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Assessment of Fission Product Decay Data for Decay Heat Calculations

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ASSESSMENT OF FISSION PRODUCT DECAY DATA FOR DECAY HEAT CALCULATIONS

A report by the Working Party
on International Evaluation Co-operation
of the NEA Nuclear Science Committee

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FOREWORD

A Working Party on International Evaluation Co-operation was established by the OECD/NEA Nuclear Science Committee (NSC) to promote the exchange of information on nuclear data evaluations, validation and related topics. Another specific aim is to provide a framework for co-operative activities between the members of the major nuclear data evaluation projects. This initiative includes the possible exchange of scientists in order to encourage co-operation. Requirements for experimental data resulting from this activity are also compiled. The working party determines common criteria for evaluated nuclear data files in order to assess and improve the quality and completeness of evaluated data.

The parties to the project are: ENDF (United States), JEFF/EFF (member countries of the NEA Data Bank) and JENDL (Japan). Co-operation with evaluation projects of non-OECD countries, specifically the Russian BROND and Chinese CENDL projects, are organised through the Nuclear Data Section of the International Atomic Energy Agency (IAEA).

This publication presents the conclusions of the work undertaken by Subgroup 25, which focused on the assessment and improvement of the evaluated decay data sub-libraries in order to obtain more accurate estimations of decay heat. Recommendations have been prepared for total absorption gamma-ray spectroscopy (TAGS) measurements of specific fission product nuclides to be undertaken in close collaboration with experimentalists in Subgroup 25.

The opinions expressed in this report are those of the authors only, and do not necessarily represent the position of any member country or international organisation. This report is published on the responsibility of the Secretary-General of the OECD.

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SUMMARY

WPEC Subgroup 25 was formed in 2005 in response to the fact that different national and international decay data sub-libraries did not generate good estimates of decay heat without the introduction of additional data produced by means of various theoretical calculations (e.g. JENDL-FPDD, JEFF-3.1/RDD and a preliminary version of the decay data sub-library in ENDF/B-VII). Although the input data to the calculations have sound theoretical bases for their adoption, specific values are not always sufficiently reliable because they are dependent on the choice of the parameters used in such studies. Calculations that adopt oft neglected mean decay data derived from total absorption gamma-ray spectroscopy (TAGS) are able to describe the total decay heat reasonably well, although the component light-particle (beta) and electromagnetic (gamma) contributions may differ significantly from equivalent benchmark measurements and decay heat standards. Two subgroup meetings were held in collaboration with the Nuclear Data Section of the International Atomic Energy Agency (IAEA). The first meeting was held in December 2005, and a follow-up meeting was held in May 2006. Various presentations and discussions at the two meetings have assisted greatly in the assembly of a list of radionuclides recommended for dedicated TAGS measurements. A TAGS research group from Valencia (Instituto de Física Corpuscular) in Spain joined and strengthened the membership of the subgroup, and described their existing measurement programme at the University of Jyväskylä in Finland, along with their capabilities and future plans. Good progress was also made in the identification of other suitable TAGS facilities in Europe. Plans for the ALTO facility at IPN-Orsay represent an additional opportunity for undertaking TAGS measurements, particularly on refractory fission products. Members of Subgroup 25 met with IPN-Orsay management to discuss the importance of new TAGS data for decay heat calculations at this new facility.

Several comparisons between the relevant files in JENDL-FPDD, JEFF-3.1/RDD and the decay data sub-library in ENDF/B-VII have been made and a need has been identified to determine the origins of some of the important mean energy data in the JENDL-FPDD files. Specific actions that arose from the two subgroup meetings are addressed in Appendix A.

Recommendations are made that focus on particular fission-product radionuclides that merit measurement by TAGS in order to improve decay heat calculations without the need to resort to theory. Progress made by the experimentalists over 2006-2008 should be monitored within the WPEC (and/or by the IAEA), with the aim of incorporating TAGS-based mean energy data into the existing decay data sub-libraries.

Web pages have been prepared at the NEA and IAEA on which presentations from the meetings can be found:

- *http://www.nea.fr/html/science/wpec/SG25/*
- *http://www-nds.iaea.org/beta_decay/*

BACKGROUND

Confident quantification of the decay heat induced by nuclear fission within a power reactor is an extremely important factor in the design of nuclear facilities for electricity generation and for the post-irradiation handling of nuclear fuels (fuel discharge, storage, transport and reprocessing, and waste handling). The total decay heat (as well as quantification of the light-particle and electromagnetic components) as a function of cooling time impacts significantly on the safe operation and various economic aspects of nuclear power generation. Such quantitative studies require comprehensive sets of nuclear data: neutron cross-sections, fission yields and decay data (primarily for fission products and actinides – half-lives, and mean light-particle and electromagnetic energies), and sound estimates of the uncertainties in these data.

During the course of the 1970s, confidence had grown sufficiently in the ability to undertake decay heat calculations with the various available databases, as demonstrated world wide (particularly in France, Japan, UK and USA). However, these calculations required the inclusion of mean beta and gamma energies derived from the Gross Theory of Beta Decay [1,2] for a significant number of fission products (e.g. in the JNDC-V2 and ENDF/B-VI databases) in order to achieve satisfactory agreement with decay heat benchmarks [3,4].

When the actinide and fission-product inventories have been calculated for the specified conditions of reactor operation and subsequent cooling period, the decay heat can be derived by summing the products of the nuclear activities in terms of the mean heavy-particle, light-particle and electromagnetic energy releases per disintegration of that nuclide:

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

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

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

where E^i_{HP}, E^i_{LP} and E^i_{EM} are the mean heavy-particle, light-particle and electromagnetic energy releases respectively per disintegration of nuclide *i*; λ^T_i 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, but are sometimes referred to collectively as "alpha"; light particles are defined as negatrons, positrons, Auger electrons and conversion electrons, but are sometimes referred to collectively as "beta"; electromagnetic radiation is defined as gamma rays, X-rays, annihilation radiation and internal bremsstrahlung, but are sometimes referred to collectively as "gamma".

The nuclear data requirements for decay-heat calculations can be determined from the information given above, and include the need to define the following parameters:

$\sigma^F_{a,k}$	effective group-averaged fission cross-section of actinide *a* in the k^{th} neutron group;
$\sigma^A_{i,j}$	total neutron absorption cross-section of fission product *i*;
$\sigma^{(n,\gamma)}_{i,j}$	(n,γ) cross-section of fission product *i*;
$\sigma^{(n,2n)}_{i,j}$	(n,2n) cross-section of fission product *i*;
$Y^i_{a,k}$	independent yields for fission product *i*;
λ_i	decay constant(s) of fission product *i*;
E^i_{HP}, E^i_{LP}, E^i_{EM}	mean heavy-particle, light-particle and electromagnetic energy releases per disintegration of nuclide *i*;

as well as:

k_α, k_{β^-}, k_{β^+}	branching fractions for α, β^- and β^+ decay to (Z,A) nuclide, as used in associated inventory calculations.

Recent years have seen the evolution of total absorption gamma-ray spectroscopy (TAGS) [5-8] at a number of experimental facilities (e.g. INEL, ISOLDE and the University of Jyväskylä) that have permitted comprehensive studies and quantification of some of the fission products whose mean energies were derived from the Gross Theory of Beta Decay. However, when the mean energies derived from the Gross Theory of Beta Decay are replaced with well-defined data derived from TAGS, there is a decline in the existing agreement between the resulting decay heat calculations and benchmark experiments [9,10].

A WPEC subgroup (SG) proposal was prepared by Yoshida to address the need for additional fission-product decay data to be derived experimentally for decay heat calculations. This proposal was accepted in April 2005, and the various outcomes of the work of the resulting subgroup (SG25) are presented in this report.

INTRODUCTION

Subgroup proposal to WPEC

Combinations of inventory and summation calculations are used to predict various aggregate properties of the fission products (FP), including FP decay heat. Such decay heat calculations are used to determine the sum total of the energy released from all the individual β-decaying nuclides that exist at well-defined cooling times. Therefore, data that characterise the β-decay of individual FP nuclides are indispensable prerequisites for summation calculations along with the fission yield data. Once a full set of fission product yields and average mean β- and γ-ray energy releases per decay ($\bar{E}_\beta$ and $\bar{E}_\gamma$) are known, decay heat as a function of cooling time can be calculated with confidence. As far as the decay data are concerned, the $\bar{E}_\beta$ and $\bar{E}_\gamma$ values are usually obtained directly from the discrete decay data and published decay schemes such as those to be found in the Refs. [11,12].

As noted in the late 1970s by Hardy, *et al.* [13], there is a danger when using conventional γ-ray spectroscopy techniques of not detecting and therefore overlooking the existence of β-feeding into the high-energy nuclear levels of daughter nuclei that are associated with decay schemes possessing significant Q-values. This phenomenon is commonly called the "pandemonium effect". Yoshida and Nakasima noted that FP decay heat calculations at that time suffered inevitably from this problem through the use of average β^- and γ-ray energies per decay derived from these incomplete decay schemes [2]. They proposed a method of compensation for these missing β-strengths on the basis of the Gross Theory of Beta Decay. Employing this method, the calculated decay heat became more consistent with measured decay heat, except for cooling times between 300 and 3 000 s. Results from comparisons between decay heat measurements and JENDL + theory data can be found in Ref. [14]. Despite this success, the introduction of theoretical data into the summation calculations was considered to be only a temporary solution. Theoretically calculated $\bar{E}_\beta$ and $\bar{E}_\gamma$ data should be replaced by values based on experimental studies when deemed appropriate.

A total absorption γ-ray spectrometer (TAGS) was developed at the Idaho National Engineering Laboratory in the 1990s [6,15], and has been applied to the study of a significant number of short-lived FP nuclides. This group determined the β-feeding transition probabilities of various nuclides, including many important isotopes in the FP region. Decay data derived from TAGS measurements are free from the pandemonium effect, and such studies are particularly powerful if they include a determination of the β-particle emission probability directly to the ground state of the daughter nuclide. A European group began a series of TAGS experiments recently for several important FP nuclides using an on-line mass separator located at the University of Jyväskylä [16].

As an exploratory test, $\bar{E}_\beta$ and $\bar{E}_\gamma$ values calculated from the INEL data were substituted into the FP decay data files to be found in JENDL, JEF-2.2 and ENDF/B-VI. Both these modified and the original decay data files were used to calculate the decay heat after a thermal fission burst, and the results compared with integral measurements from the University of Tokyo [17]. Results for ^{239}Pu are shown in Figures 1 to 6 – the most significant impact of this study can be seen in the electromagnetic (gamma) component. No theoretical corrections had been made for the missing β-strengths in the JEF-2.2 sub-library, and the inclusion of TAGS-derived data in these particular files exhibited remarkable improvements between the decay heat standards and new calculations. Such observations imply that the inclusion of TAGS data in which the higher-energy β strengths have been detected reflects these contributions correctly. However, when the correction is applied on the basis of Gross beta theory [JENDL (solid curves)], this good agreement is not maintained as the measured mean energies are subsequently introduced. The electromagnetic component is overestimated at cooling times ranging from 3 to 300 seconds – ^{141}Cs and ^{144}La are the dominant γ-ray emitting radionuclides over these cooling times when the TAGS data are introduced and replace $\bar{E}_\gamma$ values from JENDL.

On the basis of the above observations, there would appear to have been sufficient accumulation of experimental TAGS and β-feeding data over the previous 15 years for decay heat calculations to be undertaken with minimal assistance from nuclear theory (with a number of notable exceptions). The following points are proposed in order to make full use of these accumulated data, and so improve the reliability of the summation calculations.

1. Identify the nuclides which contribute significantly to FP decay heat over cooling times ranging from 10 to 5 000 seconds.

2. Assess which of these nuclides may suffer from inadequate decay data due to the “pandemonium effect”.

3. Identify the need for new β-feeding measurements using TAGS, along with the determination of β-decay directly to the daughter ground state.

Figure 1. Light-particle decay heat following thermal fission burst of ^{239}Pu – data from JENDL, JEF-2.2, JEFF-3.1 and ENDF/B-VI are shown together with experimental data from Yayoi, Lowell and Oak Ridge National Laboratory

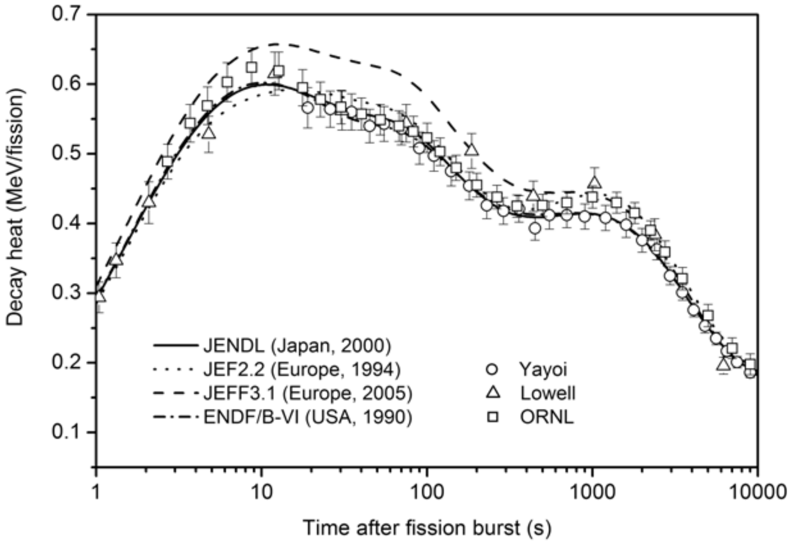


Figure 2. Light-particle decay heat following thermal fission burst of ^{239}Pu (as in Figure 1), with and without TAGS data being adopted in JENDL and JEFF-3.1

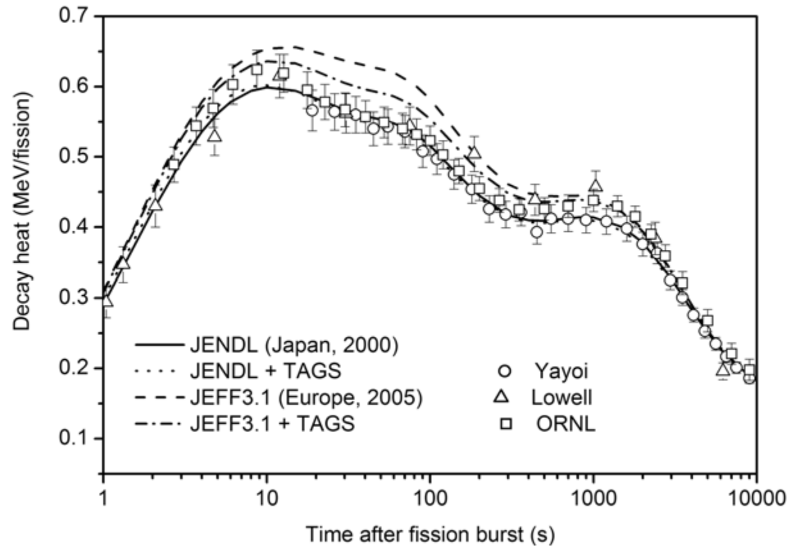


Figure 3. Electromagnetic decay heat following thermal fission burst of ^{239}Pu – data from JENDL, JEF-2.2, JEFF-3.1 and ENDF/B-VI are shown together with experimental data from Yayoi, Lowell and Oak Ridge National Laboratory

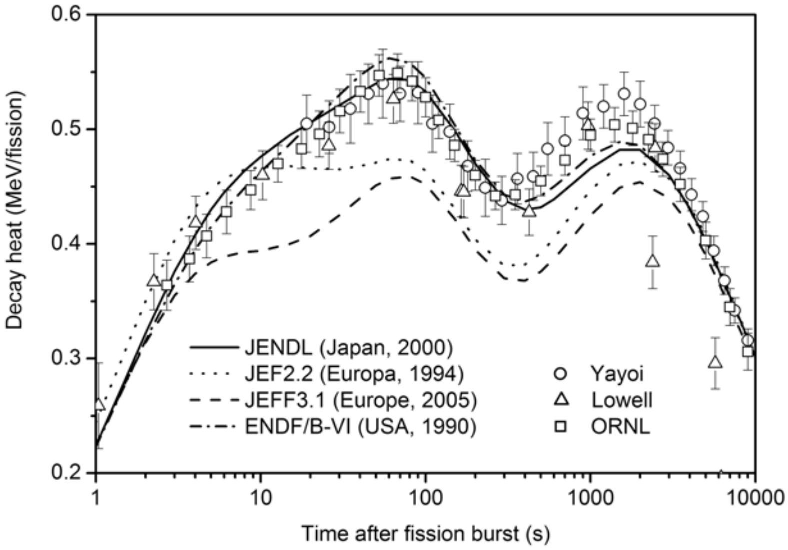


Figure 4. Electromagnetic decay heat following thermal fission burst of ^{239}Pu (as in Figure 3), with and without TAGS data being adopted in JENDL and JEFF-3.1

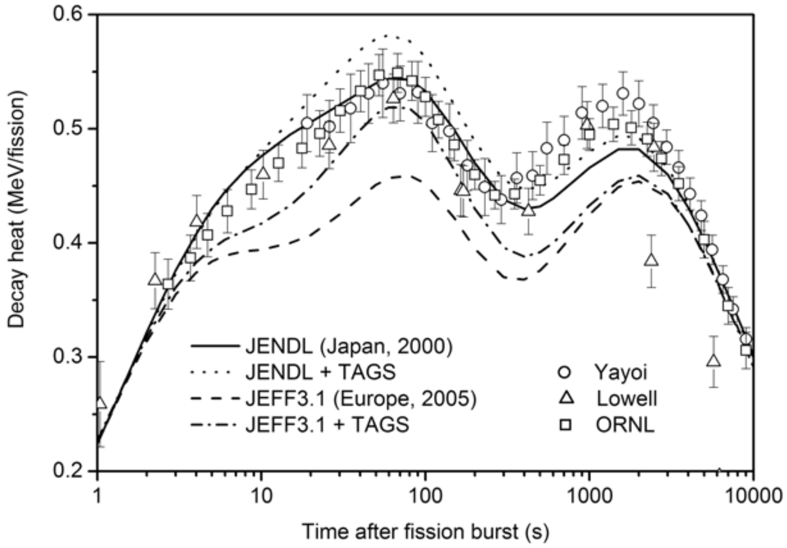


Figure 5. Total decay heat following thermal fission burst of ^{239}Pu – data from JENDL, JEF-2.2, JEFF-3.1 and ENDF/B-VI are shown together with experimental data from Yayoi and Oak Ridge National Laboratory

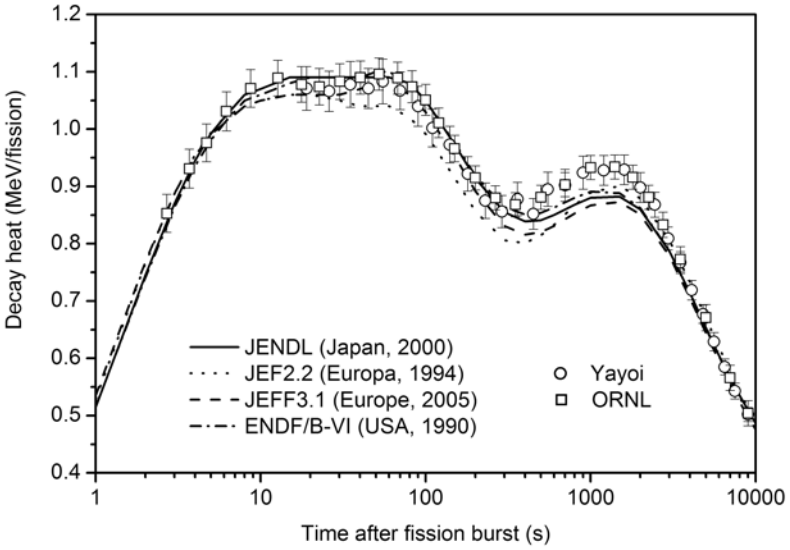
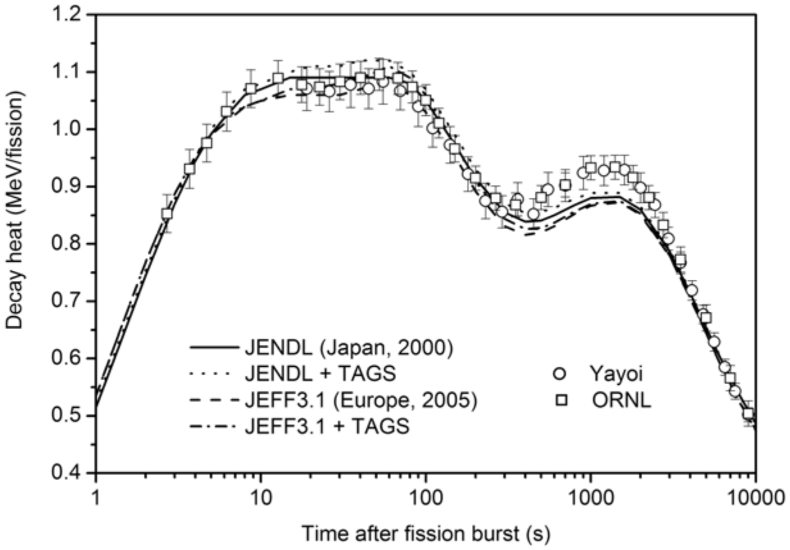


Figure 6. Total decay heat following thermal fission burst of ^{239}Pu (as in Figure 5), with and without TAGS data being adopted in JENDL and JEFF-3.1



SUBGROUP ACTIVITIES

Subgroup 25 was formed in 2005 to assess and recommend improvements to the fission product decay data for decay heat calculations. Two subgroup meetings were held to discuss the beta-decay process, disagreements between decay heat calculations and experimental data, and the feasibility of undertaking TAGS measurements to improve the existing situation. Such informed debate provided the necessary guidelines for a future programme of TAGS experiments.

Summary of the first SG25 meeting

A first meeting of WPEC Subgroup 25 entitled "Beta-decay and decay heat" was held in collaboration with the IAEA on 12-14 December 2005, in Vienna, Austria. The participants elected Professor W. Gelletly as Chairman, and A.L. Nichols as Secretary – a full Summary Report was issued as an IAEA report [INDC(NDS)-0483].

A number of presentations focused on the need for more accurate decay heat calculations. Decay heat contributes 60% of the risk of radioactive release into the environment, and a continuing requirement remains for active decay heat removal systems. Furthermore, the existing safety criteria include the need to know precisely the amount of resulting decay heat in order to assess both the core and containment strategies during the analysis and control of an abnormal event. A listing of potential sources of decay heat encompasses the quantification of unstable fission products and actinide nuclei produced by successive neutron capture on U and Pu. There are known gaps in the available data required for decay heat calculations, and consideration needs to be given to what can be successfully produced and studied at radionuclide "manufacturing" facilities such as ISOLDE at CERN.

Presentations

Gelletly described the β-decay process along with consideration of Fermi and Gamow-Teller interactions and their selection rules. There has been a significant increase in the study of exotic nuclei and their decay properties, particularly with the development of such facilities as IGISOL (Ion-guide Isotope Separator On-line) and TAGS. Various isotope production techniques were described leading up to IGISOL (development and extension of the He-jet technique at the University of Jyväskylä) which is chemistry-independent and provides ideal input to a mass separator for subsequent analysis. Refractory elements can also be studied by means of this technique, but the lack of Z discrimination poses analytical problems.

Measurements of β feeding have long been a problem in decay scheme studies. Gamma-ray emission probabilities and internal conversion coefficients can be used to derive β feeding, but the determination of direct β decay to the highly excited states of the daughter can pose serious problems for various reasons, including the unsuitability of Ge crystals for the detection and quantification of high-energy γ rays [very low intrinsic efficiency at high γ-ray energies (above ~ 1.5 MeV)]. This problem is sometimes called the "pandemonium effect", and results in significant omissions in derived decay schemes. TAGS can overcome this difficulty, and has been proposed for some of the fission products defined as problematic in decay heat calculations.

Jacqmin presented work undertaken by Roque on decay heat in reactor and fuel cycle applications, and commented on recent requests from the nuclear industry for more accurate calculations of decay heat at short and intermediate cooling times. At short cooling times (< 1 year), the demand arises from the economic desire of reactor operators for shorter refuelling times, with a target accuracy of about 10% (2σ) or better. Reactor designers are also interested in fuel behaviour at these cooling times in order to avoid excessive conservatism in plant design and construction. At intermediate and long cooling times (> 1 year), the operators of fuel cycle facilities wish to optimise the transfer/transport of spent fuel assemblies in fuel processing plants, and the storage of fuel and nuclear waste; a target accuracy of about 10% (2σ) or better is also needed in these circumstances.

The JEF-2.2 database and CEA decay heat codes have been validated at short cooling times against pulse fission experiments (Akiyama and An [17] and Dickens, *et al*. [18]), with estimated uncertainties for UOX and MOX fuels of about 15% (2σ); new experiments are planned at CEA − MERCI fuel rod irradiation in OSIRIS, followed by calorimetric measurements (2007-2010). The main radionuclide contributors at intermediate and long cooling times have been clearly identified (specific actinides and fission products); only partial validation has been achieved through post-irradiation examination (PIE), with estimated uncertainties for UOX and MOX fuels of about 12% (2σ).

New fission yield and decay data sub-libraries have recently been produced in Europe (within the JEFF-3.1 project, as released in May 2005), while the Japanese produced an equivalent file in 2000 (JENDL-FPDD). CEA are preparing production libraries on the basis of JEFF-3.1 for validation. Some shortcomings have already been recognised at short cooling times, and should be addressed through SG25. These efforts have focused on the discrepancies between JEFF-3.1/RDD and JENDL-FPDD with respect to the mean β and γ energies, β- and γ-decay heat calculations, and the need to identify and quantify the roles of the responsible radionuclides. Detailed plots of fission-product contributions to decay heat at different cooling times were presented, and illustrated the complexity of the problem.

An important question is being posed by the nuclear industry at the present time: "Is a reduction in the uncertainty in decay heat calculations to about 10% (2σ) or better achievable?" Jacqmin stressed that one important aim of SG25 must be to identify those radionuclides that need better quantified mean β and γ energy data through direct substitutions and sensitivity studies. Consideration should also be given to the adoption of the average energy data of Rudstam, *et al*. [19]. Accurate new measurements are also required to determine β and γ data for the key radionuclides by means of the most appropriate technique (i.e. TAGS).

Yoshida stressed that the primary objective of any decay heat calculation was to use an accurate combination of fission-product decay data with an appropriate summation methodology to produce calculated data that match decay heat benchmark measurements. Specialists have long appreciated the problem of obtaining the correct match of β and γ decay due to the inability to identify and measure high-energy γ rays. A lack of valid γ-ray data generates incorrect β^- data, and explains observations made in the early 1970s that adopted mean β energies determined in this manner result in overestimates of the β decay heat, while adoption of incomplete γ-ray data sets for the mean γ-energies resulted in underestimates of the γ decay heat. However, these overestimations and underestimations match, such that the total decay heat remains in good agreement with the equivalent experimental data.

The production of TAGS data in the mid-1990s created a new challenge – formulation of such data so that they provide a comprehensive description of the β and γ decay of individual radionuclides. Yoshida had inserted these new data into JENDL-FPDD (effectively replacing inaccurate/theoretical mean energies with the values derived from the TAGS measurements of Greenwood, *et al.* [6] for 49 fission products). Unfortunately, the effect of this highly appropriate action had been an observed deterioration in the overall agreement between the benchmark data and the new TAGS-based decay heat calculations for both the β and γ components (when compared with calculations based on JENDL-FPDD in which data from the Gross Theory of Beta Decay had been adopted). However, agreement was shown to improve between the decay heat benchmark data and calculations after the incorporation of TAGS-based data into the JEFF-3.1 decay data files (which does not include any theoretical β- and γ-decay data).

Bersillon commented on decay heat calculations in which the JEFF-3.1 fission yields and decay data sub-libraries had been used. He analysed the contents of individual files from the aspect of the most important contributors to decay heat. A list of the origins of the most significant radionuclides characterised in the JEFF-3.1 decay data sub-library was presented (Table 1), and he stressed that a considerable fraction of the nuclides within this database possessed no discrete spectral data whatsoever (their mean energies were derived directly from NUBASE [20]).

Table 1. Origins and contents of JEFF-3.1 decay data sub-library

Original library	No. of nuclides	Comments
UKPADD6.4	360	All with spectral data
UKHEDD2.4	116	All with spectral data
LNHB	117	All with spectral data
ENSDF 900	900	All with spectral data
NUBASE	2 359	Without spectral data [20] (61% of all the radionuclides in JEFF-3.1)

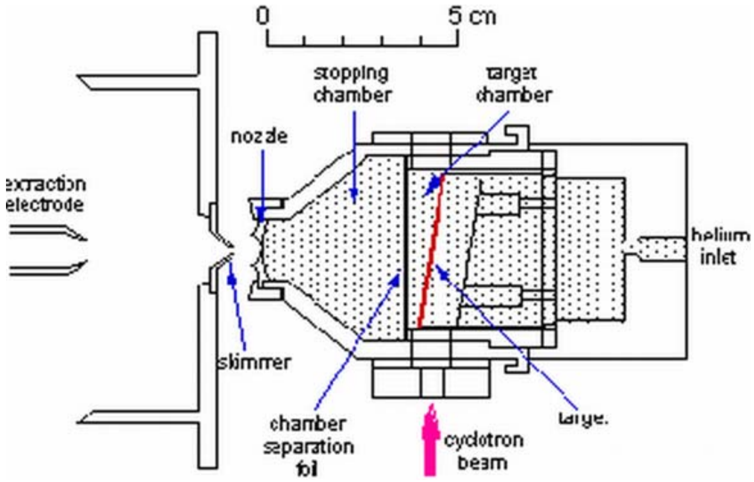
Total decay heat calculations with JEFF-3.1 were shown to be in good agreement with benchmark measurements. However, major differences occurred when β and γ decay heat components were considered separately. The mean β energies of the main contributors to β decay heat within JEFF-3.1 have also been compared with the equivalent contents of JENDL-FPDD for different cooling times. Ground and meta-stable $^{97}Y^{g,m,n}$ were highlighted for which there are three sets of recent decay data evaluations contained within JENDL-FPDD, JEFF-3.1 and UKPADD [the latter were evaluated in 2005 (but too late for inclusion in JEFF-3.1)]. Possibilities of pandemonium effect problems were also identified with ^{97}Sr, ^{96}Y, ^{99}Zr, ^{95}Rb, $^{101,102}Nb$ and ^{142}Cs. Some of these radionuclides were seen as important candidates for TAGS studies, and were subsequently included in the preliminary list of recommendations for such measurements. Bersillon also noted that the many TAGS measurements by Greenwood and co-workers [5,6] in the mid-1990s had yet to be absorbed into ENSDF in any satisfactory form by the various mass chain evaluators.

Tain presented the experimental details of TAGS as used to measure β-decay strength functions over the entire energy range through detection of the full γ-ray cascade. Thus, the problems associated with the pandemonium effect can be avoided, and β-strengths can be correctly assigned to derive valid mean β energies (and mean γ energies). Tain used EUROBALL studies of ^{150}Ho (2^-) with six detector clusters in a cubic geometry and seven Ge detectors per cluster to demonstrate the problem. Despite measurements with a 4π detector system, many high-energy γ rays remained undetected, with evidence from TAGS of a significant series of high-energy nuclear levels that were ill-defined.

Tain outlined the extraction process adopted to determine β-strengths from TAGS spectra through the formulation of the response matrix (R). Although the resulting complexity of this process can be problematic, a Monte Carlo simulation can be made of the TAGS γ-ray response (GEANT3 and GEANT4 codes). A number of possible systematic uncertainties were also noted: (a) contamination/background γ radiation; (b) studies involving (β^-n) decay, and the subsequent emission of prompt grand-daughter γ rays. These difficulties can be overcome by subtraction after measurement with a high-resolution array of γ and neutron detectors, although neutron interactions may cause further problems. A BaF_2 scintillator has been developed for such studies by the Surrey-Valencia group.

Algora presented the ongoing experiments at the University of Jyväskylä. A study of the β^- decay of ^{102,104,105}Tc by means of TAGS was launched on the IGISOL facility in late 2004. This work had been motivated by decay heat studies of ^{235,238}U and ^{239}Pu by Yoshida, *et al*. [21] that demonstrated the underestimation of γ decay heat by calculation involving radionuclides with half-lives around 1 000 secs. Therefore, TAGS was carried out to study the β^- decay of these inadequately characterised ^{102,104,105}Tc nuclides that may suffer from the Pandemonium effect. The experimental facilities were described (see Figure 7), including the differential pumping arrangement with a skimmer for ion extraction, and a rapid tape delivery system to both the TAGS and high-purity Ge detector for singles studies (IGISOL proposal 177). Beam and measuring times are chosen on the basis of the half-life of the nuclide under study, along with consideration of possible contaminants (although there was no (β^-n) decay problems with the chosen nuclides). TAGS spectra for ^{104}Tc and ^{105}Tc have been obtained, and Monte Carlo simulations have been implemented to calculate the response function prior to analysis. The TAGS data for ^{104}Tc indicate that this particular radionuclide possesses additional β-strength to produce a significant improvement in the overall fit of the measured decay heat data for ^{239}Pu around 1 000 secs cooling time. However, the lower yield of this nuclide in ^{235}U fission results in only a modest increase in the equivalent decay heat

Figure 7. IGISOL facility with the fission ion guide produces 2 700 ions/s per mb, with an efficiency of 1.6 × 10^{-4} relative to production in the target



calculations over similar time periods. Algora also noted that isotopically pure ^{105}Tc has proved to be more difficult to prepare than envisaged originally. More work is required on the resulting data and the facility to improve the technique for the study of ^{102,104}Tc. A better tape system needs to be developed, and laser ionisation should be explored to assist in obtaining isotopically clean spectra. Further measurements are also planned for ^{100,102}Tc.

Yoshida provided a brief description of the Gross Theory of Beta Decay, including the summation rules for the strength functions and the Fermi and Gamow-Teller transitions [1]. This approach was adopted in the 1970s-80s for specific fission products within the JENDL, ENDF/B-VI and JEF-2.2 decay data sub-libraries in order to introduce theoretical half-lives, mean β energies (and hence mean γ energies) that were missing from these files. The introduction of these theoretical data resulted in much closer agreement with the available benchmark data. However, one noteworthy problem that still remained was the lack of good agreement between the calculations and benchmarks for cooling times between 300 and 3 000 seconds [21].

Yoshida introduced the TAGS-based data of Greenwood, *et al.* [6] into JENDL-FPDD, and believed this approach was entirely correct despite the emergence of a lack of agreement between the decay heat calculations and benchmark experiments. The adoption of TAGS data in the files demonstrated the importance of such data as a function of the time after fission burst – however, the positive impact of some TAGS radionuclides appeared to be matched by the negative impact of others in their contributions to the total decay heat of ^{239}Pu. Yoshida reminded the meeting that the JENDL-FPDD database includes a significant amount of mean β and γ energy data from the Gross Theory of Beta Decay, unlike JEFF-3.1. Under these circumstances, he advocated the controlled introduction of TAGS-based data into JEFF-3.1 as a consequence of the "cleanliness" of this sub-library with respect to theoretical decay data. This suggestion provoked considerable debate.

Nichols presented information on the evolution in the 1990s of specific lists of important radionuclides identified with decay heat calculations. Consultations and discussions within the European community resulted in an agreed list of 27 inadequately characterised fission products, while the need for theoretical decay data was associated with another 35 neutron-rich nuclides for which there were no measurements [22]. More recently, Mills has initiated a programme of work in which JEFF-3.1 fission yields and decay data are being used in conjunction with the FISPIN10 inventory code to determine those radionuclides that contribute the greatest uncertainty to the overall uncertainty of the calculated decay heat. These calculations have been carried out on $^{235}U_{th}$, $^{239}Pu_{th}$, $^{238}U_f$ and $^{232}Th_f$ for a wide range of cooling times. Preliminary studies indicate that the major uncertainties for $^{235}U_{th}$ are identified with the following:

- short cooling times – uncertainties in fission yields;
- half-lives – $^{98}Y^m$ (± 10%), ^{100}Nb (± 13%) and ^{102}Nb (± 15%);
- energy releases – ^{87}Br (± 17%), ^{89}Sr (± 40%), ^{97}Sr (no uncertainty), ^{101}Nb (± 15%), ^{102}Nb (no uncertainty) and ^{143}La (± 53%).

Similar analyses are underway for the β and γ components of decay heat, and the calculations for $^{239}Pu_{th}$, $^{238}U_f$ and $^{232}Th_f$. These studies were seen to be a potentially important source for the identification of radionuclides for TAGS measurements, and the initial findings were introduced into the list of recommendations for TAGS.

Sonzogni described the Evaluated Nuclear Structure Data File (ENSDF) that contains the evaluated and recommended nuclear structure and decay data for 2 935 nuclei. Evaluations are undertaken at regular intervals for the various mass chains by individuals (and teams of people) within a group of 40 contributors organised under the auspices of the International Network of Nuclear Structure and Decay Data Evaluators. NuDat-2 provides users with a rapid means of accessing and viewing the data within ENSDF in a user-friendly manner. Apart from the fundamental nuclear structure and decay data, various derived information includes decay schemes, mean energies and detailed energy balances. Sonzogni indicated the value of using NuDat before undertaking any TAGS experiments. The software can be used to assist in the identification of nuclides with incomplete decay schemes, check for other missing radiation, and translate the data file to the ENDF format for nuclear applications.

Discussion

Participants brought together and assessed the various lists of important decay-heat radionuclides with respect to decay data that appeared to be inadequately quantified in the applications libraries – the original lists came from France, Japan and the UK. While these individual compilations exhibited some degree of overlap that engendered confidence in their validity, the dominant general opinion was that further detailed analyses were required before a second subgroup meeting in order to ensure that any TAGS measurements are strongly directed towards those radionuclides of importance in decay heat calculations.

Nichols believed that measurements would also be required to quantify the missing discrete γ-ray and β-particle data for these nuclides (as well as TAGS studies to determine mean β and γ energies). Detailed spectroscopic measurements were merited, including the determination of the energies and absolute emission probabilities of all β and γ transitions. Rubio stressed the importance to the TAGS studies of quantifying with confidence the absolute γ transition probabilities that populate the ground state of the daughter nuclide directly – relative emission probabilities are not sufficient for an analysis of the TAGS data to generate the necessary full set of β^- transition probabilities for the calculation of the β^- component to the mean β energy.

Possible facilities for any proposed TAGS experiments were discussed, and Gelletly proposed approaching IPN-Orsay where spectroscopic equipment already exists, or is in the process of planned assembly (including tape drive and mass separator for isobaric separation). Possible support from within Europe should be considered in conjunction with these studies. Furthermore, detailed assessment work is required in the next few months on the contents of the various sub-libraries (particularly JEFF-3.1/RDD and JENDL-FPDD) to understand and resolve some of the differences in the decay heat calculations. Bersillon also pointed out the significant differences that could be seen at longer cooling times between some of the benchmark data (e.g. in comparisons of the decay heat measurements of ORNL and Yayoi).

Participants agreed that all relevant benchmark data should be collected together, and Nichols noted that Mills had recently undertaken such an exercise.

Past and future TAGS data should be accumulated in a similar manner by the IAEA Nuclear Data Section, if possible (including Greenwood, *et al.*, known Russian experiments, and future TAGS measurements).

Recommendations and actions

Initial efforts have been made to identify the major differences in the mean energies to be found in the JENDL-FPDD-2000 and JEFF-3.1 decay data sub-libraries as a function of the cooling time of irradiated fuel. Furthermore, calculations and assessments are being undertaken to recognise those nuclides that contribute the greatest uncertainties to the decay heat as a function of cooling time.

Further assessment work is required to understand more precisely the nature of individual differences between the JENDL-FPDD-2000 and JEFF-3.1/RDD libraries as a means of improving specific decay data sub-libraries. Some discrepancies imply seriously inadequate decay schemes. These assessments will aid in the elimination of particular inadequacies (due to poorly-defined data sets). While new evaluations may also be required under these circumstances, this exercise will also identify those nuclides requiring new measurements (by TAGS).

The actions decided upon at the meeting of 12-14 December 2005 included:

- needing to understand the discrepancies between sub-libraries, along with defining suggestions for their resolution;

- determining whether there are any additional measurements of decay heat available from within Japan;

- preparing a comprehensive list of decay heat measurements, and make a suitable comparison of equivalent data sets;

- introducing requests for new decay heat measurements into the NEA High Priority Request List (HPRL);

- if possible, obtaining the original data from Greenwood, *et al*., Idaho National Laboratory (INL);

- assembling all available decay heat measurements into an appropriate database;

- assembling all available TAGS data into an appropriate database;

- contacting laboratories at which TAGS measurements could be undertaken;

- organising a meeting of representatives from the nuclear industry, laboratory managers and experimenters to resolve manpower and resource issues for TAGS studies;

- undertaking TAGS measurements for a number of key nuclides at existing facilities (e.g. ^{102}Tc, ^{145}Ba, ^{143,145}La).

Differences between decay heat benchmark experiments can be of the order of 15%. Efforts should be made to understand the cause and validity of such significant differences [e.g. 400 to 10 000 seconds in the electromagnetic (gamma) decay heat component of ^{235}U].

A provisional list of radionuclides for study by means of TAGS was prepared at the meeting, as shown in Table 2. This list was refined further at the follow-up meeting in May 2006 (see below, and Table 3).

Table 2. Radionuclides recommended for TAGS measurements (December 2005)

Radionuclide	Comments
^{87}Br	
^{92}Rb	
^{97}Sr	
^{96}Y	
^{98,101,102}Nb	
^{102,104,105}Tc	All three based on analysis of missing decay heat [21]
^{135}Te	
^{142}Cs	
^{145}Ba	Studied by Greenwood, *et al*. [6]
^{143,145}La	Both studied by Greenwood, *et al*. [6]

A detailed comparison needs to be undertaken of the 48/49 radionuclides identified as important by Yoshida with respect to their mean energies. Differences between JEFF-3.1/RDD and JENDL-FPDD-2000 also need to be tabulated and explained, and separate sets of decay heat calculations should be compared. Mean energies are available from three different sources with good pedigrees, namely JENDL-FPDD-2000, JEFF-3.1/RDD and INEL-TAGS (and possibly other experimental studies).

Accurate measurements of the average β and γ energies for the decay of fission fragments can be made by means of TAGS. Measurements of transition probabilities directly to the daughter ground state are also essential, and existing experimental facilities should be used for the study of these particular radionuclides. However, a dedicated facility should be established at a particle accelerator where sufficient beam time can be devoted to TAGS measurements in a systematic manner. Such a facility would consist of a suitable target/ ion-source, a mass separator of sufficient resolution, a sample transport system, and a total absorption spectrometer with associated equipment. Furthermore, high resolution γ-ray and internal conversion detectors should be made available for decay scheme studies. Sufficient collaborative manpower and resources would be required to exploit and realise the full potential of a dedicated facility and fulfil the desired aims of such a work programme (an additional benefit of this systematic approach would be the production of highly skilled manpower to undertake other types of nuclear data study).

Data derived from TAGS should be collected and recorded so that proper use can be made of the results of such measurements. Present databases are not suitable for this purpose. Hence, a new database should be set up for this purpose that includes all previous TAGS measurements. The contents of this new international database should be made widely available, and would not be restricted to decay heat applications. A reasonably flexible format would be required in order to contain information on maximum beta energies and intensities for both real and pseudo nuclear levels.

Significant effort should be expended to assess and compare the available decay-heat data, and relevant decay data from the JEFF-3.1/RDD, JENDL-FPDD and other newly-available libraries. Contacts should also be made with those responsible for the operational research programmes of facilities best suited for planned TAGS measurements, based on the preliminary recommendations tabulated above.

Summary of the second SG25 meeting

Specific recommendations and actions arose from the meeting held in December 2005, and were addressed further at a second meeting in May 2006. Decay-data requirements were reviewed during the course of the discussions, and a much more comprehensive list of recommended radionuclides was prepared for TAGS measurements. This meeting was also organised as a collaborative effort between the IAEA and OECD/NEA Data Bank. The full summary report of the meeting was issued as IAEA report INDC(NDS)-0499, *Summary Report of Second Consultants' Meeting: Beta Decay and Decay Heat*.

Presentations

Sonzogni described a series of decay heat calculations with a preliminary version of the US ENDF/B-VII decay data sub-library. The TAGS measurements of Greenwood, *et al*. [6] had been introduced into this database, and the resulting decay-data files were used to calculate the total, light-particle (beta) and electromagnetic (gamma) energy components of the decay heat for various fissioning actinides (^{235}U and ^{239}Pu). These data had been compared with the recommended Tobias decay-heat standards [23,24]. Sonzogni noted that the resulting decay database adopted in the ENDF/B-VII sub-library will also include the following new features:

- BRICC internal conversion coefficients (Kibédi, *et al*. analyses of Band and Raman model [25]);

- mean beta energies for a number of second forbidden non-unique transitions;

- theoretical β^- decay half-lives and β^--delayed neutron emission probabilities (P_n) for some neutron-rich nuclides, taking as their base Kratz-Hermann systematics as calculated by Pfeiffer, *et al.* [26].

Improvements will also be made to the X-ray and Auger-electron data of this sub-library.

Sensitivity studies had included controlled increases to the electromagnetic energy component by 50%, and decreases in the light-particle energy component by 50%. A check was made of the main contributors to the electromagnetic decay heat with respect to their Q-values, known/unknown decay schemes, and possible presence of the pandemonium effect, along with consideration of the TAGS measurements performed at INEL. This exercise generated two priority lists of radionuclides for ^{235}U and ^{239}Pu fission that have: a) incomplete decay schemes; and/or b) evidence of pandemonium. Any additional fission-product nuclides from these two lists were introduced into the SG25 tabulation of "Radionuclides recommended for TAGS measurements" (see Table 2).

Mills presented decay heat calculations undertaken in conjunction with the JEFF-3.1 decay data sub-library, and stressed the importance of such studies for the safe handling of spent nuclear fuel from the point of view of transport, storage and reprocessing. Validation is also required for short cooling times in terms of the total, light-particle (beta) and electromagnetic (gamma) decay heat and their uncertainties. Decay heat calculations had been carried out for ^{235,238}U and ^{239,241}Pu thermal neutron-induced fission pulses based on various JEF decay data sub-libraries (JEF-1, JEF-2.2 and JEFF-3.1) and the FISPIN inventory code. The majority of these data sets exhibited calculation/experiment (C/E) ratios that varied by factors of -0.8 to +1.15 over a cooling time of 10^5 seconds (most significant C/E deviations occurred within the ^{239}Pu light-particle and electromagnetic decay components).

Dean had assessed the TAGS data with respect to JEFF-3.1 and electromagnetic (gamma) decay heat for fission pulses on ^{235}U and ^{239}Pu and cooling times of 1 to 10^4 seconds (2.8 hours). These calculations involved a number of important variations:

- predictions using the mean gamma energies from Greenwood, *et al*. [6];
- predictions using mean gamma energies from Rudstam [19];

and their comparison with the decay heat standards of Tobias [23,24] and the recent measurements at Lowell, Yayoi and ORNL. The Tobias standards are based on least-squares fits to 54 measurements of ^{235}U and 28 measurements of ^{239}Pu. Dean noted that UK and Japanese decay heat calculations involve the adoption of thermal fission yields for both thermal and fast spectral studies, while French studies are based on fast fission yields for fast reactors and thermal fission yields for thermal reactors. Important observations included the following:

- Introduction of Rudstam mean gamma energies [19] for important radionuclides without discrete emission data exhibits no improvement over JEFF-3.1.
- Introduction of the mean gamma energies determined by Greenwood, *et al*. [6] shows a significant improvement relative to JEFF-3.1 for both ^{235}U and ^{239}Pu, particularly for cooling times from 20 to 200 seconds.
- Calculations tend to underpredict decay heat relative to the benchmark experiments.
- Recommended standards data of Tobias are generally higher than the experimental data up to cooling times of ≈100 seconds [3,4].

Bersillon reported on comparisons of JEFF-3.1 with JENDL-FPDD decay data files. The Q-values in these two sub-libraries can differ by as much as 40%, despite the known similarities in the Audi and Wapstra atomic mass tables over the previous 15 years between the data for those nuclides of greatest interest. Most up-to-date atomic mass tables should be used in the various decay data sub-libraries [27], although concomitant adjustments would also have to be made to the maximum energies of the individual beta transitions in the files. Comparisons had also been made between the equivalent Rudstam and Greenwood, *et al*. data, which were found to exhibit differences of 20-25% in their mean beta energies. Bersillon noted significant variations in the mean energy data for both ^{140}Cs and ^{144}La that could be resolved by TAGS. Furthermore, ^{144}La would appear to possess an extremely complex decay scheme that could be better addressed by adopting the TAGS data of Greenwood, *et al*. [6]. After some debate, participants agreed that specific issues noted by Bersillon after the first subgroup meeting need to be addressed in further detail.

During his studies of JENDL-FPDD and JEFF-3.1/RDD, Yoshida assessed the TAGS measurements. Decay heat calculations of the electromagnetic (gamma) component after a fission pulse in ^{239}Pu were used to determine the impact of introducing the mean energies derived from the TAGS data of Greenwood, *et al*. into JENDL-FPDD and JEFF-3.1/RDD. While these modifications to the JEFF-3.1/RDD sub-library improved the agreement between the resulting decay heat calculations and the benchmark measurements, the same additions to JENDL-FPDD resulted in significant deviations from the integral data. Yoshida was able to use these calculations and knowledge of the half-lives, Q-values of the various fission products and the following criteria to produce a list of radionuclides that merit TAGS studies:

- their contribution to the difference between JEFF-3.1/RDD and JENDL-FPDD is greater than 1% of the total difference in the beta and gamma decay-heat components;

- their highest evaluated nuclear level is less than 70% of the known Q-value.

The resulting list of radionuclides included high priority ^{98,101}Nb and ^{102,104,105}Tc, and medium priority ^{100}Zr, ^{99,102}Nb, ^{103,105}Mo and ^{103,106,107}Tc. These radionuclides were introduced into the provisional tabulation of "Radionuclides recommended for TAGS measurements" (see Table 2, with some minor modifications to the priorities).

Gelletly focused on the status of the TAGS experiments and queried the emphasis on TAGS measurements only. All participants agreed that an ideal resolution of current decay-data issues would also have to include well-defined decay schemes derived by means of high-resolution gamma-ray spectroscopy. Nevertheless, reliable mean decay data are sufficient for the resolution of decay heat issues. On this basis, TAGS measurements of the important fission-product nuclides were deemed to be critical in any decay heat calculations undertaken to assist in the confident design and safety assessments of the coolant systems for advanced reactor systems such as Generation-IV and beyond.

Tain summarised ongoing negotiations with experimental research institutes that possess or are planning to assemble facilities suitable for TAGS studies. The ALTO facility is in the process of being designed and assembled at IPN-Orsay – this apparatus should become available within two years, and has the capability of studying refractory fission products of high importance (Nb, Tc and Mo). One envisaged problem will be any request to study radionuclides that undergo delayed-neutron emission; consideration is being given to such nuclides in order to avoid possible errors in any TAGS measurements (DESPEC code).

Algora described recent TAGS measurements at the University of Jyväskylä that focused on ^{102,104,105}Tc. While difficulties had been experienced in the preparation of the ^{102}Tc source, measurements of ^{104,105}Tc were successful, and the resulting data are in the process of being analysed to produce mean beta and gamma energies. A new proposal had also been prepared to undertake further TAGS measurements at the University of Jyväskylä:

	Q_β **(keV)**	$T_{½}$ **(secs)**
^{102}Tc → ^{102}Ru	4 532	5.28
^{103}Tc → ^{103}Ru	2 662	54.2
^{103}Mo → ^{103}Tc	3 750	67.5
^{105}Mo → ^{105}Tc	4 950	35.6
^{106}Tc → ^{106}Ru	6 547	35.6

and possibly ^{98,101}Nb and ^{107}Tc. A laser ionisation process is required to produce “clean” isotopic sources of these radionuclides.

Additional discussion included the identification of suitable journals for the publication of TAGS data directed towards the resolution of decay-heat calculations for nuclear power plant: *Phys. Rev. C*, *Nucl. Sci. Eng.*, *Nucl. Instrum. Meth. Phys. Res. A and B,* and *Appl. Radiat. Isot*.

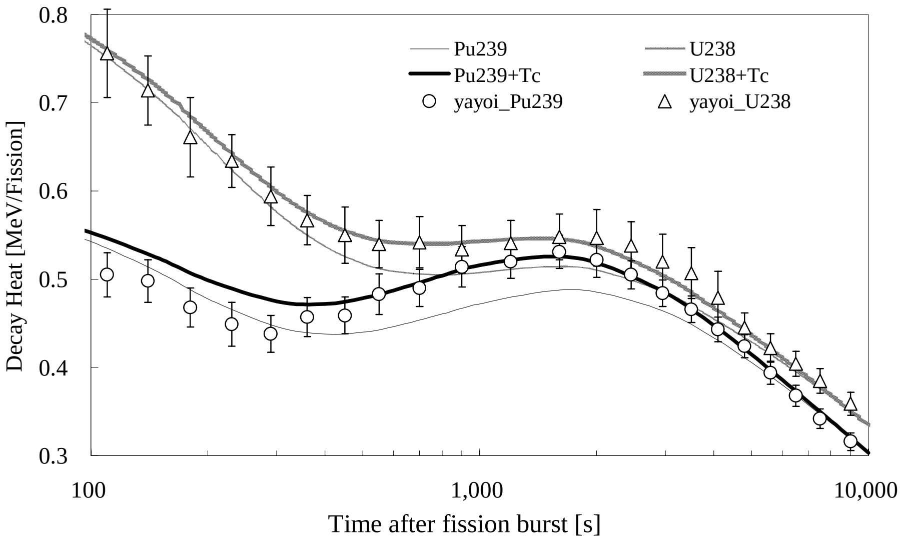
Initial TAGS measurements on the IGISOL facility at the University of Jyväskylä

TAGS measurements have recently been initiated at the University of Jyväskylä as described by Algora, *et al*. [28], focusing on the technetium fission products specified in Tables 2 and 3. As a consequence of these initial studies, the mean light-particle (E_{LP}) and electromagnetic (E_{EM}) energies of ^{104}Tc and

^{105}Tc have been determined. These nuclides possess half-lives of 1 098 and 456 s respectively, and contribute significantly to the decay heat at cooling times of 300 to 2 000 s after a fission burst when the *γ-ray discrepancy* defined in Ref. [21] is most prevalent.

The total decay heat of ^{239}Pu is seriously underestimated in the absence of the new data, as shown in Figures 5 and 6. Comparisons of Figures 1 through 6 indicate that this discrepancy can be attributed to inadequate quantification of the electromagnetic components of the decay heat. This same problem has also been observed in a significant number of other fissile nuclides ranging from ^{233}U to ^{239}Pu [21]. The introduction of the new TAGS values (E_{LP} and E_{EM}) from a preliminary analysis for ^{104}Tc and ^{105}Tc improves considerably the agreement between the decay heat summation calculations and equivalent experimental benchmarks. The impact of these data can be seen in Figure 8 for ^{238}U and ^{239}Pu, although the consistency between calculation and experiment deteriorates somewhat for cooling times less than 300 s in the case of ^{239}Pu.

Figure 8. Electromagnetic components of ^{238}U and ^{239}Pu FP decay heat showing the effect of introducing the preliminary TAGS results for ^{104,105}Tc [28] – curves are JENDL calculations before and after (+Tc) the introduction of the E_{EM} data for ^{104,105}Tc



As listed in Table 3, a range of TAGS measurements are required to ensure that all inadequately characterised fission product nuclides of significance are much better quantified with respect to E_{LP} and E_{EM}. Such a well-defined experimental programme would ensure that the decay heat of various fissile fuels can be calculated with full confidence and matched with known and future benchmark measurements over all desired cooling times. The proposed TAGS measurements specified in Table 3 need to be adequately addressed in the foreseeable future as demonstrated by the impact of the initial studies and advised by Subgroup 25.

CONCLUSIONS

Extensive discussions and analyses at the two subgroup meetings assisted greatly in the assembly of a list of radionuclides recommended for TAGS measurements (Table 3, below). This list has been refined, although studies and additional clarifications of the various decay schemes would be beneficial for ^{92}Rb, ^{89}Sr, ^{96}Y, ^{104,105}Tc and ^{142}Cs. Comparisons between the relevant files in JENDL-FPDD, JEFF-3.1/RDD and ENDF/B-VII are particularly important, and need to be made as soon as possible, particularly with respect to identifying the origins of the mean energy data in specific JENDL-FPDD files.

Good progress is being made in the identification of suitable TAGS facilities in Europe. The plans for ALTO at IPN-Orsay represent an exciting opportunity for undertaking TAGS for some of the more refractory radionuclides to be found in Table 3. Full support should be given by Subgroup 25 and WPEC to the experimental team as they begin negotiations with the management of the IPN-Orsay facility (see Appendix B).

This report has been prepared for submission to WPEC, recommending a well-defined list of fission-product radionuclides for TAGS measurements in order to improve decay heat calculations without resorting to the introduction of questionable theoretical data. Specific TAGS measurements are underway, and preparations are also being made for such studies of the refractory fission products. A significant number of the most important TAGS measurements will take at least two or three years to perform and analyse. Under these circumstances, the members of Subgroup 25 believe that they have achieved all feasible objectives over a two-year timescale through the identification of those ill-defined radionuclides that contribute significantly to the resulting decay heat of irradiated nuclear fuel, and would therefore benefit from TAGS. Specific requirements and their priority ratings could be agreed without additional analyses, while other needs required further assessment that can be found in Appendix A. The work of Subgroup 25 should be set aside until the ALTO facility is operational, since little of consequence can be achieved with respect to decay heat calculations until a substantial number of the recommended TAGS measurements have been successfully completed. Progress made by the experimentalists over 2006-2008 should be monitored by staff of the IAEA Nuclear Data Section, and further meetings are envisaged after the IPN-Orsay studies have begun, hopefully in late 2008.

Table 3. Requested TAGS measurements

Radionuclide	Priority	Q_β-value (keV)	Half-life	Comments
35-Br-86	1	7626(11)	55.1 s	
35-Br-87	1	6852(18)	55.65 s	Extremely complex decay scheme with substantial gamma component; large uncertainties in the mean gamma energy arises from significant disagreements between the various discrete gamma-ray measurements. Also (β^-,n) branch.
35-Br-88	1	8960(40)	16.36 s	(β^-,n) branch.
36-Kr-89	1	4990(50)	3.15 min	Incomplete decay scheme.
36-Kr-90	1	4392(17)	32.32 s	Incomplete decay scheme.
37-Rb-90m	2	6690(15)	258 s	Repeat of INL TAGS measurement; data check.
37-Rb-92	2	8096(6)	4.49 s	Small (β^-,n) branch.
38-Sr-89	2	1493(3)	50.53 d	
38-Sr-97	2	7470(16)	0.429 s	Extremely short half-life (0.429 s), and possible (β^-,n) branch.
39-Y-96	2	7096(23)	5.34 s	
40-Zr-99	3	4558(15)	2.1 s	
40-Zr-100	2	3335(25)	7.1 s	
41-Nb-98	1	4583(5)	2.86 s	
41-Nb-99	1	3639(13)	15.0 s	
41-Nb-100	1	6245(25)	1.5 s	
41-Nb-101	1	4569(18)	7.1 s	
41-Nb-102	2	7210(40)	1.3 s	
42-Mo-103	1	3750(60)	67.5 s	
42-Mo-105	1	4950(50)	35.6 s	
43-Tc-102	1	4532(9)	5.28 s	
43-Tc-103	1	2662(10)	54.2 s	
43-Tc-104	1	5600(50)	18.3 min	
43-Tc-105	1	3640(60)	7.6 min	
43-Tc-106	1	6547(11)	35.6 s	
43-Tc-107	2	4820(90)	21.2 s	
51-Sb-132	1	5509(14)	2.79 min	
52-Te-135	2	5960(90)	19.0 s	

Radionuclide	Priority	Q_β-value (keV)	Half-life	Comments
53-I-136	1	6930(50)	83.4 s	Incomplete decay scheme.
53-I-136m	1	7580(120)	46.9 s	
53-I-137	1	5877(27)	24.13 s	(β^-,*n*) branch.
54-Xe-137	1	4166(7)	3.82 min	Incomplete decay scheme.
54-Xe-139	1	5057(21)	39.68 s	
54-Xe-140	1	4060(60)	13.6 s	
55-Cs-142	3	7308(11)	1.69 s	(β^-,*n*) branch.
56-Ba-145	2	5570(110)	4.31 s	Repeat of INL TAGS measurement; data check.
57-La-143	2	3425(15)	14.2 min	Repeat of INL TAGS measurement; data check.
57-La-145	2	4110(80)	24.8 s	Repeat of INL TAGS measurement; data check.

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Appendix A

ADDITIONAL COMMENTS ON CANDIDATES FOR TAGS MEASUREMENTS

T. Yoshida, J. Katakura, T. Tachibana, A.L. Nichols
15 August 2007

^{87}**Br** *(Priority 1)*

A comprehensive reassessment has been made of the most relevant decay data measurements and their impact on the evaluated decay scheme of ^{87}Br:

- Sievers, H., *Nucl. Data Sheets,* 62, 327 (1991).
- Helmer, R.G., *Nucl. Data Sheets*, 95, 543 (2002).

β transitions are quantified in significant detail by both evaluators, with the only marked difference being the adopted Q-value (6 830 ± 120 keV by Sievers, and 6 852 ± 18 keV by Helmer, both from the Atomic Mass Evaluations of Audi, *et al*. at different times). The Q-value adopted in JEFF-3.1 by Nichols was 6 853 ± 18 keV (Nichols' evaluation was undertaken in late 1998). As expected, the Helmer evaluation is reflected completely in the data set to be found in the Web-based NuDat2.2 (12 May 2006).

All three decay scheme evaluations (Sievers, Helmer and Nichols) cite the measured gamma-ray emission probabilities of:

- Tovedal, H., B. Fogelberg, *Nucl. Phys*., A252, 253 (1975).
- Nuh, F.M., *et al*., *Nucl. Phys*., A293, 410 (1977).
- Raman, S., *et al*., *Phys. Rev*., C28, 602 (1983).

to assemble an extremely complex decay scheme. A detailed comparison generates the following information:

Evaluator	β transitions	γ transitions placed	Unplaced γ transitions
Sievers	160	224	138
Helmer	159	225	138
Nichols	181	374	–

The additional β transitions in the evaluated decay scheme of Nichols arise from the introduction of previously unplaced γ transitions. Note that ^{87}Br undergoes (Nichols):

$${}^{87}_{35}Br\left(\beta^-\right){}^{87}_{36}Kr \qquad \text{BF} = 0.9749\ (8)$$

$${}^{87}_{35}Br\left(\beta^- n\right){}^{86}_{36}Kr \qquad \text{BF} = 0.0251\ (8)$$

and the ($\beta^- n$) mode of decay is handled in great detail by Sievers, Helmer and Nichols based on the neutron spectrum measurements of Nuh, *et al*. (1977).

The gamma-ray measurements are particularly important in the derivation of the complex decay scheme, and the main inputs are as follows:

Reference	No. of γ	P_γ(1 419.7 keV)
H. Tovedal and B. Fogelberg (1975)	Only 19 γs	12 ± 2 (absolute)
F.M. Nuh, *et al*. (1977)*	Only 19 γs	31.2 ± 0.3 (absolute)
S. Raman, *et al*. (1983)	361 γs	22.0 ± 1.5 (absolute)
*F.M. Nuh, PhD thesis, University of California Berkeley (1975)	156 γs	100 (rel)

Nichols converted all of the absolute gamma-ray emission probabilities into values relative to the emission probability of the 1 419.6-keV gamma ray (100%), including those of Raman, *et al*.

Apart from the extreme complexity, the problem with the ^{87}Br decay scheme is immediately apparent from the significant differences/discrepancies between the various measurements of P_γ(1 419.6 keV) listed above – these values range from 12(2) to 31.2(3) for P_γ^{abs}(1 419.6 keV). Either the very large data set of Raman, *et al*. is adopted wholesale (361 γs) and the other studies are discarded, or some form of weighted mean is taken from the measurements of Tovedal and Fogelberg, Nuh, *et al*., and Raman for a limited set of γs and the large number remaining are adopted directly from Raman, *et al*. The latter approach was adopted by Nichols to give P_γ^{abs}(1 419.6 keV) of 0.20(5) [(20 ± 5)%], after adjusting the small uncertainty of Nuh, *et al*. from 0.003 to 0.03. Weighted mean values were calculated for the relative emission probabilities of a limited number of gamma rays, while the majority of the data were directly adopted from the much more extensive studies of Raman, *et al*.

As many as ≈120 gamma transitions may directly populate the ^{87}Kr ground state. Sievers and Helmer (ENSDF) ignored all studies earlier than 1983, and adopted the gamma-ray measurements of Raman, *et al*. wholesale [with P_γ^{abs}(1 419.6 keV) of 22.0 ± 1.5] that will naturally result in a lower uncertainty in the mean E_{EM} for this radionuclide based only on this one set of measurements.

Consideration of the above assessment: the mean E_{EM} value of (3 087 ± 771) keV evaluated by Nichols is not unreasonable in JEFF-3.1 and UKPADD, bearing in mind the significant disagreements in the measurements of P_γ(1 419.6 keV) to give a weighted absolute value of 0.20 ± 0.05; this uncertainty of 25% is directly reflected in the resulting large uncertainty in the mean E_{EM} value. There is a significant possibility of the pandemonium effect in ^{87}Br that, coupled with the valid existing uncertainty in the mean E_{EM} described above, presents a strong case for undertaking a TAGS study of this particular fission-product radionuclide with some priority.

^{92}Rb *(Priority 2)*

^{92}Rb is believed to generate about 6% of the total decay heat 10 secs after a fission burst in ^{235}U [A.1], representing one of the largest contributors to the decay heat at this cooling time. The electromagnetic component in JENDL (520 keV from Gross theory) is smaller than the equivalent data in JEFF-3.1 (1 750 keV) and ENDF/B-VII (1 742 keV), while the JEFF-3.1 light-particle component is consistent with ENDF/B-VII and ENSDF, but is smaller than for JENDL and ENDF/B-VI (Table A.1). This contrasts with most other known cases in which E_{EM} is larger and E_{LP} is smaller in JENDL than in JEFF-3.1.

Table A.1. Comparisons of Q_β, E_{LP} and E_{EM} (keV) within the major decay-data files for ^{92}Rb ($t_{1/2}$ = 4.492 s) [A.2]

^{92}Rb	JENDL (Gross theory)	JEFF-3.1	ENDF/B-VI	ENDF/B-VII	ENSDF Sept. 2006
Q_β	8 100	8 105	8 120	8 100	8 100
E_{LP}	3 499	2 875	3 524	2 869	2 869
E_{EM}	520	1 750	520	1 742	1 750

The highest known level in the ^{92}Sr daughter nuclide is at 7 363 keV and Q_β = 8 100 keV, implying only a small pandemonium effect. The existence of a delayed-neutron window above 7 293.74 keV [A.3] (or 7 342 keV [A.4]) complicates the situation.

According to ENSDF data, the ground-state transition is uncertain. The decay scheme on which the ENSDF evaluation is based can be used to deduce an I_β branch to the ground state of 94%. However, this large branch conflicts with the *log ft* value for a first forbidden transition. NuDat [A.2] contains an equivalent branch of only 51% if the *log ft* to the first 2+ state (first forbidden unique) is taken into account. The intensity of the ground state transition is the key factor in deducing the average light-particle and electromagnetic energies. A strong ground-state γ transition can influence E_{LP} and E_{EM}, and therefore TAGS measurements are merited to determine the absolute value of the ground-state β strength.

^{96}Y *(Priority 2)*

^{96}Y is estimated to generate about 5% of the total decay heat at 10 secs after a fission burst in ^{235}U [A.1], representing one of the largest contributors to the decay heat around this cooling time. The electromagnetic component in JENDL (1 206 keV from Gross theory) is larger than the equivalent value in JEFF-3.1 (80 keV), as listed in Table A.2, and this difference may arise from the inability of Gross theory to predict strong $0- \rightarrow 0+$ first-forbidden transitions from 0– odd-odd ground to 0+ ground states in even-even daughters (95.5% in this case).

Table A.2. Comparisons of Q_β, E_{LP} and E_{EM} (keV) within the major decay-data files for ^{96}Y ($t_{1/2}$ = 5.34 s) [A.2]

^{96}Y	JENDL (Gross theory)	JEFF-3.1	ENDF/B-VI	ENDF/B-VII	ENSDF Sept. 2006
Q_β	7 100	7 100	7 140	7 100	7 100
E_{LP}	2 657	3 205	3 229	3 183	3 183
E_{EM}	1 206	80	1 206	80	80

The highest known level in the ^{96}Zr daughter nuclide is at 6 232 keV [A.2] and Q_β = 7 100 keV, implying only a small pandemonium problem although such an effect can not be fully excluded.

Absolute β^- feeding to the ground state of ^{96}Zr was derived from the assumption of equilibrium among the decaying nuclides in the isobaric chain ^{96}Rb $\rightarrow$ ^{96}Sr $\rightarrow$ ^{96}Y $\rightarrow$ ^{96}Zr. There is a β^- delayed neutron branch within the isobaric chain, and the first excited state of ^{96}Zr is 0+, creating a difficult situation in which to determine absolute β^- feeding. The strong ground-state γ dominates E_{LP} and E_{EM}, so TAGS measurements are merited to determine the absolute value of the ground-state β strength.

^{98}**Nb** *(Priority 1)*

^{98}Nb generates about 3% of the total decay heat at 100 secs after a fission burst in ^{235}U [A.1], although the half-life of this radionuclide is only 2.86 s. The direct fission yield of this radionuclide is relatively small, and most arises from the decay of parent ^{98}Zr with a half-life of 30.7 s. The meta-stable state of ^{98}Nb^{m} ($t_{1/2}$ = 51.3 s) also has a low fission yield, and represents an insignificant contribute to the decay heat. The electromagnetic component of ^{98}Nb in JENDL (856 keV from Gross theory) is larger than the value in JEFF-3.1 (325 keV), as listed in Table A.3.

Table A.3. Comparisons of Q_β, E_{LP} and E_{EM} (keV) within the major decay-data files for ^{98}Nb ($t_{1/2}$ = 2.86 s) [A.2]

^{98}Nb	JENDL (Gross theory)	JEFF-3.1	ENDF/B-VI	ENDF/B-VII	ENSDF Sept. 2006
Q_β	4 586	4 586	4 586	4 586	4 586
E_{LP}	1 628	1 965	1 466	1 778	1 777
E_{EM}	856	325	1 190	321	321

NuDat [A.2] and *Table of Isotopes* [A.4] indicate that the highest known nuclear level fed by a β^- transition is either 2 608 or 2 206 keV, respectively, which is much smaller than the Q_β-value of 4 586 keV. This observation implies that Gross theory predictions for this nuclide suffer from the pandemonium effect.

According to ENSDF, the normalisation factor for the γ-ray emission probabilities varies significantly as a function of the method of derivation, and the value judged to be the most reliable was adopted. However, the resulting absolute emission probabilities differ by a factor of approximately four from other measurements. The β^- branch to the ground state may affect the normalisation factor, and therefore TAGS measurements are merited to determine the absolute value of the ground-state β strength.

^{102}Tc *(Priority 1)*

Although the half-life of ^{102}Tc is only 5.28 s, this radionuclide generates about 5% or more of the total decay heat at 1 000 secs after a fission burst in ^{235}U [A.1], which is a very important cooling time in the assessment of LOCAs. While there is essentially no production of ^{102}Tc by direct fission, this radionuclide is generated through the decay of ^{102}Mo which possesses a much longer half-life of 678 s. ^{102}Tc also accounts for the difference of 6% between the electromagnetic components, as calculated with JENDL and JEFF-3.1 for cooling times of 1 000 s and more after a fission burst in ^{239}Pu. As shown in Table A.4, the electromagnetic component of JENDL (119 keV from Gross theory) is much larger than the equivalent value in JEFF-3.1 (80.8 keV). Thus, ^{102}Tc is second only to ^{104}Tc in importance as the possible origin of the γ-ray discrepancy defined in Ref. [A.5].

Table A.4. Comparisons of Q_β, E_{LP} and E_{EM} (keV) within the major decay-data files for ^{102}Tc ($t_{1/2}$ = 5.28 s) [A.2]

^{102}Te	JENDL (Gross theory)	JEFF-3.1	ENDF/B-VI	ENDF/B-VII	ENSDF Sept. 2006
Q_β	4 530	4 526	4526	4 526	4 526
E_{LP}	1 420	1 945	1 420	1 945	1 945
E_{EM}	1 193	80.8	1 193	80.7	80.7

NuDat [A.2] and *Table of Isotopes* [A.4] indicate that the highest level known to be fed by a β-transition resides at an energy of 2 909 keV, which is much smaller than the Q_β-value of 4 526 keV. This observation implies that Gross theory predictions for this nuclide may suffer from the pandemonium problem.

^{104}Tc *(Priority 1)*

^{104}Tc generates about 3% of the total decay heat 1 000 secs after a fission burst in ^{235}U [A.1], which falls within the important range of cooling times for assessments of LOCAs. Furthermore, this particular radionuclide has been identified as a major potential contributor to the origins of the γ-ray discrepancy defined in Ref. [A.5].

NuDat [A.2] and *Table of Isotopes* [A.4] indicate that the highest level known to be fed by a β-transition has an energy of 4 268 keV. Although close to the Q_β-value of 5 600 keV, Gross theory implies that additional β-strength may exist within this radionuclide (see Table A.5); furthermore, the Valencia TAGS group has reported in a preliminary communication that even more β-strength exists than estimated [A.6].

Table A.5. Comparisons of Q_β, E_{LP} and E_{EM} (keV) within the major decay-data files for ^{104}Tc ($t_{1/2}$ = 1098 s) [A.2]

^{104}Te	JENDL	JEFF-3.1	ENDF/B-VI	ENDF/B-VII	ENSDF Sept. 2006
Q_β	5 603	5 600	5 620	5 600	5 600
E_{LP}	1 403[a]	1 595	1 450	1 590	1 576
E_{EM}	2 240[b]	1 890	2 245	1 890	1 885

[a] Gross theory.
[b] Experimental data taken into account.

According to the ENSDF evaluation, many unplaced γ-rays have been detected, and the normalisation factor for the γ-ray intensities was determined on the basis of only three γ-rays populating the ground state directly (with emission energies of 358, 893 and 1 515 keV).

^{105}Tc *(Priority 1)*

^{105}Tc has been identified as a major potential contributor to the origins of the γ-ray discrepancy defined in Ref. [A.5]. Although the four major decay data sub-libraries contain rather consistent data (Table A.6), the pandemonium effect still remains a possibility. The highest level known to be fed by a β-transition has an energy of 2 404 keV, which is significantly smaller than the Q_β-value of 3 640 keV.

Table A.6. Comparisons of Q_β, E_{LP} and E_{EM} (keV) within the major decay-data files for ^{105}Tc ($t_{1/2}$ = 456 s) [A.2]

^{105}Te	JENDL (ENSDF)	JEFF-3.1	ENDF/B-VI	ENDF/B-VII	ENSDF Sept. 2006
Q_β	3 640	3 640	3 585	3 640	3 640
E_{LP}	1310	1 310	1 211	1 310	1 310
E_{EM}	790	668	782	665	790

A preliminary result reported by the Valencia TAGS group suggested there may be even more β-strength in the highly excited region of the daughter [A.6]. An electromagnetic component of larger magnitude would be extremely favourable from the point of resolving the γ-ray discrepancy [A.5].

^{135}Te *(Priority 2)*

^{135}Te contributes 3% of the total decay heat at about 20 secs after a fission burst in ^{235}U [A.7], and is the biggest contributor to the difference between JENDL and JEFF-3.1 decay heat calculations around this same cooling time. Gross theory predictions adopted in JENDL indicate that ^{135}Te contributes the larger electromagnetic component (by 1.3%) and the smaller light-particle component (by 1.7%) in comparison with the equivalent decay heat calculations with JEFF-3.1 decay data. Thus, Gross theory data imply that a significant amount of β-strength exists within the high excitation energy region of ^{135}I.

Table A.7. Comparisons of Q_β, E_{LP} and E_{EM} (keV) within major decay-data files for ^{135}Te ($t_{1/2}$ = 19.0 s) [A.2]

^{135}Te	JENDL (Gross theory)	JEFF-3.1	ENDF/B-VI	ENDF/B-VII	ENSDF Sept. 2006
Q_β	5 962	5 960	5 960	5 960	5 960
E_{LP}	2 084	2 442	2 084	2 442	2 427
E_{EM}	1 478	384	1 487	384	388

The various published decay schemes imply that the highest nuclear level of ^{135}I has an energy of 4 773 keV, which is moderately lower than the Q_β-value of 5 960 keV. If the predicted β-strengths were concentrated in this narrow energy range, the resulting energy release might be judged as being too large. As shown in Table A.7, an E_{EM} in JENDL of 1 478 keV derived from Gross theory is more than a factor of four times larger that the equivalent value to be found in JEFF-3.1 (only 384 keV). This unsatisfactory situation needs to be investigated by means of TAGS.

^{142}Cs *(Priority 3)*

With a short half-life ($t_{1/2}$ = 1.68 s), ^{142}Cs contributes 4% of the total decay heat at cooling times around 1 second, which is dominated by delayed-neutron-induced fission and the release of the latent heat of the fuel. Therefore, this radionuclide is relatively unimportant from the point of decay heat calculations for LOCA studies. The highest known nuclear level has an energy of 5 280 keV, which is less than the Q_β-value of 7 307 keV, and Gross theory calculations imply that a reasonably large amount of the β-strength remains ill-defined above 5 280 keV.

Table A.8. Comparisons of Q_β, E_{LP} and E_{EM} (keV) within the major decay-data files for ^{142}Cs ($t_{1/2}$ = 1.68 s) [A.2]

^{142}Cs	JENDL (Gross theory)	JEFF-3.1	ENDF/B-VI	ENDF/B-VII	ENSDF
Q_β	7 307	7 317	7 317	7 307	7 307
E_{LP}	2 449	2 899	2 449	2 896	2 246
E_{EM}	1 787	675	1 787	675	665

As shown in Table A.8, E_{EM} of 1 787 keV in JENDL as derived from Gross theory is more than twice the value to be found in JEFF-3.1 (675 keV). According to the ENSDF evaluation, many unplaced γ-rays have been observed, implying that the decay scheme is incomplete and needs to be re-examined.

^{145}La *(Priority 2)*

^{145}La contributes between 1% and 2% of the total decay heat at 10 to 100 secs after a fission burst in ^{235}U [A.1]. Therefore, this radionuclide exerts only a modest influence on decay heat calculations.

NuDat [A.2] and *Table of Isotopes* [A.4] define the highest known nuclear level fed by a β-transition to be 2 607 keV, which is considerably less than the Q_β-value of 4 120 keV. According to the ENSDF evaluation, the normalisation factor for the γ-ray intensities was obtained from a private communication, and the detail remains uncertain. The implication of the Gross theory calculations is that a large amount of undiscovered β-strength might exist above 2 607 keV, and may be the reason why the JENDL and ENDF/B-VI values for E_{EM} are much larger than for JEFF-3.1, as listed in Table A.9 (ENDF/B-VII file adopted TAGS-INEL [A.8]). A repeat of the TAGS-INEL measurement would be highly beneficial.

Table A.9. Comparisons of Q_β, E_{LP} and E_{EM} (keV) within the major decay-data files for ^{145}La ($t_{1/2}$ = 24.8s) [A.2]

^{145}La	JENDL (Gross theory)	JEFF-3.1	ENDF/B-VI	ENDF/B-VII (TAGS INEL)	ENSDF Sept. 2006	TAGS INEL
Q_β	4 108	4 120	4 120	–	4 120	–
E_{LP}	998	1 499	877	760	1 519	762
E_{EM}	1 729	624	1 497	2 143	595	2 144

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Appendix B

NOTES FROM IPN-ORSAY MEETING TO DISCUSS THE USE OF THE ALTO FACILITY FOR TAGS MEASUREMENTS

A meeting was held at IPN-Orsay, south of Paris, on 17 July 2006. These extensive discussions focussed on the need for new TAGS measurements, possibly at the ALTO facility in Orsay.

Programme and presentations

Following a brief welcome to IPNO by D. Guillemaud-Mueller (Director), there were presentations on the various areas of concern. Firstly, Kellett and Mills introduced the various codes and data libraries on which calculations of decay heat are based. The uses of the codes, the underpinning decay data sub-libraries and their philosophies were all discussed. Deficiencies in the data were noted, along with the work of Subgroup 25 of the NEA Working Party on International Nuclear Data Evaluation Co-operation (WPEC). Emphasis was placed on discrepancies between the calculations of decay heat for various fissioning species and benchmark measurements of decay heat after a single fission pulse. Following two meetings in the previous six months, a list of radionuclides whose beta-decay characteristics need to be measured to remedy these deficiencies has been drawn up by SG25. This list was presented and discussed in some detail.

Tain and Algora described total absorption gamma-ray spectroscopy (TAGS), and the benefits of this technique for both spectroscopic measurements and studies of reactor decay heat. They reported some of the measurements made by the Valencia group using TAGS as well as the methods of analysis that they had developed. Results of TAGS measurements performed at the University of Jyväskylä for the decay of ^{104}Tc and ^{105}Tc were reported, with the radioactive species produced by means of proton-induced fission and extraction through the IGISOL system. When the resulting mean energy data were included in either the JEFF or ENDF/B decay data sub-libraries, a clear improvement occurred in the agreement between the calculations and the measured decay heat of ^{239}Pu at cooling times between 500 and 1 000 secs.

Lhuillier presented the interests of the SUBATECH-Saclay group in using the antineutrino spectrum from power reactors as the basis for determining both the reactor power and mixtures of fissioning materials in operating reactors.

Finally, Ibrahim reported on the performance characteristics of the ALTO facility and the progress made in achieving the desired operating specifications. An ambitious building programme is ongoing, and many important milestones have already been reached. The primary beams that can be used are limited in their intensity because of safety considerations, but this situation will improve with time and there is no reason to expect that the design goals cannot be reached. Both the tape transport system and the overall status of the facility were described. The meeting ended with a visit to the ALTO facility.

Status

Attendees at the IPN-Orsay concluded:

- There is a problem to be addressed in terms of the deficiencies of the decay data sub-libraries.

- TAGS measurements can contribute to the resolution of the existing discrepancies.

- The ALTO facility can provide the species of interest, although time is required to develop some of the desired beams.

- Many other experiments in nuclear spectroscopy and astrophysics require the comprehensive decay energies that can be obtainable by means of the TAGS technique – such studies would benefit considerably from the development of the ALTO facility.

Specific agreement was also reached on the following issues:

- There is a strong *prima facie* case for proceeding to consider in detail how and where to deploy the BaF_2 spectrometer at ALTO – participants felt that this endeavour had a sufficiently strong case to obtain beam-time from the Programme Advisory Committee (PAC).

- The generic case for the programme of TAGS measurements should be presented to the next PAC, which will be held in the spring of 2007. Assuming success, less detailed cases would then need to be made to

PAC in the future. This initial case should not only address decay heat applications, but also associated problems in nuclear structure and astrophysics that might be tackled by means of TAGS.

- The experimental team would be led by Tain in conjunction with Algora. Tain would be the primary point of contact for the laboratory and any potential collaborators.

- IPN staff agreed that they would be able to provide engineering and technical assistance to establish the apparatus at ALTO. Specifically, they would couple the tape system to the TAGS spectrometer.

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