Measurements of 14-MeV neutron cross-sections for the production of isomeric states in hafnium isotopes

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Introduction

Fusion reactor systems operating on the (d,t) reaction will produce copious numbers of neutrons which will, through nuclear reactions, produce activation products in material subjected to the neutron fluence. In particular, the first wall of a power reactor system and structural materials in that vicinity will be bombarded by very high fluxes which could give rise to intense radioactivity. The neutron fluence is expected to result in radiation damage to the extent that the wall will probably have to be replaced every few years. There is therefore a big incentive to minimise the activity as, if it can be restricted to a low level after a reasonable cooling time, the first wall material could be reused, with obvious economic benefits and minimising the material that might have to be put into a radioactive waste repository.

Some years ago, the UK Fusion Programme initiated a search for viable first wall materials. As activation would be an important criterion in selecting suitable elements, part of that Programme is being devoted to the establishment of a nuclear data library and associated inventory code to enable activation to be calculated with sufficient accuracy. An early library, UKCTRIIIA, was produced by Jarvis (1980) for use with the code ORIGEN.

The neutron flux at the first wall of a fusion power reactor will be of such a magnitude that sequential reactions in a given nucleus will be possible, driving the products well away from the stable region of the periodic table. For this reason, a nuclear data library must contain cross-sections for reactions in unstable nuclides as well as stable ones. To enable the search for low activity materials to be as comprehensive as possible, the nuclear data library also needs to be essentially complete; missing cross-sections could terminate a particular reaction path in calculations of activation, causing misleading conclusions to be reached regarding the suitability of some elements. In the last few years, effort has been directed towards improving UKCTRIIIA, in terms of both the quality of the data and their completeness. The result has been a new library, UKACT1, containing cross-section data in 100-group form for almost 10000 reactions. A complementary library of decay data has also been established and calculations of arisings are carried out with these two data sets using the inventory code FISPACT, a special version of the UK fission reactor inventory code, FISPIN (Burstall (1979)). Further details of the data libraries and FISPACT are to be found in Forrest et al (1988). It is worth noting that FISPACT has the capability to carry out sensitivity analysis of nuclear data and also contains a pathway analysis technique, both of which can be used to identify important reactions.

In studies of potential first wall materials, it was suggested that an alloy containing small quantities of tungsten and tantalum might be worth

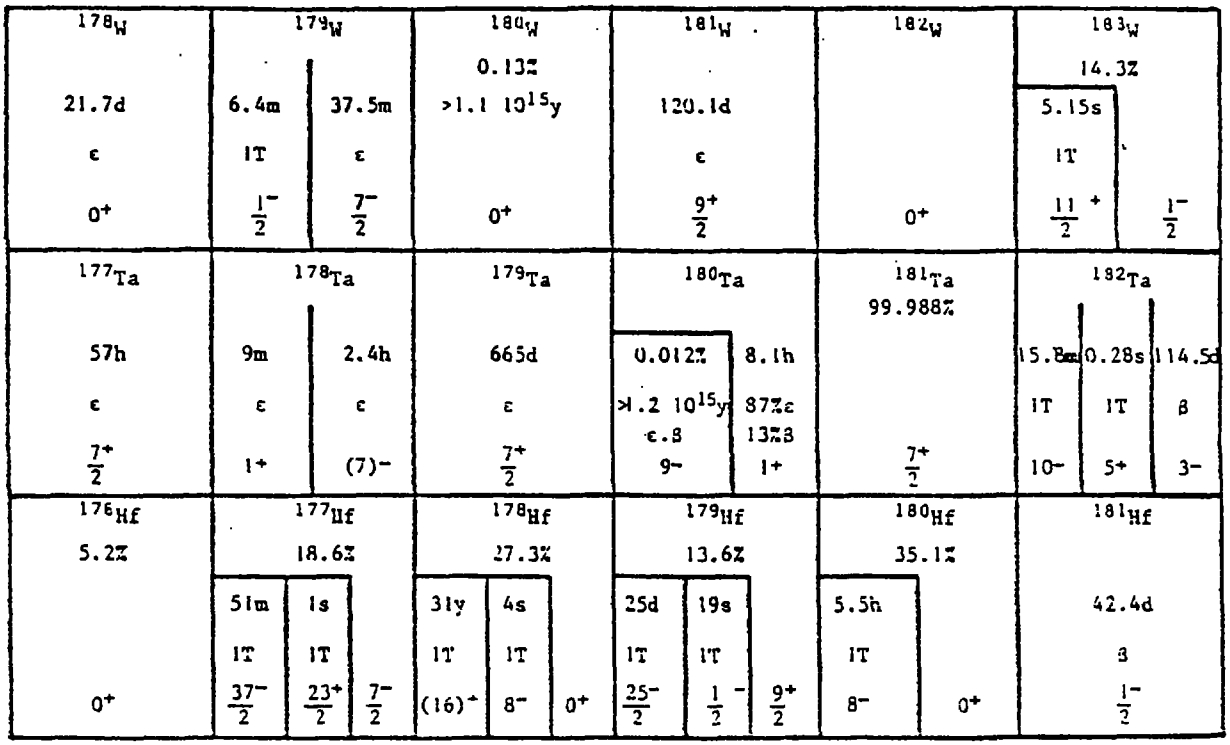

Fig 1. The periodic table in the region of tungsten, tantalum and hafnium

investigating and a request was made for the activation properties of these elements to be investigated. A look at the periodic table in the region of these elements (see Figure 1) identified a possible problem due to the presence of the 31-year isomeric state in Hf-178. The production of significant numbers of nuclei in this state could lead to the first wall being active for many years, making reuse difficult to achieve and therefore raising a potential waste disposal problem. At the time, the production of UKACT1 was still in its infancy and cross-sections for reactions in this region were not fully to hand. Some guesses were made and calculations performed, the conclusion being that production of the isomer in Hf-178 (Hf-178m2) could be sufficient to give rise to an activation problem, but in order to be more specific, better data would be required. An examination of Fig 1 shows that the situation is further complicated by the presence of a 25-day isomeric state in Hf-179 (Hf-179m2). This state could live long enough for an (n,2n) reaction to take place leading to the 31-year state in Hf-178. In fact, the nuclear physics of the situation is such that the cross-section for the reaction Hf-179m2(n,2n)Hf-178m2 in the region of 14 MeV neutron energy could be large as the incident angular momentum required for the reaction to take place is only modest, Hf-179m2 having $J^{\pi} = 25/2^-$ and Hf-178m2 $J^{\pi} = 16+$. Consequently, any Hf-179m2 formed could have a significant probability of being transformed into Hf-178m2.

A search of the literature yielded no relevant measurements or theoretical calculations of cross-sections leading to the isomeric states in Hf-178 and Hf-179, and the question then arose as to how reliable data could be derived. The first solution to this problem came when the Lawrence Livermore National Laboratory (LLNL) offered to irradiate a package of materials, including tantalum, tungsten and hafnium, on the intense 14-MeV neutron generator RTNS-II to enable activation cross-sections to be measured. Later, the co-operation of Los Alamos National Laboratory (LANL) was instrumental in enabling some theoretical calculations of relevant cross-sections to be carried out.

Table 1

Details of materials irradiated on RTNS-II

The foils, 15 mm diameter, were packaged in the order shown, sample number 1 being nearest to the neutron target.

SAMPLE NO	MATERIAL	THICKNESS (mm)	MASS (g)
1	Ni	0.01	0.016
2	Cu	1.10	0.154
3	Au	0.005	0.017
4	Ni	0.01	0.015
5	Co	0.01	0.018
6	Mn/Ni*	0.05	0.060
7	Ni	0.01	0.016
8	Hf	1.00	2.386
9	Ni	0.01	0.017
10	W	1.00	3.170
11	Ni	0.01	0.016
12	Ta	1.00	2.884
13	Ni	0.01	0.015
14	Ti	0.10	0.075
15	SS**	0.14	0.188
16	Ni	0.01	0.016
17	Mn/Ni*	0.05	0.063
18	Co	0.01	0.018
19	Ni	0.01	0.016
20	Au	0.005	0.017
21	Cu	0.10	0.154
22	Ni	0.01	0.014

* 88% Mn, 12% Ni
** stainless steel

Irradiation on RTNS-II

A package of 15 mm diameter foils of various materials was irradiated on the intense (d,t) neutron generator, RTNS-II, at LLNL in March 1987, on the 0° axis close to the tritiated target. The accelerator produced 360 keV deuterons giving a maximum neutron energy of 15.6 MeV. The package was subjected to a fluence of about 10^{18} neutrons/cm^2 over a period of 10 days.

The materials consisted of one foil each of hafnium, tantalum, tungsten, titanium and a low-activation stainless steel, together with neutron fluence monitor foils of nickel, cobalt, gold, manganese and copper, the entire package being wrapped in thin aluminium foil. The package was approximately 6 mm thick, so that there was a significant flux gradient from front to back. Details of the materials are given in Table 1. The monitor foils were chosen on the basis of the accuracy of specific 14-MeV reaction cross-sections leading to states with appropriate half-lives. The package was to be returned to the UK after irradiation and as it was not clear how long shipment would take, short half-lives were ruled out as inappropriate. The

monitor reactions to be used to determine the neutron fluence are shown in Table 2. Care was taken to prevent any possibility of cross-contamination of the foils for which cross-sections were to be measured by ensuring that the foils of these materials were sandwiched between monitor foils. Nickel was used as the primary monitor material, foils being positioned throughout the package to enable the fluence at any point in the package to be determined. The other monitor materials were used to verify the nickel results.

As an independent check on the fluence, the LLNL staff who carried out the irradiation placed foils of niobium on the front and back of the package. The activity in these was measured at LLNL before the package was returned to the UK, the estimated neutron fluences being in agreement with our measurements.

The time distribution of the neutron fluence during the 14-day irradiation period was measured at LLNL by recording the counts in half-hour intervals in a fission chamber placed some distance from the package. This enabled corrections to be made for decay of activation during the irradiation.

Measurement of the activity of the foils

The activity of each foil was measured at intervals over a period of time using a Ge(Li) detector connected to a 4096-channel pulse height analyser system attached to a VAX computer. Dead-time corrections were carried out automatically during the counting of the foils. The areas of peaks corresponding to particular gamma-rays were determined, the half-life being used as a check of the origin of each gamma-ray.

The energy calibration of the analysis system was performed using standard sources, as was the detector efficiency calibration.

The activity of the hafnium foil was dominated for the first year after irradiation by the decay of the 25-day state in Hf-179 and of Hf-175. Only after this activity had largely decayed away did the gamma-rays from the 31-year state in Hf-178 begin to show clearly. Figure 2 shows a spectrum taken several weeks after the irradiation and Figure 3 is one taken two years later. Note particularly the spectra in the region 500-650 keV. In the earlier one, there is virtually no sign of peaks, while in the later one several peaks from the decay of Hf-178m2 are prominent.

To date, only a fraction of the spectra collected have been analysed. The emphasis has been on determining hafnium cross-sections and this paper reports preliminary results. Some corrections have still to be applied to the data (notably for absorption of the gamma-rays in the foils), but these are not expected to be large. It is to be noted that, as natural elements were used in the experiment, it is not strictly possible to obtain unique cross-sections in many cases as there are a number of reactions which could lead to a given activity. However, using a priori knowledge of 14-MeV cross-sections and being guided by theory (see later), it is generally found that one cross-section dominates the route to a particular activity and it has been assumed that all the activity arises from this reaction. However, as will be shown later, this assumption is unlikely to be valid for production of the Hf-179m2 state, where the cross-sections for the reactions Hf-180(n,2n)Hf-179m2 and Hf-179(n,n')Hf-179m2 are believed to be comparable. The consequences of this are dealt with in the section on results.

So far, only the nickel foils have been used to determine the neutron fluence in the region of the hafnium foil and elsewhere in the package. The reactions used are given in Table 2, along with those in other monitor foils

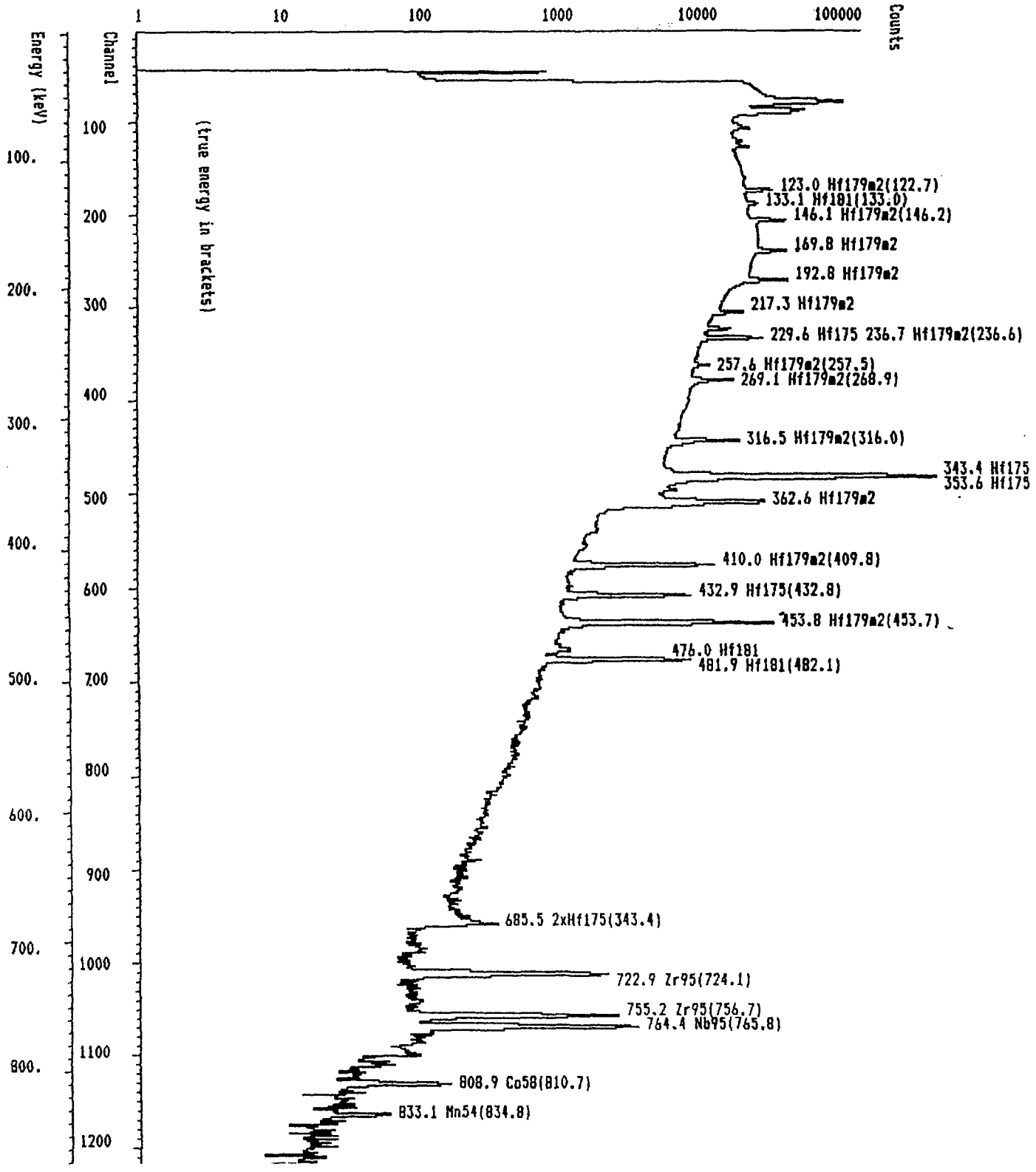

Figure 2 Hf foil spectrum taken 6 weeks after irradiation

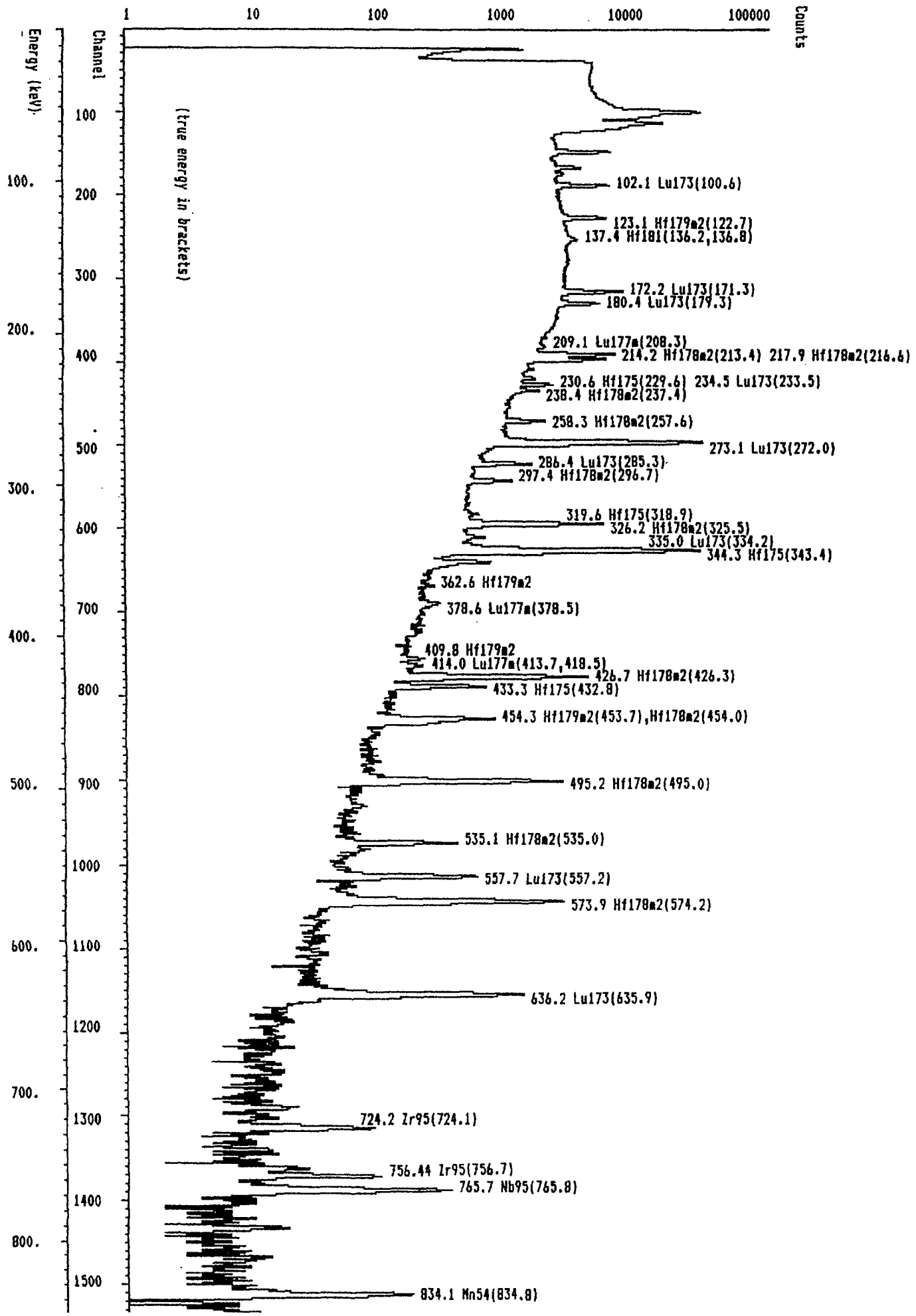

Figure 3 Hf foil spectrum taken 2 years after irradiation

Table 2 Monitor Foils - Reactions Used

Foil Material	Reactions Used
Nickel	^{58}Ni (n,p) ^{58}Co ^{58}Ni (n,pn) ^{57}Co ^{60}Ni (n,p) ^{60}Co
Copper	^{63}Cu (n,α) ^{60}Co
Gold	^{197}Au (n,2n) ^{196}Au
Cobalt	^{59}Co (n,p) ^{59}Fe ^{59}Co (n,2n) ^{58}Co
Manganese	^{55}Mn (n,2n) ^{54}Mn

Table 3 Nickel Monitor Foil Reaction Cross-sections

Reaction	Cross-section* (b)	Gamma Rays Used
^{58}Ni(n,p) ^{58}Co	0.298	511 keV,811 keV 864 keV,1675 keV
^{58}Ni (n,pn) ^{57}Co	0.651	122 keV,136 keV
^{60}Ni (n,p) ^{60}Co	0.129	1173 keV,1333 keV

(* Assuming neutron energy of 15.4 MeV)

which will be used in due course to confirm the fluence values. In order to derive an effective neutron energy for the experiment, fluence values were estimated as a function of neutron energy from the three nickel reactions shown in Table 2, using the known energy dependence of the cross-sections. The variance of the fluence values was then calculated at each energy. The minimum variance was found to lie at a neutron energy of 15.4 MeV and the corresponding cross-sections are given in Table 3. This energy is higher than expected, bearing in mind that the maximum neutron energy is 15.6 MeV. However, the minimum in the variance is fairly shallow and the data are not inconsistent with a lower effective neutron energy when the uncertainties on the cross-sections are taken into account. This point will be examined more closely before the fluences are finalised.

Results

Results of measured hafnium cross-sections are given in Table 4. In deriving these, it has been assumed that the following cross-sections are zero at a mean neutron energy of 15.4 MeV:

Hf-178(n,n′)Hf-178(31-year state)
Hf-178(n,γ)Hf-179(25-day state)

As noted above, there is some evidence that the Hf-179m2 state can be formed

Table 4 Summary of Hafnium Cross-Section Measurements

Reaction	Gamma Rays Used (keV)	Mean Cross-Section	Theoretical Cross-sections Chadwick & Young (1989)
^{176}Hf (n,2n) ^{175}Hf	230 319 343 433	1.77 ± 0.06 b	-
^{179}Hf(n,2n)^{178m2}Hf	258 326 426 495 535 574	5.91 ± 0.64 mb	2.9 mb
^{180}Hf(n,2n)^{179m2}Hf	237 269 316 363 410 454	16.7 ± 1.9 mb*	7.4 mb
^{179}Hf(n,n')^{179m2}Hf	237 269 316 363 410 454	12.8 ± 1.5 mb*	5.7 mb
^{174}Hf (n,2n) ^{173}Hf (^{173}Hf -> ^{173}Lu) β+/EC	272	2.75 ± 0.18 b	-

* The values for these cross-sections were derived from the measured data in the way described in the text.

by two routes with approximately equal cross-sections. Chadwick and Young (1989) carried out a series of calculations of hafnium cross-sections at 14 MeV using the code GNASH at LANL. This code is based on the Hauser-Feshbach statistical model with a pre-equilibrium component.

They estimated the 14-MeV cross-sections for the Hf-180(n,2n)Hf-179m2 reaction to be 7.4 mb and the Hf-179(n,n')Hf-179m2 reaction to be 5.7 mb. Assuming these values are correct in relative terms, the cross-sections for the two reactions have been derived from the measured data, taking the natural abundances of Hf-179 and Hf-180 into account. The results are included in Table 4. Besides deriving cross-sections for the formation of the long-lived isomeric states in hafnium, it has been possible to extract values for (n,2n) reactions on Hf-176 and Hf-174. The former result appears to be about the magnitude to be expected, but the latter is clearly too large. However, it should be noted that Hf-174 has an abundance of only about 0.16%, making accurate measurement difficult. The data will be scrutinised further before final results are issued.

The results of the calculations are also given in Table 4. A comparison of the measured cross-sections with the corresponding theoretical ones shows a very good level of agreement. To calculate these small isomeric cross-sections to within a factor of about two compared with the measurements has to be viewed as remarkably satisfactory and provides some degree of confidence that such calculations can be used to obtain cross-section data of this type for improving fusion cross-section files.

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