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May 2010

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Abstract

Improvements in existing national and multinational decay-data sub-libraries are merited in order to derive more accurate estimates of decay heat from irradiated Th/U fuel. Recommendations focus on particular fission-product radionuclides that warrant study and improved specification of their decay characteristics by means of total absorption gamma-ray spectroscopy (TAGS/TAS) to better define their decay properties in terms of their total energy release and component mean beta and gamma energy releases. Decay-data needs of greatest relevance to Th/U fuel have been derived, as well as requirements that are judged to be common to both the Th/U and U/Pu fuel cycles.

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1. Introduction

While formulating safe designs and procedures for the operation of nuclear facilities and the handling of irradiated fuel, a sound knowledge of the time-dependent energy release from the decay of the radioactive nuclides formed in the reactor core is critically important. Accurate estimates of this decay heat are needed for a wide range of applications including the design and transport of fuel-storage flasks and the management of the resulting radioactive waste. The nuclear power community must ensure accurate and reliable calculations of the decay heat of irradiated fuel in order to maintain credibility and confidence in the safe and reliable performance of various nuclear fuel cycles. The worldwide focus over the last six decades has been primarily on the $^{235}\text{U}/^{239}\text{Pu}$ (U/Pu) cycle resulting in a lack of specific data and dedicated knowledge for other fissioning systems. The adequate and optimal utilisation of the $^{232}\text{Th}/^{233}\text{U}$ (Th/U) fuel in the nuclear power industry over the intermediate and long term is of particular interest to India, based on the availability of significant quantities of thorium-based ore deposits.

Researchers at Manipal University have initiated a study of decay-heat calculations for thermal-neutron irradiated Th/U fuel as a function of cooling time. The present investigation is timely, given the various on-going efforts to settle existing decay heat issues with respect to U/Pu fuel. For example, WPEC Subgroup 25 (SG-25) was created in 2005 to address the need for additional experimentally-derived fission product decay data [1]. Similar analysis would be beneficial in the case of the Th/U fuel where the identities of the fission products that contribute significantly to the resulting decay heat are not nearly so well defined. Calculations of decay-heat profiles as a function of the cooling time of irradiated fuel are based on a sound quantification of the mean energies of the heavy-particle, light-particle and electromagnetic emissions of fission products and actinides within the fuel. These summation parameters are in turn dependent on a significant number of fission-product and actinide decay characteristics, including half-lives and α -particle, β -particle, conversion-electron and gamma-ray energies and emission probabilities.

The determination of β -particle emission probabilities has long been a problem in decay-scheme studies. Although γ -ray emission probabilities and internal conversion coefficients are normally used to derive β feeding to daughter nuclear levels, all forms of Ge detector possess low intrinsic efficiency for the detection of high-energy γ -ray energies above $\sim 1.5\text{MeV}$ that undermines such an exercise. Furthermore, the determination of direct β decay to the ground state of the daughter nucleus can pose serious problems for various other reasons. Hardy *et al.* [2] have demonstrated that $\sim 20\%$ of the true γ -ray intensity above 1.7 MeV for a fictional radionuclide (“Pandemonium”) would remain undetected, impacting significantly on the use of γ -ray singles data to calculate the β transitions by means of gamma population-depopulation balances of the various proposed nuclear levels. The problem has been labelled “the Pandemonium effect”, and results in decay-scheme omissions. TAGS (Total Absorption Gamma-Ray Spectroscopy) measurements can overcome these difficulties, and provide the necessary mean beta and gamma decay energies for decay heat calculations. Thus, TAGS measurements have improved the quantification of decay heat calculations in many instances where experimentally measured β -decay schemes were previously incomplete (or non-existent) for a number of important fission products relevant to ^{235}U and ^{239}Pu reactor fuel [3, 4].

WPEC SG-25 sought to characterize the β -decay of individual fission product nuclides required for decay heat summation calculations: highly-relevant studies were proposed [5] and some TAGS experiments have been undertaken or are in progress [6]. Thus, the existing request list for TAGS measurements can be appropriately supplemented by including the important contributors to decay heat specific to ^{233}U , the fissile material bred from ^{232}Th in the Th/U cycle.

An independent assessment to validate the JEFF-3.1.1 Radioactive Decay Data and Fission Yield sub-libraries [7] involved the incorporation of existing TAGS data from Greenwood *et al* [3, 4]. These studies have shown that JEFF-3.1.1 is an improvement over the JEFF-3.1 database (without TAGS

data) for fission-pulse studies of the ^{235}U mean electromagnetic and light-particle components*. An almost equivalent improvement was seen in the case of ^{239}Pu ; however, the ^{238}U and ^{241}Pu fissioning systems remained largely unaffected. Despite TAGS measurements of specific fission-product nuclides of importance in irradiated $^{235}\text{U}/^{239}\text{Pu}$ fuel, their relative contribution to the decay heat in other systems may not be as significant owing to the variation in their fission yields across different fissioning systems. Appendix A shows the independent fission yields for the thermal neutron induced fission of three fissioning systems, and the variation of the yields are clear to see, particularly for the light mass peak. An illustration of this behaviour was discussed during the SG-25 assessment, where addition of preliminary TAGS data for ^{104}Tc and ^{105}Tc made a significant improvement to the ^{239}Pu decay-heat predictions, but had little effect on ^{235}U . Such disparate findings support the need for dedicated decay heat calculations for ^{233}U . The decay data of each contributing nuclide can then be examined on a case-by-case basis and assessed as prospective candidates for TAGS. This form of study is merited to determine how much improvement can be achieved in decay heat calculations for the Th/U fuel cycle following TAGS measurements of specific ^{233}U -based nuclides.

Full summation calculations for ^{233}U fuel following a fission burst have been carried out in the present assessment by means of the DECROI code [8]. Fission products have been analysed in detail at five cooling times of interest. The results have been compared for similarities and differences with equivalent calculations carried out for ^{235}U . We present quantified percentage contributions to the decay heat from each fission product nuclide formed by neutron irradiation on ^{233}U . Differences between U/Pu and Th/U decay heat calculations are presented including the mean light-particle and electromagnetic components. As expected, some fission products appear in common for both ^{233}U and ^{235}U fuels and their appearance in both the SG-25 assessment and this Th/U decay heat list of important and inadequately-defined radionuclides underlines the importance of their selection for TAGS measurements.

A further important aspect of the current study was to identify inadequately characterized fission products specific to ^{233}U fuel which have not been highlighted by SG-25 as important for U/Pu fuel, nor studied by Greenwood *et al.* Additional TAGS candidates unique to ^{233}U fuel are itemised and priorities are assigned in perceived order of importance based on well-defined criteria. Therefore, this analytical approach defines further needs for improved decay-scheme data to assist in decay heat calculations, and extends the list of requirements for additional TAGS studies.

2. Methodology

Total decay heat and other aggregate properties of fission products may be predicted through the use of a combination of inventory and summation calculations. Neutron cross sections, fission yields and decay data represent the input to summation calculations used to determine the release of decay heat over an extended period of time after reactor shutdown (*i.e.*, following termination of neutron-induced fission). Despite actinide decay contributing to the total decay heat of irradiated nuclear fuel, this contribution has not been considered owing to the need to resolve and reduce the more significant uncertainties attributed to specific fission products. The DECROI code was used to determine the evolving inventory of fission product nuclides as a function of neutron irradiation for both ^{235}U and ^{233}U fuels. A detailed explanation of the calculations follows.

2.1 DECROI Inventory Formation and Decay Code

DECROI calculates the time evolution of a variety of radioactive nuclei, whether they are present at the outset, or are formed during irradiation by a particle flux. The inventory of nuclides as a function of time and irradiation history is governed by the following matrix equation:

* The terms “mean electromagnetic” and “mean light-particle” are often quoted as “mean gamma” and “mean beta” respectively. However, nuclear data files used for applications contain mean electromagnetic and light-particle energies including for example, Auger and conversion electrons in the “light-particle” component, with its concomitant impact and appropriate reduction in the “electromagnetic” component. The equivalent quantities to those stored in the data files are actually determined in decay heat measurements. Nevertheless, these terms tend to be used interchangeably, and can cause confusion.

$$\frac{dN}{dt} = M.N \quad (1)$$

where N is the matrix of the numbers of nuclei, and M is the matrix governing the time evolution of the system. The matrix M is composed of two parts:

$$M = M(\text{decay}) + M(\text{reaction}) \quad (2)$$

where $M(\text{decay})$ contains the natural decay properties of all nuclei required, and $M(\text{reaction})$ describes the reactions due to the irradiation. $M(\text{decay})$ is constructed from the half-lives, decay modes and branching fractions. These data are taken from NUBASE [9], but equivalent data from any appropriately-formatted data file could be used. NUBASE contains data for 3852 nuclei, but memory restrictions limit this number to only 3000 nuclei in DECROI calculations, which are appropriately selected at the outset to suit the system to be considered, *i.e.*, limited by mass to the fission product region of interest.

$M(\text{reaction})$ constitutes a selection of parameters describing the transmutation of the nuclei present as a consequence of the specified irradiation conditions. Hence the elements of this matrix are the product of the irradiating flux and cross sections of various transmutation reactions. DECROI does not possess a specific cross section library, hence this matrix is either specified or calculated independently, based on the individual case to be analysed. The irradiation flux may vary in time in the form of a histogram, or may be constant. The flux may also be set to zero for any given time interval.

The initial conditions are defined in terms of the inventory of stable and unstable nuclei at time $t = 0$, as provided by the user. Based on this initial inventory, the matrix is calculated from the combination of decay data and other data related to the irradiation conditions, but restricted to only the relevant nuclei (as limited by the matrix dimensions). The differential matrix as defined by equation (1) is solved exactly by calculating the precise eigenvalues and eigenvectors of the transfer matrix M . Suitable sub-routines for this procedure come from the LAPACK library of linear algebra, which are sufficiently stable numerically to treat the large span of half-lives and/or irradiating flux.

The results are calculated at a certain number of times (maximum of 30 per calculation) after time 0, at times defined by the user in standard units, *e.g.*, minutes, seconds, milli-seconds, *etc.* Solution of the transfer matrix in terms of exact eigenvalues and eigenvectors is carried out once per irradiation step, or only once if there is no irradiation, and the result is the simple solution of a combination of linear exponentials. Number density and activity are given as output for each user-specified time and for each nuclide (stable, unstable, ground or isomer). All other data manipulations are carried out from adoption of this output, based on user needs, *e.g.* a separate file containing average energies can be used to find the decay heat by combining with the activity output.

Decay heat as a function of cooling time may be calculated with confidence once the complete set of fission product yields and the mean energy released per decay through light-particle and electromagnetic emissions (\bar{E}_{LP} and \bar{E}_{EM} , respectively) have been determined.

2.2 Fission Product Yields for ^{233}U

Fission-yield files are normally assembled from evaluations of individual fission-yield measurements combined with interpolation of these available data to produce comprehensive data sets based on fits to the familiar double humped curve (see Appendix A). The fission yields adopted in the calculation of Th/U fuel inventories as a function of irradiation history were taken from the fission-yields sub-library of JEFF-3.1.1 [7]. Variations in the thermal fission yields of ^{233}U , ^{235}U and ^{239}Pu are shown in Appendix A. While significant differences can be seen between ^{239}Pu and $^{233,235}\text{U}$ in the location of the lower-mass peak, much less pronounced differences occur between the lower-mass peaks of ^{233}U and ^{235}U , particularly with respect to the rising and falling shoulders.

2.3 Decay Heat Calculations

When the actinide and fission-product inventories have been calculated for the specified conditions of reactor operation and cooling period, the decay heat can be derived by summing the products of the

nuclear activities expressed in terms of the mean heavy-particle, light-particle and electromagnetic energy releases per disintegration of each specific nuclide:

$$H_{HP}(t) = \sum_{i=1}^M \lambda_i^T N_i(t) E_{HP}^i \quad (3)$$

$$H_{LP}(t) = \sum_{i=1}^M \lambda_i^T N_i(t) E_{LP}^i \quad (4)$$

$$H_{EM}(t) = \sum_{i=1}^M \lambda_i^T N_i(t) E_{EM}^i \quad (5)$$

where E_{HP}^i, E_{LP}^i and E_{EM}^i are the mean heavy-particle, light-particle and electromagnetic energy releases respectively per disintegration of nuclide i ; λ_i^T is the total decay constant of nuclide i ; and $H_{HP}(t), H_{LP}(t)$ and $H_{EM}(t)$ are the total heavy-particle, light-particle and electromagnetic decay heat respectively at time t after reactor shutdown. Heavy particles are defined as alpha particles, recoil nuclei, protons, neutrons and spontaneous fission fragments (sometimes referred to collectively as the ‘‘alpha’’ component); light particles are defined as negatrons, positrons, Auger electrons and conversion electrons (sometimes referred to collectively as the ‘‘beta’’ component); electromagnetic radiation is defined as gamma rays, X-rays, annihilation radiation and internal bremsstrahlung (sometimes referred to collectively as the ‘‘gamma’’ component). The total decay heat at a particular cooling time (t) is given by simple summation of these terms:

$$H_{total}(t) = H_{HP}(t) + H_{LP}(t) + H_{EM}(t) \quad (6)$$

Attention has focused in this study on the main contributors to the decay heat, *i.e.*, fission products, while the lesser contributions arising from the actinides have been ignored.

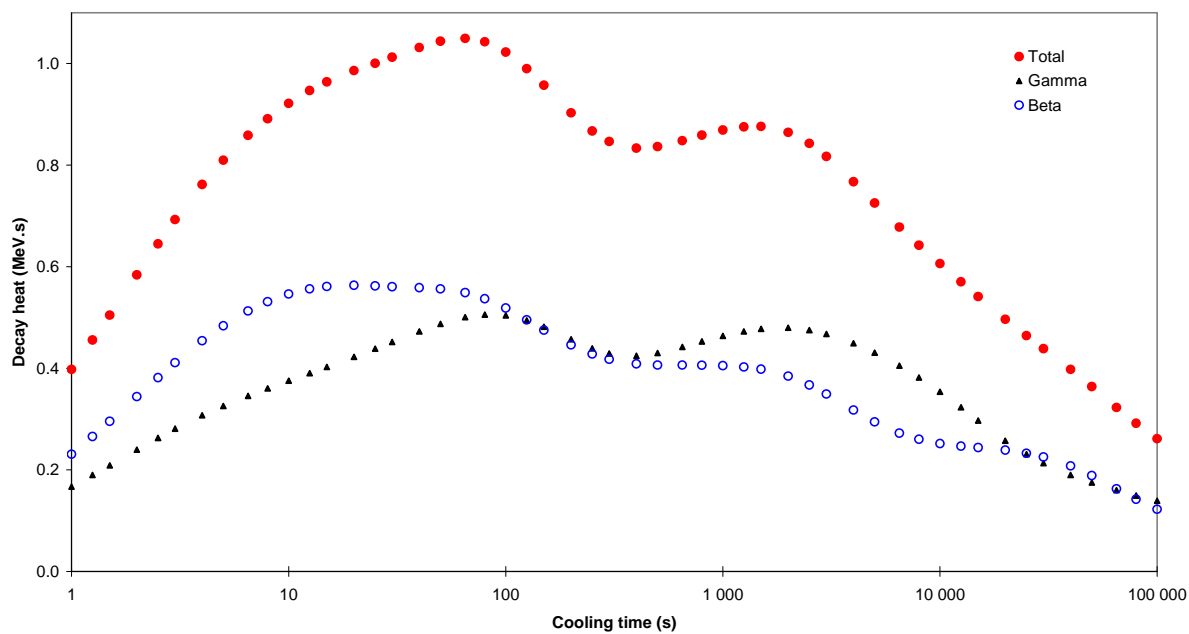
3. Decay Heat Calculations for Th/U Fuels - Results

Calculated inventories and all relevant radionuclidic decay energies were brought together in decay-heat calculations for thermal neutron irradiated Th/U fuel. The primary aims were to assess and determine the main contributors to the resulting decay heat as a function of the cooling time of the fuel, and to compare these findings with similar studies of U/Pu fuel in order to identify both significant differences and radionuclides common to equivalent lists. The major fission products in order of importance expressed as percentage contribution to their total decay heat are given in Tables 1, 2, 3, 4 and 5 for irradiated fuel cooling times of 10, 100, 1000, 5000 and 10 000 s, respectively.

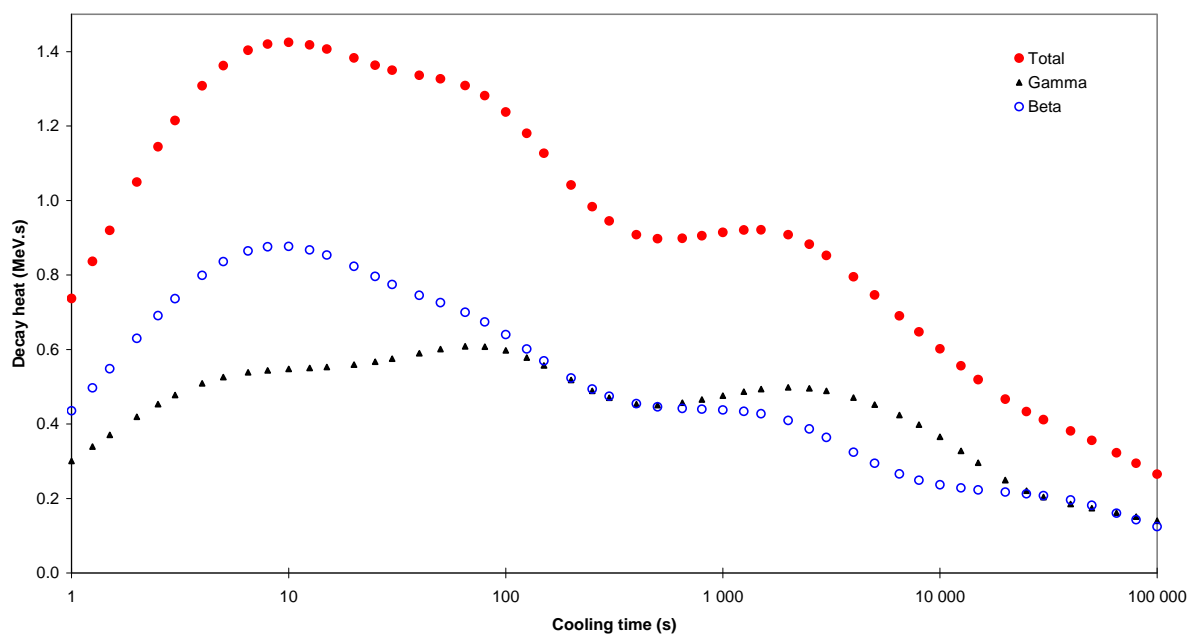
3.1 Major contributors to the decay heat of irradiated Th/U fuel

Decay-heat calculations were undertaken for ^{233}U , ^{235}U and ^{239}Pu over a wide range of comparable cooling times, providing supportive evidence of differences, similarities and trends that are explored more fully below. Figures 1 (a), (b), and (c) depict \bar{E}_{total} , \bar{E}_{LP} and \bar{E}_{EM} : at cooling times up to 10^5 s for ^{233}U , ^{235}U and ^{239}Pu . The varied and interlinked importance of the main components of decay heat across such a range of cooling times can be clearly observed from such plots. Thus, the light-particle decay component for both ^{233}U and ^{235}U fuel is seen to contribute more to the total decay heat than the electromagnetic radiation at cooling times from approximately 1 and 100 s, both components are equally important from 100 to 1000 s, while the electromagnetic radiation predominates from 1000 to 20 000 s. Whereas for ^{239}Pu fuel, the light-particle decay component contributes more from 1 s through to 1000 s, when the electromagnetic radiation predominates to ~ 20 000 s, with approximately equal contributions to 100 000 s.

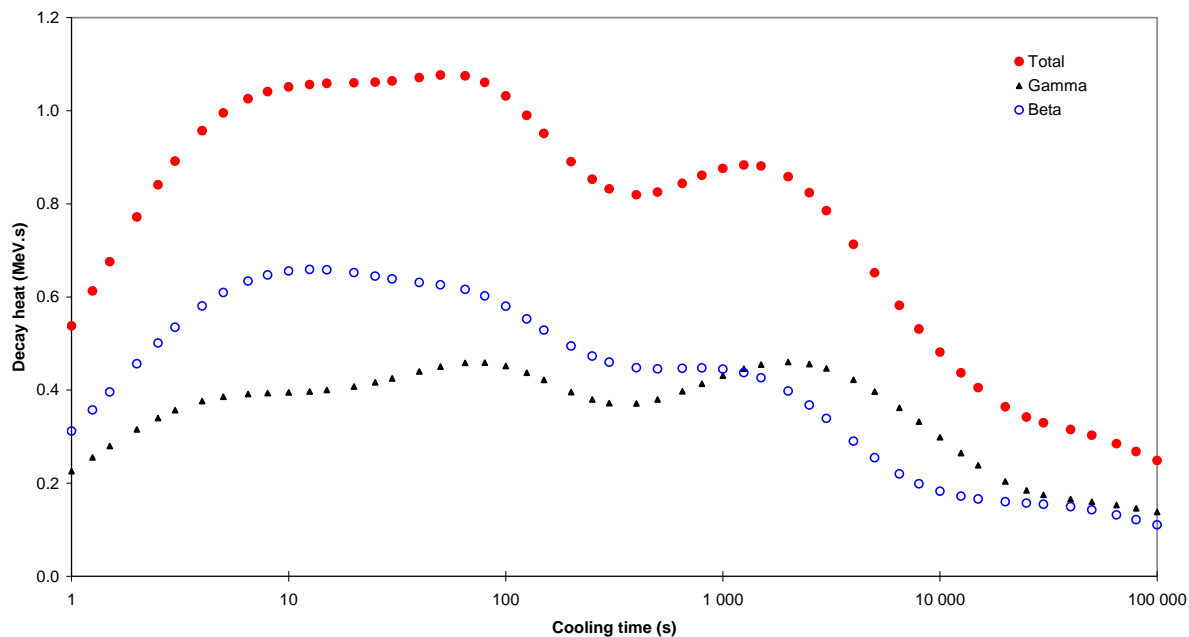
Figure 1: Calculated decay heat from (a) ^{233}U , (b) ^{235}U , and (c) ^{239}Pu .



(a) ^{233}U



(b) ^{235}U

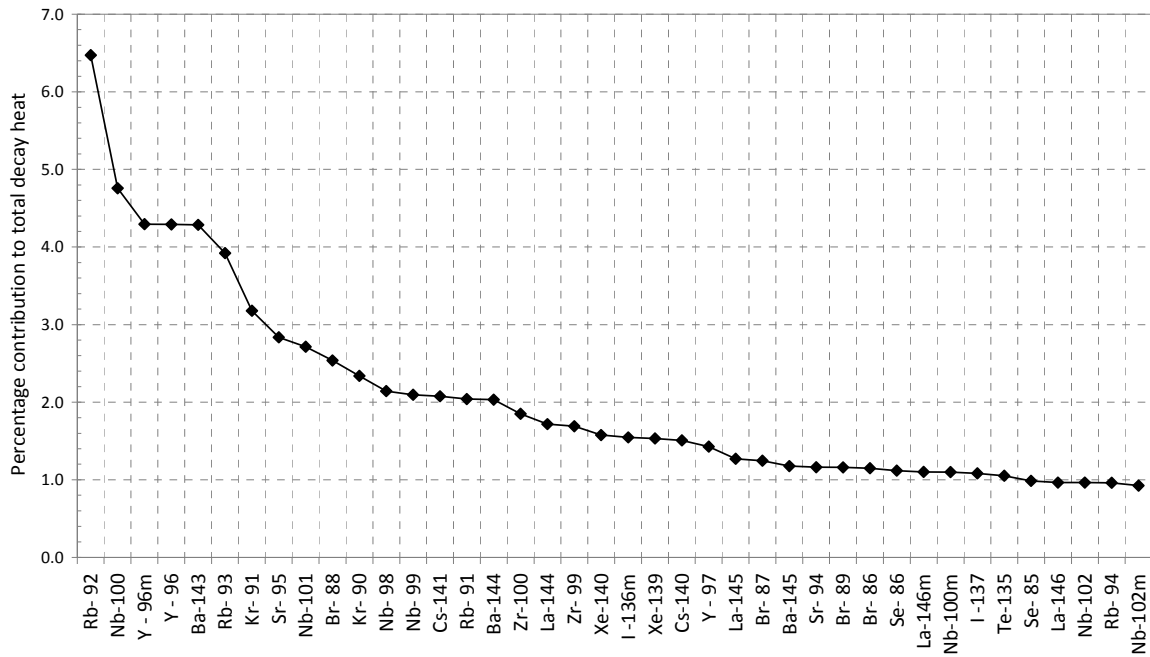


(c) ^{239}Pu

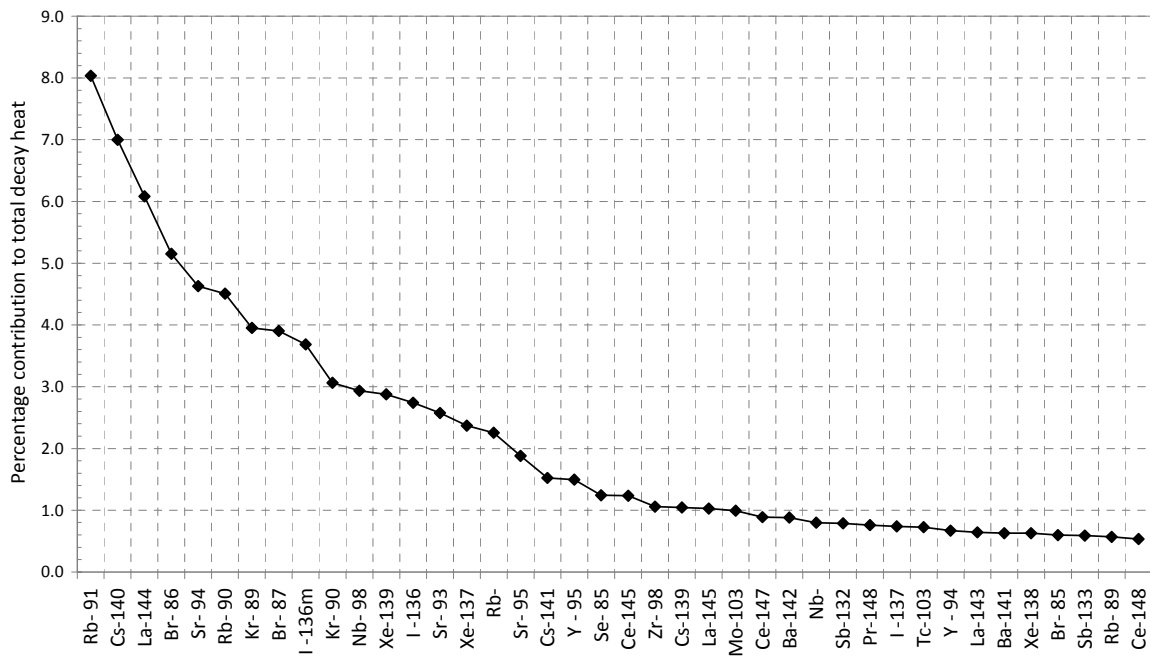
Contributions to decay heat as a function of the cooling time of the thermal-neutron irradiated Th/U fuel are given in Figures 2 (a), (b), (c), (d) and (e) and Tables 1-5. For example, at a cooling time of 1000 s, ^{138}Cs , contributes only 4.25% to the total fission-product decay heat of thermal-neutron irradiated Th/U fuel, but contributes 13.09% at 5000 s. Comparisons have been made with equivalent $^{235}\text{U}/^{239}\text{Pu}$ -based listings to assist in defining the emphasis and choice of radionuclides for dedicated TAGS studies of highest relevance to Th/U fuel. These initial assessments involved consideration of the contributions and identities of only those forty radionuclides calculated to contribute most significantly to the total decay heat for both Th/U and U/Pu types of reactor fuel. Requirements for TAGS measurements were also determined with respect to the following: (a) commonality between Th/U and U/Pu fuels; (b) fission-product decay data needs judged as “more important” for Th/U fuel studies (compared with U/Pu fuel); and (c) TAGS studies undertaken by Greenwood and co-workers at the Idaho National Engineering Laboratory in the 1990s [3, 4].

These comparisons are noted in the Comments columns of the tables for fission products ordered in terms of their estimated percentage contribution to the total decay heat. As noted in Section 1, the TAGS measurements of Greenwood *et al.* [4] have a profound influence on the perceived requirements for improved mean energy and decay scheme data. Their work has addressed and resolved certain inadequacies in decay heat calculations (although not all), and impacts significantly on any request list for further TAGS and decay-data studies.

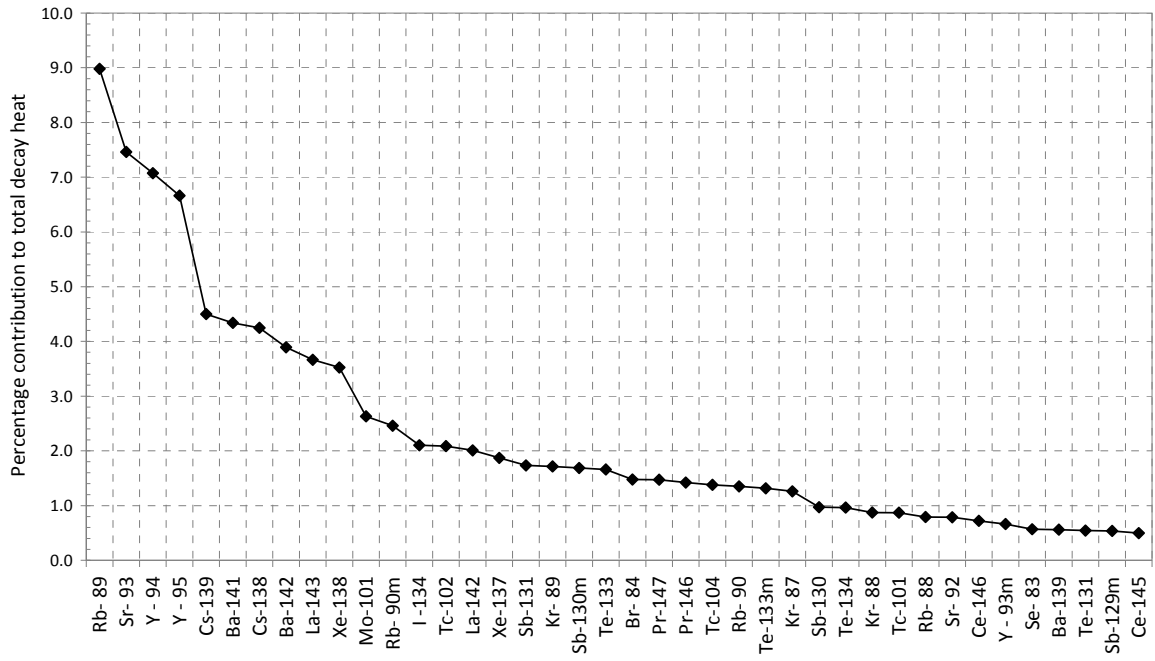
Figure 2: Main fission-product contributors to the decay heat of ^{233}U at cooling times of (a) 10 s, (b) 100 s, (c) 1000 s, (d) 5000 s, and (e) 10 000 s.



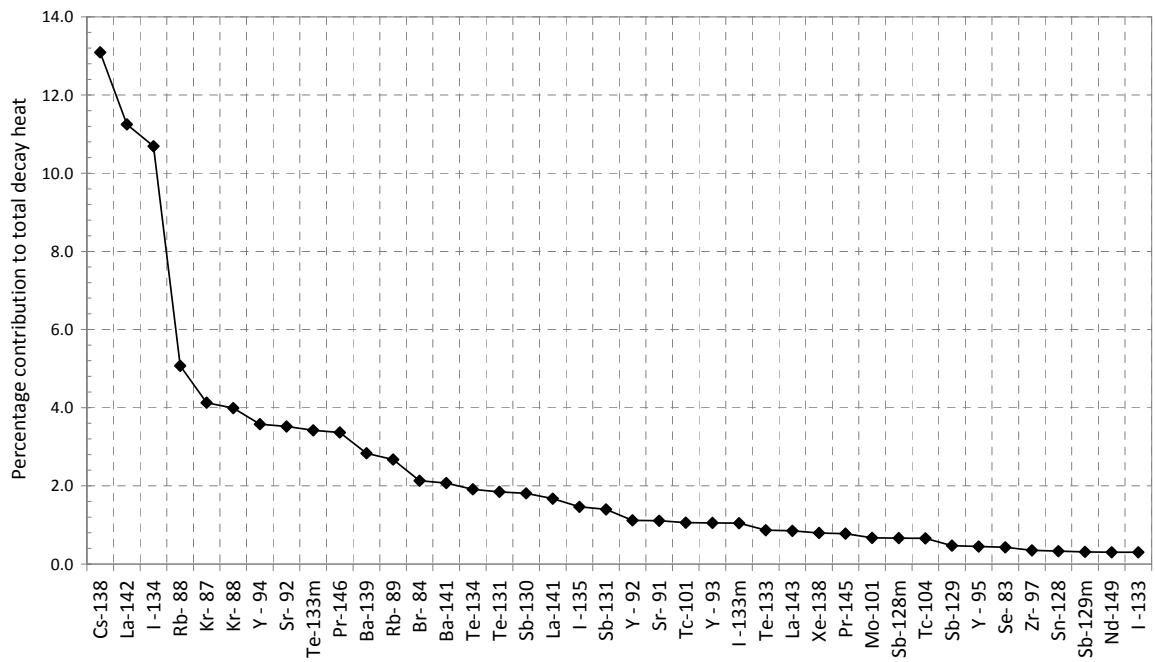
(a) 10 s



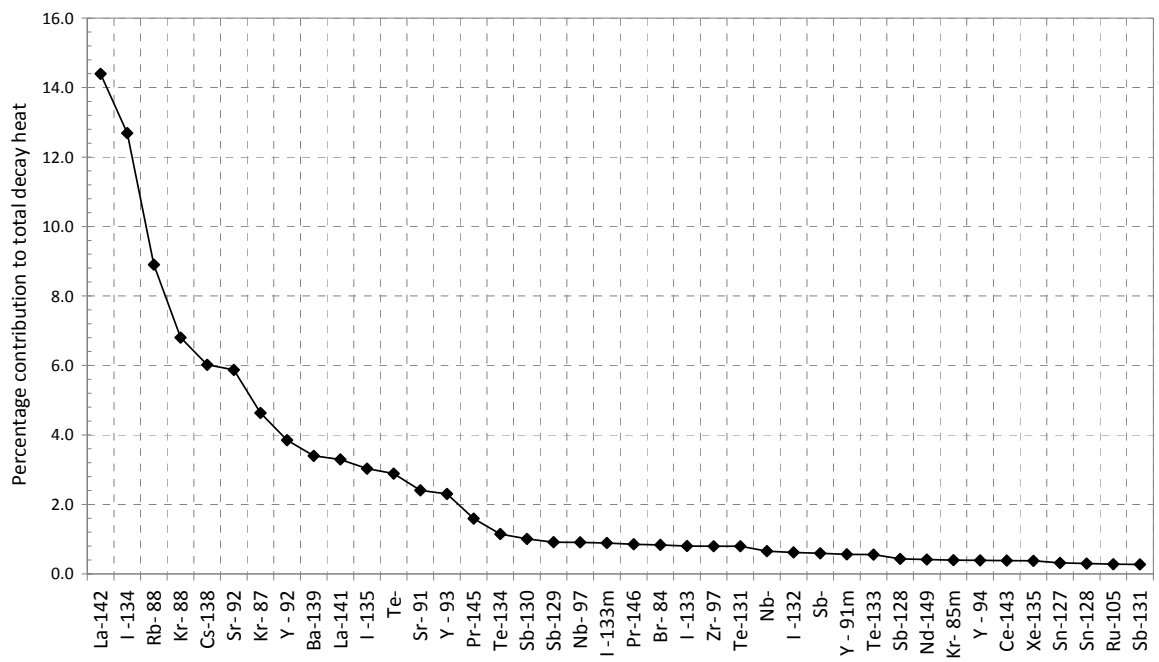
(b) 100 s



(c) 1000 s



(d) 5000 s



(e) 10 000 s

Table 1: Contribution of fission-product radionuclides to the total decay heat of thermal-neutron irradiated Th/U fuel after a cooling time of 10 s.

Radionuclide	Contribution (%)	Comments
37-Rb-92	6.47	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
41-Nb-100	4.76	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
39-Y-96m	4.29	
39-Y-96	4.29	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
56-Ba-143	4.29	TAGS measurements by Greenwood <i>et al</i> [4]
37-Rb-93	3.92	TAGS measurements by Greenwood <i>et al</i> [4]
36-Kr-91	3.18	
38-Sr-95	2.84	TAGS measurements by Greenwood <i>et al</i> [4]
41-Nb-101	2.71	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
35-Br-88	2.54	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
36-Kr-90	2.34	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
41-Nb-98	2.14	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
41-Nb-99	2.10	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
55-Cs-141	2.08	TAGS measurements by Greenwood <i>et al</i> [4]
37-Rb-91	2.04	TAGS measurements by Greenwood <i>et al</i> [4]
56-Ba-144	2.03	TAGS measurements by Greenwood <i>et al</i> [4]
40-Zr-100	1.85	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
57-La-144	1.72	TAGS measurements by Greenwood <i>et al</i> [4]
40-Zr-99	1.69	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
54-Xe-140	1.58	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
53-I-136m	1.55	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
54-Xe-139	1.53	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
55-Cs-140	1.51	TAGS measurements by Greenwood <i>et al</i> [4]
39-Y-97	1.43	
57-La-145	1.27	TAGS measurements by Greenwood <i>et al</i> [4]; also highlighted as repeat by WPEC SG-25 for irradiated U/Pu fuel [1]
35-Br-87	1.24	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
56-Ba-145	1.18	TAGS measurements by Greenwood <i>et al</i> [4]; also highlighted as repeat by WPEC SG-25 for irradiated U/Pu fuel [1]
38-Sr-94	1.16	TAGS measurements by Greenwood <i>et al</i> [4]
35-Br-89	1.16	
35-Br-86	1.15	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
34-Se-86	1.12	
57-La-146m	1.10	
41-Nb-100m	1.10	
53-I-137	1.08	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
52-Te-135	1.05	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
34-Se-85	0.99	
57-La-146	0.96	
41-Nb-102	0.96	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
37-Rb-94	0.96	
41-Nb-102m	0.93	

Table 2: Contribution of fission-product radionuclides to the total decay heat of thermal-neutron irradiated Th/U fuel after a cooling time of 100 s.

Radionuclide	Contribution (%)	Comments
37-Rb-91	8.04	TAGS measurements by Greenwood <i>et al</i> [4]
55-Cs-140	7.00	TAGS measurements by Greenwood <i>et al</i> [4]
57-La-144	6.08	TAGS measurements by Greenwood <i>et al</i> [4]
35-Br-86	5.15	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
38-Sr-94	4.63	TAGS measurements by Greenwood <i>et al</i> [4]
37-Rb-90	4.51	TAGS measurements by Greenwood <i>et al</i> [4]
36-Kr-89	3.95	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
35-Br-87	3.90	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
53-I-136m	3.69	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
36-Kr-90	3.06	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
41-Nb-98	2.94	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
54-Xe-139	2.88	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
53-I-136	2.74	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
38-Sr-93	2.57	TAGS measurements by Greenwood <i>et al</i> [4]
54-Xe-137	2.37	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
37-Rb-90m	2.25	TAGS measurements by Greenwood <i>et al</i> [4]; also highlighted as repeat by WPEC SG-25 for irradiated U/Pu fuel [1]
38-Sr-95	1.88	TAGS measurements by Greenwood <i>et al</i> [4]
55-Cs-141	1.52	TAGS measurements by Greenwood <i>et al</i> [4]
39-Y-95	1.50	TAGS measurements by Greenwood <i>et al</i> [4]
34-Se-85	1.24	
58-Ce-145	1.24	TAGS measurements by Greenwood <i>et al</i> [4]
40-Zr-98	1.06	
55-Cs-139	1.04	TAGS measurements by Greenwood <i>et al</i> [4]
57-La-145	1.03	TAGS measurements by Greenwood <i>et al</i> [4]; also highlighted as repeat by WPEC SG-25 for irradiated U/Pu fuel [1]
42-Mo-103	0.99	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
58-Ce-147	0.89	TAGS measurements by Greenwood <i>et al</i> [4]
56-Ba-142	0.88	TAGS measurements by Greenwood <i>et al</i> [4]
41-Nb-99m	0.80	
51-Sb-132	0.79	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
59-Pr-148	0.76	TAGS measurements by Greenwood <i>et al</i> [4]
53-I-137	0.74	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
43-Tc-103	0.73	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
39-Y-94	0.67	TAGS measurements by Greenwood <i>et al</i> [4]
57-La-143	0.64	TAGS measurements by Greenwood <i>et al</i> [4]; also highlighted as repeat by WPEC SG-25 for irradiated U/Pu fuel [1]
56-Ba-141	0.63	TAGS measurements by Greenwood <i>et al</i> [4]
54-Xe-138	0.63	
35-Br-85	0.60	
51-Sb-133	0.59	
37-Rb-89	0.57	TAGS measurements by Greenwood <i>et al</i> [4]
58-Ce-148	0.54	TAGS measurements by Greenwood <i>et al</i> [4]

Table 3: Contribution of fission-product radionuclides to the total decay heat of thermal-neutron irradiated Th/U fuel after a cooling time of 1000 s.

Radionuclide	Contribution (%)	Comments
37-Rb-89	8.98	TAGS measurements by Greenwood <i>et al</i> [4]
38-Sr-93	7.46	TAGS measurements by Greenwood <i>et al</i> [4]
39-Y-94	7.08	TAGS measurements by Greenwood <i>et al</i> [4]
39-Y-95	6.66	TAGS measurements by Greenwood <i>et al</i> [4]
55-Cs-139	4.50	TAGS measurements by Greenwood <i>et al</i> [4]
56-Ba-141	4.34	TAGS measurements by Greenwood <i>et al</i> [4]
55-Cs-138	4.25	TAGS measurements by Greenwood <i>et al</i> [4]
56-Ba-142	3.89	TAGS measurements by Greenwood <i>et al</i> [4]
57-La-143	3.66	TAGS measurements by Greenwood <i>et al</i> [4]; also highlighted as repeat by WPEC SG-25 for irradiated U/Pu fuel [1]
54-Xe-138	3.53	
42-Mo-101	2.63	
37-Rb-90m	2.46	TAGS measurements by Greenwood <i>et al</i> [4]; also highlighted as repeat by WPEC SG-25 for irradiated U/Pu fuel [1]
53-I-134	2.10	
43-Tc-102	2.09	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
57-La-142	2.01	TAGS measurements by Greenwood <i>et al</i> [4]
54-Xe-137	1.87	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
51-Sb-131	1.74	
36-Kr-89	1.71	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
51-Sb-130m	1.69	
52-Te-133	1.66	
35-Br-84	1.48	
59-Pr-147	1.47	TAGS measurements by Greenwood <i>et al</i> [4]
59-Pr-146	1.42	TAGS measurements by Greenwood <i>et al</i> [4]
43-Tc-104	1.38	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
37-Rb-90	1.35	TAGS measurements by Greenwood <i>et al</i> [4]
52-Te-133m	1.31	
36-Kr-87	1.26	
51-Sb-130	0.97	
52-Te-134	0.96	
36-Kr-88	0.87	
43-Tc-101	0.87	
37-Rb-88	0.79	
38-Sr-92	0.79	
58-Ce-146	0.72	TAGS measurements by Greenwood <i>et al</i> [4]
39-Y-93m	0.66	
34-Se-83	0.57	
56-Ba-139	0.56	
52-Te-131	0.54	
51-Sb-129m	0.54	
58-Ce-145	0.50	TAGS measurements by Greenwood <i>et al</i> [4]

Table 4: Contribution of fission-product radionuclides to the total decay heat of thermal-neutron irradiated Th/U fuel after a cooling time of 5000 s.

Radionuclide	Contribution (%)	Comments
55-Cs-138	13.09	TAGS measurements by Greenwood <i>et al</i> [4]
57-La-142	11.25	TAGS measurements by Greenwood <i>et al</i> [4]
53-I-134	10.69	
37-Rb-88	5.07	
36-Kr-87	4.13	
36-Kr-88	3.99	
39-Y-94	3.58	TAGS measurements by Greenwood <i>et al</i> [4]
38-Sr-92	3.52	
52-Te-133m	3.42	
59-Pr-146	3.37	TAGS measurements by Greenwood <i>et al</i> [4]
56-Ba-139	2.84	
37-Rb-89	2.67	TAGS measurements by Greenwood <i>et al</i> [4]
35-Br-84	2.13	
56-Ba-141	2.07	TAGS measurements by Greenwood <i>et al</i> [4]
52-Te-134	1.91	
52-Te-131	1.85	
51-Sb-130	1.81	
57-La-141	1.67	
53-I-135	1.46	
51-Sb-131	1.40	
39-Y-92	1.12	
38-Sr-91	1.11	
43-Tc-101	1.06	
39-Y-93	1.05	
53-I-133m	1.05	
52-Te-133	0.86	
57-La-143	0.85	TAGS measurements by Greenwood <i>et al</i> [4]; also highlighted as repeat by WPEC SG-25 for irradiated U/Pu fuel [1]
54-Xe-138	0.79	
59-Pr-145	0.78	
42-Mo-101	0.67	
51-Sb-128m	0.66	
43-Tc-104	0.66	Also highlighted by WPEC SG-25 for irradiated U/Pu fuel [1]
51-Sb-129	0.47	
39-Y-95	0.45	TAGS measurements by Greenwood <i>et al</i> [4]
34-Se-83	0.43	
40-Zr-97	0.35	
50-Sn-128	0.33	
51-Sb-129m	0.31	
60-Nd-149	0.30	TAGS measurements by Greenwood <i>et al</i> [4]
53-I-133	0.30	

Table 5: Contribution of fission-product radionuclides to the total decay heat of thermal-neutron irradiated Th/U fuel after a cooling time of 10 000 s.

Radionuclide	Contribution (%)	Comments
57-La-142	14.40	TAGS measurements by Greenwood <i>et al</i> [4]
53-I-134	12.69	
37-Rb-88	8.90	TAGS measurements by Greenwood <i>et al</i> [4]
36-Kr-88	6.81	
55-Cs-138	6.02	
38-Sr-92	5.87	
36-Kr-87	4.63	
39-Y-92	3.85	
56-Ba-139	3.40	
57-La-141	3.30	
53-I-135	3.03	
52-Te-133m	2.89	
38-Sr-91	2.40	TAGS measurements by Greenwood <i>et al</i> [4]
39-Y-93	2.30	
59-Pr-145	1.59	
52-Te-134	1.15	
51-Sb-130	1.00	
51-Sb-129	0.91	
41-Nb-97	0.91	
53-I-133m	0.89	
59-Pr-146	0.85	
35-Br-84	0.83	
53-I-133	0.80	
40-Zr-97	0.80	
52-Te-131	0.80	
41-Nb-97m	0.65	
53-I-132	0.62	
51-Sb-128m	0.59	
39-Y-91m	0.56	
52-Te-133	0.56	
51-Sb-128	0.43	
60-Nd-149	0.41	
36-Kr-85m	0.40	TAGS measurements by Greenwood <i>et al</i> [4]
39-Y-94	0.39	
58-Ce-143	0.38	TAGS measurements by Greenwood <i>et al</i> [4]
54-Xe-135	0.38	
50-Sn-127	0.32	
50-Sn-128	0.30	TAGS measurements by Greenwood <i>et al</i> [4]
44-Ru-105	0.28	
51-Sb-131	0.27	

3.2 Data assessment of fission-product contributors to the decay heat of irradiated Th/U fuel

All relevant decay schemes were assessed to ascertain their complexity and potential for emitting undetected high-energy γ rays from daughter nuclear levels populated by β^- decay. The decay schemes of a significant number of fission products listed in Tables 1-5 are well defined and do not merit either TAGS measurements or further singles γ studies to improve decay heat calculations.

Table 6 constitutes five individual lists of fission products defined in terms of the cooling time that contribute significantly to the decay heat of Th/U fuel and for which TAGS measurements have been successfully undertaken by Greenwood *et al* [4]. While some of these radionuclides would benefit from improved decay-scheme data (e.g. gamma-ray energies and emission probabilities, and beta-particle energies and emission probabilities (especially characterisation of beta decay directly to the ground state of the daughter nucleus)), their mean beta and gamma energies have been reasonably well defined by means of TAGS, and further such studies are not believed to be necessary at this time other than to check the correctness of the existing data.

Table 6: TAGS measurements performed by Greenwood *et al.* [4] of relevance to Th/U fuel.

Cooling time (s)	Radionuclides – TAGS measurements by Greenwood <i>et al.</i> (1997)
10	^{91}Rb , ^{93}Rb , ^{94}Sr , ^{95}Sr , ^{140}Cs , ^{141}Cs , ^{143}Ba , ^{144}Ba , ^{145}Ba , ^{144}La , ^{145}La
100	^{89}Rb , ^{90}Rb , $^{90}\text{Rb}^m$, ^{91}Rb , ^{93}Sr , ^{94}Sr , ^{95}Sr , ^{94}Y , ^{95}Y , ^{139}Cs , ^{140}Cs , ^{141}Cs , ^{141}Ba , ^{142}Ba , ^{143}La , ^{144}La , ^{145}La , ^{145}Ce , ^{147}Ce , ^{148}Ce , ^{148}Pr
1 000	^{89}Rb , ^{90}Rb , $^{90}\text{Rb}^m$, ^{93}Sr , ^{94}Y , ^{95}Y , ^{138}Cs , ^{139}Cs , ^{141}Ba , ^{142}Ba , ^{142}La , ^{143}La , ^{145}Ce , ^{146}Ce , ^{146}Pr , ^{147}Pr
5 000	^{89}Rb , ^{94}Y , ^{95}Y , ^{138}Cs , ^{141}Ba , ^{142}La , ^{143}La , ^{146}Pr , ^{149}Nd
10 000	^{94}Y , ^{138}Cs , ^{142}La , ^{146}Pr , ^{149}Nd

Table 7 lists those fission products of significant importance that merit TAGS measurements within the context of the Th/U and U/Pu fuel cycles. The decay characteristics of these radionuclides are judged to be inadequately known, and need to be much better defined for their confident adoption in decay heat calculations for both fuel types. These radionuclides are important contributors to the decay heat of thermal-neutron irradiated ^{233}U fuel, as used in the Th/U cycle, and were also assessed as ill-defined and in need of TAGS studies by WPEC SG-25 (see Table 3 in Ref. [1]). Thus, the requirements for TAGS measurements and associated decay scheme data for these particular fission products are seen to have further increased in importance because of the common needs arising from power applications of the two differing fuel cycles.

Table 7: Requested TAGS measurements of common importance to both Th/U and U/Pu fuel (see also Table 3 in Ref. [1]).

Cooling time (s)	Radionuclides – Th/U and U/Pu fuel
10	^{86}Br , $^{87}\text{Br}(\beta^-, n)$, $^{88}\text{Br}(\beta^-, n)$, ^{90}Kr , ^{92}Rb , ^{96}Y , ^{99}Zr , ^{100}Zr , ^{98}Nb , ^{99}Nb , ^{100}Nb , ^{101}Nb , ^{102}Nb , ^{135}Te , $^{136}\text{I}^m$, $^{137}\text{I}(\beta^-, n)$, ^{139}Xe , ^{140}Xe
100	^{86}Br , $^{87}\text{Br}(\beta^-, n)$, ^{89}Kr , ^{90}Kr , ^{98}Nb , ^{103}Mo , ^{103}Tc , ^{132}Sb , ^{136}I , $^{136}\text{I}^m$, $^{137}\text{I}(\beta^-, n)$, ^{137}Xe , ^{139}Xe
1 000	^{89}Kr , ^{102}Tc , ^{104}Tc , ^{137}Xe
5 000	^{104}Tc
10 000	–

Fission products assessed in terms of their primary importance to the Th/U fuel cycle are defined in Table 8. The decay schemes of all radionuclides listed in Tables 1-5 were assessed further from the point of view of comparisons of their Q-values (Audi *et al.* [10]) with the level energies of the daughter nucleus populated by β^- -decay (ENSDF [11]). Energy differences of greater than ~ 1 MeV between the reasonably well-defined Q-value and the highest known level energy of a specific radionuclide were seen as supportive evidence for the need to undertake exploratory TAGS measurements in order to quantify the extent of ‘‘Pandemonium’’ and derive the necessary mean beta and gamma energies for decay heat calculations. Assessments of the relevant decay schemes were also

carried, and a refined list of fission products was produced in terms of the radionuclides of importance to the Th/U fuel cycle that would or might benefit from TAGS studies (Table 9).

Table 8: Radionuclides of high relevance to Th/U decay heat calculations, but not included in Tables 6 and 7, above.

Cooling time (s)	Radionuclides
10	^{85}Se , ^{86}Se , ^{89}Br , ^{91}Kr , ^{94}Rb , $^{96}\text{Y}^m$, ^{97}Y , $^{100}\text{Nb}^m$, $^{102}\text{Nb}^m$, ^{146}La , $^{146}\text{La}^m$
100	^{85}Se , ^{85}Br , ^{98}Zr , $^{99}\text{Nb}^m$, ^{133}Sb , ^{138}Xe
1 000	^{83}Se , ^{84}Br , ^{87}Kr , ^{88}Kr , ^{88}Rb , ^{92}Sr , $^{93}\text{Y}^m$, ^{101}Mo , ^{101}Tc , $^{129}\text{Sb}^m$, ^{130}Sb , $^{130}\text{Sb}^m$, ^{131}Te , ^{133}Te , $^{133}\text{Te}^m$, ^{134}Te , ^{138}Xe , ^{139}Ba
5 000	^{83}Se , ^{84}Br , ^{87}Kr , ^{88}Kr , ^{88}Rb , ^{91}Sr , ^{92}Sr , ^{92}Y , ^{93}Y , ^{97}Zr , ^{101}Mo , ^{101}Tc , ^{128}Sn , $^{128}\text{Sb}^m$, ^{129}Sb , $^{129}\text{Sb}^m$, ^{130}Sb , ^{131}Sb , ^{131}Te , ^{133}Te , $^{133}\text{Te}^m$, ^{134}Te , ^{133}I , $^{133}\text{I}^m$, ^{134}I , ^{135}I , ^{138}Xe , ^{139}Ba , ^{141}La , ^{145}Pr
10 000	^{84}Br , $^{85}\text{Kr}^m$, ^{87}Kr , ^{88}Kr , ^{88}Rb , ^{91}Sr , ^{92}Sr , $^{91}\text{Y}^m$, ^{92}Y , ^{93}Y , ^{97}Zr , ^{97}Nb , $^{97}\text{Nb}^m$, ^{105}Ru , ^{127}Sn , ^{128}Sn , ^{128}Sb , $^{128}\text{Sb}^m$, ^{129}Sb , ^{130}Sb , ^{131}Sb , ^{131}Te , ^{133}Te , $^{133}\text{Te}^m$, ^{134}Te , ^{132}I , ^{133}I , $^{133}\text{I}^m$, ^{134}I , ^{135}I , $^{135}\text{I}^m$, ^{135}Xe , ^{139}Ba , ^{141}La , ^{143}Ce , ^{145}Pr

Table 9: Recommended TAGS measurements for radionuclides with decay schemes judged to possess the potential to suffer from ‘‘Pandemonium’’ – important to Th/U decay-heat calculations, and not included in Tables 6 and 7, above.

Cooling time (s)	Radionuclides	
	Priority 1	Priority 2
10	^{89}Br , ^{91}Kr , ^{94}Rb , $^{96}\text{Y}^m$, ^{97}Y , $^{100}\text{Nb}^m$, $^{102}\text{Nb}^m$	^{86}Se , $^{146}\text{La}^m$
100	^{85}Se , ^{98}Zr (?)	$^{99}\text{Nb}^m$, ^{133}Sb
1 000	^{101}Mo , $^{130}\text{Sb}^m$, ^{138}Xe	^{84}Br , ^{87}Kr , ^{92}Sr , $^{129}\text{Sb}^m$
5 000	–	^{87}Kr , ^{88}Rb , ^{92}Sr , $^{128}\text{Sb}^m$, $^{129}\text{Sb}^m$, ^{139}Ba , ^{141}La
10 000	–	^{87}Kr , ^{88}Rb , ^{92}Sr , $^{128}\text{Sb}^m$, ^{139}Ba , ^{141}La

4. Discussion

A significant difficulty in an attempt to assess decay-heat calculations for the Th/U fuel cycle is the lack of any suitable benchmark measurements of well-characterised fuel. While such studies have been extensively undertaken for U/Pu, decay-heat measurements are relatively sparse for thermal-neutron irradiated Th/U fuel [12]. Yarnell and Bendt have determined the total decay heat for thermal-neutron irradiated ^{233}U at cooling times from 20 to 100 000 s and total uncertainties ranging from 5.3% to 6.5%, respectively, by means of a fast-response cryogenic boil-off calorimeter [13]. Equivalent studies were undertaken by Journey *et al.* to quantify the total gamma energy for ^{233}U thermal-neutron fission at twelve different cooling times ranging from 29 to 146500 s [14]. Measured and calculated total gamma energies were approximately the same at cooling times longer than 10 000 s, while the measured values were approximately 14% larger than calculated energies over shorter cooling periods. A combination of NaI(Tl) and well-type plastic scintillators has been used by Akiyama *et al.* to measure the mean beta and gamma components of decay heat, respectively, following the fast-neutron irradiation of ^{233}U at cooling times from 19 to 24 000 s. It is seen that uncertainties in the measured beta energy release per fission varied between 3.3% and 5.8%, while uncertainties in the measured gamma energy release per fission varied between 2.6% and 6.0% [15, 16, 17].

The limited experimental study of ^{233}U decay heat is rather unfortunate, and makes a clearly focused review of decay-data needs more difficult to carry out with confidence. Nevertheless, available fission-yield data and known Q-values have been adopted to generate what are believed to be reasonable estimates of the total decay heat as a function of cooling time along with the relative contributions of individual fission products. The apportioning of decay energy between the light-

particle and electromagnetic components is much more uncertain, and requires improvement through a cycle of (a) comparison of calculations against benchmarks, followed by (b) TAGS and decay-scheme measurements to define (c) adjustments in the relevant decay-data files, and (d) re-calculation and comparison.

Despite the existing situation, decay-heat calculations for thermal neutron induced fission of ^{233}U fuel were performed, and a review was undertaken of the quality of the known decay data for the forty main contributors at a given cooling time to the calculated total decay heat. These calculations were compared with equivalent lists for U/Pu fuels. Judgements were made with respect to those radionuclides which are (a) common to both fuel types, and (b) of greater significance to Th/U fuel decay-heat calculations. This exercise also involved consideration of an important set of TAGS measurements undertaken in the 1990s by Greenwood *et al.* [4] and radionuclides were removed from the evolving TAGS request list for Th/U fuel if they had already been studied by Greenwood and co-workers.

The fission products listed in Table 7 are deemed to be important for both Th/U and U/Pu decay heat studies and would benefit from TAGS measurements. Much of the focus of this request list and the similar tabulation of WPEC SG-25 remain on the radioisotopes of a limited number of specific elements: Br-Kr, Zr-Nb-Tc, and I-Xe. The contents of Table 7 represent a contrasting combination of volatile (*i.e.*, Br, Kr, I and Xe) and refractory elements (*i.e.*, Zr, Nb and Tc), of which the latter have historically proven more difficult to purify and prepare as suitable source materials. This problem has been largely overcome [6], and the expectation would be that TAGS measurements could be successfully performed. Evidence that all fuel types would benefit from the same improved decay data adds considerable support to the need to carry out TAGS studies on these particular radionuclides

Additional fission products of high significance in Th/U decay-heat calculations are listed separately in Table 8. Nuclear structure analyses were carried out for all of these fission products in order to derive some guidance concerning those radionuclides that would benefit from TAGS measurements. In effect, the aim was to identify the nuclides most likely to exhibit “Pandemonium”. As described in Section 3, comparisons were made of evaluated Q-values for β^- decay [10] and the high-energy nuclear levels of the daughter as determined from the evaluation of measured spectral data [11]. This situation was assessed with respect to the completeness and overall consistency of the known decay scheme, and avoidance of the artificial creation of forbidden β^- transitions. While the full validity of such subjective analytical assessments is debatable and open to criticism involving the lack of known nuclear properties, this approach is believed to furnish some useful guidance to experimentalists. The final recommendations and proposed priorities are based on this qualitative assessment, and are given in Table 9.

A number of specific observations affecting the assembly of Table 9 are worthy of note to explain the exclusion of some metastable states and the uncertain inclusion of a radionuclide possessing an extremely simple decay scheme:

- 1). Some of the main contributors to the fission-product decay at cooling times above 100 s are metastable states that undergo 100% isomeric transition decay – $^{93}\text{Y}^m$, $^{97}\text{Nb}^m$ and $^{133}\text{I}^m$ – and arise directly from parent β^- decay. Their decay characteristics are inappropriate for TAGS studies, and are not included for this reason in Table 9.
- 2). $^{91}\text{Y}^m$ undergoes greater than 98.5% isomeric-transition decay, and therefore this radionuclide does not merit TAGS measurements.
- 3). ^{98}Zr is calculated to contribute 1.06% of the total fission-product decay heat at a cooling time of 100 s. The decay scheme for this radionuclide is a single beta-particle emission directly to the ground state of daughter ^{98}Nb , with a Q-value of 2250(20) keV. No gamma rays have been observed, and TAGS measurements could well furnish no evidence for “Pandemonium” (hence the uncertainty registered against this radionuclide in Table 9).

5. Conclusions

Calculations and analyses of the fission products that contribute significantly to the decay heat of thermal-neutron irradiated ^{233}U fuel have been conducted and a list of radionuclides suitable for TAGS measurements (Table 9) has been recommended with the aim of improving the data input to such calculations. As noted above, a subjective set of judgements were made to identify those fission products with decay scheme characteristics that would/might benefit significantly from TAGS and γ -ray singles studies. A combination of factors were considered: (a) percentage contribution to the total fission-product decay heat at well-defined cooling times; (b) likelihood and extent of adopting ill-defined mean beta-particle and gamma-ray energies (“Pandemonium”); and (c) assessment of the nature and perceived completeness of the evaluated decay scheme. All three factors were combined together to formulate the final set of recommendations to be found in Table 9. The refined contents of this table are based on assessments of the known nuclear structure of all possible candidates listed in Table 8, and the inclusion of only those radionuclides that might exhibit significant inadequacies identified with the known inability to detect and quantify high-energy γ rays from their β^- decay. Along with the contents of Table 7, the radionuclides listed in Table 9 represent the requirements for improved nuclear decay data in order to achieve improvements in the decay heat calculations for thermal-neutron irradiated ^{233}U fuel.

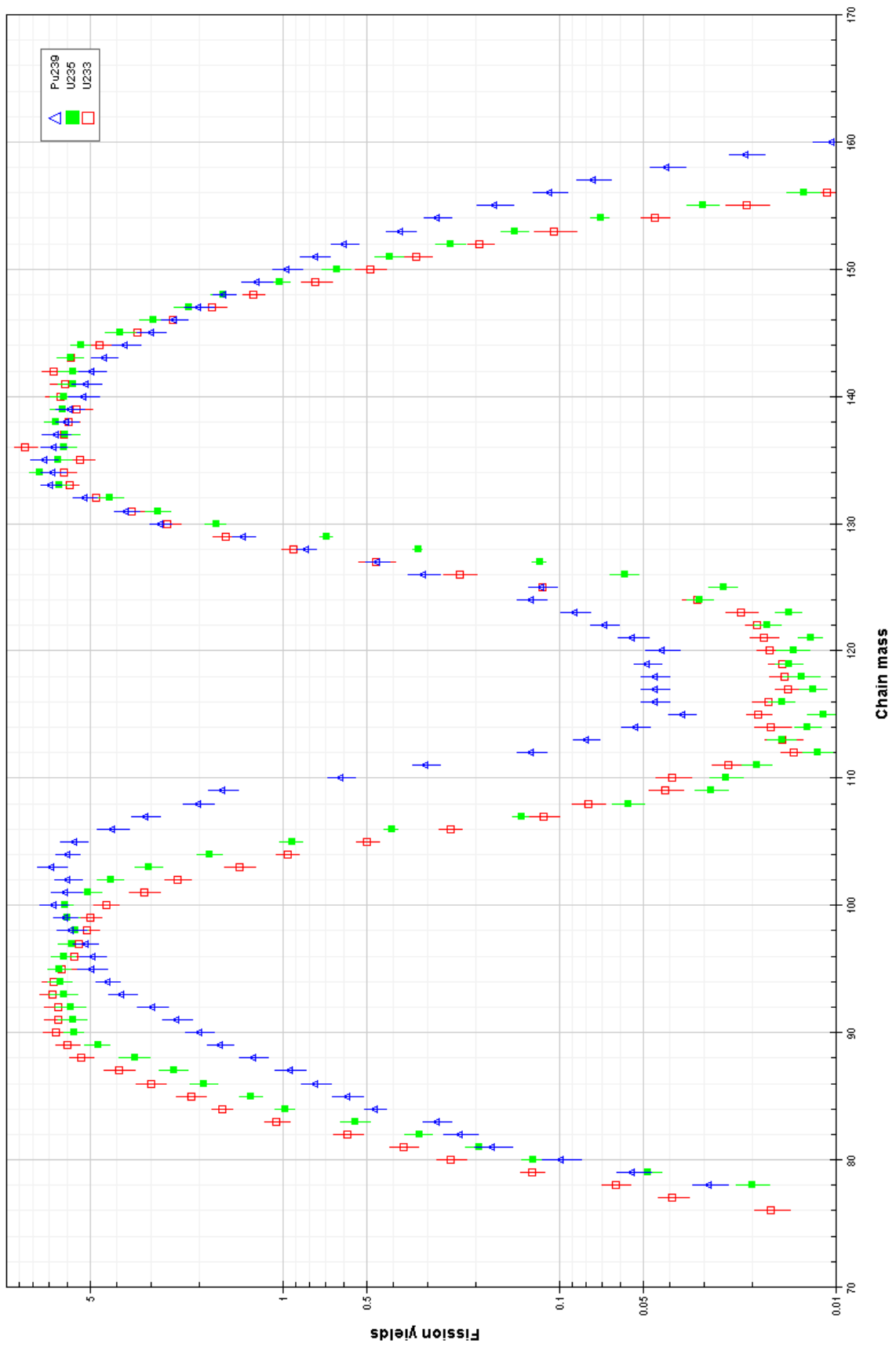
A major inadequacy in the assessment exercise for the Th/U fuel cycle has been the serious lack of appropriate decay-heat measurements. These studies would have greatly benefited from an extensive series of benchmark experiments under well-controlled irradiation conditions. Comparative studies between decay-heat calculations and substantial benchmark data would aid immensely in identifying and focusing on the most likely fission products that contribute significantly to ^{233}U decay heat and possess incomplete decay-scheme data within the multinational and national nuclear applications files. Well-defined benchmark experiments need to be made of the total decay heat and component contributions (\overline{E}_{LP} and \overline{E}_{EM}) generated during the Th/U fuel cycle.

Suitable TAGS facilities are operational in Europe, and are in the process of being established elsewhere (India and USA). The assembly of a TAGS spectrometer at VECC, Kolkata, India, represents an exciting opportunity to undertake such studies of some of the radionuclides of greater importance to the Th/U fuel cycle [5]. Full support should be given to the experimental team responsible for the measurement programmes to be undertaken at this facility. Such work would also benefit from the forging of strong links with other relevant on-going studies worldwide, as encouraged by staff at the IAEA Nuclear Data Section and OECD Nuclear Energy Agency. Further discussions and analyses should be initiated when judged appropriate, after substantial progress has been made in the experimental studies of relevance to the Th/U fuel cycle.

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Independent Chain Mass Fission Yields for ^{233}U , ^{235}U and ^{239}Pu .

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