Reactor Decay Heat in ^{239}Pu: Solving the γ Discrepancy in the 4–3000-s Cooling Period

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The β feeding probability of ^{102,104,105,106,107}Tc, ^{105}Mo, and ^{101}Nb nuclei, which are important contributors to the decay heat in nuclear reactors, has been measured using the total absorption technique. We have coupled for the first time a total absorption spectrometer to a Penning trap in order to obtain sources of very high isobaric purity. Our results solve a significant part of a long-standing discrepancy in the γ component of the decay heat for ^{239}Pu in the 4–3000 s range.

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Nuclear reactors provide a significant fraction of the world's electricity. A burgeoning population and an associated growth in economic activity suggest that world demand will double by 2050. Until now, the bulk of this has come from the burning of fossil fuels. There is general concern that reserves of fossil fuels are limited and their burning damages the environment. In particular, it contributes to the emission of large amounts of CO_2. In this context, nuclear power, based on the fission process, will be less damaging to the environment. Accordingly there is now a renaissance in the building of nuclear power stations around the world. Modern reactor designs, based on many years of operating experience, are much more efficient, more economical, and safer than earlier designs. Although the basic principles are well established, we still lack certain information, such as a knowledge of the decay properties of specific nuclei that are important contributors to the heating of the reactor during and after operation. The estimation and control of the heat emitted by the decay of fission products plays a key role in the safe operation of reactors. The primary aim of this work is to study the decay properties of specific nuclei that are important contributors to this source of heat.

Approximately 8% of the total energy generated during the fission process is related to the energy released in the natural decay of fission products, and is commonly called decay heat [1]. Once the reactor is shut down, the energy released in radioactive decay provides the main source of heating. Hence, coolant needs to be maintained after termination of the neutron-induced fission process in a reactor, and the form and extent of this essential requirement needs to be specified on the basis of decay-heat summation calculations. Decay heat varies as a function of time after shutdown and can be determined theoretically from known nuclear data. Such computations are based on the inventory of nuclei created during the fission process and after reactor shutdown and their radioactive decay characteristics:

$$\mathbf{f}(\mathbf{t}) = \sum_i (\bar{\mathbf{E}}_{\beta,\mathbf{i}} + \bar{\mathbf{E}}_{\gamma,\mathbf{i}} + \bar{\mathbf{E}}_{\alpha,\mathbf{i}})\lambda_\mathbf{i}\mathbf{N_i}(\mathbf{t}), \qquad (1)$$

where $f(t)$ is the power function, $\bar{E}_i$ is the mean decay energy of the ith nuclide (β, γ, and α components), λ_i is the decay constant of the ith nuclide, and $N_i(t)$ is the number of nuclide i at cooling time t. These calculations require extensive libraries of cross sections, fission yields, and decay data. Obviously, an accurate assessment of the decay heat is highly relevant to the design of nuclear facilities. Calculations of the decay heat are also important for the design of the shielding of discharged fuel, the design and transport of fuel-storage flasks, and the management of the resulting radioactive waste. This assessment is not only relevant to safety, it also has economic and legislative consequences. For example, the accuracy of the presently available decay data is not high enough, and this situation translates into higher safety margins implying larger economic costs. Reducing the uncertainty of available decay data is one of the main objectives of this work.

Most nuclear applications involving β decay rely on data available from databases (see, for example, [2]). The compiled data are typically the result of the evaluation of

different measurements, but until now they have been mainly based on the use of Ge detectors (high resolution technique). As a result, and 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'' [3]. In a high resolution experiment the feeding probability to a certain level is deduced from the γ intensity balance of the γ rays feeding and deexciting the level. If the β decay has a large Q_β value, levels fed at high excitation can deexcite by the emission of weak and (or) high energy γ rays. The detection of such γ rays is experimentally difficult because of limitations in sensitivity and the relatively poor efficiency of Ge detectors. As a consequence, a resulting level scheme is incomplete and, in particular, the β feeding is incorrectly assigned to levels at low energy. This has serious consequences for decay-heat calculations: because of the omission of nuclear levels fed in β decay there is an underestimate of the total γ energy and an overestimate of the total β energy released in the decay process.

In contrast to the high resolution technique, the total absorption technique is based on the detection of the γ cascades that follow the β decay instead of detecting the individual γ rays. With the use of a highly efficient device, in essence a calorimeter placed around the source, an almost 100% efficiency for detecting γ cascades can be achieved and then the pandemonium effect can be avoided. We have applied this technique in several experiments at GSI and ISOLDE [4], and new reliable methods of analysis of the resulting data have been developed recently [5,6].

In recent years improvements in the world's major data libraries for decay-heat summation calculations have resulted in fairly good reproducibility of the integral-type measurements for different fissioning nuclei from ^{233}U to ^{241}Pu. However, there remains a substantial discrepancy between calculation and experiments for the electromagnetic component of the decay heat over cooling times that range from 300 to 3000 s after an instantaneous fission event [7]. This effect has been called by the authors of Ref. [7] ''the γ-ray discrepancy.'' The discrepancy occurs for ^{233,235,238}U as well as for ^{239}Pu independently of the database used [JEF2.2, JNDC-V2 (actually JENDL), ENDF/B-IV] [7]. In particular the discrepancy was larger for the JEF database, and it extended over a much larger time interval. The larger discrepancy is related to the philosophy underlying JEF, namely, that only experimental values are used when available and they are not augmented by theoretical estimations even though they may be incomplete. A careful study by Yoshida *et al.* [7] has shown that among several possible reasons for the discrepancy the most plausible explanation is the underestimation of the $\bar{E}_\gamma$ for some nuclei that have a half-life of about 1000 s or that have a shorter half-life, but with a precursor that has a half-life of about 1000 s in the β decay chain. The most important, although not the only candidates are ^{102,104,105}Tc. A set of additional nuclides was subsequently identified, resulting in a high priority list containing nuclei that are large contributors to the decay heat and also to the inconsistencies in the different decay data libraries used for summation calculations [8]. We selected from these nuclei the cases with highest priority to be measured using the total absorption technique. The results of the measurements of the β decay of the selected nuclei (^{102,104,105,106,107}Tc, ^{105}Mo, and ^{101}Nb) and their impact in the decay-heat summation calculations will be presented here for the first time.

The above-mentioned 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. In this facility nuclear reaction products recoiling out of a thin target are stopped in a gas (helium) and are transported by gas flow through a differential pumping system directly into the acceleration stage of a mass separator. This system is chemically insensitive.

The mass resolving power of IGISOL is not sufficient to produce isotopically pure sources, of great importance in these types of studies. Therefore, in our experiment we have combined for the very first time a total absorption spectrometer with the available Penning trap at this facility (JYFLTRAP) [10], which provides a mass resolving power of the order of 10^5.

To produce the isotopes of interest, proton beams of 30 and 50 MeV were delivered by the JYFL cyclotron to induce fission in a natural U target of 15 mg/cm^2. Typical primary beam currents were about 4 μA. The activity extracted from JYFLTRAP was then 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 with dimensions of $\oslash = 200$ mm $\times$ $l = 200$ mm and $\oslash = 200$ mm $\times$ $l = 100$ mm. The longer crystal has a longitudinal hole of $\oslash = 43$ mm. The crystals were separated by 0.5 cm. This setup has a 57% peak and 92% total efficiency for the 662 keV ^{137}Cs γ line and 27% peak and 70% total efficiency for a 5 MeV γ line [estimated using Monte Carlo (MC) simulations]. At the measuring point, inside the TAGS, a silicon detector was placed. In our setup a Ge detector was also placed at the collection point to monitor the purity of the sources.

Analysis of the total absorption spectra requires the solving of the ''inverse problem,'' $\mathbf{d} = \mathbf{R}(B)\mathbf{f}$, 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 (B), and can only be calculated using MC techniques. The analysis of the data presented here was carried out using the methods of analysis established by the Valencia group [5,6]. The response function was determined by means of the GEANT4 code [11]. The quality of the MC simulations was tested by comparing the result of

the simulations with measured spectra for radioactive sources of ^{22,24}Na, ^{60}Co, and ^{137}Cs.

The first step in the analysis of the TAGS data is the elimination of possible contaminants and distortions of the measured spectra. The extremely good separation of the isobars obtained with JYFLTRAP solves a major part of the problem, but during the measurements the daughter decay can also contaminate our spectra. The contribution of the daughter activity can be reduced in general by proper choice of collection and measuring cycles of the decay of interest, but in some cases it cannot be avoided and has to be evaluated and subtracted. For that, in cases where a contribution from the daughter was expected, separate measurements of the daughter activity were performed. Another possible source of contamination is the background in the measurements. During our cycle of measurements the background was measured every two hours for one hour. In principle the TAGS coincidences can be used with β coincidences in the Si detector to obtain the spectrum and avoid the contribution of the background. However, this type of analysis requires a very careful evaluation of the Si efficiency, and as the precision of the positioning of the tape was not sufficiently accurate for this purpose, we decided to use the singles spectra for the analysis. This also has the advantage that much higher statistics can be used. On the other hand, the Si-TAGS coincidence spectra can be used to evaluate the correctness of the background subtraction coefficients by comparing the coincidence spectra with the subtracted one. Another distortion of the spectra is related to the pulse pileup. We have calculated the pileup distortion as described in [5].

We will discuss briefly the analysis of the ^{104}Tc case. More details of the analysis as well as that of the other cases will be presented in a forthcoming publication [12]. As mentioned earlier, the response matrix of the detector depends on the level scheme of the daughter nucleus. Typically several assumptions can be made about the level schemes. In the decay of ^{104}Tc we have adopted the proposed nuclear level scheme up to an excitation of 1515 keV in the daughter ^{104}Ru [13], and from the 1720 keV excitation up to the Q_β value [14] we have used the statistical nuclear model, that is, level densities and E1, M1, E2 γ-decay strength functions to generate an average branching ratio matrix. The parameters for the level density function and for the γ-decay strength functions were taken from Ref. [15]. Once the level scheme and the branching ratio matrix are defined, $\mathbf{R}(B)$ is constructed recursively from the individual γ and β responses calculated in the MC simulations as explained in Ref. [5]. Only allowed transitions were considered, and the spin-parity of the ground state of ^{104}Tc was assumed to be 3^+. For the solution of the inverse problem we have used the expectation maximization algorithm [6]. This algorithm can be implemented iteratively, and it provides positive solutions for the feeding distribution. At every iteration (k), the quality of the solution $\mathbf{f}_k$ can be tested by comparing the spectrum generated by $\mathbf{R}(B)\mathbf{f}_k$ with the measured spectrum $\mathbf{d}$. The results obtained for ^{104}Tc are presented in Fig. 1.

The results of our analysis are the feeding distributions $\mathbf{f}$ of the decay of the studied nuclei. They were used to calculate the mean γ and β energies released in the decay using the following relations: $\bar{E}_\gamma = \sum_i E_i I_i$, and $\bar{E}_\beta = \sum_i I_i \langle E_\beta \rangle_i$, where E_i is the energy of the level i, I_i is the normalized feeding to level i, and $\langle E_\beta \rangle_i$ is the mean energy of the β continuum populating level i. The deduced values for the mean energies are listed in Table I, compared to the values taken from the ENDF/B-VII database, which are based on high resolution measurements (the values of the new JEFF-3.1 database are very similar to the ENDF/B-VII database). Table I shows a large increase in the mean γ energy released in the decay of ^{104,105,106,107}Tc and ^{105}Mo compared with the high resolution results, which indicates that the original data suffered seriously from the pandemonium effect. In contrast, the mean energies from the decay of ^{102}Tc and ^{101}Nb are comparable with the high resolution results and will not have an impact on the problem. The large increase in the mean γ energy released in the decays of ^{104,105}Tc has a large effect on the discrepancy between 300 and 3000 s in the decay heat of ^{239}Pu. Our results solve a significant part of the discrepancy in the γ component of the decay heat for ^{239}Pu in this cooling time interval. Figure 2 presents the comparison of the calculated decay heat based on the data from the ENDF/B-VII database before and after the inclusion of our new results, with the measurements of Tobias [16] for ^{239}Pu. A striking feature is that the discrepancy which also existed for the shorter cooling times (4–200 s) with this database is practically solved. Among the many fission products that contribute to the decay heat in ^{239}Pu, the seven decays

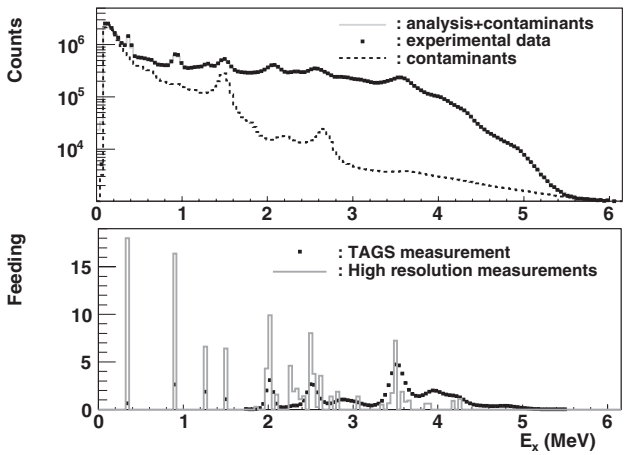

FIG. 1. Upper panel: Comparison of the reconstructed spectrum from our analysis (gray) with the measured spectrum (black dots) for the decay of ^{104}Tc (practically undistinguishable). The dotted line represents the contribution of contaminants. Lower panel: Deduced feeding from the current analysis (black dots) compared with previously known feeding from high resolution measurements (gray histogram lines) [13].

TABLE I. Comparison of mean γ and β energies included in the ENDF/B-VII database with the results of the analysis of our measurements (in keV). The errors of our analysis come mainly from different assumptions on the level schemes and parameters of the statistical model.

Nuclide	$T_{1/2}$ (s)	$\bar{E}_\gamma$ ENDF	$\bar{E}_\gamma$ TAGS	$\bar{E}_\beta$ ENDF	$\bar{E}_\beta$ TAGS
^{101}Nb	7.1(3)	270(22)	445(279)	1966(307)	1797(133)
^{105}Mo	35.6(16)	552(24)	2407(93)	1922(122)	1049(44)
^{102}Tc	5.28(15)	81(5)	106(23)	1945(16)	1935(11)
^{104}Tc	1098(18)	1890(31)	3229(24)	1595(75)	931(10)
^{105}Tc	456(6)	668(19)	1825(174)	1310(205)	764(81)
^{106}Tc	35.6(6)	2191(51)	3132(70)	1906(67)	1457(30)
^{107}Tc	21.2(2)	515(11)	1822(450)	2054(254)	1263(212)

measured in this work account for more that 30% of the cumulative fission yield in ^{239}Pu. Our work shows that the data for five of them were affected by a large error due to the pandemonium effect, an error large enough to explain a significant part of the discrepancy that existed in the ENDF-based summation calculations. We have demonstrated that identifying major contributions and measuring the associated decays with the proper technique can significantly improve the decay-heat predictions.

In this Letter we have presented the results of TAGS measurements of nuclei that are important contributors to the decay heat in reactors. In the measurements we have combined three experimental techniques for the very first time to study a long-standing problem in the description of the decay-heat data: IGISOL, to produce and separate refractory elements, JYFLTRAP, as a high resolution isobaric separator, and the total absorption technique. The results of our measurements solve a large part of the discrepancy in the γ component of the decay-heat data of ^{239}Pu and show the importance of total absorption measurements to address this problem. Our results and the application of these techniques can also have an impact in related fields such as the prediction of neutrino spectra from reactors, which can be important in both nonproliferation applications and the study of neutrino oscillation experiments [17].

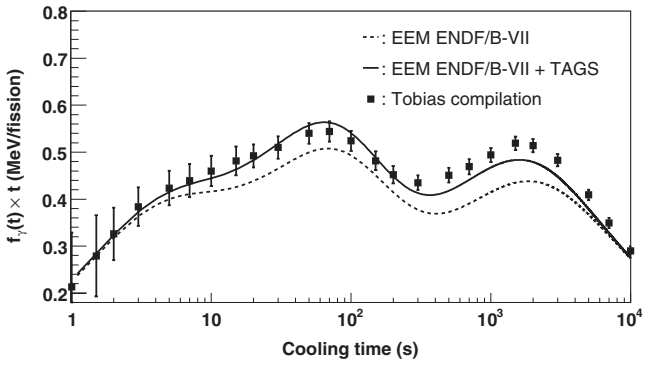

FIG. 2. Comparison of the calculated electromagnetic decay-heat component for ^{239}Pu before and after the inclusion of our measurements with the data of Tobias *et al.* [16], which is considered the standard of the field.

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