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Current Status of Decay Heat Measurements, Evaluations, and Needs

J. K. Dickens

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Engineering Physics and Mathematics Division

CURRENT STATUS OF DECAY HEAT MEASUREMENTS, EVALUATIONS, AND NEEDS

J. K. Dickens

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CURRENT STATUS OF DECAY HEAT MEASUREMENTS, EVALUATIONS, AND NEEDS

J. K. Dickens

This report is dedicated to John C. Connor (1923-1986)
An able colleague and a cherished friend

ABSTRACT

Over a decade ago serious concern over possible consequences of a loss-of-coolant accident in a commercial light-water reactor prompted support of several experiments designed specifically to measure the latent energy of beta-ray and gamma-ray emanations from fission products for thermal reactors. This latent energy was termed Decay Heat. At about the same time the American Nuclear Society convened a working group to develop a standard for use in computing decay heat in real reactor environs primarily for regulatory requirements. This working group combined the new experimental results and best evaluated data into a standard which was approved by the ANS and by the ANSI. The primary work since then has been (a) on improvements to computational efforts and (b) experimental measurements for fast reactors. In addition, the need for decay-heat data has been extended well beyond the time regime of a loss-of-coolant accident; new concerns involve, for example, away-from-reactor shipments and storage. The efficacy of the ANS standard for these longer time regimes has been a subject of study with generally positive results. However, a specific problem, namely, the consequences of fission-product neutron capture, remains contentious. Satisfactory resolution of this problem merits a high priority.

INTRODUCTION

At the conference on Nuclear Cross Sections for Technology held in Knoxville, Tennessee, in October 1979, I presented a paper[1] reviewing the status of decay heat up to that time. The present report has the goal of providing a report on some of the important activities concerning decay heat in the intervening seven years.

In an operating reactor, power is obtained following the nuclear transmutation of the fuel material, usually ^{235}U, and the subsequent conversion of mass to energy. The energy obtained in this fission process is divided among the kinetic energies of the fission products (including neutrons), energy of gamma radiation from "prompt" decay of highly-excited fission products, and finally the time-dependent beta- plus gamma-ray energy released following "delayed" decay of fission products. This last-described source contributes about 7% of the total energy obtainable in the fission process, and because of its time-dependent nature has been labelled "Decay Heat."

When a reactor is shut down, the prompt sources of energy die with fission rate, but there remains the decay heat source of energy. The amount of energy available from this source decreases with time following shutdown, of course, as the fission products decay. As some fission products have very long lifetimes, one may be concerned about this source of energy for a very long time. Consequently, there have been continuing efforts to determine values of decay heat as a function of time following shutdown. In this country these efforts have resulted in an American Nuclear Society (ANS) standard[2] on decay heat. A preliminary report on this standard was given in my report to the Knoxville conference.

From a decay heat point of view, fuel management can be divided into three categories which might be called (a) in-core utilization (or burnup), (b) on-site storage, and (c) off-site storage. These three categories involve different time regimes with regard to decay heat. Values of decay heat for times immediately following fission to "cooling" times of a few days after fission are applicable to in-core utilization, not only to determine the decay-heat contribution to the reactor power production, but also to assess the consequences of a loss-of-coolant accident (LOCA). For on-site storage, one needs to know the decay heat for times following fission consistent with the storage time which at present may be several years. For off-site storage (a feature still in the future!), the consequences of much longer-term decay heat will be important.

In principle, decay heat can be computed using known nuclear data. The technique, as exemplified by the ORIGEN family of codes,[3] is to compute the inventory of fission products created during some irradiation history and cooling period, then to use evaluated radionuclide decay characteristics (e.g., beta-ray branching probabilities, etc.) to determine the energy release rate for each fission product, and finally to determine the total decay heat by summing the energy release rate for each fission product weighted by the amount of that fission product in the computed inventory. Needed, of course, are data files[4] containing fission-product yields for each fissioning isotope in the fuel and files[5,6] containing evaluated nuclear data for decay of each radionuclide produced in the fission process. Also needed are cross-section data involving neutron interactions with all isotopes created during the irradiation. Indeed, the amount of data required to do the computation is substantial.

Finally, in a real reactor, the fuel is contained in a rod made of a light element (usually a zirconium-based alloy), and several rods are held together in a unit (a bundle) also made of light elements (e.g., stainless steel). Neutrons will interact with these structural elements creating radionuclides which, when they decay, will contribute to the decay heat. In addition, neutron interactions with the fuel will also result in production of heavy actinides (e.g., Am and Cm isotopes), and decay of these heavy elements will also contribute to the decay heat.

DECAY HEAT FOR SHORT COOLING TIMES

Most of the experimental emphasis has been for short cooling times, i.e., for times after fission between ~1 sec and ~1 day. The impetus[1,7] for these experiments was the concern about consequences of a LOCA coupled with the difficulty in calculating decay heat for very short times because of lack of experimental decay data on short-lived fission products. By 1979, several measurements[8-12] of short-cooling-time decay heat for thermal-neutron fission of several fuel isotopes were essentially completed and were reviewed in my previous report. In addition, these experimental data were combined with calculated decay heat data to provide the basis for an American Nuclear Society standard[2] on decay heat for thermal-neutron fission, and this standard also was discussed in my previous report. Since then the ORNL experimental data have been published,[13,14] and the ANS standard was approved by the

American National Standards Institute (ANSI) as an U.S. Decay Heat Standard. In addition, measurements made at the National Engineering Research Laboratory (NERL) at the University of Tokyo of decay heat following fast-neutron fission[15] have become available. Work is continuing on improving knowledge, and evaluated data files of nuclear data for short-lived radionuclides.

At first it was thought that, because the various experiments utilized different irradiation, cooling, and counting times (as well as different measuring equipment), the experimental data could not be directly compared to each other, but would require comparisons with computed decay heat to determine if there were inconsistencies among the various experiments. However, we deduced a method for making direct comparisons of the various experimental data sets (see ref. 14 for details). Results from the several experiments and the ANSI-ANS standard are presented in Table 1 for fuel materials ^{235}U and ^{239}Pu. The overall agreement among the various data sets is quite good, particularly in view of the quite different measurement configurations used. The data from the Los Alamos group[8] (LANL) generally have the largest values, while our own data (ORNL) have the smallest values. Indeed, the differences between the LANL data and the ORNL data are rather larger than the combined assigned experimental uncertainties. These differences have been of some concern, particularly in Great Britain, for the ^{239}Pu decay heat. The NERL experiment for fast-fission decay heat is the most similar to the ORNL experiment. The NERL and ORNL data for ^{239}Pu decay heat are exhibited in Figure 1 for the separate components of decay heat, viz., beta-ray decay heat in the upper half of the figure, and gamma-ray decay heat in the lower half of the figure. One would expect that the difference in the incident neutron energy would result in less than 1% difference in decay heat, and so values from the two measurements should be very similar. And, in fact, to within the uncertainties assigned to individual data points in the figure the two measurements do provide essentially equivalent results. This comparison thus supports the ORNL data with regard to the "LANL-ORNL" discrepancy; but, as one might well expect, there is a difficulty, which is readily observed in Figure 2 showing the NERL and ORNL results for decay heat for ^{235}U. The beta-ray decay heat in the upper half of the figure are in quite good agreement, but there is a clear discrepancy in the gamma-ray decay heat data in the lower half of the figure for cooling times between 600 and 6000 seconds. At present writing (May 1986) the observed differences are not understood. Besides the incident neutron energy (which should not yield the observed result) the only substantive difference between the two experiments is in the sample: the NERL ^{235}U sample was about 1000 times more massive than the ORNL sample and was mounted on a metal backing whereas the ORNL sample had a polyethylene backing. The ^{239}Pu measurements had the same sample differences, however, and it seems unlikely that sample characteristics would affect one set of measurements and not the other. In addition, for cooling times <600 secs, as exhibited in Table 1, the NERL ^{235}U gamma-ray data are in better agreement with the ORNL data than with the LANL results.

As mentioned above, research designed to improve computed decay heat for short cooling times has continued, including improvements in nuclear data files. Perhaps the most "successful" at present is the Japanese Nuclear Data Committee (JNDC) data set[6] assembled by researchers at the Japanese Atomic Energy Research Institute (JAERI). It is the only available evaluated data set that results in calculated data that are in good agreement with experimental data for cooling times <10 seconds (see Figures 1 and 2). There are, however, still some problems. One problem is readily apparent in the lower half of Figure 2; the calculation does not agree with either set of experimental data for cooling times >3000 secs. The second problem is that evaluated data for very short-lived fission products (particularly those for which there are as yet no experimental data) are deduced[16] from a theoretical framework[17] that is not expected to (and does not) provide accurate data for individual radionuclides. Recently, theorists at the Max-Planck-Institut fur Kernphysik (MPK) have studied beta decay of very short-lived fission products from a more fundamental theoretical framework,[18] and have demonstrated calculated beta-ray decay heat results[19] quite consistent with the experimental data exhibited in the upper halves of Figures 1 and 2. Again, however, although detailed comparisons of calculated beta-ray decay characteristics with experimental beta-ray decay characteristics are more favorable for the MPK

Table 1. Comparisons of Measured Decay Heat for Recent Experiments and the Current ANSI-ANS Decay Heat Standard

Time interval (sec)	LANL[a] (thermal)	IRT[b] (thermal)	CEN[c] (thermal)	ORNL[d] (thermal)	NERL[e] (fast)	ANSI-ANS[f]
			Total $^{235}U(\beta+\gamma)$ Decay Heat (MeV/fission)			
2.2 − 10.2		2.107 ± 0.042		1.983 ± 0.076		2.105 ± 0.044
10.2 − 20.2	1.147 ± 0.040	1.024 ± 0.020		0.941 ± 0.025		1.023 ± 0.021
20.2 − 100	2.251 ± 0.045	2.195 ± 0.043		2.088 ± 0.052	2.141 ± 0.090	2.242 ± 0.044
100 − 600	1.930 ± 0.033	1.903 ± 0.038	1.818 ± 0.073	1.808 ± 0.048	1.853 ± 0.037	1.916 ± 0.035
600 − 1200	0.663 ± 0.010	0.657 ± 0.013	0.646 ± 0.026	0.616 ± 0.019	0.668 ± 0.011	0.666 ± 0.012
1200 − 6000	1.386 ± 0.021	1.353 ± 0.027	1.379 ± 0.055	1.282 ± 0.040	1.347 ± 0.027	1.382 ± 0.025
6000 − 14000	0.510 ± 0.010	0.487 ± 0.010	0.525 ± 0.021	0.510 ± 0.030	0.515 ± 0.010	0.513 ± 0.009
			Total $^{239}Pu(\beta+\gamma)$ Decay Heat (MeV/fission)			
2.2 − 10.2		1.627 ± 0.044		1.475 ± 0.050		1.510 ± 0.065
10.2 − 35.2		1.447 ± 0.039		1.327 ± 0.037		1.377 ± 0.058
35.2 − 130	1.613 ± 0.048	1.457 ± 0.039		1.397 ± 0.035	1.360 ± 0.058	1.435 ± 0.060
130 − 600	1.482 ± 0.044	1.407 ± 0.037	1.465 ± 0.105	1.363 ± 0.030	1.329 ± 0.061	1.415 ± 0.061
600 − 2500	1.421 ± 0.042	1.344 ± 0.035	1.424 ± 0.095	1.296 ± 0.030	1.276 ± 0.047	1.338 ± 0.059
2500 − 14000	1.104 ± 0.031	1.032 ± 0.027	1.174 ± 0.078	1.061 ± 0.028	1.056 ± 0.025	1.078 ± 0.052

[a] Ref. 8.
[b] Ref. 9.
[c] Ref. 12.
[d] Ref. 10, 13, and 14.
[e] Ref. 15.
[f] Ref. 2.

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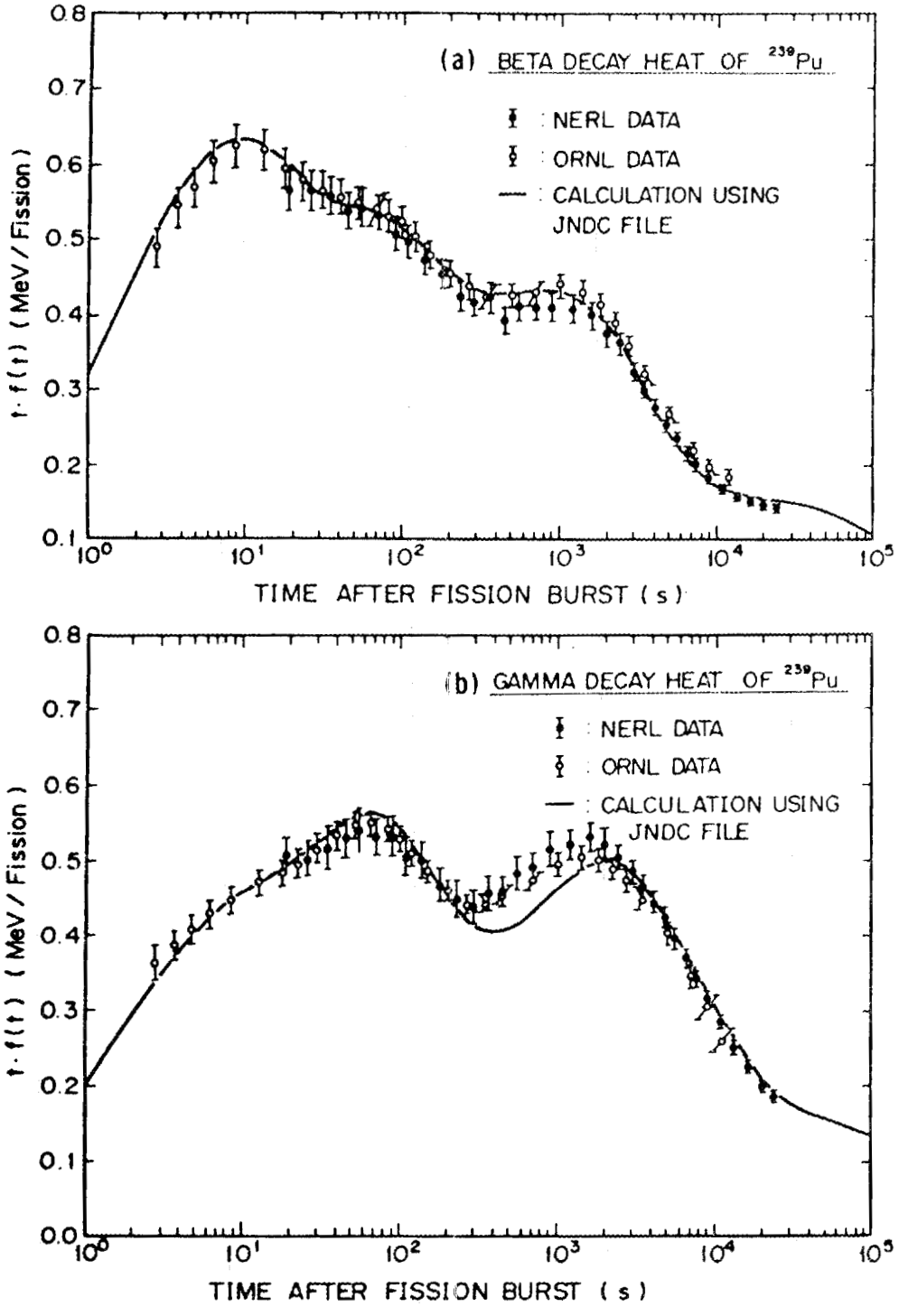

Fig. 1. Beta decay heat (upper figure) and gamma decay heat experimental data for two recent experiments (refs. 14 and 15) on ^{239}Pu. Also shown are results of a summation calculation using the JNDC data file (ref. 6). The function *f(t)* is the decay heat function following an instantaneous burst of fissions. These data sets are in excellent agreement for both components, beta-ray and gamma-ray decay heat.

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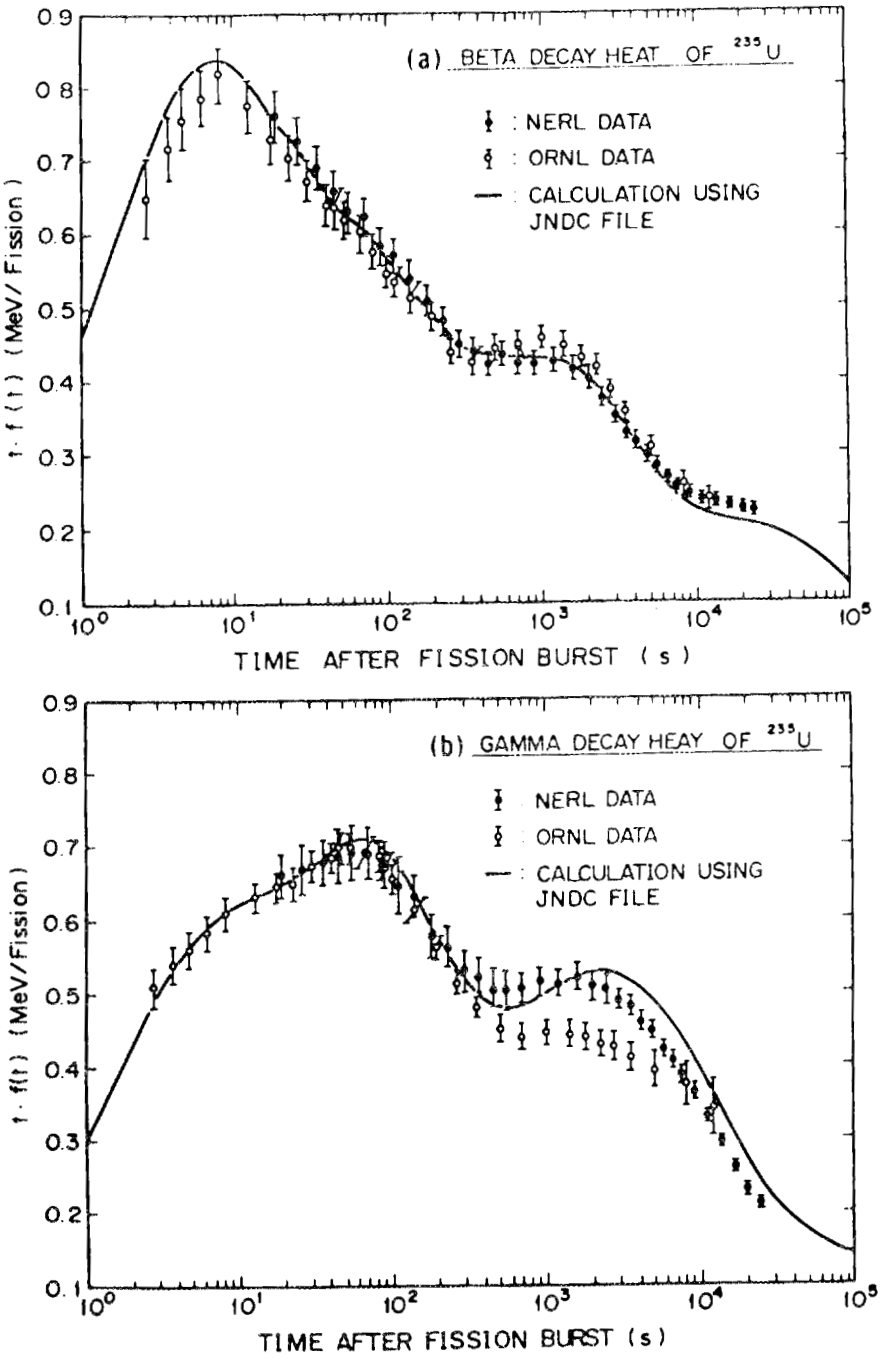

Fig. 2. As Fig. 1, only for experiments (refs. 13 and 15) on ^{235}U. The major discrepancy in the lower figure around $t = 10^3$ s is not understood, as discussed in the text.

framework than for the JNDC data file, still there are cases for individual radionuclides for which the MPK theory does not agree well with experiment. Partly because of this situation, the suggestion[20] by the MPK group that commercial power reactors could be safely operated at an increase in average operating power of 5% has not been accepted by the responsible regulatory bodies.

DECAY HEAT FOR INTERMEDIATE COOLING TIMES

Our knowledge of decay heat for cooling times longer than a few days and up to about 3 years (10^8 sec) is based almost entirely on summation calculations of the type described above. Early in this time regime decay heat is due primarily to fission-product radionuclide decay; this condition is certainly the case for the ANSI-ANS standard. Later in this time regime fission-product radionuclide decay is still substantially dominant over the decay of the heavy isotopes (created by sequential capture processes) and decay of the light radionuclides created in the structural elements; however, decay of radionuclides created by neutron interactions with fission products becomes important. Indeed, measurements[21] of radionuclides created by long-term irradiations of fuel elements indicate relatively substantial amounts of ^{134}Cs which can be made only by neutron capture by the stable fission product ^{133}Cs. Present-day sophisticated summation computer programs can determine inventories of such radionuclides given the fuel irradiation history and a file of nuclear data containing the excitation function describing the reaction cross sections, for example, for the ^{133}Cs$(n,\gamma)^{134}$Cs capture process.

The ANSI-ANS standard, however, was prepared to be applicable to any (reasonable) reactor operating history. It was necessary, then, to supply the user of the standard with a method of determining decay heat from such radionuclides as ^{134}Cs without detailed knowledge of the reactor operating history. This facet of the standard was accomplished by a single-valued multiplicative function of cooling time which had been determined by doing a complete decay heat computation using a reactor power history that would produce the maximum decay heat from such "capture" radionuclides and yet could still be likely in the actual operation of present-day reactors. Clearly, then, application of the standard to fuel burned at less than the nominal maximum power density ought to result in overestimating the total decay heat. An idea of how much of an overestimation would result is indicated in Figure 3. The data shown in this figure are documented in a report[22] of summation calculations performed in support of a U.S. Nuclear Regulatory Commission guideline discussed in the next paragraph. The calculations were for a "typical" albeit somewhat optimistic operation history, and they were performed for a variety of ^{235}U enrichments and total fuel burnup, as indicated in the figure. For cooling times <3 years the ANSI-ANS standard overestimates the total decay heat, and so using the standard would result in a conservative design of, let us suppose, the on-site pool to hold the spent fuel.

DECAY HEAT FOR LONG COOLING TIMES

As mentioned above, two years ago the U.S. Nuclear Regulatory Commission prepared a guideline[23] for off-site spent reactor fuel storage. The guideline used the calculations in the report[22] quoted above. The calculated decay-heat values included decay heat from the heavy actinide radionuclides as well as from the light-element radionuclides in the structural materials. These latter two sources are not considered in the ANSI-ANS standard for these long cooling times, and so, as shown in Figure 3, the standard slightly underpredicts decay heat for long cooling times. One should observe, however, that at present the standard is applicable for cooling times only up to 30 years, although data are given in the standard for longer cooling times. An important feature of this guideline,[23] however, is the allowed alternative use of the ANSI-ANS standard for computations to which the standard applies.

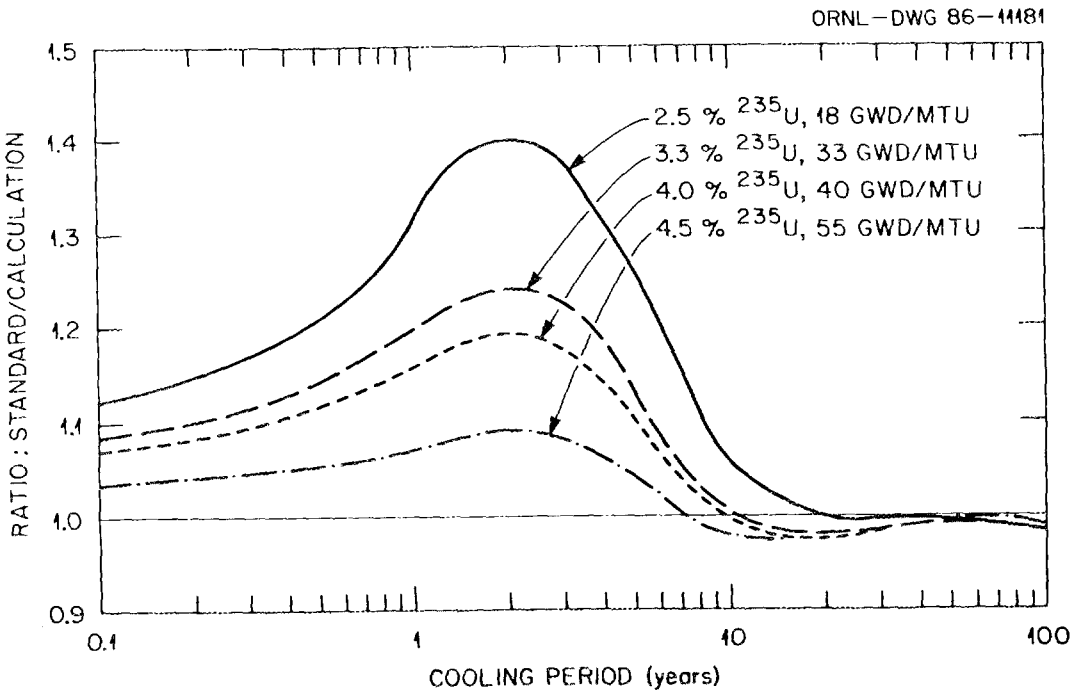

Fig. 3. Ratio of decay heat deduced from the ANSI-ANS standard to that determined from calculations (ref. 22) using the ORIGEN code (ref. 3) for several different fuel enrichments and total fuel utilization (burnup).

Although there are no specific decay heat experimental data for these longer time periods, there are a few calorimetric measurements of decay heat from older spent-fuel assemblies. Such measured data have been compared[22,24] with predictions based on summation calculations. The comparisons are generally quite favorable, particularly considering that details of the irradiation history of the subject fuel assemblies are often difficult to obtain.

CONCERNING POWER-PRODUCTION-REACTOR ACCIDENTS

In my 1979 report to the Knoxville conference I discussed a hypothetical LOCA in order to provide a rationale for the ongoing measurements. Figure 4 gives a representation of part of an analysis of a typical "worst-case" LOCA, showing the expected temperature of the cladding due to the indicated decay-heat source. The major phases of a LOCA are indicated in the figure, as the hypothetical LOCA progresses from pipe rupture at $T = 0$, through blowdown, emergency core-cooling refill and reflood, and then finally reduction of the temperature of the fuel rods.[25] Although the example is quite simplified, and quite schematic, the point was that properly handled it was *at that time* anticipated that one could expect to bring the emergency under control within 15 minutes or so.

Interestingly, this 1979 report was given some six months after the accident at one of the Three-Mile Island (TMI) reactors. Even then, some six months later, not much was known about that

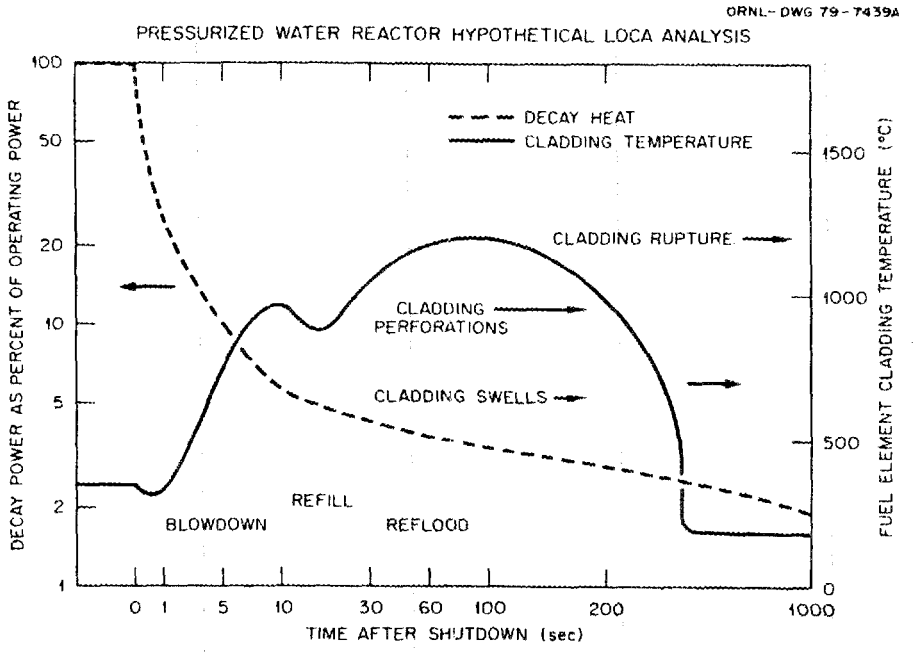

Fig. 4. Fuel element cladding temperature as a function of shutdown time for a typical "worst-case" LOCA. The time scale (abscissa) is intended only to be schematic; maximum cladding temperatures may occur earlier or later than shown depending upon assumptions made.

accident beyond the operators' log book entries of actions taken. By now we have a fairly complete picture of the course of that accident.[26] Indeed, the initiation of the TMI accident was less severe than the "hypothetical" scenario discussed in the previous paragraph. However, the problem persisted because the cause was not immediately recognized. One aspect of interest for the present discussion was that the consequences of the accident evolved over a period of about four hours, and not 15 minutes as our earlier best estimates had suggested. However, it appears that the reactor did not always behave as it had been expected to behave following operator actions. Clearly the decay heat in the fuel rods was the driving source. One may reasonably inquire about the importance of knowledge of values of decay heat during the evolution of the accident, for, in fact, the actual heat generation rates in the early phases of the accident were some 20% smaller than Federal regulations[27] specify and so increases in temperature would be less than (or not as rapid as) anticipated on the bases of calculated pre-accident scenarios. Such is, of course, a moot inquiry insofar as the TMI accident is concerned. For the future, however, I suggest that we should question the practice of "conservative" estimates (which are really overestimates) of the consequences of decay heat (or any other parameter, for that matter) when developing and training personnel.

The very recent reactor accident at Chernobyl in the Ukraine (only two weeks ago as I write this report) appears to be an example of an unexpected non-nuclear disruption very severely exacerbated by subsequent destruction of a nearby nuclear reactor. The initial problem at Chernobyl apparently involved the electric-power generation equipment, and this problem could have occurred whether the heat source was a nuclear reactor (as it was) or was a coal-burning or oil-burning furnace. The Russian authorities reported a fire and an explosion[28] in the building housing the reactor and the

generating equipment. It is apparent from a picture of the damaged building that the explosion was substantial. What is not known at the present writing is the mechanism which caused the reactor to be damaged. Certainly one possibility, especially considering the observed damage to the building, is that the reactor was damaged by the explosion. Once the cooling mechanism of the reactor was destroyed, as it surely must have been as a consequence of the explosion, decay heat in the fuel elements provided the heat to increase the temperature of the fuel elements and then of the graphite moderator to ignition temperature. This heating process took some time, however, at least several hours, and so burning graphite was not the initial mechanism of fission-product release into the atmosphere, nor was it the process by which hydrogen gas was generated that caused the initial explosion.

SYNOPSIS AND RECOMMENDATIONS

Much of the experimental and theoretical work leading to an understanding of decay heat was completed by 1980; that research has been summarized in several review articles.[29] Since then the primary experimental efforts at NERL[15] have been for fast-neutron fission of fuel elements, not only ^{235}U and ^{239}Pu, but also ^{238}U and ^{232}Th. The primary theoretical efforts have been to improve the several evaluated nuclear data files[30] and, in particular, to try to understand why using the JNDC file results in the best computed results especially for the very short cooling times.[31] Despite the fact that decay heat has been a very important problem with important repercussions, there has been very little support in this country during the past seven years for additional research to solve the remaining problems. At the very minimum the current ANSI-ANS standard should be improved by (a) including specific decay-heat data for thermal-neutron fission of ^{241}Pu (and perhaps also ^{233}U) and improving the data file for ^{238}U by including the new experimental data[15] for fast-neutron fission of ^{238}U, (b) extending the standard beyond its current limitation of 30 years, and (c) including decay heat from long-lived heavy elements and from light-element structural materials. Use of this standard (which I should note has been subjected to extensive peer review) in the regulatory process should be strongly encouraged, and its use in the regulatory guide[23] mentioned above should be recognized as an important action. Indeed, one may observe that the decay-heat problem has been an interesting and instructive problem from both engineering and physics points of view, an excellent example of the melding of two disciplines toward a common goal. The remaining tasks ought to be completed.

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