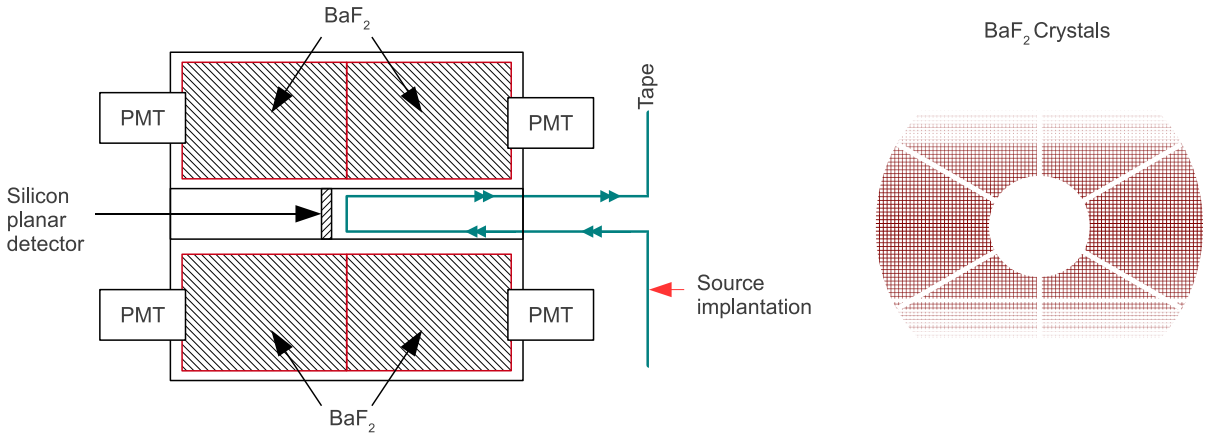


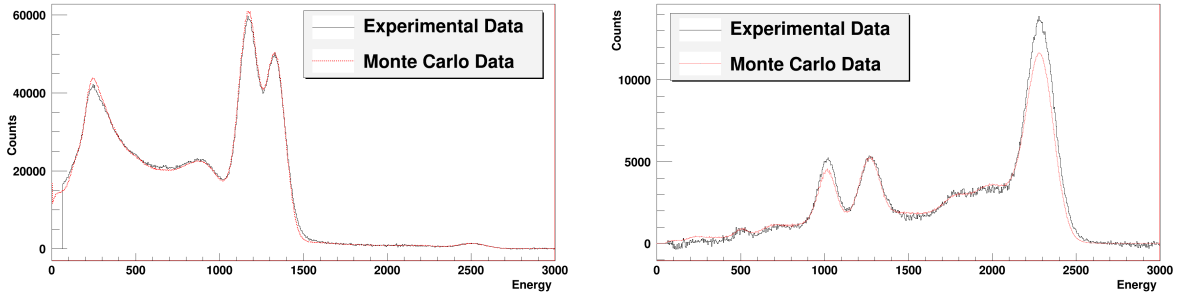
Figure 3: Valencia-Surrey TAS detector, $(left)$ longitudinal cross section showing the locations of the tape and planar silicon detector; $(right)$ cross section of the crystal arrangement.

The separated source from JYFL was deposited onto a tape delivery system which moved regularly to place the source at the centre of the TAS detector, simultaneously removing decayed sources and thus reducing the background produced by any daughter products. Each measuring cycle of the tape was optimised to the half-life of the decay being measured. Background measurements were taken at regular intervals between source measurement. Each segment of the TAS detector was readout separately producing many different output options. For the initial stage of the analysis two outputs were created: sum of each segment (E-crystal); and an online summation of the signals (Software Sum), which collects the signal as if the detector system was a single crystal (see Fig.4).

Standard sources of ^{22}Na, ^{137}Cs and ^{60}Co were used to calibrate and characterise the detector (however, these sources were sealed as a safety requirement, and therefore the silicon detector could not be applied for beta tagging in the initial set-up). ^{24}Na created by the JYFL was also used as a calibrant. The TAS data were cleaned-up and improved by undertaking both background and pile-up subtraction, prior to implementing energy and resolution calibrations. The detector manufacturer's specifications and the resulting energy and resolution calibrations were adopted as input to the GEANT4 code in order to create an appropriate Monte-Carlo (MC) model. This model is presently being used to characterise the detector on the basis of the calibrant sources.

4. Current State of Data Analysis

The spectra produced by the MC model for the calibration sources are currently being compared with the equivalent TAS data for each source (Fig.4). MC spectral data are not in satisfactory agreement with the TAS calibration data. Therefore, the existing MC code is being adjusted with respect to detector geometry, based on known differences between the original specified geometry and that subsequently quoted by the manufacturer. Physics processes within the *GEANT4* MC code will also be assessed for validity at a later stage. When the MC model is able to replicate a good agreement to the calibration data, attention will be turned with confidence towards analyses of the spectral data of interest.



(a) E-crystal of ^{60}Co data (b) Software sum of ^{22}Na data

Figure 4: Comparison of the MC and the experimental data: (a) γ-rays from ^{60}Co calibration source collected in each crystal and then summed. (b) γ-rays collected from ^{22}Na source calibration source as if the complete detector array was a single crystal.

MC models are required for the measured nuclei of interest. These model are created as before, with adjustments to the decay read-in files as created for each source from the available decay schemes (taken from nuclear data sets). A statistical model will be used to extend the calculations to high energies. Since the experimental sources are unsealed, gating on the silicon detector should be possible in order to eliminate most of the spectral background. The effects

of pile-up and any contamination from daughter decay will also be removed. These cleaned-up spectra can then be compared with the MC spectra to show whether the *pandemonium effect* affects the nuclear data set.

Assuming that the original measurements are affected by the *pandemonium effect* as postulated in Ref.[6], a new decay scheme will be created for the MC model. This new decay scheme will be created from the recorded nuclear data up to an energy threshold set to exclude regions of excitation energy in the daughter that might have been affected by the *pandemonium effect*. Above this new threshold the decay scheme will consist of "pseudo levels" added with their corresponding γ-branching ratios on the basis of the adopted a statistical model. Once this is completed an iterative process of adjusting the feeding to all the levels will be carried out so that the MC spectra gives good agreement with the experimental data.

5. Conclusions

Total absorption γ-ray spectroscopy can be used to avoid the *pandemonium effect* found in some HPGe gamma measurements of beta feeding distributions. The analysis of the TAS data is non-trivial and accurate MC models of the system are needed to extract the beta feeding distributions. Hopefully, with the measurement of selected nuclei by means of TAS, more accurate decay heat calculations can be achieved, and so assist in providing even greater confidence in operational procedures involving irradiated fuel.

6. Acknowledgements

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