Practical applications of the total absorption technique

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We present two lines of research that can be considered practical applications of the total absorption technique (TAS). The first application is related to measurements of the beta decay of specific isotopes playing an important role in the heating of nuclear fuel after reactor shutdown. The second concerns the search for optimal beta decay candidates for the construction of a monochromatic neutrino beam facility. The results of the analysis of TAS measurements relevant to these applications will be discussed.

Keywords: Reactor decay heat; beta decay; total absorption spectroscopy; monoenergetic neutrino beam facility.

En esta contribución presentamos dos líneas de investigación que pueden considerarse aplicaciones prácticas de la técnica de absorción total (TAS por sus siglas en inglés). La primera aplicación se relaciona con la medición de la desintegración beta de isótopos específicos que juegan un papel importante en el calentamiento del combustible nuclear después de apagar un reactor. La segunda aplicación se relaciona con la búsqueda de candidatos óptimos para la construcción de un haz monocromático de neutrinos. Los resultados de los análisis de las medidas TAS relevantes a estas aplicaciones se discutirán.
1. Introduction

Most nuclear applications involving beta decay rely on data available from databases (see for example Ref. 1). The compiled data is typically the result of the evaluation of many different measurements, using different techniques, but until now they are mainly based on the use of Ge detectors (high resolution technique). Depending on the case, the decay data for a specific isotope can suffer from systematic uncertainties. One common problem is the existence of decay data that suffer from the “Pandemonium effect”. This effect, first introduced by Hardy and coworkers [2], is related to the difficulties we face when building a level scheme in a beta decay study which relies on high resolution detectors. In a high resolution experiment the feeding probability to a certain level is deduced from the gamma intensity balance of the gammas feeding and de-exciting the level. If the beta decay has a large Q value, beta feeding at high excitation energies in the daughter can occur, where there is a high level density. As a consequence the feeding probability can be very fragmented, which means that there may be many weak decay paths from the high-lying levels to the low-lying ones. The decay from the high-lying levels may also proceed by the emission of high energy gamma rays.

The detection of gammas, which are weak or of high energy, with Ge detectors presents problems because of limitations in sensitivity and the relatively poor efficiency. If we fail to detect gamma rays from these high-lying levels, the resulting level scheme is incomplete and, in particular, the beta feeding is incorrectly assigned to levels at low energy.

The total absorption technique is based on the detection of the gamma cascades that follow the beta decay instead of detecting the individual gamma rays. With the use of a highly efficient device, in essence a calorimeter placed around the source, an almost 100% efficient γ for detecting gamma cascades can be achieved and then the Pandemonium effect can be avoided. In this contribution two applications of the total absorption technique will be presented. The first is related to measurements of the beta decay of isotopes relevant to the prediction of the heat-up of nuclear fuel after reactor shutdown. The second application concerns the search for optimal beta decay candidates for the construction of a monoenergetic neutrino beam facility.

2. Application to the reactor decay heat

There is renewed interest in the use of nuclear power due to an increased need for “clean” energy sources that do not contribute to the contamination of the atmosphere, and hence to global warming. A major aspect of public concern related to the nuclear power industry is based on the perception of the operational safety of such facilities. Safe operational procedures are a major pre-requisite of the design and development of nuclear power plants. The primary aim of this work was to study the decay properties of specific nuclei that are important contributors to the reactor decay heat emitted from the core immediately after shutdown.

Approximately 8% of the total energy generated during the fissio process is related to gamma and beta energy released in the natural decay of fissio products, and it is commonly called decay heat [3]. Once the reactor is shutdown, the energy released in radioactive decay provides the main source of heating. Hence, coolant needs to be maintained after termination of the fissio process in a reactor, and the form and extent of this essential requirement needs to be specified on the basis of decay-heat calculations. Decay heat varies as a function of time after shutdown. In principle it can be determined theoretically from known nuclear data, with computations based on the inventory of nuclei created during the fissio process and after reactor shutdown, and their radioactive decay characteristics. These calculations require extensive libraries of cross-sections, fission-yield and decay-data.

As mentioned earlier some of the data in the decay databases may suffer from the so-called Pandemonium effect. This effect has serious consequences for decay heat calculations: because of the erroneous omission of nuclear levels fed in beta decay there is an underestimate of the total γ energy and an overestimate of the total β energy released in the decay process. The only way to avoid this problem is the application of total absorption techniques to β-decay studies. We have performed several experiments at GSI and ISOLDE using this technique [4], and new methods of analysis of the resulting data have been developed recently [5].

Improvements in the World’s major data libraries for summation calculations have resulted in fairly good reproducibility of the integral-type measurements for different fissioning nuclei from 233U to 241Pu. However, there remains a substantial discrepancy between calculation and experiments for the electromagnetic component of the decay over cooling times that range from 300 to 3000 s after an instantaneous fission event [6]. This effect has been called by the authors [6] the γ-ray discrepancy.

The γ-ray discrepancy occurs for 233,235,238U as well as for 239Pu [6]. This is the reason why the identification of the nuclei responsible for the discrepancy was considered an important task by the authors. A careful study has shown that the best although not the only candidates are 102,104,105Tc. The choice of these nuclei is justified by the following:

a) they have a sizable amount of fissio yield,

b) their fissio yields in the different fissionable nuclei are correlated as required to solve the discrepancy,
c) their half-lives satisfy the time condition given above and

d) the $\beta$-decay $Q$-value is large enough to allow additional $\beta$-feeding in the high excitation region of the decay scheme of the daughter nuclei.

A sizable amount of $\beta$-strength was artificially added to the JENDL database at an excitation energy $\geq 4.5$ MeV for $^{104}\text{Tc}$ and $\geq 2.5$ MeV for $^{105}\text{Tc}$ in order to test the validity of the hypothesis [6]. As a result, the values of $E_\gamma$ increased by 0.3 and 0.7 MeV for $^{104}\text{Tc}$ and $^{105}\text{Tc}$. The total effective increase solves two-thirds of the $\gamma$-ray discrepancy. Yoshida et al. also demonstrated that the strength required is roughly comparable to what is predicted by Gross Theory [7,8]. However we can only decide on the basis of experiment, and the decision was therefore taken to measure the $\beta$-decay of $^{102,104,105}\text{Tc}$ to the Ru isotopes by means of the total absorption technique.

The above mentioned Tc isotopes are refractory elements which are difficult to extract from conventional ion sources. Therefore, our experiments were performed at the Ion-Guide Isotope Separator On-Line (IGISOL) [9] facility of the University of Jyväskylä. Nuclear reaction products recoiling out of a target are stopped in a gas (usually helium) and are transported by a gas flow through a differential pumping system directly into the acceleration stage of a mass separator. This process can be made fast enough for some reaction products to survive as singly-charged ions. Applying this technique, no ion source in the classical sense is used. The system is chemically insensitive which allows the extraction of refractory elements.

We have performed two experiments at this facility to study the beta decay of isotopes related to the quantification of the decay heat of irradiated nuclear fuel. The first experiment was carried out to measure the beta decay of $^{104,105}\text{Tc}$ isotopes. Our second set of experiments involved combining for the first time a total absorption spectrometer with the Penning trap at IGISOL (JYFLTRAP) [10] to measure the beta decay of $^{101}\text{Nb}$, $^{104,105,106,107}\text{Tc}$ and $^{101,102,105,106}\text{Mo}$. We also repeated the measurements of the $^{104,105}\text{Tc}$ isotopes in order to test the purity of the sources used in our first experiment. The Penning trap is an excellent isotope separator for this type of study (very high resolution "isobaric" separator) where the purity of the sources is of great importance.

We present the results of our analysis for $^{102,104,105}\text{Tc}$ isotopes using the data from the second experiment. A proton beam of 30 MeV was delivered by the cyclotron at the University of Jyväskylä to produce fission in a natural U target of 15 mg/cm$^2$. Typical currents were about 4 $\mu$A. The separated activity produced by the IGISOL facility was further purified using the JYFLTRAP (Penning Trap) as a high resolution separator (isobaric separator). Then the activity was carried to the Total Absorption Gamma Spectrometer (TAGS) by a tape transport system. The Tags was designed at the Nuclear Institute of St. Petersburg and consists of two NaI(Tl) cylindrical crystals (larger crystal has dimension: $\varnothing=200$ mm $\times$ $l=200$ mm, and has a longitudinal hole of $\varnothing=43$ mm, smaller crystal has dimensions:

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$E_\gamma$ (JEFF-3.1)</th>
<th>$E_\gamma$ (TAGS)</th>
<th>$E_\beta$ (JEFF-3.1)</th>
<th>$E_\beta$ (TAGS)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{104}\text{Tc}$</td>
<td>1890(30)</td>
<td>3229(35)</td>
<td>1595(75)</td>
<td>931(15)</td>
</tr>
<tr>
<td>$^{105}\text{Tc}_a$</td>
<td>668(19)</td>
<td>1825(174)</td>
<td>1310(173)</td>
<td>764(81)</td>
</tr>
<tr>
<td>$^{105}\text{Tc}_b$</td>
<td>668(19)</td>
<td>1999(174)</td>
<td>1310(173)</td>
<td>684(81)</td>
</tr>
</tbody>
</table>

**Figure 1.** Upper panel: comparison of the results of the reconstructed spectrum (black) from our analysis with the measured spectrum (grey) for $^{104}\text{Tc}$. The lower line of the upper panel represents the contribution of contaminants (background and pileup). Lower panel: deduced feeding from the current analysis (dots), compared with previously known feeding from high resolution measurements (discrete lines).
\( r = 200 \text{ mm} \times l = 100 \text{ mm} \). This setup has an estimated total gamma efficiency of 70\% at 5 MeV.

A Ge detector was placed at the collection point of the setup in order to monitor the composition of the sources.

One significant problem with TAGS measurements is the successful elimination of possible contaminants. A TAGS is a highly efficient summing device with modest resolution compared with a high resolution setup (Ge detector). Therefore, a clean, high-purity source is of paramount importance. Isobaric contamination was removed by means of the JYFLTRAP and the selection of appropriate collection/measuring cycles. Of the isotopes studied, \(^{102}\text{Tc}\) was the most problematic, since the yield is very low and, even when using the Penning trap, difficulties were experienced in separating it from the parent \(^{102}\text{Mo}\). We produced the \(^{102}\text{Tc}\) activity through the parent \(^{102}\text{Mo}\) and then the parent activity was subtracted as a contaminant.

Analysis of the total absorption spectra requires the solving of the “inverse problem” \( \mathbf{d} = \mathbf{R} (\mathbf{B}) \mathbf{f} \) where \( \mathbf{d} \) represents the measured data, \( \mathbf{R} \) is the response matrix of the detector, and \( \mathbf{f} \) is the feeding distribution we wish to determine. The response function \( \mathbf{R} \) depends on the detector and branching ratios of the levels in the daughter nucleus (\( \mathbf{B} \)), and can only be calculated using Monte Carlo (MC) techniques. The analysis of the data presented here was carried out using the methods of analysis established by the Valencia group [5]. The response function was determined by means of the GEANT4 code [11]. The quality of the Monte Carlo simulations was tested by comparing the result of the simulations with measured radioactive sources of \(^{22,24}\text{Na}\).

We have used the Expectation Maximization Algorithm in the analysis of the \(^{102,104,105}\text{Tc}\) decay. As mentioned earlier the response matrix of the detector depends on the level scheme of the daughter nucleus, we are analysing. Several assumptions can be made about the level scheme. Details of the \(^{102}\text{Tc}\) analysis will be given in Ref. 12. We have adopted the proposed nuclear level scheme of Ref. 12 up to an excitation of 1515 keV in the case of \(^{104}\text{Tc}\) and from the 1720 keV excitation up to the the Q value of 5516(6) keV [14] we have used the statistical nuclear model to generate an average branching ratio matrix. Once the level scheme and the branching ratio matrix are defined \( \mathbf{R} (\mathbf{B}) \) is constructed from the individual gamma responses calculated in the MC simulations as explained in Ref. 5. Only allowed Gamow-Teller transitions were considered and the spin-parity of the ground state of \(^{104}\text{Tc}\) was assumed to be \( 3^+ \). The results of our preliminary analysis for \(^{104}\text{Tc}\) are presented in Figs. 1 and 2.

We have assumed a known level scheme for \(^{105}\text{Tc}\) up to an excitation energy of 1325 keV [15] and the statistical nuclear model was used to generate the average branching ratio matrix from 1360 keV up to the Q value of 3746(6) keV [14]. Compared with \(^{104}\text{Tc}\), the \(^{105}\text{Tc}\) level scheme is more uncertain because the spin-parity assignment of the ground state of the parent is \( 3/2^- \) and approximately 50\% of the low lying levels of \(^{103}\text{Ru}\) fed in the decay according to [15] have uncertain spin-parity assignments (up to 1325 keV). Thus, several level schemes were assumed and the uncertainty of the analysis presented here reflect this effect. Another uncertainty
in this analysis is related to the fact that we were not able to determine the feeding to the ground state, and only an upper limit is given (< 9%) in the latest compilation [15]. Thus, the results of two analyses as presented as limiting cases in which feeding to the $^{105}$Ru ground state is first set to 9%, and then 0%. The results of our preliminary analysis for $^{105}$Tc are shown in Figs. 3 and 4 for the first assumption in which the ground state feeding is first set at 9%.

The analysis spectra (grey) in the upper panels of Fig. 1 and 3 are compared to the spectra generated from the feeding pattern determined in our analysis (black) for $^{104}$Tc and $^{105}$Tc respectively. Results for the beta feeding deduced are compared in the lower panels with the feeding derived previously from high resolution measurements. Both figures show that a large amount of beta feeding is observed in our experiment at high excitation in the daughter which was not previously seen in high resolution experiments [13, 15].

The strengths deduced from our analysis for the $^{104,105}$Tc cases are presented in Figs 2 and 4, and compared with the strength predicted by Gross Theory and deduced from high resolution measurements. These comparisons show that a large amount of strength is observed in our experiment which was not previously detected in high resolution measurements. This pattern resembles what is needed to explain the gamma-ray discrepancy, and is comparable with the predictions of the Gross Theory for $^{104,105}$Tc decay.

The results of our preliminary analysis were used to calculate the mean gamma and beta energies released in the decay. These values are listed in Table I, compared to the values taken from the JEFF-3.1 database, which are based on high resolution measurements. The quoted uncertainties include the influence of several experimental effects. Our results show a large increase in the mean gamma energy released in the decay of $^{104,105}$Tc compared with the high resolution results. However, the decay of $^{102}$Tc is comparable with the high resolution results, and is not a major contributor to the existing problem. When our mean gamma energies are incorporated in the ENDF decay data library [16], a significant part of the discrepancy in the gamma component of the decay heat for $^{239}$Pu is resolved, although this effect is less dramatic for $^{235}$U because of the differences in fission yield data.

3. Application to the monoenergetic neutrino facility

The demonstration of possible non-zero neutrino masses has been one of the most exciting problems of the last years in physics, since its evidence puts the Electroweak Theory of the Standard Model under scrutiny. Flavour mixing is a related problem that still lacks a deep understanding: if the last unknown mixing parameter $|U_{e3}|$ is non-zero, the possibility is open for Charge Conjugation-Parity violation in the lepton sector, which opens a window for new physics. To study these topics it will be desirable to have an intense tuneable neutrino source. In Ref. 17 a novel concept for constructing a neutrino/antineutrino factory was proposed. The facility, which is presently under study, could provide high intensity neutrinos of a single flavour with tunable energy. The idea is to obtain a collimated neutrino beam by means of accelerating to high energy radioactive ions that will decay through the beta process in a storage ring with long arms. In Ref. 17 two possible radioactive nuclei were proposed: $^{6}$He a beta minus decay case, and $^{13}$Ne a beta plus case.

In an additional twist to the original idea, Bernard et al. [18], have proposed to construct a monochromatic neutrino beam facility. A monochromatic neutrino beam can be created by means of accelerated nuclei that decay through electron capture (EC). Compared with the conventional beta decay process where three particles (an electron, an antineutrino, and the daughter nucleus) are produced, the EC process is kinematically a two-body problem, which means that the neutrino energy is well defined. The idea presented in [18] is based on the acceleration of radioactive isotopes that decay mainly by EC to one specific state in the daughter. Tuning the energy of such a radioactive beam (Lorentz boost) provides a tuneable monoenergetic neutrino source when the radioactive nuclei decay along the straight sections of the storage ring.

The potential of such a facility is clear. The unprecedented beam flavour, and the purity of the beam makes it attractive for the study of both appearance and disappearance oscillation experiments and for new precision neutrino physics.

If we concentrate now in the search for possible candidates for the monoenergetic neutrino beam facility there are some basic constraints on the properties of the beta decaying nuclei. The electron-capture facility will require a different approach to acceleration and storage compared to the standard beta-beam [17] since the nuclei can not be fully stripped. For both facilities the half-life of the decaying isotopes should be in a reasonable interval, not too short to allow reasonable intensities, and in the case of the electron-capture facility not too long since partially charged ions will have a short vacuum life-time. Another important requirement is to find nuclei with a strong EC component to one level.

![Figure 4. Comparison of the deduced strength for $^{105}$Tc (grey) with the strength predicted by Gross Theory (dotted line), assuming 9% feeding to the ground state of $^{105}$Ru. Discrete lines represent the strength derived from high resolution studies.](image-url)
We have made a systematic study of possible candidates for such a facility in the Gd region. For example, $^{152}\text{Yb}$ could be a good candidate. $^{152}\text{Yb}$ has a half-life of 3.1 s, which is a reasonable compromise for the half-life taking into account the constraints already mentioned. Since until now only high resolution measurements of its beta decay were available and they show that there is a large beta feeding to one level, it is worthwhile confirming that result using total absorption spectroscopy.

The experimental data presented here were obtained during an experiment performed at GSI in 2001. The measurement was performed at the On-Line Mass Separator using a TAS setup described in [19]. The experiment was optimized for the study of $^{152}\text{Tm}$ decay [20], but the separated mass ($A=152$) also included $^{152}\text{Yb}$, which was produced via the reaction $^{96}\text{Ru}(^{58}\text{Ni},2\text{p})^{152}\text{Yb}$ at 4.53 MeV/u (beam energy at the target).

In this contribution we present a preliminary analysis of the TAS data. The EC component of the decay in the TAS spectrum is selected by requiring coincidences with the X-rays of the daughter nuclei. This is done for the nucleus of interest and for each possible contaminant nucleus of the decay chain to separate their contributions.

In the upper part of Fig. 5, the grey line represents the analyzed TAS spectrum which was obtained by gating on the X-rays from $^{152}\text{Tm}$, the daughter of Yb, including some contribution from contaminants. The black line is the spectrum reproduced after the analysis, which shows a nice agreement. The red line represents the contribution of the contaminants (background of the X-ray spectra and pileup) to the analyzed spectrum. The analysis was performed using the same techniques as discussed for the Tc cases. In particular we have assumed a known level scheme up to an excitation of 1090 keV, and the statistical nuclear model was used to generate the average branching ratio matrix from the excitation of 1120 keV up to the Q value. More details will be given in a forthcoming publication [21].

The lower part of Fig. 5 shows the feeding distribution obtained from the TAS analysis. Our preliminary results confirm that a large amount of feeding is concentrated in the levels 458.6 and 482.4 (88 (1%) of the total), to be compared with the earlier high resolution results of 8.0(6) and 87.2(5) % in the levels 458.6 and 482.4 respectively, which makes $^{152}\text{Yb}$ a possible candidate for the monoenergetic neutrino beam facility from the point of view of nuclear structure.

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