Nuclear decay heat - the energy released by fission fragments

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Decay heat is an important issue that has to be dealt with after a nuclear reactor is shut down. Following the fission of an actinide nucleus, such as ^{235}U or ^{239}Pu, about 800 neutron-rich nuclides are produced. Most of these nuclides are unstable, that is, they will undergo nuclear decay, releasing energy in the process.

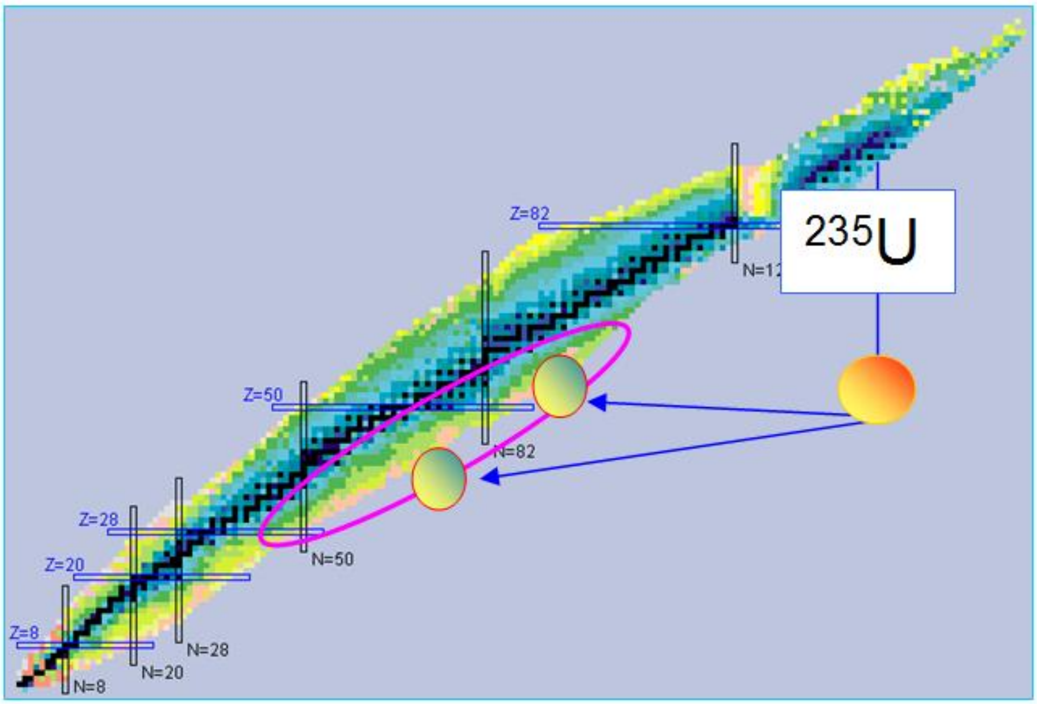

Figure 1: Schematic representation of the fission of ^{235}U. The magenta contour represents the region of the chart of nuclides that will mainly be populated in the process.

The total energy released in nuclear decay can be divided into three groups: (i) that carried out by photons (electromagnetic waves: gammas and X-rays), (ii) light particles (electrons and positrons from beta decay as well as conversion and Auger electrons), and (iii) heavy particles (neutrons and fission fragments). Neutrinos take a good share of the energy, roughly one third; however, since they have a negligible interaction with matter, their effect can be ignored. In a nuclear reactor, the energy released in the decay will be thermalized, that is, converted to heat. As a result, reactors must be cooled down even after shutdown. The same is true for spent fuel rods.

The fission fragments and their daughter nuclides form a decay network. Assuming no feeding from fission, the network follows these linearly coupled differential equations:

$$dN_k(t)/dt = -\lambda_k\ Ni(t) + \Sigma\ \lambda_{ik}\ N_i(t). \qquad (1)$$

where $\lambda_k = \ln(2)/T_{1/2k}$, with $T_{1/2k}$ the half-life of the k-th nuclide in the network; additionally, $\lambda_{ik} = b_{ik}\, \lambda_k$, where b_{ik} is the probability of the i-th nuclide decaying to the k-th one. The decay heat as a function of time is given by:

$$DH(t) = \Sigma\, N_k(t) \langle E_k \rangle, \quad (2)$$

where the $\langle E_k \rangle$ is the average energy released by the k-th nuclide. The Evaluated Nuclear Structure Data File (ENSDF) containing recommended decay data is the main and often the sole source for λ_k, b_{ik} and $\langle E_k \rangle$ values. These equations can be solved numerically for a given initial condition, that is a set of $N_i(t{=}0)$ values. For a single fission event, the $N_i(t{=}0)$ are the fission yields that can be obtained from the Evaluated Nuclear Data File (ENDF) library.

In a nuclear reactor, fission fragments are produced while it operates. Additionally, radioactive nuclides will be produced by neutron capture on the structural materials. In order to gauge our understanding, we will solve a simpler problem -- the decay heat following a single fission event. A plot of decay heat multiplied by the time in such a scenario can be seen in Fig. 2.

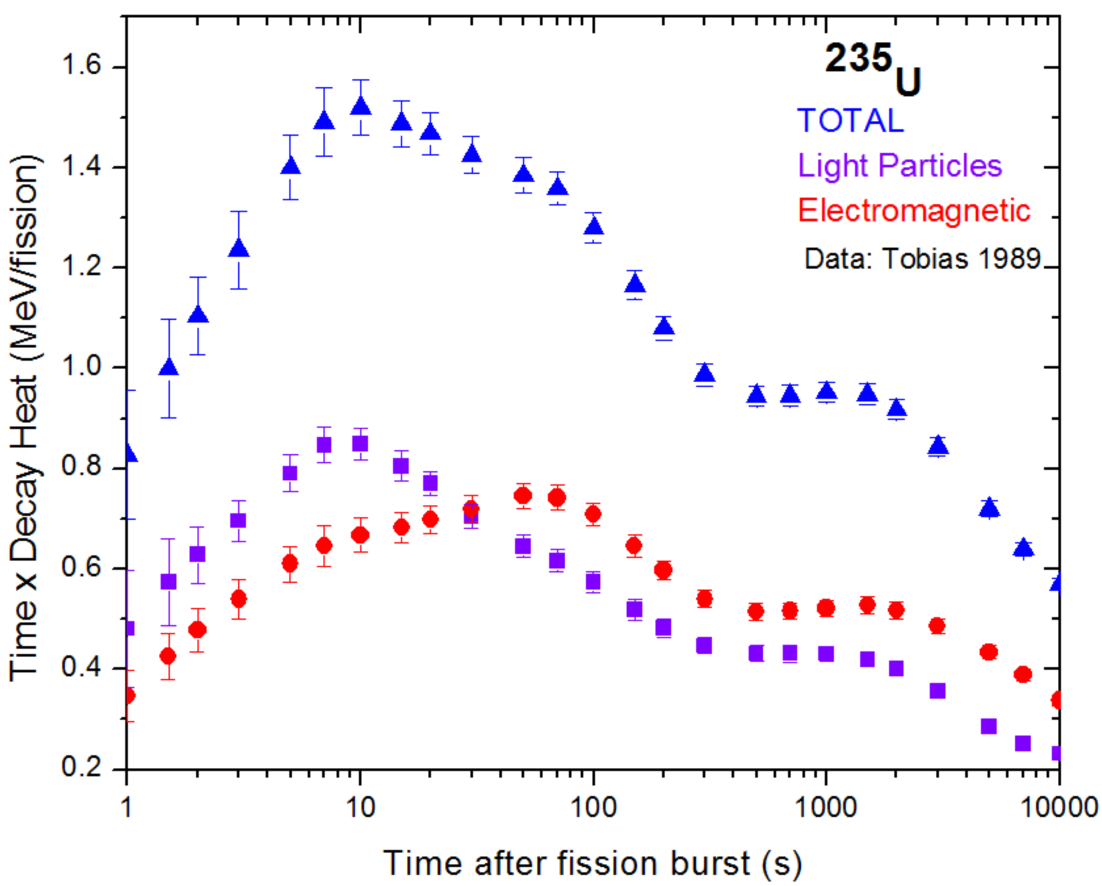

Figure 2: Decay heat multiplied by time after a single fission burst. The total decay heat (blue symbols) decreases quickly as a function of time and is basically the sum of the light particle term (purple symbols) and the electromagnetic term (red).

Many of the fission fragments are neutron-rich nuclides that will undergo beta-minus decay. In this decay, a neutron inside the nucleus is transformed into a proton plus an electron plus an antineutrino. As the energy available for the decay increases, more higher-lying energy levels in the daughter nuclide can be populated, resulting in a larger number of less intense gamma rays. The weaker gamma rays can be missed in experiments using high-resolution Germanium detectors, or not properly assigned to a level, resulting in incomplete decay schemes.

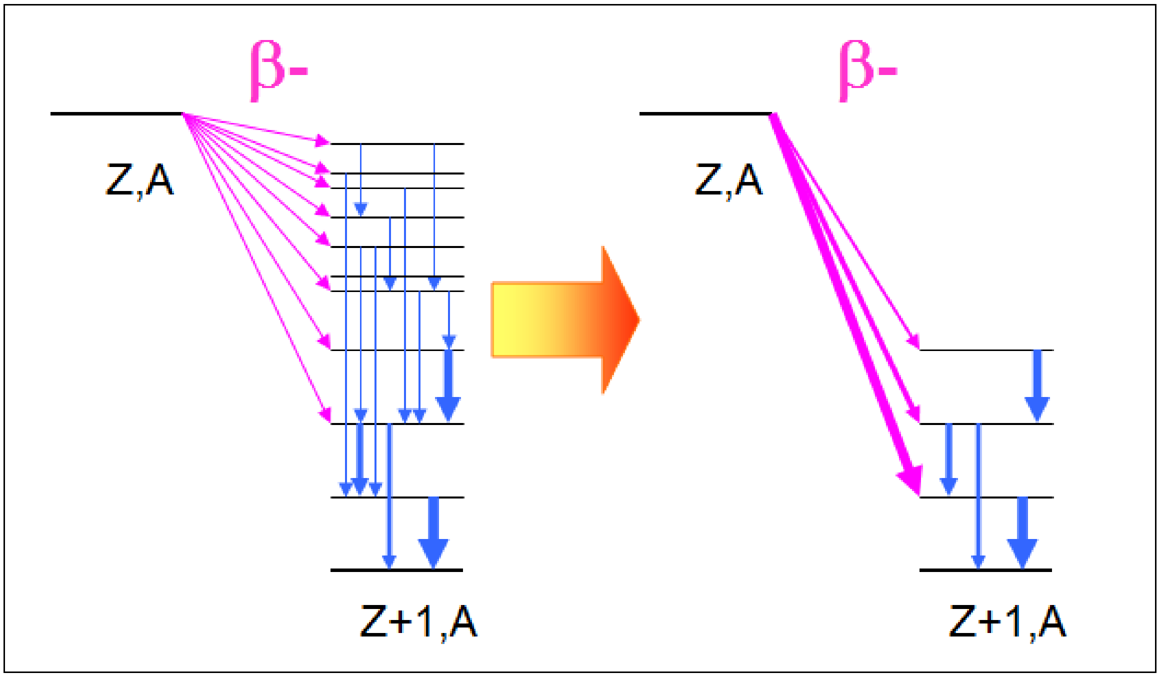

Figure 3: Incomplete decay scheme (right) resulting from missing the weak gamma rays in the real decay scheme (left)

The decay scheme on the right of Fig. 3 will have a lower average gamma energy and higher average beta and neutrino energies than the one on the left. The term "Pandemonium" was coined to describe this effect by John Hardy and collaborators [1].

When the decay information was combined with fission probabilities to obtain a value of the decay heat as a function of time, because of the Pandemonium effect, the gamma decay heat was under-predicted while the beta was over-predicted. Because photons can travel farther through matter than electrons, the underestimation of the electromagnetic decay heat can have serious consequences in shielding calculations.

This problem was first realized during the '70s [2] and a series of experiments were performed using a Total Absorption Gamma Spectrometer (TAGS), which is basically a high-efficiency NaI(Tl) crystal acting as a calorimeter. The fission fragments were obtained from a research reactor. These are not easy experiments since the high efficiency of the crystal comes at the cost of poor energy resolutions, and gamma spectroscopy cannot be used to identify the nucleus undergoing beta minus decay. Nevertheless, the $\langle E_k \rangle$ values from TAGS will be far more realistic.

A second generation of experiments was performed during the '90s by a group at Idaho National Laboratory (INL) led by R. Greenwood [3]. The fission fragments were obtained from a ^{252}Cf source. The usefulness of these data in decay heat calculations was first demonstrated by T. Yoshida and collaborators [4]. Additionally T. Yoshida was instrumental in setting up an international collaboration of experts under the auspices of the OECD Nuclear Energy Agency that studied the problem in detail and made recommendations for future experimental work [5].

In Fig. 4 we plot the decay heat for thermal-neutron induced fission on ^{235}U. The calculations without INEL TAGS data (dashed lines) can be compared with those including INEL TAGS data (full lines). A good description of the electromagnetic decay heat (red symbols) is obtained.

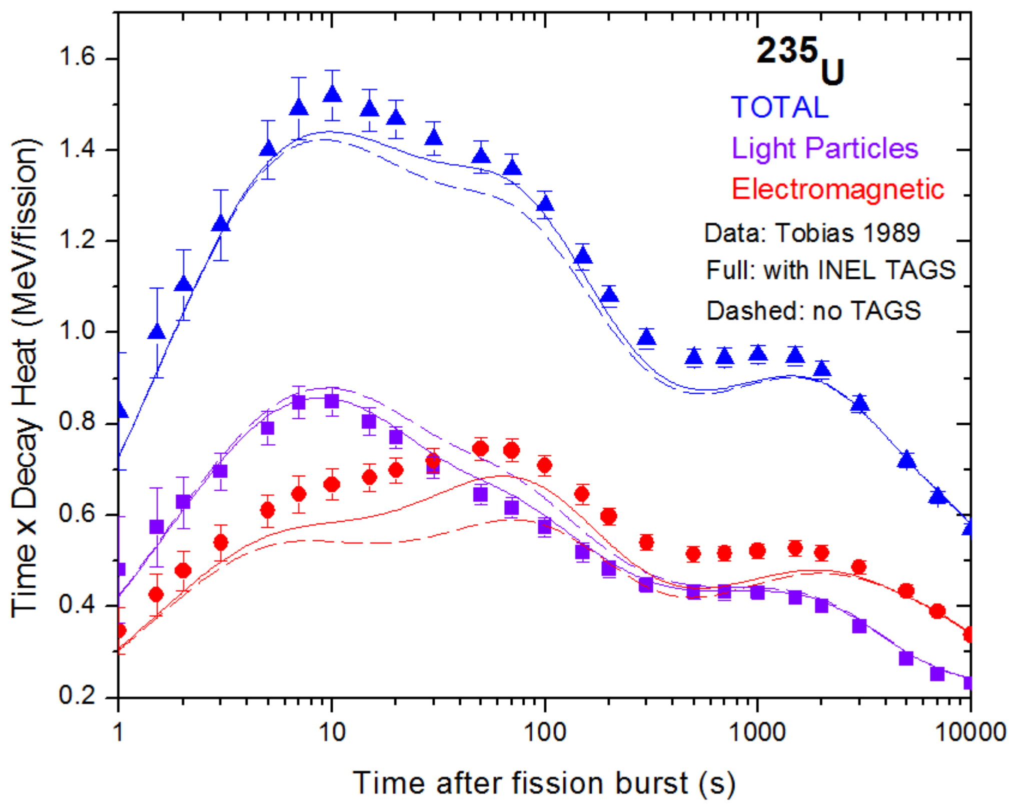

Figure 4: Decay heat multiplied by time after a single fission burst.

A third generation of experiments was performed in Europe, using radioactive beams from the facilities at CERN-ISOLDE and Jyvaskyla. Seven isotopes were studied. The results, which had a definite impact on the decay heat of ^{239}Pu, were published in Physical Review Letters [6] by Algora and collaborators. An accompanying Viewpoint article [7] was written about it.

The NNDC contribution to this work was to perform the decay heat calculations, and as the custodian of the ENSDF and ENDF databases, to provide recommended nuclear decay and fission yield data. A plot of the electromagnetic decay heat as a function of time can be seen in the Fig. 5 below.

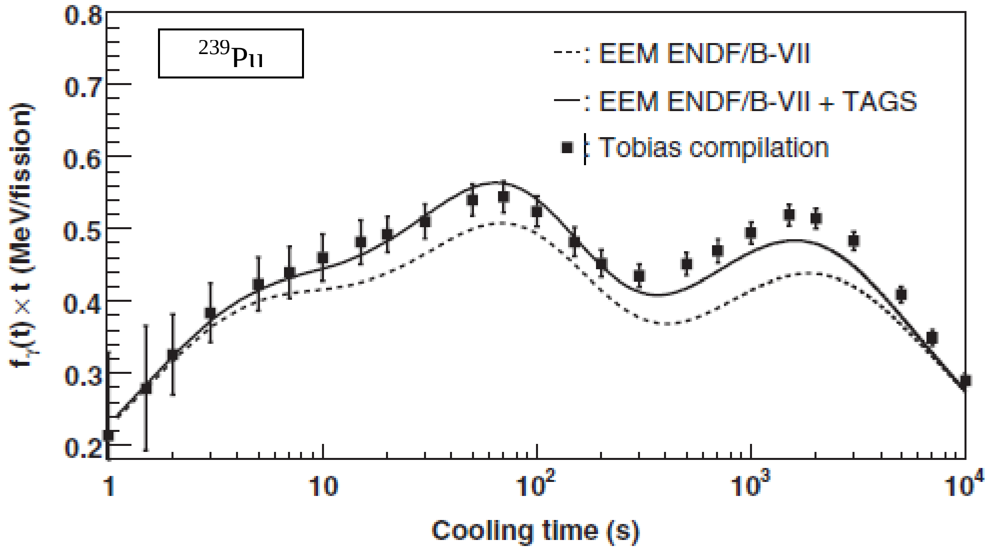

Figure 5: Electromagnetic decay heat multiplied by time for ^{239}Pu. The dashed line includes the TAGS INL measurements, while the full line includes the newly measured values.

The fission yields for the seven studied isotopes are considerably larger for ^{239}Pu than for ^{235}U, see the Fig. 6 below, which explains why the effect was more important for ^{239}Pu.

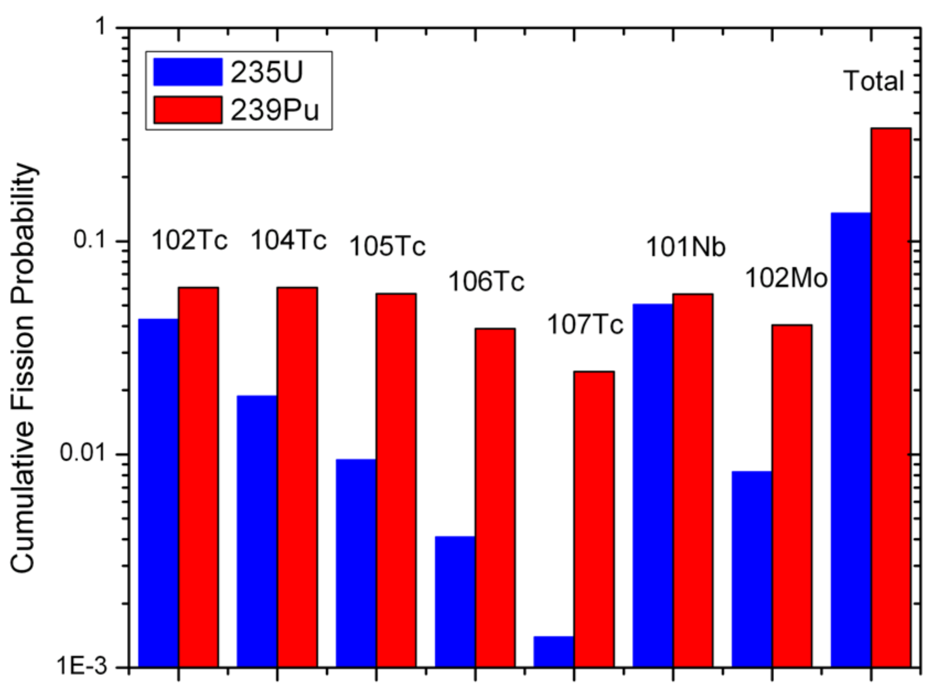

Figure 6: Cumulative fission fragment yields for the different nuclides studied in the Algora *et al.* article.

In addition to improving the calculation of decay heat, which is of great importance in existing as well as next generation nuclear reactors, these experimental results are of great theoretical interest. TAGS experiments measure the average beta feeding as a function of excitation energy. This feeding is basically the product of two terms, the Gamow-Teller distribution times the level density. The modeling of both quantities for neutron rich nuclides can be quite challenging but current understanding of them is adequate for safe operation of a nuclear reactor.

References

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