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The ^{180}Lu Beta Decay to the ^{180}Hf 8^- Isomeric State and the r-Process Formation of ^{180}Ta^m

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ABSTRACT

The fractional decay of ^{180}Lu ($T_{1/2}$ = 5.7 min) to the 8^- isomeric state $^{180}Hf^m$ ($T_{1/2}$ = 5.5 h) is investigated, applying multi nucleon transfer reactions between a 11.7 MeV/u ^{136}Xe beam and a tantalum/tungsten target, on-line mass separation and γ-ray spectroscopy. The branching ratio of ^{180}Lu ß-decays to the 8^- $^{180}Hf^m$ state is estimated to be 5.7 % which leads to the interpretation of a dominant formation of $^{180}Ta^m$ by r-process nucleonsynthesis.

Der ^{180}Lu Beta Zerfall zu dem Isomer in ^{180}Hf und die r-Prozeß-Bildung von $^{180}Ta^m$

Zusammenfassung

Der partielle Zerfall von ^{180}Lu ($T_{1/2}$ = 5,7 min) zu dem 8^- isomeren Zustand $^{180}Hf^m$ ($T_{1/2}$ = 5,5 h) wurde unter Anwendung von Multinukleon-Transferraktionen zwischen einem 11,7 MeV/u ^{136}Xe Strahl mit einem Tantal/Wolfram Target, on-line Massenseparation und γ-Strahl-Spektroskopie untersucht. Das Verzweigungsverhältnis des ^{180}Lu β-Zerfalls zum 8^- $^{180}Hf^m$ Zustand wurde zu 5,7 % abgeschätzt. Dies führt zu der Interpretation, daß $^{180}Ta^m$ vorwiegend über die r-Prozeß-Nukleosynthese gebildet wurde.

INTRODUCTION

According to the recent investigations of Beer and Ward (1) and Beer and Macklin (2) the extremely small solar abundance of $^{180}Ta^{m}$ can be explained by small neutron capture and/or ß-decay branchings in the common neutron capture nucleosynthesis of the s- and/or r-processes. The essential point of the arguments refers to an isomeric state in $^{180}Hf^{m}$ which has an allowed-hindered beta decay to $^{180}Ta^{m}$. In this way a branching is established large enough to account for the small solar abundance of $^{180}Ta^{m}$ (2.46×10^{-6}, Si ≡ 10^{6}). In Fig. 1 the s- and r-process synthesis paths to generate $^{180}Ta^{m}$ are illustrated. They differ in the way the isomeric state in $^{180}Hf^{m}$ is populated. In s-process nucleosynthesis, this population is due to neutron capture in ^{179}Hf and was determined [2] to be (1.24 ± 0.06) %. In post r-process nucleosynthesis, ^{180}Lu can have a fractional ß-decay branch f_m^{180} to the $^{180}Hf^{m}$ isomeric state. Beer and Macklin [2] have estimated that in order to reproduce the solar $^{180}Ta^{m}$ abundance totally by the r-process, f_m^{180} should be as large as 2.5 % provided the fractional ß-decay branch h from $^{180}Hf^{m}$ to $^{180}Ta^{m}$ amounts to 0.34 % (see Fig. 1). It is the aim of this paper to present a measurement of f_m^{180} and to discuss the conclusions with respect to nucleosynthesis formation of $^{180}Ta^{m}$.

EXPERIMENTAL METHOD

The difficulty in measuring f_m^{180} is that nuclear reactions suitable for producing ^{180}Lu will in general yield $^{180}Hf^{m}$ simultaneously, therefore, one has to detect weak decay branches

of the short-lived nucleus ^{180}Lu in the presence of large quantities of its long-lived daughter activities of interest, $^{180}Hf^{m}$. For example, the relatively extensive decay scheme of ^{180}Lu, obtained by Swindle et al. (3) via the $^{180}Hf(n,p)^{180}Lu$ reaction is incomplete with regard to transitions to the 8^- $^{180}Hf^{m}$ isomeric state because this state is also populated by the $^{179}Hf(n,\gamma)^{180}Hf^{m}$ reaction due to the presence of ^{179}Hf in the natural hafnium target.

In the present investigation, a 11.7 MeV/u ^{136}Xe beam from the UNILAC was used for irradiation of a tantalum/tungsten target (4) positioned inside the ion source of the GSI on-line mass separator (5) One of the mass-separated beams was collected on an aluminium foil in front of a Ge(Li) detector for on-line and subsequent off-line γ-counting.

It is important to note that the overall efficiency of the on-line mass separator depends upon both the chemical properties and the half-life of the isotope of interest. Both lutetium and hafnium isotopes are released from the ion source, however with different overall-efficiencies. (The release of hafnium appears to contrast with earlier ^{136}Xe on tantalum-tungsten experiments (4,6), in which ^{181}Hf and ^{182}Hf have not been observed in the mass-181 and mass-182 beams, respectively. The different behaviour is most probably due to target ion-source modificiations). The idea of the present determination of f_m^{180} is to deduce this quantity from a γ-ray measurement on a mass-180 sample and to correct for the part of the $^{180}Hf^{m}$

activity, which is not due to ^{180}Lu decay in this sample, but collected as $^{180}Hf^{m}$. This isobaric contamination is estimated by a separate measurement of $^{177}Hf^{m}$ which is shielded against any ß-decay. In doing this, we have to rely upon the following approximate assumptions:

- Production cross sections are taken for $^{177}Hf^{m}$ $(37/2^{-})$ and $^{180}Hf^{m}(8^{-})$ to be 5.4 ± 0.5 mb and 6.1 ± 0.3 mb as measured [7] for 8.5 MeV/u ^{136}Xe on tungsten reactions, and for ^{180}Lu to be 0.27 mb as estimated (4,7) for 9.1 MeV/u ^{136}Xe on tantalum-tungsten reactions. We assume thereby, that the cross section ratios determined in these experiments remain unchanged under the conditions of the present experiment, that is for 11.7 MeV/u ^{136}Xe energy and for the actual target containing a sandwich array if thin tantalum and tungsten foils of 34 mg/cm² and 14 mg/cm² total thickness, respectively.
- We assume identical overall-efficiencies for 51-min $^{177}Hf^{m}$ and 5.5-h $^{180}Hf^{m}$, whereas the "fractionation" effect due to difference in overall-efficiency between ^{180}Lu and $^{180}Hf^{m}$ will be determined experimentally.

The determination of the fractional ^{180}Lu decay required, therefore, two measurements. First the $^{177}Hf^{m}(T_{1/2} = 51$ min) nuclei are collected and their activity is counted by the characteristic γ-line at 277 keV. Then the reaction products of mass 180, ^{180}Lu and $^{180}Hf^{m}$, are measured. The ^{180}Lu nuclei are counted on-line with a Ge(Li) detector via the strongest γ-ray line at 408 keV. The $^{180}Hf^{m}$ nuclei which are produced directly in the transfer reaction as well as by ^{180}Lu decay

during the collection period are measured through the 332 keV γ-line. For the number of counts C_{Hf}^{177}, C_{Lu}^{180} and C_{Hf}^{180} from the activated $^{177}Hf^{m}$, ^{180}Lu and $^{180}Hf^{m}$ nuclei the following relations can be specified under the above-mentioned approximations:

$$C_{Hf}^{177} = N_{Hf}^{177}\, g_{Hf}^{177} \varepsilon_{Hf}^{177} \frac{1}{\lambda_{Hf}^{177}} (1-e^{-\lambda_{Hf}^{177} T'_b})(1-e^{-\lambda_{Hf}^{177} T'_c})\, e^{-\lambda_{Hf}^{177} T'_w} \quad (1)$$

$$C_{Lu}^{180} = N_{Lu}^{180}\, g_{Lu}^{180} \cdot \varepsilon_{Lu}^{180} \left[T_b - \frac{1}{\lambda_{Lu}^{180}} (1-e^{-\lambda_{Lu}^{180} T_b})\right] \quad (2)$$

$$C_{Hf}^{180} = g_{Hf}^{180}\, \varepsilon_{Hf}^{180} \frac{1}{\lambda_{Hf}^{180}} (1-e^{-\lambda_{Hf}^{180} T_b})(1-e^{-\lambda_{Hf}^{180} T_c})\, e^{-\lambda_{Hf}^{180} T_w} \left[N_{Hf}^{180} + N_{Lu}^{180}\, f_m^{180}\, S_{Lu}^{Hf}\right] \quad (3)$$

$$S_{Lu}^{Hf} = \frac{\lambda_{Hf}^{180}}{\lambda_{Hf}^{180} - \lambda_{Lu}^{180}} \frac{e^{-\lambda_{Lu}^{180} T_w} - e^{-\lambda_{Lu}^{180}(T_b+T_w)}}{e^{-\lambda_{Hf}^{180} T_w} - e^{-\lambda_{Hf}^{180}(T_b+T_w)}} - \frac{\lambda_{Lu}^{180}}{\lambda_{Hf}^{180} - \lambda_{Lu}^{180}} \quad (4)$$

where C and N designate numbers and rates, respectively, of radioactive nuclei collected on the aluminium foil for a given current of ^{136}Xe projectiles, G is the number of γ-disintegrations of the analyzed line per decay, ε the Ge(Li) efficiency of the γ-line. T_b is the time of collecting the mass-separated beam on the aluminium collector, with the ^{136}Xe beam irradiating the tantalum-tungsten target. T_w is the waiting time between end of collection and start of counting for the $^{177}Hf^{m}$ and $^{180}Hf^{m}$ samples. λ stands for the decay rate and T_c is the Ge(Li) counting time. The wanted unknown fraction of ^{180}Lu nuclei which decay to $^{180}Hf^{m}$

is designated by f_m^{180}.

N_{Hf}^{180} can be related to N_{Hf}^{177} by means of the production cross sections σ^m_{Hf180} and σ^m_{Hf177} under the assumption that the target thickness does not vary with time (4)

$$N_{Hf}^{177} = \frac{\sigma^m_{Hf177}}{\sigma^m_{Hf\ 180}} N_{Hf}^{180} \qquad (5)$$

and N_{Hf}^{180} is related to N_{Lu}^{180} by:

$$N_{Hf}^{180} = F \frac{\sigma^m_{Hf180}}{\sigma_{Lu180}} N_{Lu}^{180}, \qquad (6)$$

where σ_{Lu180} is the ^{180}Lu production cross section and F a factor which takes into account that $^{180}Hf^m$ is suppressed compared to ^{180}Lu in the mass-180 beam. From eqs. (1) to (5) we find the following relation for f_m^{180} :

$$f_m^{180} = \left[\frac{C_{Hf}^{180}\ \lambda_{Hf}^{180}}{g_{Hf}^{180}\ \varepsilon_{Hf}^{180}\ (1-e^{-\lambda_{Hf}^{180} T_b})(1-e^{-\lambda_{Hf}^{180} T_c}) e^{-\lambda_{Hf}^{180} T_w}} - \frac{(\sigma^m_{Hf180}/\sigma^m_{Hf177}) C_{Hf}^{177}\ \lambda_{Hf}^{177}}{g_{Hf}^{177}\ \varepsilon_{Hf}^{177}\ (1-e^{-\lambda_{Hf}^{177} T'_b})(1-e^{-\lambda_{Hf}^{177} T'_c}) e^{-\lambda_{Hf}^{177} T'_w}} \right] \cdot \frac{\varepsilon_{Lu}^{180}\ g_{Lu}^{180}\ [T_B - \frac{1}{\lambda_{Lu}^{180}} (1-e^{-\lambda_{Lu}^{180} T_b})]}{C_{Lu}^{180}} \frac{1}{S_{Lu}^{Hf}} \qquad (7)$$

Eq. (7) is correct for the ideal condition of a constant ^{136}Xe beam intensity during the collection time T_b and before, so that the nuclei of interest are in saturation in the target catcher, before the collection period starts. If the time

dependence of the ^{136}Xe beam $\phi(t)$ and the release properties of the ion source were known, eq. (7) could be corrected for beam time fluctuations.

For the present evaluation, we did not apply such a correction, but derived simple time-averaged values from the measured (5) ^{136}Xe beam current (see Table 1).

The factor F defined in eq. (6) can be determined from measured quantities using eqs. (1), (2) and (5)

$$F = \frac{\sigma_{Lu180}}{\sigma^{m}_{Hf177}} \cdot \frac{C^{177}_{Hf}}{C^{180}_{Lu}} \cdot \frac{g^{180}_{Lu}}{g^{177}_{Hf}} \cdot \frac{\varepsilon^{180}_{Lu}}{\varepsilon^{177}_{Hf}} \quad \frac{[T_B - \frac{1}{\lambda^{180}_{Lu}}(1-e^{-\lambda^{180}_{Lu} T_b}]}{\frac{1}{\lambda^{177}_{Hf}}(1-e^{-\lambda^{177}_{Hf} T_b})(1-e^{-\lambda^{177}_{Hf} T_c})e^{-^{177}_{Hf} T_w}} \qquad (3)$$

RESULTS AND DISCUSSION

The measured γ-ray spectra are shown in Fig. 2. In Table 1 the relevant quantities from the measurement and from literature are summarized, yielding f^{180}_{m}=6.0 % and F = 0.23%. The latter value corresponds to overall efficiencies of approximately 5 % for ^{180}Lu and 10^{-4} for $^{180}Hf^{m}$.

In this calculation it is assumed that $g^{180}_{Lu} = 0.5$ [8] represents the fraction of <u>total</u> ^{180}Lu decay via the 408 keV γ line, but as in the ^{180}Lu decay scheme of ref. [8] no branching to the $^{180}Hf^{m}$ isomeric state is indicated we have

to assume that this fraction f_m^{180} was neglected. If it is taken into account we have to replace $g_{Lu}^{180} \longrightarrow g_{Lu}^{180}\,(1-f_m^{180})$ in eqs. (2), (6) and (8). This leads to $f_m^{180} = 5.7$ % and $F = 2.2 \times 10^{-3}$. The present result is of importance as it allows now the decision of whether $^{180}Ta^m$ is of s- or r-process origin.

In ref. (2) it was demonstrated that the s-process might give only a negligible amount of $^{180}Ta^m$ compared to r-process formation. In this discussion the fractional ß-decay from $^{180}Hf^m$ to $^{180}Ta^m$ was taken to be 3.4×10^{-3} assuming an allowed-hindered ß-decay. Under this assumption the presently determined fractional ^{180}Lu decay to $^{180}Hf^m$ allows the calculation of the r-process abundance $N_r(^{180}Ta^m)$:

$$N_r(^{180}Ta^m) = N_r(^{180}Hf)\,\frac{h}{1-h}\cdot\frac{f_m^{180}}{1-f_m^{180}} \qquad (9)$$

where $$N_r(^{180}Hf) = N_\odot(^{180}Hf) - \frac{\sigma N_s(^{180}Hf)}{\sigma(^{180}Hf)} \qquad (10)$$

$N_r(^{180}Hf)$ represents the r-process abundance of ^{180}Hf which is calculated [2] by subtracting the s-process abundance

$$N_s = \sigma N_s(^{180}Hf) \;/\; \sigma(^{180}Hf) \qquad (11)$$

from the solar abundance $N_\odot(^{180}Hf)$. σ is the Maxwellian averaged capture cross section of ^{180}Hf reported in ref. (2).

From the present f_m^{180} result and the $N_r(^{180}Hf)$ value (2) of 0.029 (Si≡10^6), we have estimated $N_r(^{180}Ta^m) \approx 5.95 \times 10^{-6}$

(Si≡10^6). Therefore the solar abundance of ^{180}Ta (2.46×10^{-6}, Si≡10^6) can obviously be explained by r-process nucleonsynthesis.

Finally, we would like to stress that the present rough estimate of the ^{180}Lu ——> ^{180}Hf branching is based on assumptions concerning production cross section ratios and separation efficiencies. Furthermore, time fluctuations of the ^{136}Xe beam intensity during the collection period have not been taken into account as well as possible differences in the intensities of mass separated ^{177}Hf^m and ^{180}Hf^m beams due to accumulation of these isotopes in the ion source during pre-collection irradiations.

It is challenging to improve our understanding of the ^{180}Ta^m formation by remeasuring the branching f_m^{180} under improved conditions, and by additionally determining the ^{180}Hf^m ——> ^{180}Ta^m decay parameters, in order to verify h = 3.4×10^{-3}.

If it should turn out that h is really as high as 3.4×10^{-3} and f_m^{180} = 0.057 then ^{180}Ta^m is overproducted by a factor of 2.4 This could mean that additionally to the formation of ^{180}Ta^m some process of destruction (i.e. thermalization of the isomeric and ground state) has to be considered.

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The critical comments of Dr. O. Klepper are gratefully acknowledged.

TABLE I

	$^{177}Hf^{m}$	$^{180}Hf^{m}$	^{180}Lu
$T_{1/2}$ [+)]	51 min	5.5 h	5.7 min
$\lambda(s^{-1})$	$2.2652 \cdot 10^{-4}$	$3.5007 \cdot 10^{-5}$	$2.0267 \cdot 10^{-3}$
$E\gamma$ (keV) [+)]	277	332	408
g [+)]	0.75	0.944	0.50
ε	$8.6 \cdot 10^{-2}$	$7.4 \cdot 10^{-2}$	$1.1 \cdot 10^{-2}$
Average beam current (particle·nA)	10.7	14.0	14.0
T_b(s)	6192	11014	11014
T_w(s)	2951	13467	-
T_c(s)	7200	3600	-
σ^{m}_{Hf177}(mb)	5.4±0.05 [++)]		
σ^{m}_{Hf180}(mb)		6.1±0.3 [++)]	
σ^{m}_{Lu180}(mb)			0.27 [++)]

+) From Ref. (8)

++) The production cross sections were measured at ^{136}Xe energies of 8.5 MeV/U and 9.1 MeV/U (see text)

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FIGURE CAPTIONS

Fig. 1 The nuclear level structure along the A=180 isobaric chain of significance to the stellar nucleosynthesis of $^{180}Ta^{m}$ ($>10^{13}$ yr). The most important quantities are the fractional branchings into $^{180}Hf^{m}$ (5.5 h) through both: 30 keV neutron capture by ^{179}Hf (denoted by B) for the s-process and the β-decay of ^{180}Lu (denoted by f_m^{180}) for the r-process. These are then followed by the possible β-decay branching, h from $^{180}Hf^{m}$ into the isomer $^{180}Ta^{m}$. We have also shown the spins, parities, and energies (in MeV) of many levels as well as their relevant decay properties and half lives (when known).

Fig. 2 Parts of the γ-ray spectra measured at mass 180 (in-beam and off-line) and mass 177 (off-line). Two Ge(Li) detectors with efficiencies of 12 and 40 % were used for the γ-ray in-beam and off-line measurements, respectively.

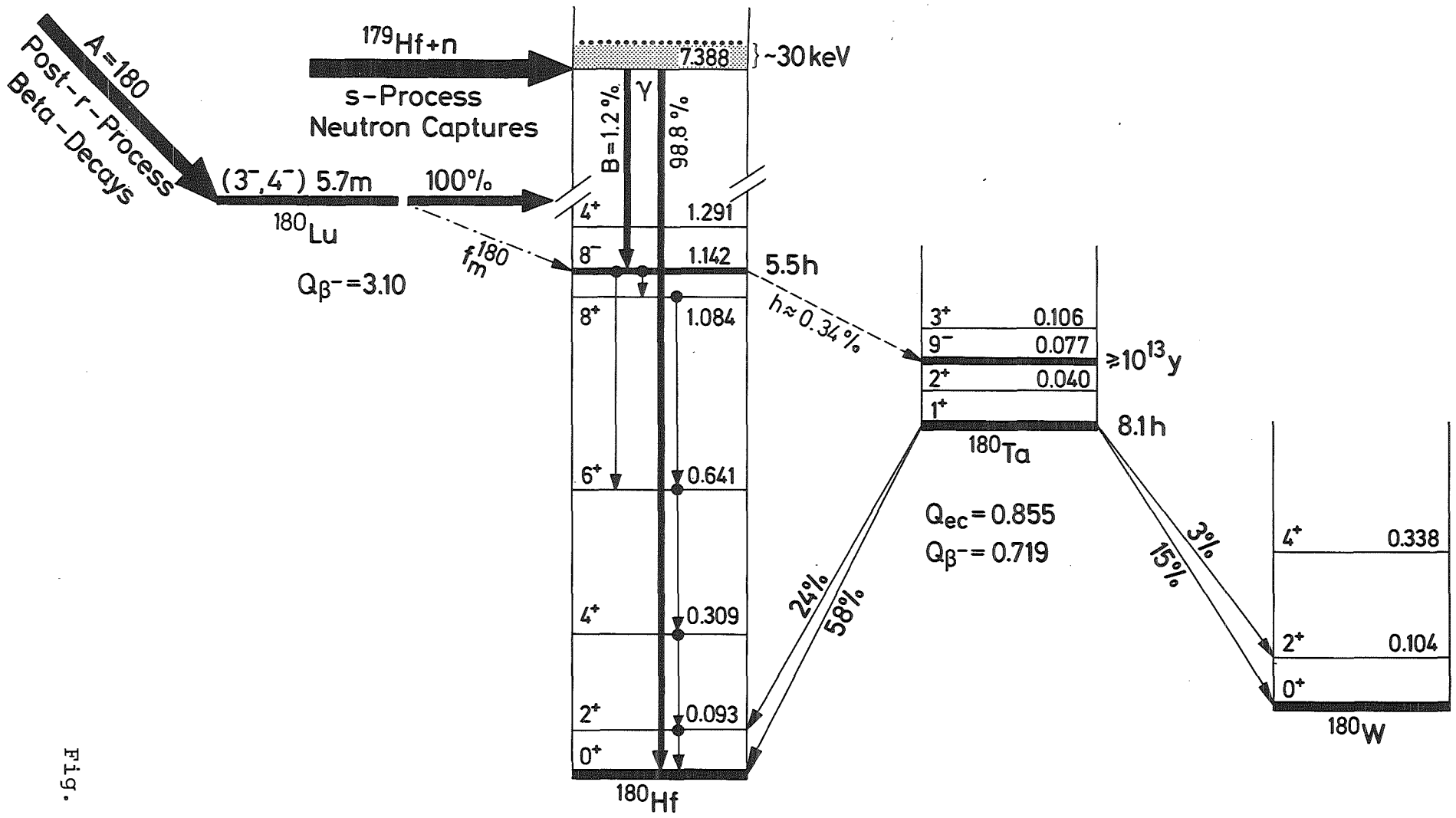
A=180
Post-r-Process
Beta-Decays
179Hf+n
s-Process
Neutron Captures
7.388
~30keV
B=1.2%
γ
98.8%
(3⁻,4⁻) 5.7m
100%
180Lu
Qβ⁻=3.10
f_m^180
4⁺ 1.291
8⁻ 1.142
5.5h
h≈0.34%
8⁺ 1.084
6⁺ 0.641
4⁺ 0.309
2⁺ 0.093
0⁺
180Hf
3⁺ 0.106
9⁻ 0.077
≥10¹³y
2⁺ 0.040
1⁺
8.1h
180Ta
Qec=0.855
Qβ⁻=0.719
24%
58%
3%
15%
4⁺ 0.338
2⁺ 0.104
0⁺
180W

Fig. 1

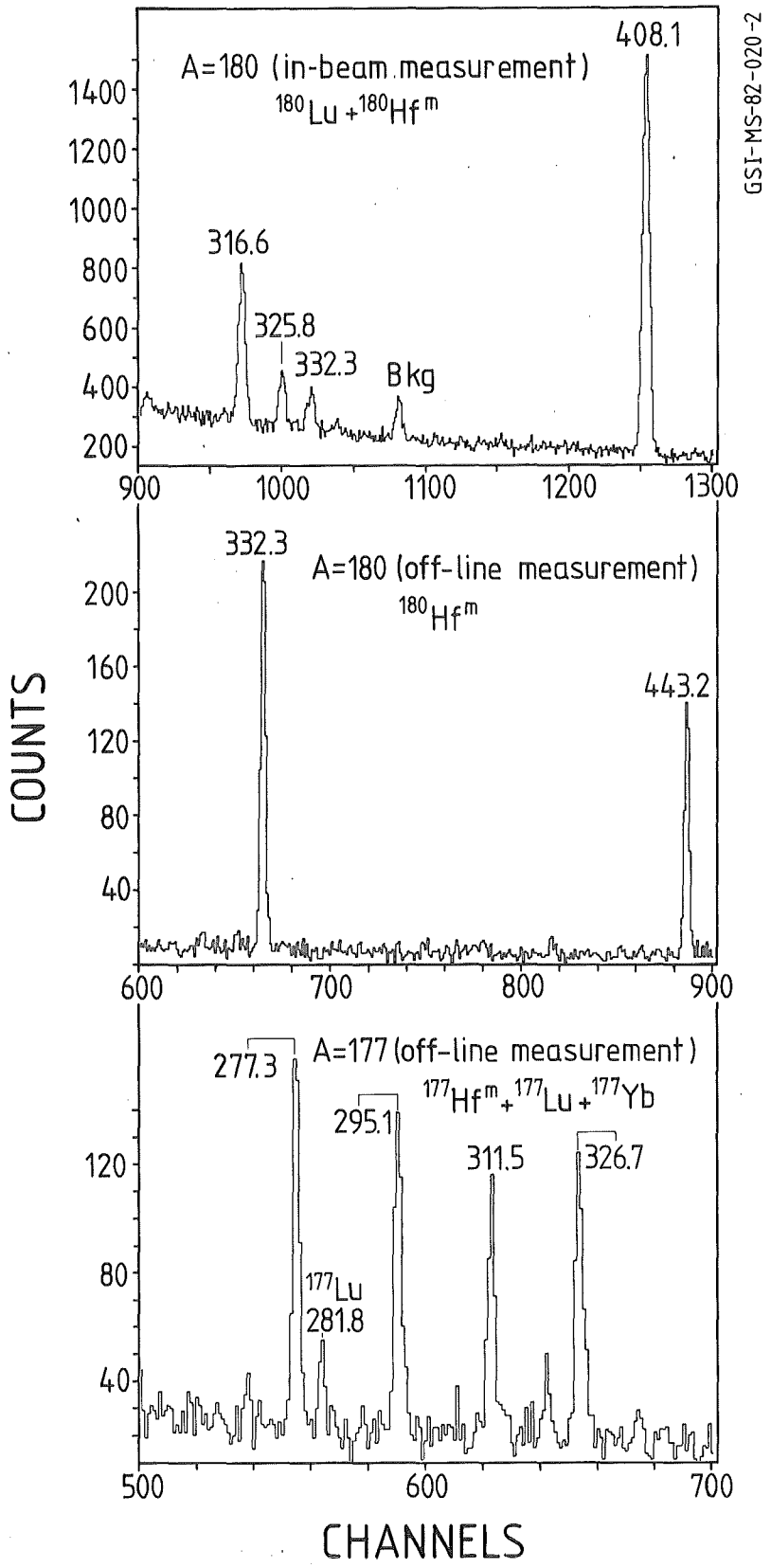
A=180 (in-beam measurement)
180Lu + 180Hf m
408.1
316.6
325.8
332.3
Bkg
A=180 (off-line measurement)
180Hf m
332.3
443.2
A=177 (off-line measurement)
177Hf m + 177Lu + 177Yb
277.3
295.1
311.5
326.7
177Lu
281.8
COUNTS
CHANNELS
GSI-MS-82-020-2

Fig. 2